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Universal Electromagnetic Response Relations

applied to the free homogeneous electron gas

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Thesis

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Publications

While I have compiled this thesis by myself, parts of the work presented here have been done in collaboration with the authors mentioned below and have already been published or submitted for publication or are currently in preparation for imminent submission. States are indicated accordingly.

Journal articles

- *Wavevector-dependent refractive indices from ab initio calculated dielectric tensor in optical limit* [(*)]
  with R. Starke, G.A. H. Schober and J. Kortus
  In preparation.

- *Wavevector-dependent optical properties from wavevector-independent proper conductivity tensor* [(**)]
  with R. Starke, G.A.H. Schober, N. Bulut and J. Kortus
  Eur. Phys. J. B **93**, 54 (2020) [1]

- *Fundamental response tensor of the free electron gas* [(**)]
  with R. Starke, G.A.H. Schober and N. Bulut
  Preprint. [2]

- *Ab initio electronic structure and optical conductivity of bismuth tellurohalides*
  with S. Schwalbe, R. Starke, G.A.H. Schober and J. Kortus
  Phys. Rev. B **94**, 205130 (2016) [3]

Proceedings

- *Wavevector-dependent optical properties from wavevector-independent ab initio conductivity tensor*
  DPG Spring Meeting 2018 – Proceedings (KFM 27.13)

- *Wavevector-dependent dielectric function and ab initio materials physics*
  DPG Spring Meeting 2017 – Proceedings (DF 9.1)

- *Theoretical investigation of BiTeX (X=Cl,Br,I): Crystal structure and optical conductivity*
  DPG Spring Meeting 2016 – Proceedings (MM 15.15)

Software

- *Elk Optics Analyzer (ElkOA)* © 2017-2020 R. Wirnata [4]
  available as FLOSS[2] under GPLv3 on:
  github : `https://github.com/PandaScience/ElkOpticsAnalyzer`
  PyPI : `https://pypi.org/project/elkoa/`

[1] (*) first author, (**) corresponding author
[2] Free/Libre and Open Source Software

Publications unrelated to this thesis

- *Heat capacity of η-AlFe (Fe_2Al_5)*
  with T. Zienert, L. Amirkhanyan, J. Seidel, T. Weissbach, T. Gruber, O. Fabrichnaya, and J. Kortus
  Intermetallics **77**, 14 (2016) [5]

- *A density functional theory study on elastic and thermodynamic characteristics of Al_5Fe_2*
  DPG Spring Meeting 2015 – Proceedings (MM 17.9)

Further, this thesis can be regarded as a logical extension of a series of publications centered around the *Functional Approach to electrodynamics in media* [6–15], which was recently developed by R. Starke and G.A.H. Schober and to which the author of this thesis also contributed.

Abstract

Universal Electromagnetic Response Relations applied to the free homogeneous electron gas.—This thesis is concerned with the application of the recently developed "Functional Approach" to electrodynamics of media to the model of the free homogeneous electron gas. Based on an exclusively microscopic field theory it is shown that with the help of universally valid relations between response functions, all relevant optical and magnetic (linear) materials properties can be extracted from the mere current-current response. For this purpose, it is essential to base all calculations on the *full* current density operator, i. e. the sum of diamagnetic, orbital *and* spinorial contributions. Furthermore, we use the example of the magnetic susceptibility to demonstrate that the distinction between proper and direct response functions is in general crucial. Lastly, with the "Lindhard integral theorem" we prove that not only the longitudinal but also the transverse part of the full frequency- and wavevector-dependent fundamental response tensor of the free electron gas is completely determined by the characteristic Lindhard integral.

Kurzdarstellung

Anwendung universeller Relationen zwischen elektromagnetischen Antwortfunktionen auf das freie homogene Elektronengas.—Die vorliegende Arbeit befasst sich mit der Anwendung des kürzlich entwickelten „Functional Approach" zur Elektrodynamik in Medien auf das Modell des freien homogenen Elektronengases. Basierend auf einer ausschließlich mikroskopischen Feldtheorie wird gezeigt, dass mittels universell gültiger Relationen zwischen Antwortfunktionen sowohl alle relevanten optischen als auch magnetischen (linearen) Materialeigenschaften allein aus der Strom-Strom-Korrelation gewonnen werden können. Dabei ist es essentiell, alle Berechnungen auf dem *vollen* Stromdichteoperator aufzubauen, also auf der Summe aus diamagnetischem, orbitalem *und* spinoriellem Anteil. Weiterhin wird anhand der magnetischen Suszeptibilität demonstriert, dass im Allgemeinen die Unterscheidung zwischen eigenen und direkten Antwortfunktionen nicht zu vernachlässigen ist. Schließlich wird mit dem „Lindhard-Integral-Theorem" bewiesen, dass nicht nur der longitudinale, sondern auch der transversale Anteil des vollen frequenz- und wellenvektorabhängigen fundamentalen Antworttensors des freien Elektronengases komplett durch das charakteristische Lindhard-Integral bestimmt ist.

Contents

Introduction

> The career of a young theoretical physicist consists of treating the harmonic oscillator in ever-increasing levels of abstraction.
>
> *Sidney Coleman (1937-2007)*

Materials science is one of the largest fields of study nowadays. From fracture mechanics to high-energy and plasma physics and from ab initio physics to chemical synthesis, all these branches of science sooner or later have to refer to some kind of categorizing models.

While early studies in the 20th century spawned (from today's perspective) relatively easy microscopic descriptions of matter like the Rutherford and Bohr model, more abstract theoretical approaches were discovered quickly. Most important in this context has been the constitution of electrodynamics and quantum mechanics, which in fact dates back to the 19th century, where prominent experimental physicists like Hertz and Röntgen made their first discoveries with electromagnetic radiation. On the theoretical side, Maxwell completed the so-called Maxwell equations (to which he actually contributed only a single term), while Schrödinger discovered the probabilistic regime of physics with his famous equation of motion. Paired with the advent of special and general relativity, the foundation of virtually all relevant theories in modern physics has been laid and this is also where this thesis builds upon.

Coming back to materials models, quantum theory seeked to replace classical mechanics in the microscopic description of matter, since the former was not able to reproduce significant effects discovered by spectroscopical measurements like the Stern-Gerlach experiment. While classical models still find application primarily in engineering, quantum (field) theory has taken over the condensed matter field of research in theoretical physics.

So given a basic quantum system, the first thing an undergraduate student learns is that physical properties in form of (in principle) measurable quantities are mathematically represented by hermitian operators. Prime examples for such observables are position and momentum, usually introduced first by using the model of a simple harmonic oscillator. Unfortunately, this insight cannot be used for any practical application, say, for calculating optical or magnetic properties of the said quantum system. Now the question arises how physical quantities like the dielectric function should be accessible from quantum mechanics instead, when there is no suitable "dielectric operator" to take the expectation value from. The answer was given by the Japanese mathematical physicist Ryogo Kubo during his study of Green function approaches to linear response theory. The formalism discovered by him connects (in its spectral representation) single particle quantum states to so-called retarded response functions, which in turn can be converted to the searched-for dielectric function and

other integral kernels.

In the following sections, this concept is applied to the free, non-relativistic and homogeneous electron gas, which is probably the most essential materials model in solid state physics. Because of the missing interaction terms (hence it is called "free"), the entire physics can be expressed in terms of single-particle operators, very much like the simple harmonic oscillator from "quantum mechanics 101". In fact, it can be shown that even the higher quantum field theories eventually describe nothing but simple oscillators, although it is not some enigmatic pseudo-particle which oscillates, but the fields themselves.

In Chpt. 1 we start with basic electrodynamics, i. e. the description of electromagnetic fields in terms of *their own sources*. A central object in this respect is the *free* electromagnetic Green function, which in the so-called temporal gauge has only a spatial component.

Gauge Claim. *In response theory, the temporal gauge condition $\varphi \equiv 0$ seems to be the preferred one. In particular, it allows to work exclusively in the Cartesian space as opposed to Minkowskian spacetime.*

Chpt. 2 by contrast introduces the essential concepts of linear response theory, where the electromagnetic fields are expressed in terms of *external sources*. Here, the key quantity is the fundamental response tensor. Based on an exclusively microscopic field theory, we provide the framework for the universal response relations, which will be used throughout this thesis to convert one response function into another. Further, we illustrate the difference of proper and direct response functions and show how the dispersion relation is connected to the *full* electromagnetic Green function.

Central Claim. *In its rôle as spatial part of the fundamental response tensor, the frequency- and wavevector-dependent current response function determines all linear electromagnetic materials properties.*

In Part II Chpt. 3, we first derive the fundamental response tensor of the free electron gas within the Kubo-Greenwood formalism and learn, that it is composed of a diamagnetic, orbital and spinorial contribution. Since the isolated diamagnetic part is simultaneously known as the London model, we spend entire Chpt. 4 on optical and magnetic properties which can be derived already from this most simple toy model. We then continue with the study of the full current response in Chpt. 5, where we repeat all considerations of the London model and make out similarities and differences in these two models.

Part I.

Microscopic electrodynamics in media

1. Classical electrodynamics

This first chapter introduces the basic formalism of electrodynamics based on the following set of coupled differential equations,

$$\nabla \cdot \boldsymbol{E}(\boldsymbol{x},t) = \varepsilon_0^{-1}\rho(\boldsymbol{x},t)\,, \tag{1.1}$$

$$\nabla \times \boldsymbol{B}(\boldsymbol{x},t) - \mu_0\varepsilon_0\,\partial_t \boldsymbol{E}(\boldsymbol{x},t) = \mu_0 \boldsymbol{j}(\boldsymbol{x},t)\,, \tag{1.2}$$

$$\nabla \cdot \boldsymbol{B}(\boldsymbol{x},t) = 0\,, \tag{1.3}$$

$$\nabla \times \boldsymbol{E}(\boldsymbol{x},t) + \partial_t \boldsymbol{B}(\boldsymbol{x},t) = 0\,, \tag{1.4}$$

commonly referred to as Maxwell equations.[1] Concretely, they consist of Gauß' law for magnetic and electric fields (Eqs. (1.1) and (1.3)), Ampère's circuit law including Maxwell's displacement current (Eq. (1.2)) and Faraday's law of induction (Eq. (1.4)). Mathematically, they pose a linear system of partial differential equations in the electric and magnetic fields $\boldsymbol{E}$ and $\boldsymbol{B}$ with sources ρ and $\boldsymbol{j}$. In order to solve this system, initial conditions for $\boldsymbol{E}$ and $\boldsymbol{B}$ would need to be fixed and the spatial charge density ρ as well as the current density $\boldsymbol{j}$ must be known a priori for all times and positions. On a fundamental, microscopic level, these equations are always valid. In particular, they hold independently of any scale or material. Part I of this thesis is exclusively based on these four equations with all their implications.

In Sct. 1.1, the physics behind these Maxwell equations will be reformulated in a covariant manner, i. e. in a form that is invariant under Lorentz transformation. Effectively, this translates Eqs. (1.1) to (1.4) into a coupled set of wave equations for the potentials $\boldsymbol{A}$ and φ.[2] Fixing the temporal gauge $\varphi \equiv 0$ in Sct. 1.2 will allow us to proceed predominantly in the Euclidean regime, whereas in general all equations would have to be analyzed on a Minkowskian manifold. In particular electrodynamics in media in Chpt. 2 is immensely simplified by this rather unusual gauge condition. This substantiates our Gauge Claim. Sct. 1.3 then provides the (Cartesian) free Green function as a tool to express electromagnetic fields in terms of *their own* sources. By contrast, expressing fields within a material in terms of *external sources* is content of linear response theory introduced in Chpt. 2. The chapter closes with a general outline on the correct usage of total and partial functional derivatives as used heavily throughout this thesis.

In the following sections and in general for this part of the thesis, we will follow the lines and conventions of Ref. [6] and especially Ref. [11].

[1] This is actually the simplified 4-set of vector equations introduced by Heaviside, Gibbs and Hertz in 1884 on the basis of Maxwell's originally set of 20 equations published in the groundbreaking work [16].

[2] An alternative Lagrangian formulation based on the field strength tensor is given in App. C.1.

1.1. Covariant formulation

As a classical relativistic field theory, electromagnetism in form of the Maxwell Eqs. (1.1) to (1.4) hold independently of the inertial frame under consideration, i. e. certain equations retain their form under Lorentz transformation, provided they have been formulated in a covariant manner. For our purpose, the relativistic notation will merely provide a convenient way to simplify subsequent equations, whereas the discussion of transformational properties of e. g. response tensors lies outside the scope of this thesis, albeit quite important when it comes to applications like the Fizeau experiment [12, 17] or the discussion of the relativistic covariance of Ohm's law [7].

In the following (and in general for all tensors throughout this thesis), we will follow the conventions of a zero-based index notation as used in the Ricci calculus [18] and general relativity theory [19]. In particular, we will adopt the East Coast convention $(-,+,+,+)$ for the metric signature, which allows one to simply read-off the spatial part of any four-tensor without changing signs. Further, we will spare function arguments at times in favor of easier readability where it is implicitly understood, that tensor fields depend on the four-vector $x^\mu = (ct, \boldsymbol{x})^\mathsf{T}$ in real space or $k^\mu = (\omega/c, \boldsymbol{k})^\mathsf{T}$ in the Fourier domain.

As coupled set of partial differential equations, the Maxwell equations are used to determine expressions for the electric and magnetic fields in terms of their sources. However, because $\boldsymbol{E}$ and $\boldsymbol{B}$ are not indepentent of each other, there are in fact not six but only three degrees of freedom to be determined. A convenient way to eliminate this redundancy is to insert the relation between fields and potentials (which follow directly from Eqs. (1.3) and (1.4), cf. App. B.1),

$$\boldsymbol{E}(\boldsymbol{x},t) = -\nabla\varphi(\boldsymbol{x},t) - \partial_t \boldsymbol{A}(\boldsymbol{x},t)\,, \tag{1.5}$$

$$\boldsymbol{B}(\boldsymbol{x},t) = \nabla \times \boldsymbol{A}(\boldsymbol{x},t)\,, \tag{1.6}$$

into the inhomogeneous Maxwell equations (1.1) and (1.2). This yields the coupled equations of motion for φ and $\boldsymbol{A}$,

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \Delta\right)\varphi(\boldsymbol{x},t) - \frac{\partial}{\partial t}\left(\frac{1}{c^2}\frac{\partial}{\partial t}\varphi(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{A}(\boldsymbol{x},t)\right) = \frac{\rho(\boldsymbol{x},t)}{\varepsilon_0}\,, \tag{1.7}$$

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \Delta\right)\boldsymbol{A}(\boldsymbol{x},t) + \nabla\left(\frac{1}{c^2}\frac{\partial}{\partial t}\varphi(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{A}(\boldsymbol{x},t)\right) = \mu_0\boldsymbol{j}(\boldsymbol{x},t)\,, \tag{1.8}$$

where in the first one the identity $\nabla\times\nabla\times\boldsymbol{A} = \nabla(\nabla\cdot\boldsymbol{A}) - \Delta\,\boldsymbol{A}$ has been used. Thus, the problem of finding six different (but dependent) field components for $\boldsymbol{E}$ and $\boldsymbol{B}$ has been reduced to finding only the four components of the corresponding potentials. As will be explained in the next section, this number can be reduced even further to only three degrees of freedom after fixing the gauge.

By introducing the potential and current four-vectors as $A^\mu = (\varphi/c, \boldsymbol{A})^\mathsf{T}$ and $j^\mu = (c\rho, \boldsymbol{j})^\mathsf{T}$, Eqs. (1.7) and (1.8) can further be combined into a single covariant wave equation for the

four-potential,

$$(\eta^{\mu}{}_{\nu}\,\Box + \partial^{\mu}\partial_{\nu})A^{\nu} = \mu_0 j^{\mu}\,, \tag{1.9}$$

where

$$\Box \coloneqq -\partial^{\mu}\partial_{\mu} = \left(\frac{1}{c^2}\,\frac{\partial^2}{\partial t^2} - \Delta\right) \tag{1.10}$$

denotes the d'Alembert operator and

$$(\eta^{\mu}{}_{\nu}) = (\eta^{\mu\alpha}\eta_{\alpha\nu}) = \overset{\leftrightarrow}{\mathbb{1}}_{4\times 4} \tag{1.11}$$

is the mixed tensor version of the metric defined in Eq. (B.27). A third way to describe the electromagnetic propagation is by defining the exterior derivative of the four-potential as so-called field strength tensor,

$$F^{\mu\nu} \overset{\text{def}}{=} \partial^{\mu}A^{\nu} - \partial^{\nu}A^{\mu}\,, \tag{1.12}$$

such that the wave equation (1.9) can be expressed even more compactly as

$$\partial_{\nu}F^{\mu\nu} = \mu_0 j^{\mu}\,. \tag{1.13}$$

Now taking the mixed second partial derivative of the field strength tensor implies charge conservation in (flat)[3] space-time via the Lorentz-covariant continuity equation,

$$\partial_{\mu}j^{\mu} = 0\,. \tag{1.14}$$

In order to close the circle w. r. t. the free wave equation, we apply the d'Alembert operator to the field strength tensor,

$$\Box\, F^{\mu\nu} = \mu_0(\partial^{\mu}j^{\nu} - \partial^{\nu}j^{\mu})\,, \tag{1.15}$$

which then translates under the identifications given in App. B.2 back to the respective wave equations for the electric and magnetic fields in their Cartesian form,

$$\Box\, \boldsymbol{E} = -\frac{1}{\varepsilon_0}\nabla\rho - \mu_0\,\partial_t \boldsymbol{j}\,, \tag{1.16}$$

$$\Box\, \boldsymbol{B} = \mu_0 \nabla \times \boldsymbol{j}\,. \tag{1.17}$$

The latter two equations could have been found as well by taking the curl of Ampère's and Faraday's law and subsequently inserting the two Gauß' laws, Eqs. (1.1) and (1.3), respectively.

Although we only used the inhomogeneous Maxwell equations (1.1) and (1.2) in the derivation of the two Lorentz-covariant wave equations (1.9) and (1.13), the latter both are fully equivalent to the *entire* set of Maxwell equations. This circumstance can be explained by the fact that the homogeneous equations do not contain any information on the dynamics but

[3] We only consider special relativity in this thesis. See App. B.2 for details on the notation conventions used throughout this thesis.

rather represent initial conditions on the longitudinal field components at the starting time,

$$\nabla \cdot \boldsymbol{E}(\boldsymbol{x}, t_0) = \nabla \cdot \boldsymbol{E}_0(\boldsymbol{x}) = \rho_0(\boldsymbol{x})/\varepsilon_0 \,,$$
$$\nabla \cdot \boldsymbol{B}(\boldsymbol{x}, t_0) = \nabla \cdot \boldsymbol{B}_0(\boldsymbol{x}) = 0 \,,$$

which have to apply at all later times $t > t_0$ as well. In particular, they imply

$$\boldsymbol{E}_{\mathrm{L}}(\boldsymbol{x}, t) = \frac{1}{4\pi\varepsilon_0} \int \mathrm{d}^3\boldsymbol{x}' \; \rho(\boldsymbol{x}', t) \, \frac{\boldsymbol{x} - \boldsymbol{x}'}{|\boldsymbol{x} - \boldsymbol{x}'|^3} \,, \tag{1.18}$$

$$\boldsymbol{B}_{\mathrm{L}}(\boldsymbol{x}, t) = 0 \,, \tag{1.19}$$

for the respective longitudinal parts, whereas the corresponding transverse parts are exclusively determined by the field dynamics.[4]

Nevertheless, the homogeneous Maxwell equations are contained within the covariant formulation as well, namely in form of the geometric Bianchi identity [19, Eq. 10.65],

$$\partial_\gamma F_{\alpha\beta} + \partial_\alpha F_{\beta\gamma} + \partial_\beta F_{\gamma\alpha} = 0 \,, \tag{1.20}$$

which is always solved for the field strength tensor as defined in Eq. (1.12). In particular, by setting any of α, β or γ to zero, Eq. (1.20) reduces to Gauß' law for the electric field (Eq. (1.1)), and in case the tuple (α, β, γ) is any permutation from $\mathrm{S}_3(\{1, 2, 3\})$, Ampère's law in form of Eq. (1.2) is retained.

The fact that the physics behind the Maxwell equations can be expressed equivalently by $\{\boldsymbol{E}, \boldsymbol{B}\}$, $F^{\mu\nu}$ or A^μ is an extremely important result which will be exploited later in the discussion of the free and full electromagnetic Green functions. For the sake of this thesis, the most important consequence of this section is that we may work with the potentials $\boldsymbol{A}$ and φ instead of the electromagnetic fields $\boldsymbol{E}$ and $\boldsymbol{B}$ directly. This facilitates upcoming discussions tremendously, especially after fixing the temporal gauge (see Sct. 1.2), whereby it is even save to work exclusively with the Cartesian vector potential. Therefore, we will use covariant notation in the following only when necessary or when emphasizing particular (covariant) aspects of relations, and otherwise resort to the Cartesian representation whenever possible.

1.2. Temporal gauge

Rewriting the Maxwell equations in terms of the potentials greatly reduced the complexity of the initial value problem as shown in the previous section. It further allowed to work with (Lorentz) covariant equations which can be crucial, depending on the exact problem. The four-potential A^μ itself, however, is still subject to a so-called gauge freedom, i. e. replacing

[4] This is an astonishing discovery which has a huge impact on the entire way electrodynamics is treated in higher theories like (relativistic) quantum electrodynamics, where only the transverse parts $\boldsymbol{E}_{\mathrm{T}}$ and $\boldsymbol{B}_{\mathrm{T}}$ are quantized via the Yang-Mills Lagrange density (C.21), and on the other hand many-body theory which then closes the circle by providing a quantized (albeit usually non-relativistic) field theory for the Schrödinger field and thereby for the charge density $\rho(\boldsymbol{x}) = (-e)\,\langle\hat{n}(\boldsymbol{x})\rangle$ (see Sct. 3.2) which in turn determines the remaining longitudinal part $\boldsymbol{E}_{\mathrm{L}}$. Only the much later discovered non-abelian SU(2) and SU(3) gauge theories treat all parts equally and eventually leads to the standard model of (particle) physics.

it according to

$$A^\mu \longmapsto A^\mu + \partial^\mu f\,, \tag{1.21}$$

with an arbitrary scalar function f, or equivalently in Cartesian form,

$$\boldsymbol{A}(\boldsymbol{x},t) \longmapsto \boldsymbol{A}(\boldsymbol{x},t) + \nabla f(\boldsymbol{x},t)\,, \tag{1.22}$$

$$\varphi(\boldsymbol{x},t) \longmapsto \varphi(\boldsymbol{x},t) - \partial_t f(\boldsymbol{x},t)\,, \tag{1.23}$$

has no impact on the defining equation of the field strength tensor (1.12) and hence produces the same set of electric and magnetic fields, $\{\boldsymbol{E},\boldsymbol{B}\}$. In the theory of differential equations this characteristic is reflected by the fact that adding a homogeneous solution to any other one always constructs a new solution. A pure gauge, i. e. $A^\mu \equiv \partial^\mu f$, is exactly such a homogeneous solution for the wave equation (1.9). The equation of motion for potentials therefore has to be complemented by an additional gauge condition in order to receive definite results. That way, the number of redundand degrees of freedom can be reduced once more leaving only three of the initial six degrees as anticipated in the beginning of the previous section.

The most popular gauge choices are undoubtly the Lorenz[5] gauge, $\partial_\mu A^\mu = 0$, which is unique in retaining manifest Lorentz covariance, and the Coulomb gauge $\nabla\cdot\boldsymbol{A} = 0$, which restricts the vector potential to its transverse part, $\boldsymbol{A} \mapsto \boldsymbol{A}_\mathrm{T}$. As a consequence of the latter, longitudinal and transverse degrees of freedom of the potentials fully decouple and thereby reduce the wave equations (1.7) and (1.8) to

$$\Delta\,\varphi(\boldsymbol{x},t) = -\rho(\boldsymbol{x},t)/\varepsilon_0\,, \tag{1.24}$$

$$\Box\,\boldsymbol{A}(\boldsymbol{x},t) = \mu_0\,\boldsymbol{j}_\mathrm{T}(\boldsymbol{x},t)\,. \tag{1.25}$$

The solution of the first equation is known explicitly and together with the gauge condition $\boldsymbol{A}_\mathrm{L} \equiv \boldsymbol{0}$ and the transformation from the Coulomb gauge to any other, $A^\mu \mapsto A^\mu_\mathcal{C} + \partial^\mu f$, the general form of φ and $\boldsymbol{A}_\mathrm{L}$ may be expressed gauge-independently as [10, §4.1.3]

$$\varphi(\boldsymbol{x},t) = \partial_t f(\boldsymbol{x},t) + \frac{1}{4\pi\varepsilon_0}\int \mathrm{d}^3\boldsymbol{x}'\,\frac{\rho(\boldsymbol{x}',t)}{|\boldsymbol{x}-\boldsymbol{x}'|}\,, \tag{1.26}$$

$$\boldsymbol{A}_\mathrm{L}(\boldsymbol{x},t) = \nabla f(\boldsymbol{x},t)\,. \tag{1.27}$$

In other words, the scalar potential generally consists of a pure gauge and the Coulomb potential (which is the general solution of Poisson's equation (1.24)), while the longitudinal vector potential is always given by a pure gauge. In order to find the remaining transverse part of the vector potential, the Coulomb gauge is again employed in the framework of quantum electrodynamics and eventually yields the following expression (cf. [20, Eq. (1.154)] or [21, Eq. (1.89)]),

$$\boldsymbol{A}_\mathrm{T}(\boldsymbol{x},t) = \sum_{\lambda=1,2}\sqrt{\frac{\hbar}{\varepsilon_0}}\int\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^{3/2}}\,\frac{1}{\sqrt{2\omega_{\boldsymbol{k}}}}\left(a_{\boldsymbol{k}\lambda}\boldsymbol{e}_{\boldsymbol{k}\lambda}e^{-\mathrm{i}\omega_{\boldsymbol{k}}t+i\boldsymbol{k}\cdot\boldsymbol{x}} + a^*_{\boldsymbol{k}\lambda}\boldsymbol{e}^*_{\boldsymbol{k}\lambda}e^{\mathrm{i}\omega_{\boldsymbol{k}}t-i\boldsymbol{k}\cdot\boldsymbol{x}}\right)\,, \tag{1.28}$$

[5] Named after the Danish physicist Ludvig Lorenz, not after the much more popular Dutch Hendrik Antoon Lorentz, and actually introduced by neither of both, but by the Irish academic and Professor of Natural Philosophy George Francis FitzGerald.

which in this form is known as mode expansion. Here, the two possible values for λ denote the so-called polarization with corresponding (transverse) polarization vectors $\boldsymbol{e}_{\boldsymbol{k},\lambda} \perp \boldsymbol{k}$ and Fourier amplitudes $\boldsymbol{a}_{\boldsymbol{k},\lambda}$, whereas the occuring frequencies $\omega_{\boldsymbol{k}}$ need to be determined through a fitting dispersion relation. Interestingly, this result can likewise be derived in an entirely classic picture as well, a fact that already holds for the creation and annihilation operator $\hat{a}^{(\dagger)}$ of the simple (quantum mechanical) harmonic oszillator.

While the Coulomb gauge certainly has the largest scope of application, in the specific case of (linear) response theory it proved not to be the best choice. Instead, for the remaining part of this thesis we will adhere to the temporal gauge (a. k. a. Weyl gauge) defined by

$$\varphi \equiv 0\,, \tag{1.29}$$

which turned out to work particularly well for the study of electrodynamics in media. Applying the temporal gauge condition to the wave equations (1.7) and (1.8) yields

$$-\frac{\partial}{\partial t}\left(\nabla \cdot \boldsymbol{A}(\boldsymbol{x},t)\right) = \frac{1}{\varepsilon_0}\,\rho(\boldsymbol{x},t)\,, \tag{1.30}$$

$$\left(\frac{1}{c^2}\,\frac{\partial^2}{\partial t^2} - \Delta\right)\boldsymbol{A}(\boldsymbol{x},t) + \nabla\left(\nabla \cdot \boldsymbol{A}(\boldsymbol{x},t)\right) = \mu_0\,\boldsymbol{j}(\boldsymbol{x},t)\,, \tag{1.31}$$

or equivalently in the Fourier domain,

$$-\omega\left(\boldsymbol{k} \cdot \boldsymbol{A}(\boldsymbol{k},\omega)\right) = \frac{1}{\varepsilon_0}\,\rho(\boldsymbol{k},\omega)\,, \tag{1.32}$$

$$\left(-\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2\right)\boldsymbol{A}(\boldsymbol{k},\omega) - \boldsymbol{k}\left(\boldsymbol{k} \cdot \boldsymbol{A}(\boldsymbol{k},\omega)\right) = \mu_0\,\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.33}$$

where the convention compiled in App. B.3 have been employed. Obviously, the first equation follows from the second one when taking the scalar product with the vector $\boldsymbol{k}$. Therefore we may safely discard the first one and keep working exclusively with the second equation. Nonetheless, we may still combine them in order to eliminate the divergence term in favour of the charge density and thereby recast Eq. (1.33) into

$$\left(-\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2\right)\boldsymbol{A}(\boldsymbol{k},\omega) = \mu_0\left(\boldsymbol{j}(\boldsymbol{k},\omega) - \frac{c\boldsymbol{k}}{\omega}\,c\rho(\boldsymbol{k},\omega)\right)\,. \tag{1.34}$$

Inserting in this equation the relations between fields and potentials, Eqs. (1.5) and (1.6), whose Fourier equivalent simplify in temporal gauge to

$$\boldsymbol{E}(\boldsymbol{k},\omega) = \mathrm{i}\omega\boldsymbol{A}(\boldsymbol{k},\omega)\,, \tag{1.35}$$

$$\boldsymbol{B}(\boldsymbol{k},\omega) = \mathrm{i}\boldsymbol{k} \times \boldsymbol{A}(\boldsymbol{k},\omega)\,, \tag{1.36}$$

results in the standard wave equations for electromagnetic fields in Fourier space (cf. Eqs. (1.16)

and (1.17)),

$$\left(-\frac{\omega^2}{c^2}+|\boldsymbol{k}|^2\right)\boldsymbol{E}(\boldsymbol{k},\omega)=-\frac{1}{\varepsilon_0}\,\mathrm{i}\boldsymbol{k}\,\rho(\boldsymbol{k},\omega)+\mu_0\,\mathrm{i}\omega\,\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.37}$$

$$\left(-\frac{\omega^2}{c^2}+|\boldsymbol{k}|^2\right)\boldsymbol{B}(\boldsymbol{k},\omega)=\mu_0\,\mathrm{i}\boldsymbol{k}\times\boldsymbol{j}(\boldsymbol{k},\omega)\,. \tag{1.38}$$

For the electric field, one could have alternatively started from Eq. (1.33), which can also be written as

$$\left(-\frac{\omega^2}{c^2}+|\boldsymbol{k}|^2-|\boldsymbol{k}|^2\overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{k})\right)\boldsymbol{A}(\boldsymbol{k},\omega)=\mu_0\,\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.39}$$

leading to an equation of motion for $\boldsymbol{E}$ exclusively in terms of the spatial current $\boldsymbol{j}$,

$$\left(-\frac{\omega^2}{c^2}+|\boldsymbol{k}|^2\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\right)\boldsymbol{E}(\boldsymbol{k},\omega)=\mu_0\,\mathrm{i}\omega\,\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.40}$$

similar to Eqs. (1.33) and (1.38). The same result is obtained when inserting Gauß' law (which follows directly from the combination of Eq. (1.32) and Eq. (1.35)),

$$\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{E}(\boldsymbol{k},\omega)=\frac{\rho(\boldsymbol{k},\omega)}{\varepsilon_0}\,, \tag{1.41}$$

into Eq. (1.37) in order to eliminate the charge density again.

1.3. Free Green function

An enormous advantage of working in Fourier space is that differential equations can be formally solved for the respective fields just by algebraically inverting the corresponding differential operators. Moreover, functional derivatives reduce to regular ones, as we will see in the next section. In case of Eqs. (1.38) to (1.40), we hence find the following concise expressions in Fourier space,

$$\boldsymbol{A}(\boldsymbol{k},\omega)=\overleftrightarrow{D}_0(\boldsymbol{k},\omega)\,\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.42}$$

$$\boldsymbol{E}(\boldsymbol{k},\omega)=\mathrm{i}\omega\overleftrightarrow{D}_0(\boldsymbol{k},\omega)\,\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.43}$$

$$\boldsymbol{B}(\boldsymbol{k},\omega)=\mathrm{i}|\boldsymbol{k}|\overleftrightarrow{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k},\omega)\,\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.44}$$

where the *free electromagnetic Green function* $\overleftrightarrow{D}_0$ is explicitly given by (cf. [22, §2.1.7])

$$\overleftrightarrow{D}_0(\boldsymbol{k},\omega)=\mathbb{D}_0(\boldsymbol{k},\omega)\left(\overleftrightarrow{\mathbb{1}}-\frac{c^2|\boldsymbol{k}|^2}{\omega^2}\overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{k})\right)\,, \tag{1.45}$$

with $\mathbb{D}_0$ being the *scalar Green function* of the d'Alembert operator (up to the factor μ_0),

$$\mathbb{D}_0(\boldsymbol{k},\omega)=\frac{\mu_0}{-\omega^2/c^2+|\boldsymbol{k}|^2}=-\frac{1}{\varepsilon_0\omega^2}\,\frac{\omega^2}{\omega^2-c^2|\boldsymbol{k}|^2}\,. \tag{1.46}$$

The latter both quantities emerge as the formal solutions of their defining equations in Fourier space,

$$\frac{1}{\mu_0}\left(-\frac{\omega^2}{c^2}+|\boldsymbol{k}|^2\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\right)\overleftrightarrow{D}_0(\boldsymbol{k},\omega)\stackrel{\mathrm{def}}{=}\overleftrightarrow{\mathbb{1}}\,, \tag{1.47}$$

$$\frac{1}{\mu_0}\left(-\frac{\omega^2}{c^2}+|\boldsymbol{k}|^2\right)\mathbb{D}_0(\boldsymbol{k},\omega)\stackrel{\mathrm{def}}{=}1\,, \tag{1.48}$$

where the factor μ_0 was chosen to be included in both definitions such that Eq. (1.42) is retained in this simple form. In addition, it can be shown that this free Green function embeds trivially in Minkowskian space,

$$(D_0)^{\mu}{}_{\nu}(\boldsymbol{k},\omega)=\begin{pmatrix}0 & \mathbf{0}^{\mathsf{T}}\\ \mathbf{0} & \overleftrightarrow{D}_0(\boldsymbol{k},\omega)\end{pmatrix}\,, \tag{1.49}$$

provided the temporal gauge is used. Generally, using the latter obviously simplifies equations in electromagnetic response theory considerably. This substantiates our Gauge Claim, according to which the temporal (a. k. a.) Weyl gauge is the natural one to use in this field of study

Further, the so-called electric and magnetic solution generators will play an important rôle in subsequent sections since they can be used to comfortably express universal response relations (cf. Sct. 2.3). In terms of the free electromagnetic Green function, these two dimensionless operators read

$$\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega)=-\varepsilon_0\omega^2\overleftrightarrow{D}_0(\boldsymbol{k},\omega)\,, \tag{1.50}$$

$$\overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega)=-\varepsilon_0\omega c|\boldsymbol{k}|\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\overleftrightarrow{D}_0(\boldsymbol{k},\omega)\,, \tag{1.51}$$

such that the solutions Eqs. (1.43) and (1.44) can be expressed equivalently as

$$\boldsymbol{E}(\boldsymbol{k},\omega)=\frac{1}{\mathrm{i}\omega\varepsilon_0}\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega)\boldsymbol{j}(\boldsymbol{k},\omega)\,, \tag{1.52}$$

$$c\boldsymbol{B}(\boldsymbol{k},\omega)=\frac{1}{\mathrm{i}\omega\varepsilon_0}\overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega)\boldsymbol{j}(\boldsymbol{k},\omega)\,. \tag{1.53}$$

For later purpose, it is also convenient to recast both solution generators solely in terms of projection and rotation operators instead of the bulkier free Green function:

$$\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega)=\overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{k})+\frac{\omega^2}{\omega^2-c^2|\boldsymbol{k}|^2}\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{1.54}$$

$$\overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega)=\frac{\omega c|\boldsymbol{k}|}{\omega^2-c^2|\boldsymbol{k}|^2}\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,. \tag{1.55}$$

In the second equation, the identity $\overleftrightarrow{R}_{\mathrm{T}}\overleftrightarrow{D}_0\equiv\mathbb{D}_0\overleftrightarrow{R}_{\mathrm{T}}$ has been used. On the other hand, $\overleftrightarrow{D}_0$ can be brought into isotropic form,

$$\overleftrightarrow{D}_0(\boldsymbol{k},\omega)=-\frac{1}{\varepsilon_0\omega^2}\overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{k})+\mathbb{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\,. \tag{1.56}$$

which holds even outside of the isotropic limit. In the so-called optical limit, $\boldsymbol{k} \to \boldsymbol{0}$, all these operators simplify even further,

$$\lim_{\boldsymbol{k}\to\boldsymbol{0}} \overleftrightarrow{D}(\boldsymbol{k},\omega) = -\frac{1}{\varepsilon_0\omega^2}\,\overleftrightarrow{\mathbb{1}}\,, \tag{1.57}$$

$$\lim_{\boldsymbol{k}\to\boldsymbol{0}} \overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}}\,, \tag{1.58}$$

$$\lim_{\boldsymbol{k}\to\boldsymbol{0}} \overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega) = 0\,. \tag{1.59}$$

The latter relations will prove handy in Sct. 2.7.

1.4. Total functional derivatives

In this section, we will shortly introduce the highly effective formalism of total functional derivatives that will put us in a position to derive relations between arbitrary response functions with minimal effort.

First consider the partial composition $f = f(x, y)$ with $y = y(x)$. Its total derivative w. r. t. the variable x reads (cf. Eq. (B.6))

$$\frac{\mathrm{d}}{\mathrm{d}x} f(x,y) = \frac{\partial}{\partial x} f(x,y) + \frac{\partial}{\partial y} f(x,y)\, y'(x)\,. \tag{1.60}$$

By inserting the abbreviation $f(x, y(x)) =: \tilde{f}(x)$, the functional relation between f and x stays unaltered, whereas the exact form of f simultaneously changes in a way that the total derivative can be expressed alternatively in terms of $\tilde{f}$ as

$$\frac{\mathrm{d}}{\mathrm{d}x} f(x,y) = \frac{\mathrm{d}}{\mathrm{d}x} \tilde{f}(x) \equiv \frac{\partial}{\partial x} \tilde{f}(x) \neq \frac{\partial}{\partial x} f(x,y)\,. \tag{1.61}$$

The particular meaning of partial or total derivatives obviously depends on the exact form of the function. Strictly mathematical speaking, it would be necessary to introduce a new symbol for each such "function form" as done in this example with f and $\tilde{f}$. However, in physics this is usually done implicitly by a change of function arguments, which especially apply for the Fourier transform (see App. B.3). This makes it all the more important to always include function arguments in equations, except when the meaning is absolutely unambiguous.

Functional derivatives behave in a very similar way, especially in Fourier space, where the general real space formalism reduces to mere ordinary (partial) derivatives (cf. App. B.4). In this context, the wave equation for the electric field will serve as an instructive example, since it can be expressed either in terms of the charge density *and* the spatial current (Eq. (1.37)), or alternatively solely as function of the latter (Eq. (1.40)). Applying the regular (i. e. partial) functional derivative w. r. t. the spatial current to the second variant $\boldsymbol{E} = \boldsymbol{E}[\boldsymbol{j}]$ as given by Eq. (1.52), we find

$$\frac{\delta \boldsymbol{E}[\boldsymbol{j}]}{\delta \boldsymbol{j}} \equiv \frac{\mathrm{d}\boldsymbol{E}[\boldsymbol{j}]}{\mathrm{d}\boldsymbol{j}} = \mathrm{i}\omega \overleftrightarrow{D}_0(\boldsymbol{k},\omega)\,. \tag{1.62}$$

By algebraically solving the wave equation (1.37) for the electric field $\boldsymbol{E} = \boldsymbol{E}[\rho, \boldsymbol{j}]$ with

$\rho = \rho[\boldsymbol{j}]$,

$$\boldsymbol{E}[\rho, \boldsymbol{j}](\boldsymbol{k}, \omega) = i\omega\, \mathbb{D}_0(\boldsymbol{k}, \omega)\, \boldsymbol{j}(\boldsymbol{k}, \omega) - c^2\, \mathrm{i}\boldsymbol{k}\, \mathbb{D}_0(\boldsymbol{k}, \omega)\, \rho(\boldsymbol{k}, \omega)\,, \tag{1.63}$$

and applying again the regular functional derivative, we find on the other hand

$$\frac{\delta \boldsymbol{E}[\rho, \boldsymbol{j}]}{\delta \boldsymbol{j}} = \mathrm{i}\omega\, \mathbb{D}_0(\boldsymbol{k}, \omega)\, \overleftrightarrow{\mathbb{1}}\,, \tag{1.64}$$

which is apparently not equal to Eq. (1.62).

Up to this point, there are already several things to note: First of all, we always use the symbol d for *total* derivatives independently of their functional or primitive character, which is in particular transparent in Fourier space. Secondly, the result of a functional derivative obviously depends on the exact form of the functional, or in other words on the total number and types of functional dependencies, just like the regular partial derivative depends on the number of function arguments. Consequently, the partial functional derivatives of different forms of the same functional do not coincide in general. However, their total functional derivatives do always, just like for regular functions. For the above example, this will be shown explicitly in a moment. A more severe problem is given by the fact that contrary to arguments of functions, functional dependencies are not noted explicitly, which can lead to fatal errors when taking functional derivatives. Thus it is essential to always indicate the associated defining equations. Throughout the formulae in this thesis, there is however not much potential for confusion in this regard, since more often than not, the missing partial derivatives emerge from implicit functional dependencies given either by the continuity equation,

$$\rho[\boldsymbol{j}](\boldsymbol{k}, \omega) = \frac{\boldsymbol{k} \cdot \boldsymbol{j}(\boldsymbol{k}, \omega)}{\omega}\,. \tag{1.65}$$

which expresses the charge density as a functional of the spatial current, or Faraday's law,

$$\boldsymbol{B}[\boldsymbol{E}](\boldsymbol{k}, \omega) = \frac{\boldsymbol{k} \times \boldsymbol{E}(\boldsymbol{k}, \omega)}{\omega}\,, \tag{1.66}$$

relating the magnetic field to the transverse electric field. Finding the correct functional dependencies in each case is hence more of a physical than a mere mathematical task. In some cases total and partial functional derivatives always coincide. This is particularly true for the functional $\rho[\boldsymbol{j}]$, since the continuity equation does not allow for any ambiguity (except for an additional constant term, which has no impact on the derivative). Likewise, in the temporal gauge all functional derivatives of the trivial potential functional $\varphi[\boldsymbol{A}] = 0$ vanish, such that in particular the total and partial derivatives w. r. t. the vector potential,

$$\frac{\mathrm{d}}{\mathrm{d}\boldsymbol{A}} = \frac{\delta}{\delta \boldsymbol{A}} + \frac{\delta \varphi}{\delta \boldsymbol{A}} \frac{\delta}{\delta \varphi} \overset{\varphi=0}{\equiv} \frac{\delta}{\delta \boldsymbol{A}}\,, \tag{1.67}$$

coincide.

Putting all this together, the total functional derivative of the electric field w. r. t. the

spatial current is given by

$$\frac{\mathrm{d}\boldsymbol{E}[\boldsymbol{j}]}{\mathrm{d}\boldsymbol{j}} = \frac{\mathrm{d}\boldsymbol{E}[\rho,\boldsymbol{j}]}{\mathrm{d}\boldsymbol{j}} = \frac{\delta\boldsymbol{E}[\rho,\boldsymbol{j}]}{\delta\boldsymbol{j}} + \frac{\delta\boldsymbol{E}[\rho,\boldsymbol{j}]}{\delta\rho}\frac{\delta\rho[\boldsymbol{j}]}{\delta\boldsymbol{j}}\,, \tag{1.68}$$

and thus exactly yields the already familiar relation between scalar and tensorial electromagnetic Green function (1.45),

$$\mathrm{i}\omega\overleftrightarrow{D}_0 = \mathrm{i}\omega\,\mathbb{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{\mathbb{1}} - c^2\,\mathrm{i}\boldsymbol{k}\,\mathbb{D}_0(\boldsymbol{k},\omega)\,\frac{\boldsymbol{k}^{\mathsf{T}}}{\omega}\,. \tag{1.69}$$

Similarly, by inserting the two inhomogeneous Maxwell equations

$$\rho(\boldsymbol{k},\omega) = \varepsilon_0\,\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{E}(\boldsymbol{k},\omega)\,, \tag{1.70}$$

$$\boldsymbol{j}(\boldsymbol{k},\omega) = \frac{1}{\mu_0}\,\mathrm{i}\boldsymbol{k}\times\boldsymbol{B}(\boldsymbol{k},\omega) - \varepsilon_0\,\mathrm{i}\omega\,\boldsymbol{E}(\boldsymbol{k},\omega)\,, \tag{1.71}$$

into the wave equation (1.34) and solving for $\boldsymbol{A}$, the vector potential can be expressed as a functional of $\boldsymbol{E}$ and $\boldsymbol{B}$,

$$\boldsymbol{A}[\boldsymbol{E},\boldsymbol{B}](\boldsymbol{k},\omega) = -\frac{1}{\mathrm{i}\omega}\,\varepsilon_0\,\mathbb{D}_0(\boldsymbol{k},\omega)\Big(\omega^2\boldsymbol{E}(\boldsymbol{k},\omega) - c^2\boldsymbol{k}\,(\boldsymbol{k}\cdot\boldsymbol{E}(\boldsymbol{k},\omega)) + \omega c^2\boldsymbol{k}\times\boldsymbol{B}(\boldsymbol{k},\omega)\Big)\,. \tag{1.72}$$

which is by Eqs. (1.50) and (1.51) and the relation $\overleftrightarrow{R}_{\mathrm{T}}\overleftrightarrow{D}_0 \equiv \mathbb{D}_0\overleftrightarrow{R}_{\mathrm{T}}$ identical to

$$\boldsymbol{A}[\boldsymbol{E},\boldsymbol{B}](\boldsymbol{k},\omega) = \frac{1}{\mathrm{i}\omega}\left(\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega)\,\boldsymbol{E}(\boldsymbol{k},\omega) + \overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega)\,c\boldsymbol{B}(\boldsymbol{k},\omega)\right)\,. \tag{1.73}$$

The partial derivatives w. r. t. the electromagnetic fields can now be read-off in a most convenient way from the latter equation. With the two identities

$$\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega) + \frac{c|\boldsymbol{k}|}{\omega}\,\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,\overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}}\,, \tag{1.74}$$

and

$$\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega) - \frac{c|\boldsymbol{k}|}{\omega}\,\overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega) = \overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{1.75}$$

as well as the partial derivatives

$$\frac{\delta\boldsymbol{B}(\boldsymbol{k},\omega)}{\delta\boldsymbol{E}(\boldsymbol{k},\omega)} = +\frac{|\boldsymbol{k}|}{\omega}\,\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{1.76}$$

$$\frac{\delta\boldsymbol{E}(\boldsymbol{k},\omega)}{\delta\boldsymbol{B}(\boldsymbol{k},\omega)} = -\frac{\omega}{|\boldsymbol{k}|}\,\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{1.77}$$

where Eq. (1.76) follows directly from Faradays law Eq. (1.66), and for Eq. (1.77) one has to insert the decomposition $\boldsymbol{E} = \boldsymbol{E}_{\mathrm{L}} + \boldsymbol{E}_{\mathrm{T}}$ (cf. Eq. (B.171)) and use that $\boldsymbol{E}_{\mathrm{L}} \neq \boldsymbol{E}_{\mathrm{L}}[\boldsymbol{B}]$. we find

the corresponding total derivatives in the Fourier domain to be

$$\frac{\mathrm{d}\boldsymbol{A}[\boldsymbol{E},\boldsymbol{B}]}{\mathrm{d}\boldsymbol{E}} = \frac{\delta\boldsymbol{A}}{\delta\boldsymbol{E}} + \frac{\delta\boldsymbol{A}}{\delta\boldsymbol{B}}\frac{\delta\boldsymbol{B}}{\delta\boldsymbol{E}} = \frac{1}{\mathrm{i}\omega}\,, \tag{1.78}$$

$$\frac{\mathrm{d}\boldsymbol{A}[\boldsymbol{E},\boldsymbol{B}]}{\mathrm{d}\boldsymbol{B}} = \frac{\delta\boldsymbol{A}}{\delta\boldsymbol{B}} + \frac{\delta\boldsymbol{A}}{\delta\boldsymbol{E}}\frac{\delta\boldsymbol{E}}{\delta\boldsymbol{B}} = -\frac{1}{\mathrm{i}|\boldsymbol{k}|}\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,. \tag{1.79}$$

This is in perfect accord with the relation between fields and potentials in the temporal gauge, Eqs. (1.35) and (1.36).

2. Electrodynamics in media

In condensed matter physics, (linear) response theory aims to describe the reaction of a material to (small) external perturbations. Thereby, the induced fields are considered to be generated exclusively by the applied perturbations, namely in form of a redistribution of charges and currents within the sample. Matching the experimental situation, the external fields are regarded as fully controllable, whereas the material-dependent induced fields are subject to theoretical studies. The relation between both contributions is introduced as so-called response function and as long as the external fields are small compared to the undisturbed ones within the material, the induced response is assumed to be linear in the perturbation. Specifically for electric and magnetic fields, the associated response functions can be identified with the inverse permittivity $\varepsilon_{\mathrm{r}}^{-1}$ and the permeability μ_{r}, respectively. As we will see later, the latter two are not independent from each other. Quite to the contrary, it has been shown that all response functions can be traced back to a single *fundamental response tensor*. In fact, it is even only its spatial part, the so-called *current response tensor*, that comprises the entire information about all electromagnetic material properties.

This chapter presents the basic formalism of response theory in terms of external, induced and total fields while following closely the notations and conventions from Ref. [6]. In Sct. 2.1, the modern (microscopic) splitting of fields and sources is introduced and compared to the commonly employed traditional (macroscopic) notation. Sct. 2.2 continues with the introduction of the current response χ_{ij} as the spatial part of the *fundamental response tensor* $\chi^{\mu}{}_{\nu}$, in terms of which all other linear electromagnetic response functions may be expressed using the framework given by Sct. 2.3. In Sct. 2.4, the essential difference between direct and proper response functions is illustrated. While magnetic properties are usually derived from a direct response, optical properties are by contrast accessed via proper functions. Sct. 2.5 continues with the decomposition of response tensors in longitudinal and transverse parts in the isotropic limit. Optical, static and combined limites are covered as well. Scts. 2.6 and 2.7 finally deal with the question how the wave equation in media can be solved in the framework of linear response theory and how this is connected to longitudinal and transverse electromagnetic dispersion relations.

2.1. Field identifications

The linear structure of Maxwell's equations allow us to arbitrarily split the two source terms in Eqs. (1.1) and (1.2) while retaining their functional relations to electric and magnetic fields for each part. In ab initio materials physics it is common practice to mimic the experimental setup in form of externally adjustable fields and detectable (induced) changes in a specimen.

This suggests a splitting into external and internal contributions,

$$\rho(\boldsymbol{x},t) = \rho_{\mathrm{int}}(\boldsymbol{x},t) + \rho_{\mathrm{ext}}(\boldsymbol{x},t)\,, \tag{2.1}$$
$$\boldsymbol{j}(\boldsymbol{x},t) = \boldsymbol{j}_{\mathrm{int}}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{ext}}(\boldsymbol{x},t)\,, \tag{2.2}$$

where the internal ones further split into contributions present without any perturbation and contributions induced by perturbation,

$$\rho_{\mathrm{int}}(\boldsymbol{x},t) = \rho_{\mathrm{int},0}(\boldsymbol{x},t) + \rho_{\mathrm{ind}}(\boldsymbol{x},t)\,, \tag{2.3}$$
$$\boldsymbol{j}_{\mathrm{int}}(\boldsymbol{x},t) = \boldsymbol{j}_{\mathrm{int},0}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{x},t)\,. \tag{2.4}$$

As we will see shortly, it is most convenient to additionally introduce total sources,

$$\rho_{\mathrm{tot}}(\boldsymbol{x},t) = \rho_{\mathrm{ext}}(\boldsymbol{x},t) + \rho_{\mathrm{ind}}(\boldsymbol{x},t)\,, \tag{2.5}$$
$$\boldsymbol{j}_{\mathrm{tot}}(\boldsymbol{x},t) = \boldsymbol{j}_{\mathrm{ext}}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{x},t)\,, \tag{2.6}$$

which correspond to electric and magnetic fields of the form

$$\boldsymbol{E}_{\mathrm{tot}}(\boldsymbol{x},t) = \boldsymbol{E}_{\mathrm{ext}}(\boldsymbol{x},t) + \boldsymbol{E}_{\mathrm{ind}}(\boldsymbol{x},t)\,, \tag{2.7}$$
$$\boldsymbol{B}_{\mathrm{tot}}(\boldsymbol{x},t) = \boldsymbol{B}_{\mathrm{ext}}(\boldsymbol{x},t) + \boldsymbol{B}_{\mathrm{ind}}(\boldsymbol{x},t)\,. \tag{2.8}$$

On the other hand, both electromagnetic fields are fully determined by their potentials $\boldsymbol{A}$ and φ according to Eqs. (1.5) and (1.6), such that the source splitting carries over to them as well,

$$\boldsymbol{A}_{\mathrm{tot}}(\boldsymbol{x},t) = \boldsymbol{A}_{\mathrm{ext}}(\boldsymbol{x},t) + \boldsymbol{A}_{\mathrm{ind}}(\boldsymbol{x},t)\,, \tag{2.9}$$
$$\varphi_{\mathrm{tot}}(\boldsymbol{x},t) = \varphi_{\mathrm{ext}}(\boldsymbol{x},t) + \varphi_{\mathrm{ind}}(\boldsymbol{x},t)\,. \tag{2.10}$$

This partitioning will prove extremely important in the derivation of response relations in the remaining part of this chapter as it offers an easy possibility to split or substitute factors in functional derivatives.

The Maxwell equations in form of Eqs. (1.1) to (1.4) including all their implications are valid independent of any space-time scale or a specific application, i. e. they are in particular valid for electrodynamics in media. Historically, however, these equations were regarded as valid only in vacuo, while nowadays they are more or less known as the "macroscopic" theory. Since some authors adopt the "macroscopic" style but still treat the fields microscopically, we will refer to this notation as *traditional partitioning* as opposed to the *modern partitioning* introduced in this section. Although there are some subtle differences regarding the averaging part (see App. D.2 for details), the traditional approach can be translated into its modern counterpart quite flawlessly by suitably defining all occuring fields in terms of the components of Eqs. (2.7) and (2.8) as will be shown in this section.

In macroscopic electrodynamics of media, fields and sources are typically split into free and bound[1] contributions instead of internal and external. Additional, so-called *material*

relations are fixed, (cf. [23, p. 3], [24, Eq. (28.1)]),

$$\boldsymbol{D} = \varepsilon_0 \varepsilon_r \boldsymbol{E}\,, \tag{2.11}$$

$$\boldsymbol{H} = \mu_0^{-1} \mu_r^{-1} \boldsymbol{B}\,, \tag{2.12}$$

which connect the *electric field* $\boldsymbol{E}$ and the *magnetic induction* $\boldsymbol{B}$ to the *displacement field* $\boldsymbol{D}$ and the *magnetic field* $\boldsymbol{H}$ via the material properties ε_r and μ_r.[2] The inhomogeneous Maxwell equations (1.1) and (1.2) are then replaced by

$$\nabla \cdot \boldsymbol{D}(\boldsymbol{x}, t) = \rho_\mathrm{f}(\boldsymbol{x}, t)\,, \tag{2.13}$$

$$\nabla \times \boldsymbol{H}(\boldsymbol{x}, t) = \boldsymbol{j}_\mathrm{f}(\boldsymbol{x}, t) + \partial_t \boldsymbol{D}(\boldsymbol{x}, t)\,, \tag{2.14}$$

for the *free sources* ρ_f and $\boldsymbol{j}_\mathrm{f}$, and

$$\nabla \cdot \boldsymbol{P}(\boldsymbol{x}, t) = \rho_\mathrm{b}(\boldsymbol{x}, t)\,, \tag{2.15}$$

$$\nabla \times \boldsymbol{M}(\boldsymbol{x}, t) = \boldsymbol{j}_\mathrm{b}(\boldsymbol{x}, t) + \partial_t \boldsymbol{P}(\boldsymbol{x}, t)\,, \tag{2.16}$$

for the *bound sources* ρ_b and $\boldsymbol{j}_\mathrm{b}$. These are known as Maxwell equations in media and again constitute pairs of inhomogeneous differential equations, but now in the fields $\boldsymbol{D}$ and $\boldsymbol{H}$, and $\boldsymbol{P}$ and $\boldsymbol{M}$, respectively. The *polarization* $\boldsymbol{P}$ and the *magnetization* $\boldsymbol{M}$ are defined by

$$\boldsymbol{P}(\boldsymbol{x}, t) = -\varepsilon_0 \boldsymbol{E}(\boldsymbol{x}, t) + \boldsymbol{D}(\boldsymbol{x}, t)\,, \tag{2.17}$$

$$\boldsymbol{M}(\boldsymbol{x}, t) = \mu_0^{-1} \boldsymbol{B}(\boldsymbol{x}, t) - \boldsymbol{H}(\boldsymbol{x}, t)\,, \tag{2.18}$$

which in turn implies

$$\nabla \cdot \boldsymbol{E}(\boldsymbol{x}, t) = \rho_\mathrm{tot}(\boldsymbol{x}, t)\,, \tag{2.19}$$

$$\nabla \times \boldsymbol{B}(\boldsymbol{x}, t) - \mu_0 \varepsilon_0 \, \partial_t \boldsymbol{E}(\boldsymbol{x}, t) = \mu_0 \boldsymbol{j}_\mathrm{tot}(\boldsymbol{x}, t)\,, \tag{2.20}$$

for the *total sources* $\rho_\mathrm{tot} = \rho_\mathrm{f} + \rho_\mathrm{b}$ and $\boldsymbol{j}_\mathrm{tot} = \boldsymbol{j}_\mathrm{f} + \boldsymbol{j}_\mathrm{b}$.

Although this treatment appears to be completely equivalent to the partitioning from Eqs. (2.7) and (2.8), it is in fact not. This is because according to the fundamental theorem of vector analysis (Eqs. (B.167) to (B.169)) both, transverse (via curl) *and* the longitudinal parts (via divergence) of a vector field are required for its full description. In the traditional splitting, we find by contrast that the field equations for $\boldsymbol{D}$, $\boldsymbol{P}$, $\boldsymbol{H}$ and $\boldsymbol{M}$ are *incomplete*, because Eqs. (2.14) and (2.16) only define longitudinal parts whereas Eqs. (2.13) and (2.15) similarly only define transverse parts of the respective fields. Consequently, the fields $\boldsymbol{E}$ and $\boldsymbol{B}$ are not properly defined as well. This is a mathematical fact which cannot be argued away

[1] As of today, there is in fact no consensus on how the terms "free" and "bound" have to be interpreted, which becomes immediately obvious when comparing the statements given in different contemporary text books on electrodynamics. For a thourough discussion see [9, §2.2.2] and especially all references specified therein.

[2] All italic terms refer to the historical naming scheme and do not necessarily reflect the real nature of the respective fields. For example, the *magnetic field* $\boldsymbol{H}$ is not the field that enters Lorentz' force law, $\boldsymbol{F} = q\boldsymbol{v} \times \boldsymbol{B}$, but the *real* magnetic field $\boldsymbol{B}$ (historically: *magnetic induction*) does instead.

by physically asserting a "macroscopic" nature for these fields.

It is often claimed (e. g. [25]), that by Eqs. (2.11) and (2.12) the fields $\boldsymbol{H}$ and $\boldsymbol{D}$ were fully defined. From an ab initio point of view however, it is clear that this cannot be a valid argument since the response functions ε_r and μ_r are not known in general. In fact, in the framework of linear response theory, they are actually *defined* by these very equations. Since a single equation cannot be used to define two otherwise independent quantities, this is obviously not a workable solution. Alternatively, some authors (e. g. [26]) state that the incomplete fields are subject to a gauge freedom. Consequently, this would have to apply to ε_r and μ_r as well which does not seem to be very reasonable either, since one of the great challenges in materials science is to calculate response functions from first principles and compare the results with experimental measurements. A possible gauge freedom would make this an inherently impossible task. The third objection, which is simultaneously the most popular one, is the introduction of $\boldsymbol{P}$ and $\boldsymbol{M}$ as (sometimes macroscopically averaged) *dipole densities*. Again, this leads to a whole series of problems, of which one is even more severe than the other. For example, this assumption seems especially far-fetched in context of the otherwise highly delocalized free electron gas. In fact, such a Clausius-Mossotti-type model [27] fails even for real materials as has already been discussed by Resta and Vanderbilt in their "modern theory of polarization"[28]. Moreover, for the macroscopically averaged version one would be limited to the macroscopic domain right from the start, which seems not very productive in context of microscopic ab initio theories. At the end of the day, this argument ultimately fails for the simple reason "that the attempt to define the polarization and magnetization as continuous densities of electric and magnetic *point dipoles* leads precisely back to the original problem of insufficiently defined fields"[9, p.13]. A more detailed discussion with a sensible selection of references is given in [9, §2.2.1].

All this given, if one still insists on the traditional *notation*, it still can be translated into its modern counterpart by simply postulating the following *fundamental field identifications* (see [6] or e. g. [24, Eq. 29.20])

$$\boldsymbol{P}(\boldsymbol{x},t) = -\varepsilon_0 \boldsymbol{E}_{\mathrm{ind}}(\boldsymbol{x},t)\,, \tag{2.21}$$

$$\boldsymbol{D}(\boldsymbol{x},t) = +\varepsilon_0 \boldsymbol{E}_{\mathrm{ext}}(\boldsymbol{x},t)\,, \tag{2.22}$$

$$\boldsymbol{E}(\boldsymbol{x},t) = \boldsymbol{E}_{\mathrm{tot}}(\boldsymbol{x},t)\,, \tag{2.23}$$

and

$$\boldsymbol{M}(\boldsymbol{x},t) = \mu_0^{-1} \boldsymbol{B}_{\mathrm{ind}}(\boldsymbol{x},t)\,, \tag{2.24}$$

$$\boldsymbol{H}(\boldsymbol{x},t) = \mu_0^{-1} \boldsymbol{B}_{\mathrm{ext}}(\boldsymbol{x},t)\,, \tag{2.25}$$

$$\boldsymbol{B}(\boldsymbol{x},t) = \boldsymbol{B}_{\mathrm{tot}}(\boldsymbol{x},t)\,, \tag{2.26}$$

where the total fields $\boldsymbol{E}_{\mathrm{tot}}$ and $\boldsymbol{B}_{\mathrm{tot}}$ are given by Eqs. (2.7) and (2.8), respectively. As discussed before, the Maxwell equations in form of Eqs. (1.1) to (1.4) apply to each contribution

Tab. 2.1.: Traditional field equations versus modern field identifications. Where the former lacks definitions of transverse parts of $\boldsymbol{D}$ and $\boldsymbol{P}$ and longitudinal parts of $\boldsymbol{H}$ and $\boldsymbol{M}$, the latter provides well defined expressions for both parts such that all fields are uniquely defined.

traditional (macroscopic)	$\nabla \cdot \boldsymbol{E} = \rho_{\text{tot}}/\varepsilon_0$	$\nabla \times \boldsymbol{E} = -\partial_t \boldsymbol{B}$	$\nabla \cdot \boldsymbol{B} = 0$	$\nabla \times \boldsymbol{B} = \mu_0 \boldsymbol{j}_{\text{tot}} + \partial_t \boldsymbol{E}/c^2$
	$\nabla \cdot \boldsymbol{D} = \rho_{\text{f}}$	$\nabla \times \boldsymbol{D} = ?$	$\nabla \cdot \boldsymbol{H} = ?$	$\nabla \times \boldsymbol{H} = \boldsymbol{j}_{\text{f}} + \partial_t \boldsymbol{D}$
	$\nabla \cdot \boldsymbol{P} = -\rho_{\text{b}}$	$\nabla \times \boldsymbol{P} = ?$	$\nabla \cdot \boldsymbol{M} = ?$	$\nabla \times \boldsymbol{M} = \boldsymbol{j}_{\text{b}} - \partial_t \boldsymbol{P}$
modern (microscopic)	$\nabla \cdot \boldsymbol{E} = \rho_{\text{tot}}/\varepsilon_0$	$\nabla \times \boldsymbol{E} = -\partial_t \boldsymbol{B}$	$\nabla \cdot \boldsymbol{B} = 0$	$\nabla \times \boldsymbol{B} = \mu_0 \boldsymbol{j}_{\text{tot}} + \partial_t \boldsymbol{E}/c^2$
	$\nabla \cdot \boldsymbol{D} = \rho_{\text{ext}}$	$\nabla \times \boldsymbol{D} = -\partial_t \boldsymbol{H}/c^2$	$\nabla \cdot \boldsymbol{H} = 0$	$\nabla \times \boldsymbol{H} = \boldsymbol{j}_{\text{ext}} + \partial_t \boldsymbol{D}$
	$\nabla \cdot \boldsymbol{P} = -\rho_{\text{ind}}$	$\nabla \times \boldsymbol{P} = \partial_t \boldsymbol{M}/c^2$	$\nabla \cdot \boldsymbol{M} = 0$	$\nabla \times \boldsymbol{M} = \boldsymbol{j}_{\text{ind}} - \partial_t \boldsymbol{P}$

separately and hence result in

$$\nabla \cdot \boldsymbol{D}(\boldsymbol{x}, t) = \rho_{\text{ext}}(\boldsymbol{x}, t)\,, \tag{2.27}$$
$$\nabla \times \boldsymbol{D}(\boldsymbol{x}, t) = -\mu_0 \varepsilon_0 \partial_t \boldsymbol{H}(\boldsymbol{x}, t)\,, \tag{2.28}$$
$$\nabla \cdot \boldsymbol{H}(\boldsymbol{x}, t) = 0\,, \tag{2.29}$$
$$\nabla \times \boldsymbol{H}(\boldsymbol{x}, t) = \boldsymbol{j}_{\text{ext}}(\boldsymbol{x}, t) + \partial_t \boldsymbol{D}(\boldsymbol{x}, t)\,, \tag{2.30}$$

for the external fields and

$$\nabla \cdot \boldsymbol{P}(\boldsymbol{x}, t) = -\rho_{\text{ind}}(\boldsymbol{x}, t)\,, \tag{2.31}$$
$$\nabla \times \boldsymbol{P}(\boldsymbol{x}, t) = \mu_0 \varepsilon_0 \partial_t \boldsymbol{M}(\boldsymbol{x}, t)\,, \tag{2.32}$$
$$\nabla \cdot \boldsymbol{M}(\boldsymbol{x}, t) = 0\,, \tag{2.33}$$
$$\nabla \times \boldsymbol{M}(\boldsymbol{x}, t) = \boldsymbol{j}_{\text{ind}}(\boldsymbol{x}, t) - \partial_t \boldsymbol{P}(\boldsymbol{x}, t)\,, \tag{2.34}$$

for the induces ones, such that all introduced fields are now uniquely defined by their own set of equations. An overview of all field definitions of both approaches is given in Table 2.1.

2.2. Fundamental response tensor

When applying electromagnetic fields to materials, the initial (undisturbed) charges and currents will respond to these external perturbations in form of an internal reordering of these very sources, which in turn generates so-called induced fields. Thus, a natural starting point of response theory would be to express the induced charges and currents in terms of the externally applied fields $\{\boldsymbol{E}_{\text{ext}}, \boldsymbol{B}_{\text{ext}}\}$. The problem with this approach is, that this functional would not be well-defined since electric and magnetic fields can not be varied independently from each other as shown in Sct. 1.4 and discussed in more detail in [6, §4.3]. At the same time, it is the electromagnetic potential A^μ which enters the fundamental quantum field theoretical Lagrangian via minimal coupling principle instead of the fields $\boldsymbol{E}_{\text{ext}}$ and $\boldsymbol{B}_{\text{ext}}$,

and hence we choose [6, Eq. 5.1]

$$j^\mu_{\text{int}} = j^\mu_{\text{int}}[A^\nu_{\text{ext}}] \tag{2.35}$$

as the fundamental functional which connects the four-current in the material to the four-potential of the perturbation. This choice simultaneously allows for a manifestly Lorentz-covariant formulation of response theory. In particular to first-order Taylor expansion, one finds for the above functional the approximation

$$j^\mu_{\text{int}}(x) = j^\mu_{\text{int},0}(x) + \int \mathrm{d}^4x' \, \frac{\delta j^\mu_{\text{int}}(x)}{\delta A^\nu_{\text{ext}}(x')} \left(A^\nu_{\text{ext}}(x') - A^\nu_{\text{ext},0}(x') \right) , \tag{2.36}$$

which is usually evaluated at vanishing reference potential $A^\nu_{\text{ext},0}$.[3] By the partitioning (2.4), the difference between the perturbed and unperturbed internal currents can be replaced with the corresponding induced contribution. Thus, Eq. (2.36) simplifies in compact notation to

$$j^\mu_{\text{ind}} = \chi^\mu{}_\nu \, A^\nu_{\text{ext}} \, , \tag{2.37}$$

by which we have introduced the *fundamental response tensor*,

$$\chi^\mu{}_\nu(x, x') \overset{\text{def}}{=} \frac{\delta j^\mu_{\text{ind}}(x)}{\delta A^\nu_{\text{ext}}(x')} \, , \tag{2.38}$$

as the functional derivative of the induced four-current w. r. t. the external four-potential.

In order to account for current conservation in each domain, the continuity equation (1.14) must hold for total, external and induced sources separately, such that

$$0 \overset{!}{=} \partial_\mu j^\mu_{\text{ind}}(x) = \int \mathrm{d}^4x \, \partial_\mu \chi^\mu{}_\nu(x, x') \, A^\nu_{\text{ext}}(x') \, . \tag{2.39}$$

Further, the gauge invariance of the four-potential (1.21) should not lead to physical currents or put differently, as an observable field the four-current should be invariant under gauge transformation. Therefore, we additionally require

$$0 = \int \mathrm{d}^4x \, \chi^\mu{}_\nu(x, x') \partial'^\nu f(x') = - \int \mathrm{d}^4x \, \partial'^\nu \chi^\mu{}_\nu(x, x') \, f(x') \, . \tag{2.40}$$

Here, the closed surface integral emerging during the partial integration in the second step vanished because the response to an infinitely distant perturbation in spacetime should do as well. Due to the arbitrariness of A^μ in Eq. (2.39) and likewise for f in Eq. (2.40), both requirements can be combined into the concise restriction [30, p. 390]

$$\partial_\mu \chi^\mu{}_\nu(x, x') = \partial'^\nu \chi^\mu{}_\nu(x, x') = 0 \, . \tag{2.41}$$

Expanding the latter equation immediately reveals that the nine remaining independent com-

[3] There are certain situations where a non-vanishing reference potential is preferrable, e. g. when studying the Hall conductivity, where a voltage is applied while the material is under influence of an otherwise constant magnetic field. [29]

ponents can all be expressed explicitly in terms of the *current response tensor*,

$$\overleftrightarrow{\chi}(\boldsymbol{x},\boldsymbol{x}';t-t') = \frac{\delta \boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{x},t)}{\delta \boldsymbol{A}_{\mathrm{ext}}(\boldsymbol{x}',t')}\,, \tag{2.42}$$

namely by means of the relations

$$\chi^{0}{}_{\nu}(\boldsymbol{x},\boldsymbol{x}';\omega) = \frac{c}{\mathrm{i}\omega}\sum_{i}\frac{\partial}{\partial x^{i}}\,\chi_{ij}(\boldsymbol{x},\boldsymbol{x}';\omega)\,, \tag{2.43}$$

$$\chi^{\mu}{}_{0}(\boldsymbol{x},\boldsymbol{x}';\omega) = \frac{c}{\mathrm{i}\omega}\sum_{j}\frac{\partial}{\partial x'^{j}}\,\chi_{ij}(\boldsymbol{x},\boldsymbol{x}';\omega)\,, \tag{2.44}$$

$$\chi^{0}{}_{0}(\boldsymbol{x},\boldsymbol{x}';\omega) = -\frac{c^{2}}{\omega^{2}}\sum_{ij}\frac{\partial}{\partial x^{i}}\frac{\partial}{\partial x'^{j}}\,\chi_{ij}(\boldsymbol{x},\boldsymbol{x}';\omega)\,. \tag{2.45}$$

In Fourier domain, this is equivalent to the following (3+1)-representation,

$$\chi^{\mu}{}_{\nu}(\boldsymbol{k},\boldsymbol{k}';\omega) = \begin{pmatrix} -\dfrac{c^{2}}{\omega^{2}}\,\boldsymbol{k}^{T}\overleftrightarrow{\chi}\,\boldsymbol{k}' & \dfrac{c}{\omega}\,\boldsymbol{k}^{T}\overleftrightarrow{\chi} \\ -\dfrac{c}{\omega}\,\overleftrightarrow{\chi}\,\boldsymbol{k}' & \overleftrightarrow{\chi} \end{pmatrix}, \tag{2.46}$$

where the transformation convention for response functions from App. B.3 has been applied. The latter was chosen in a way that leaves response laws like Eq. (2.37) invariant under Fourier transformation and simultaneously is in accordance with the functional chain rule as well as numerical implementations like "The Elk Code" [31]. Eq. (2.46) displays explicitly, that also in Fourier space the entire information on the electromagnetic response is already contained within the Cartesian current response χ_{ij}, which is just the spatial part of the corresponding fundamental tensor. Another consequence of Eq. (2.46) is, that there can be at most nine independent electromagnetic linear response functions for any material as a matter of principle [32–34], provided one accepts that the fundamental response tensor already comprises already the entire (electromagnetic) response information. In other words, every possible electromagnetic response function can be expressed in terms of $\chi_{ij} \equiv \chi^{i}{}_{j}$. This fact is explicitly shown in Sct. 2.3.

For homogeneous "materials" like the free electron gas where the tensor in Eq. (2.42) only depends on the difference of the two spatial arguments as discussed in App. B.3, the corresponding Fourier representation becomes a function of a single argument and therefore further simplifies to (cf. [32, p. 145])

$$\chi^{\mu}{}_{\nu}(\boldsymbol{k},\boldsymbol{k}';\omega) = \chi^{\mu}{}_{\nu}(\boldsymbol{k},\omega)\,\delta^{3}(\boldsymbol{k}-\boldsymbol{k}')\,. \tag{2.47}$$

The same form emerges naturally for periodic crystals provided that all frequencies corresponding to the applied electromagnetic fields do not fall into the X-ray or gamma region of the spectrum, which is certainly the case for those experiments which are our concern here. Correspondingly, all wavevectors are from now on assumed to lie in the first Brillouin zone (see App. D.2)

In the beginning of this chapter, response functions have been introduced as integral kernels

relating the induced fields of a material to the externally applied field. Alternatively, by replacing external with total fields, *proper* response functions can be defined alongside their *direct* counterparts. In particular regarding the fundamental response tensor as given by Eq. (2.38) and Eq. (2.42), we define the *proper fundamental response tensor* and its spatial part by

$$\widetilde{\chi}^{\mu}{}_{\nu}(x,x') = \frac{\delta j^{\mu}_{\mathrm{ind}}(x)}{\delta A^{\nu}_{\mathrm{tot}}(x')}\,, \qquad \overset{\leftrightarrow}{\widetilde{\chi}}(\boldsymbol{x},\boldsymbol{x}',t-t') = \frac{\delta \boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{x},t)}{\delta \boldsymbol{A}_{\mathrm{tot}}(\boldsymbol{x}',t')}\,, \tag{2.48}$$

while both tensors are related via the Dysonian equation

$$\chi^{\mu}{}_{\nu}(x,x') = \widetilde{\chi}^{\mu}{}_{\nu}(x,x') + \int \mathrm{d}^4y \int \mathrm{d}^4y'\, \widetilde{\chi}^{\mu}{}_{\lambda}(x,y)\,(D_0)^{\lambda}{}_{\rho}(y,y')\,\chi^{\rho}{}_{\nu}(y',x')\,, \tag{2.49}$$

or in compact notation,

$$\chi = \widetilde{\chi} + \widetilde{\chi} D_0 \chi\,. \tag{2.50}$$

The same relation carries over to their respective spatial parts. In Fourier space, this can be explicitly shown using the formalism of total functional derivatives, where we first note that in temporal gauge χ can be identified with the *total* functional derivative,

$$\frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{k},\omega)}{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}(\boldsymbol{k},\omega)} \overset{(1.67)}{=} \frac{\delta \boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{k},\omega)}{\delta \boldsymbol{A}_{\mathrm{ext}}(\boldsymbol{k},\omega)} = \overset{\leftrightarrow}{\chi}(\boldsymbol{k},\omega)\,. \tag{2.51}$$

Combining the functional chain rule with an expansion of total fields into induced and external contributions then leads to

$$\frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}} = \frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{A}_{\mathrm{tot}}}\left(\frac{\mathrm{d}\boldsymbol{A}_{\mathrm{tot}}}{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}}\right) = \frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{A}_{\mathrm{tot}}}\left(\frac{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}} + \left(\frac{\mathrm{d}\boldsymbol{A}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}\right)\frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}}\right)\,, \tag{2.52}$$

which is by Eqs. (1.42), (2.42) and (2.48) equivalent to

$$\overset{\leftrightarrow}{\chi} = \overset{\leftrightarrow}{\widetilde{\chi}} + \overset{\leftrightarrow}{\widetilde{\chi}}\overset{\leftrightarrow}{D_0}\overset{\leftrightarrow}{\chi}\,. \tag{2.53}$$

Here, we particularly note that Eq. (1.42) is valid for all vector potentials (induced, external, total) in terms of *their* sources. By contrast, the corresponding Green function that relates *total* potentials to *external* currents will be introduced in the next section.

Finally, expanding Eq. (2.48) to linear order yields the response law

$$j^{\mu}_{\mathrm{ind}} = \widetilde{\chi}^{\mu}{}_{\nu} A^{\nu}_{\mathrm{tot}}\,, \tag{2.54}$$

similarly to Eq. (2.37).

2.3. Universal response relations

In this section the general approach of how to find arbitrary response relations will be exerted on the example of the electromagnetic fields themselves. Assuming homogeneity in space and time from now on (see App. D.2 for details), we start by inserting the representation of the

electric field in temporal gauge, Eq. (1.35), into Eq. (2.51),

$$\frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{k},\omega)}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}(\boldsymbol{k},\omega)} = \frac{1}{\mathrm{i}\omega}\overleftrightarrow{\chi}(\boldsymbol{k},\omega)\,. \tag{2.55}$$

By comparing the latter equation with the microscopic Ohm law,

$$\boldsymbol{j}_{\mathrm{ind}}(\boldsymbol{k},\omega) = \overleftrightarrow{\sigma}(\boldsymbol{k},\omega)\,\boldsymbol{E}_{\mathrm{ext}}(\boldsymbol{k},\omega) \tag{2.56}$$

which relates the induced current in a material to an externally applied electric field by means of the conductivity tensor $\overleftrightarrow{\sigma}$, the identity

$$\overleftrightarrow{\chi}(\boldsymbol{k},\omega) = \mathrm{i}\omega\overleftrightarrow{\sigma}(\boldsymbol{k},\omega) \tag{2.57}$$

can be read-off (cf. [32, Eq. (3.185)] and [35, Eq. (4.13)]). Thus, all linear electromagnetic response functions can alternatively be expressed in terms of the conductivity instead of the current response tensor. It is important to note that in case of Ohm's law, the functional derivative of the current w. r. t. the electric field has again to be identified with a total one, i. e. $\boldsymbol{j}_{\mathrm{ind}} = \boldsymbol{j}_{\mathrm{ind}}[\boldsymbol{E}_{\mathrm{ext}}]$ in Eq. (2.56). Consequently the conductivity already accounts for all effects that can be ascribed to the presence of magnetic fields due to time-dependent electric fields. A fully relativistic and gauge-independent derivation of Eq. (2.57) is given in Ref. [7], where it is explicitly shown that Ohm's law can in fact be derived from the linear response law Eq. (2.37) directly.

Next we want to find the aforementioned universal response relations between the conductivity and other linear electromagnetic response tensors as stated in our Central Claim. For that reason, we define all possible responses of induced electromagnetic fields w. r. t. their external counterparts by the following dimensionless total functionals,

$$\overleftrightarrow{\chi}_{EE}(\boldsymbol{k},\omega) \overset{\mathrm{def}}{=} \frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}} = \frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}\left(\frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}\right) = \frac{1}{\mathrm{i}\omega\varepsilon_0}\,\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega)\overleftrightarrow{\sigma}(\boldsymbol{k},\omega)\,, \tag{2.58}$$

$$\overleftrightarrow{\chi}_{EB}(\boldsymbol{k},\omega) \overset{\mathrm{def}}{=} \frac{1}{c}\frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}} = \frac{1}{c}\frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}\left(\frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}}\right) = \frac{\mathrm{i}}{c|\boldsymbol{k}|\varepsilon_0}\,\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega)\overleftrightarrow{\sigma}(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.59}$$

$$\overleftrightarrow{\chi}_{BE}(\boldsymbol{k},\omega) \overset{\mathrm{def}}{=} c\,\frac{\mathrm{d}\boldsymbol{B}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}} = c\,\frac{\mathrm{d}\boldsymbol{B}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}\left(\frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}\right) = \frac{1}{\mathrm{i}\omega\varepsilon_0}\,\overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega)\overleftrightarrow{\sigma}(\boldsymbol{k},\omega)\,, \tag{2.60}$$

$$\overleftrightarrow{\chi}_{BB}(\boldsymbol{k},\omega) \overset{\mathrm{def}}{=} \frac{\mathrm{d}\boldsymbol{B}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}} = \frac{\mathrm{d}\boldsymbol{B}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}\left(\frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}}\right) = \frac{\mathrm{i}}{c|\boldsymbol{k}|\varepsilon_0}\,\overleftrightarrow{\mathbb{B}}(\boldsymbol{k},\omega)\overleftrightarrow{\sigma}(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.61}$$

where Ohm's law together with Eqs. (1.52) and (1.53) have been used in Eqs. (2.58) and (2.60) and the latter results together with the derivatives Eqs. (1.76) and (1.77) have been inserted in turn into Eqs. (2.59) and (2.61). By response relation (2.57), these four functional derivatives can be alternatively expressed in terms of the current response χ instead of the conductivity σ. Similarly, the electric and magnetic solution generators $\mathbb{E}$ and $\mathbb{B}$ may be replaced with the free Green function D_0 by virtue of Eqs. (1.50) and (1.51). In particular, we find for the

first and last equations

$$\overleftrightarrow{\chi}_{EE}(\boldsymbol{k},\omega) = \overleftrightarrow{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{\chi}(\boldsymbol{k},\omega)\,, \tag{2.62}$$

and

$$\overleftrightarrow{\chi}_{BB}(\boldsymbol{k},\omega) = -\mathbb{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{\chi}_{RR}(\boldsymbol{k},\omega)\,, \tag{2.63}$$

respectively, with the abbreviation

$$\overleftrightarrow{\chi}_{RR}(\boldsymbol{k},\omega) \coloneqq \overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\overleftrightarrow{\chi}(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,. \tag{2.64}$$

We particularly note, that the response relations Eqs. (2.57) to (2.63) are similarly valid for the corresponding proper versions of χ_{EE} etc. under the replacement $\sigma \to \widetilde{\sigma}$ and $\chi \to \widetilde{\chi}$.

2.4. Direct and proper response

As already mentioned in Sct. 2.1, in the traditional approach to electrodynamics in media constutive laws are introduced next to the Maxwell equations. Among them are usually

$$\boldsymbol{P} = \varepsilon_0 \overleftrightarrow{\chi}_{\mathrm{e}} \boldsymbol{E}\,, \tag{2.65}$$

relating the polarization in terms of the (total) electric field via the electric susceptibility χ_{e}, and

$$\boldsymbol{M} = \overleftrightarrow{\chi}_{\mathrm{m}} \boldsymbol{H}\,, \tag{2.66}$$

giving the magnetization in terms of the (external) magnetic field with help of the magnetic susceptibility χ_{m}, together with the already introduced Ohm law Eq. (2.56) and the two material relations (2.11) and (2.12). By the fundamental field identifications given in Sct. 2.1, we can directly read off the identities

$$\overleftrightarrow{\chi}_{\mathrm{m}} \equiv \overleftrightarrow{\chi}_{BB}\,, \tag{2.67}$$

$$-\overleftrightarrow{\chi}_{\mathrm{e}} \equiv \overleftrightarrow{\widetilde{\chi}}_{EE}\,. \tag{2.68}$$

Interestingly, the constutive law for the magnetization translates to a direct response tensor whereas the one for the polarization corresponds to a proper tensor. If we still wanted to express the latter in terms of a direct tensor, the formalism of total functional derivatives has to be employed once again, yielding the relation

$$(\overleftrightarrow{\chi}_{EE})^{-1} - (\overleftrightarrow{\widetilde{\chi}}_{EE})^{-1} = \overleftrightarrow{\mathbb{1}}\,. \tag{2.69}$$

For the electric permittivity and the magnetic permeability from Eqs. (2.11) and (2.12) on the other hand, we find

$$\frac{1}{\varepsilon_0}\frac{\mathrm{d}\boldsymbol{D}}{\mathrm{d}\boldsymbol{E}} \equiv \frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{tot}}} = \overleftrightarrow{\varepsilon}_{\mathrm{r}}\,, \tag{2.70}$$

$$\frac{1}{\mu_0}\frac{\mathrm{d}\boldsymbol{B}}{\mathrm{d}\boldsymbol{H}} \equiv \frac{\mathrm{d}\boldsymbol{B}_{\mathrm{tot}}}{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}} = \overleftrightarrow{\mu}_{\mathrm{r}}\,. \tag{2.71}$$

For these response functions it is noteworthy that they cannot be classified as proper or direct since they represent functional derivatives of *total* (instead of induced) fields w. r. t. external ones. Further, it is in fact not ε itself but its inverse which is a response function. The dielectric tensor is related to the corresponding susceptibility via

$$(\overleftrightarrow{\varepsilon}_{\mathrm{r}})^{-1} = \frac{\mathrm{d}\boldsymbol{E}_{\mathrm{tot}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}} = \frac{\mathrm{d}(\boldsymbol{E}_{\mathrm{ext}}+\boldsymbol{E}_{\mathrm{ind}})}{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}} = \overleftrightarrow{\mathbb{1}} + \overleftrightarrow{\chi}_{EE}\,, \tag{2.72}$$

$$\overleftrightarrow{\varepsilon}_{\mathrm{r}} = \frac{\mathrm{d}\boldsymbol{E}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{E}_{\mathrm{tot}}} = \frac{\mathrm{d}(\boldsymbol{E}_{\mathrm{tot}}-\boldsymbol{E}_{\mathrm{ind}})}{\mathrm{d}\boldsymbol{E}_{\mathrm{tot}}} = \overleftrightarrow{\mathbb{1}} - \overleftrightarrow{\widetilde{\chi}}_{EE}\,, \tag{2.73}$$

and similar for the two magnetic response functions,

$$\overleftrightarrow{\mu}_{\mathrm{r}} = \frac{\mathrm{d}\boldsymbol{B}_{\mathrm{tot}}}{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}} = \frac{\mathrm{d}(\boldsymbol{B}_{\mathrm{ext}}+\boldsymbol{B}_{\mathrm{ind}})}{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}} = \overleftrightarrow{\mathbb{1}} + \overleftrightarrow{\chi}_{BB}\,, \tag{2.74}$$

$$(\overleftrightarrow{\mu}_{\mathrm{r}})^{-1} = \frac{\mathrm{d}\boldsymbol{B}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{B}_{\mathrm{tot}}} = \frac{\mathrm{d}(\boldsymbol{B}_{\mathrm{tot}}-\boldsymbol{B}_{\mathrm{ind}})}{\mathrm{d}\boldsymbol{B}_{\mathrm{tot}}} = \overleftrightarrow{\mathbb{1}} - \overleftrightarrow{\widetilde{\chi}}_{BB}\,, \tag{2.75}$$

from which we can immediately read off the two familiar relations

$$\overleftrightarrow{\varepsilon}_{\mathrm{r}} = \overleftrightarrow{\chi}_{\mathrm{e}} + \overleftrightarrow{\mathbb{1}}\,, \tag{2.76}$$

$$\overleftrightarrow{\mu}_{\mathrm{r}} = \overleftrightarrow{\chi}_{\mathrm{m}} + \overleftrightarrow{\mathbb{1}}\,. \tag{2.77}$$

Moreover, from the universal response relations Eqs. (2.62) and (2.63) follow the alternative representations

$$(\overleftrightarrow{\varepsilon})^{-1}_{\mathrm{r}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}} + \overleftrightarrow{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{\chi}(\boldsymbol{k},\omega)\,, \tag{2.78}$$

$$\overleftrightarrow{\varepsilon}_{\mathrm{r}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}} - \overleftrightarrow{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{\widetilde{\chi}}(\boldsymbol{k},\omega) \tag{2.79}$$

and

$$\overleftrightarrow{\mu}_{\mathrm{r}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}} - \mathbb{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\overleftrightarrow{\chi}(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.80}$$

$$(\overleftrightarrow{\mu})^{-1}_{\mathrm{r}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}} + \mathbb{D}_0(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\overleftrightarrow{\chi}(\boldsymbol{k},\omega)\overleftrightarrow{R}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.81}$$

which in turn lead back to the Dysonian Eq. (2.50) or equivalently (cf. [32, Eq. 5.22])

$$\overleftrightarrow{\widetilde{\sigma}}(\boldsymbol{k},\omega) = \overleftrightarrow{\sigma}(\boldsymbol{k},\omega)\overleftrightarrow{\varepsilon}_{\mathrm{r}}(\boldsymbol{k},\omega)\,. \tag{2.82}$$

in the first case, and to

$$\overleftrightarrow{\sigma}_{RR}(\boldsymbol{k},\omega) = \overleftrightarrow{\widetilde{\sigma}}_{RR}(\boldsymbol{k},\omega)\,\overleftrightarrow{\mu}_{\mathrm{r}}(\boldsymbol{k},\omega) \tag{2.83}$$

in the second one. By combining Eq. (2.78) and Eq. (2.80) it is obvious, that the permittivity and permeability cannot be chosen independent from each other. Instead, both are connected via

$$\overleftrightarrow{\mu}_{\mathrm{r}} = \overleftrightarrow{P}_{\mathrm{L}} - (\overleftrightarrow{\varepsilon}_{\mathrm{r}})^{-1}_{RR} \tag{2.84}$$

In the latter two equations, σ_{RR}, $\widetilde{\sigma}_{RR}$ and ε^{-1}_{RR} have been defined similar to χ_{RR} in Eq. (2.64).

In Ref. [6], it is systematically shown that the universal response relations Eqs. (2.58) to (2.61) and the ones introduced in this section reduce in suitable limiting cases to standard relations which are well known and frequently employed in theoretical materials science. By contrast, given any response function in such a limiting case, it is in general not possible to derive arbitrary other ones. For instance, the frequency dependent dielectric function $\varepsilon_{\mathrm{r,L}}(\omega)$ is not even close to enough to construct the conductivity tensor. In fact, with the mere longitudinal part it is not even sufficient to construct its own tensor. This is due to the fact that the entire functional information w. r. t. the arguments $\boldsymbol{k}$ *and* ω of the full *tensor-valued* function is required for the universal response relation to be valid. However, in the isotropic limit discussed in Sct. 2.5 this is relaxed to the level of longitudinal and transverse parts of the respective response functions.

Considering the response relations given in this section, it is clear that a proper tensor cannot be simply replaced by its direct counterpart and vice versa. For example the proper current response is related to the dielectric tensor "only" by a (frequency- and wavevector-dependent) prefactor and a trivial constant, whereas its direct version is connected to the *inverse* dielectric tensor by a comparable relation. Evidently, the dielectric tensor is in general (not even approximately) equal to its inverse. Consequently, there is a huge difference in identitifying a, say, empirically modelled response function as being proper or direct. Especially regarding response theory in ab initio materials physics, direct and proper response functions are usually referred to as "reducible" and "irreducible" (or screened) in analogy to the self-energy $\Sigma(\boldsymbol{k},\omega)$ [32, §5.2.2]. Both terms originate in fact in the Feynman graph representation of Green function theory.

In this context the random phase approximation (RPA) plays an important rôle which essentially states that a response function calculated in a non-interacting approximation like the Hartree theory has to be identified with a *proper* function from fundamental theory (eventhough it is the functional derivative w. r. t. an *external* field). In textbooks, this is usually discussed on the example of the density response

$$\widetilde{\chi} \overset{(2.54)}{=} \frac{1}{c^2}\,\widetilde{\chi}^0{}_0 \overset{\mathrm{def}}{=} \frac{\delta\rho_{\mathrm{ind}}}{\delta\varphi_{\mathrm{tot}}}\,, \tag{2.85}$$

in terms of which Hedin's equation for the screened potential w in the so-called GW-approximation be derived,

$$w = v + v\,\widetilde{\chi} w\,. \tag{2.86}$$

Here, $\widetilde{\chi}$ is also known as irreducible polarization (see [6, §5.2] for a more detailed explanation)

and v denotes the Coulomb kernel which reads in Fourier space

$$v(\boldsymbol{k},\omega) \equiv v(\boldsymbol{k}) = \frac{1}{\varepsilon_0 |\boldsymbol{k}|^2}\,, \tag{2.87}$$

and translates in real space to the instantaneous interaction

$$v(\boldsymbol{x}-\boldsymbol{x}', t-t') = \frac{1}{4\pi\varepsilon_0}\,\frac{\delta(ct-ct')}{|\boldsymbol{x}-\boldsymbol{x}'|}\,. \tag{2.88}$$

We remark however that by the universal response relations the identification of ab initio screened response functions with proper fundamental functions applies to the other responses as well.

2.5. Isotropic and combined limits

Isotropic limit.—A most essential limit which is regularly assumed next to temporal and spatial homogeneity is isotropy. On the level of response tensors this premise is reflected in the ability to deconstruct three-tensors in terms of their corresponding *scalar* longitudinal and transverse functions (cf. [22, §1.6.1] or [32, Eq. (3.173)]),

$$\overleftrightarrow{\chi}(\boldsymbol{k},\omega) = \chi_{\mathrm{L}}(\boldsymbol{k},\omega)\overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{k}) + \chi_{\mathrm{T}}(\boldsymbol{k},\omega)\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.89}$$

such that e. g. the current induced by a purely longitudinal vector potential is purely longitudinal itself and analogously for purely transverse potentials. By comparing this expression with the full decomposition of a spatial response tensor,

$$\overleftrightarrow{\chi} = \overleftrightarrow{\mathbb{1}}\overleftrightarrow{\chi}\overleftrightarrow{\mathbb{1}} = \overleftrightarrow{P}_{\mathrm{L}}\overleftrightarrow{\chi}\overleftrightarrow{P}_{\mathrm{L}} + \overleftrightarrow{P}_{\mathrm{L}}\overleftrightarrow{\chi}\overleftrightarrow{P}_{\mathrm{T}} + \overleftrightarrow{P}_{\mathrm{T}}\overleftrightarrow{\chi}\overleftrightarrow{P}_{\mathrm{L}} + \overleftrightarrow{P}_{\mathrm{T}}\overleftrightarrow{\chi}\overleftrightarrow{P}_{\mathrm{T}}\,, \tag{2.90}$$

which is the tensorial version of Eq. (B.171) valid for field quantities, it is obvious that the isotropic limit indeed represents a drastic approximation to the general case. Simultaneously, it allows for a reduction of all tensorial response relations exclusively in terms of their scalar longitudinal and transverse parts. Since all relations in the isotropic limit can be easily derived with help of the projector formalism explained in App. B.5, we will plainly state the most important ones in the following. A more detailed discussion of this limit is presented in [8, App. D1].

We start with the Dysonian equation (2.53) for the current response. After some algebra, this tensorial equation reduces to

$$\chi_{\mathrm{L}}(\boldsymbol{k},\omega) = \widetilde{\chi}_{\mathrm{L}}(\boldsymbol{k},\omega) + \widetilde{\chi}_{\mathrm{L}}(\boldsymbol{k},\omega)\left(-\frac{1}{\varepsilon_0\omega^2}\right)\chi_{\mathrm{L}}(\boldsymbol{k},\omega)\,, \tag{2.91}$$

$$\chi_{\mathrm{T}}(\boldsymbol{k},\omega) = \widetilde{\chi}_{\mathrm{T}}(\boldsymbol{k},\omega) + \widetilde{\chi}_{\mathrm{T}}(\boldsymbol{k},\omega)\,\mathbb{D}_0(\boldsymbol{k},\omega)\,\chi_{\mathrm{T}}(\boldsymbol{k},\omega)\,, \tag{2.92}$$

for the respective scalar parts. By Eq. (2.85), which in the homogeneous and isotropic limit

simplifies to

$$\widetilde{\chi}(\boldsymbol{k},\omega) \overset{(2.46)}{=} -\frac{c^2}{\omega^2}\,\boldsymbol{k}^{\mathsf{T}}\overset{\leftrightarrow}{\widetilde{\chi}}(\boldsymbol{k},\omega)\,\boldsymbol{k} = -\frac{|\boldsymbol{k}|^2}{\omega^2}\widetilde{\chi}_{\mathrm{L}}(\boldsymbol{k},\omega)\,, \tag{2.93}$$

and similarly holds for the direct versions of the involved response functions, the equation for the longitudinal current response can be converted into a corresponding relation for the density response,

$$\chi(\boldsymbol{k},\omega) = \widetilde{\chi}(\boldsymbol{k},\omega) + \widetilde{\chi}(\boldsymbol{k},\omega)\,v(\boldsymbol{k})\chi(\boldsymbol{k},\omega)\,. \tag{2.94}$$

Further, all these Dyson-like equations may be rewritten into the equivalent "kernel form"

$$\widetilde{\chi}^{-1} = \chi^{-1} + v\,, \tag{2.95}$$

and similar for Eqs. (2.91) and (2.92) with v replaced by $(-\varepsilon_0\omega^2)^{-1}$ or $\mathbb{D}_0$, respectively. With Eqs. (2.92) and (2.94), the universal response relations between fundamental and dielectric tensor (2.79) and (2.78) then translate to

$$\varepsilon_{\mathrm{r,L}}^{-1}(\boldsymbol{k},\omega) = 1 + v(\boldsymbol{k})\chi(\boldsymbol{k},\omega)\,, \tag{2.96}$$

$$\varepsilon_{\mathrm{r,L}}(\boldsymbol{k},\omega) = 1 - v(\boldsymbol{k})\widetilde{\chi}(\boldsymbol{k},\omega)\,, \tag{2.97}$$

for the longitudinal, and

$$\varepsilon_{\mathrm{r,T}}^{-1}(\boldsymbol{k},\omega) = 1 + \mathbb{D}_0(\boldsymbol{k},\omega)\,\chi_{\mathrm{T}}(\boldsymbol{k},\omega)\,, \tag{2.98}$$

$$\varepsilon_{\mathrm{r,T}}(\boldsymbol{k},\omega) = 1 - \mathbb{D}_0(\boldsymbol{k},\omega)\,\widetilde{\chi}_{\mathrm{T}}(\boldsymbol{k},\omega)\,, \tag{2.99}$$

for the transverse part. These are standard relations and at least the first is well known in electronic structure theory. Decomposing the general relation between the magnetic susceptibility and the dielectric tensor (2.84) in the same way yields

$$\mu_{\mathrm{r,L}}(\boldsymbol{k},\omega) \equiv 1\,. \tag{2.100}$$

This is not surprising since according to Eq. (1.19), magnetic fields have no longitudinal component in the first place such that Eq. (2.100) is trivially fulfilled. Hence, we may always identify the magnetic permeability with its transverse part, $\mu_{\mathrm{r}} \equiv \mu_{\mathrm{r,T}}$, and thus Eq. (2.84) actually implies

$$\mu_{\mathrm{r}}(\boldsymbol{k},\omega)\,\varepsilon_{\mathrm{r,T}}(\boldsymbol{k},\omega) \equiv 1 \tag{2.101}$$

in the isotropic limit. As shown in Ref. [10, §3.2.4], this result can be derived as well directly from Faraday's law for (purely transverse) light waves. Combining Eq. (2.101) with Eq. (2.77) and Eq. (2.78) yields an expression for the the magnetic susceptibility in the isotropic limit,

$$\overset{\leftrightarrow}{\chi}_{\mathrm{m}}(\boldsymbol{k},\omega) = \mathbb{D}_0(\boldsymbol{k},\omega)\,\chi_{\mathrm{T}}(\boldsymbol{k},\omega)\,\overset{\leftrightarrow}{P}_{\mathrm{T}}(\boldsymbol{k}) = \chi_{\mathrm{m}}(\boldsymbol{k},\omega)\,\overset{\leftrightarrow}{P}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.102}$$

which is via the Dysonian relation (2.92) valid for the respective proper response functions as well. Alternatively, Eq. (2.102) can be derived from the universal response relation Eq. (2.61) (or even more directly from Eq. (2.63)). In fact, it can be shown that the entire transverse

electromagnetic response can be expressed solely in terms of the transverse current response [10, §3.2.3]. In particular we obtain the relations

$$\chi_{EE,\mathrm{T}}(\boldsymbol{k},\omega) = \chi_{BB,\mathrm{T}}(\boldsymbol{k},\omega) = \chi_{\mathrm{m}}(\boldsymbol{k},\omega) = \mathbb{D}_0(\boldsymbol{k},\omega)\,\chi_{\mathrm{T}}(\boldsymbol{k},\omega)\,. \tag{2.103}$$

which we will refer to again in a moment.

Optical limit.—On top of the isotropic limit there are two further limits that have to be named already for their experimental significance. Firstly, the *dynamical long-wavelength* or *optical limit*, which is defined by the condition $|\boldsymbol{k}| \to 0$. Here, the material appears all the more homogeneous as assumed already in the homogeneous limit, and for even smaller wavevectors the longitudinal and transverse parts of a response function roughly fulfill

$$\chi_{\mathrm{L}}(\boldsymbol{k},\omega) \approx \chi_{\mathrm{T}}(\boldsymbol{k},\omega)\,. \tag{2.104}$$

The justification for this repeatedly drastic assumption is that for $|\boldsymbol{k}| \to 0$, the direction of the wavevector is not defined anymore. Consequently, a distinction between the components of a response function parallel and orthognal to the wavevector $\boldsymbol{k}$ is pointless. In literature, the explicit dependence on the wavevector is usually omitted. More often than not, response functions then have to be interpreted in the long-wavelength limit in the sense of

$$\lim_{|\boldsymbol{k}|\to 0} \varepsilon_{\mathrm{r,L}}(\boldsymbol{k},\omega) = \varepsilon(\omega)\,. \tag{2.105}$$

Further, in this limit the electric solution operator approaches identity,

$$\lim_{|\boldsymbol{k}|\to 0} \overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}}\,, \tag{2.106}$$

and thereby simplifies the general relations between the dielectric tensor and (optical) conductivity (2.78) and (2.79) to the well known standard relations

$$(\overleftrightarrow{\varepsilon}_{\mathrm{r}})^{-1}(\boldsymbol{k},\omega) \approx \overleftrightarrow{\mathbb{1}} + \frac{1}{\mathrm{i}\omega\,\varepsilon_0}\,\overleftrightarrow{\sigma}(\boldsymbol{k},\omega)\,, \tag{2.107}$$

$$\overleftrightarrow{\varepsilon}_{\mathrm{r}}(\boldsymbol{k},\omega) \approx \overleftrightarrow{\mathbb{1}} - \frac{1}{\mathrm{i}\omega\,\varepsilon_0}\,\overleftrightarrow{\widetilde{\sigma}}(\boldsymbol{k},\omega)\,, \tag{2.108}$$

which are commonly used and implemented in electronic structure physics codes (e. g. [31]).

At this point we emphasise again that not all response relations can be applied in this limit because the entire information in the sense of full wavevector- and frequency-dependence of a response function is required in general.

Static limit.—Another quite important case is the static limit, $\omega \to 0$, where the response of the medium to an external perturbation is instantaneous and relates the static parts of the respective field quantities. By Faraday's law, static electric fields are purely longitudinal, i. e. they are generated exclusively by static charge densities. On the other hand, magnetic fields are always transverse by Gauß' law. Consequently, in the isotropic and static limit

any cross-coupling between electric and magnetic fields should vanish. This can be explicitly shown with the universal response relations Eqs. (2.58) to (2.61), which simplify under these conditions to

$$\chi_{EE}(\boldsymbol{k},0) \equiv \chi_{EE,\mathrm{L}}(\boldsymbol{k}) = v(\boldsymbol{k})\chi(\boldsymbol{k})\,, \tag{2.109}$$

$$\chi_{BB}(\boldsymbol{k},0) \equiv \chi_{BB,\mathrm{T}}(\boldsymbol{k}) = \frac{1}{c^2}\,v(\boldsymbol{k})\,\chi_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.110}$$

and

$$\overleftrightarrow{\chi}_{EB}(\boldsymbol{k},0) = \overleftrightarrow{\chi}_{BE}(\boldsymbol{k},0) \equiv 0\,. \tag{2.111}$$

Thus, in the static limit the medium is effectively described by only two independent scalar response functions relating induced electric fields to external ones and similarly for magnetic fields. Furthermore, for $\omega \to 0$ the formula for the isotropic magnetic susceptibility, Eq. (2.102), simplifies to

$$\chi_{\mathrm{m}}(\boldsymbol{k}) = \mu_0\,\frac{\chi_{\mathrm{T}}(\boldsymbol{k})}{|\boldsymbol{k}|^2}\,, \tag{2.112}$$

where $\chi_{\mathrm{T}}(\boldsymbol{k}) = \lim_{\omega\to 0}\chi_{\mathrm{T}}(\boldsymbol{k},\omega)$ is the transverse current response in the optical limit. This is yet again an important standard relation in electronic structure physics (cf. [32, Eq. (3.183)]) which holds likewise for the respective proper functions.

Combined limits—Both, the long-wavelength and the static limit can even be combined such that isotropic response functions may be reduced to a single scalar number. In case of the permittivity, this is known as the dielectric constant defined by

$$\lim_{\omega\to 0}\varepsilon_{\mathrm{r}}(\omega) \equiv \varepsilon_{\mathrm{r}}\,. \tag{2.113}$$

Despite the fact, that such a material constant can never contain all the relevant information a full response tensor comprises, it is common practice to report this number when performing ab initio calculations or even spectroscopic measurements of a material (cf. e. g. Ref. [36]). In fact, the importance of the dielectric constant originates from simple capacitor experiments, in which a dielectric is placed between two conducting electrodes and a voltage is applied. The permittivity for an idealized plate capacitor is then given by

$$C = \varepsilon_0\varepsilon_{\mathrm{r}}\,\frac{A}{d}\,, \tag{2.114}$$

where A is the area of each of the plate's electrodes, d is the distance between the electrodes and C is the capacitance. For real materials in real experimental situations, this approximation is of course not adequate. However, the combined static and optical limit has another entirely different meaning on the conceptional side. In general, the order in which these two limits are taken affects the resulting "response number", i. e. in general

$$\lim_{|\boldsymbol{k}|\to 0}\lim_{\omega\to 0}\chi(\boldsymbol{k},\omega) \neq \lim_{\omega\to 0}\lim_{|\boldsymbol{k}|\to 0}\chi(\boldsymbol{k},\omega)\,. \tag{2.115}$$

Even worse, the direction along the limit $|\boldsymbol{k}| \to 0$ is taken comes into play as well. Conse-

quently, the resulting number depends on the direction in the four-dimensional hyperplane spanned by the arguments $\boldsymbol{k}$ and ω. This is particularly important when a response function has to be evaluated along a dispersion relation like $\omega = c|\boldsymbol{k}|$ as is the case for the dielectric tensor as explained in Sct. 2.7.

Further empirical limiting cases are thoroughly discussed in [6, §7].

2.6. Full Green function

As already mentioned before, all equations given in Chpt. 1 are valid for induced, external and total fields separately, as long as they are given as functionals of *their own sources*. Hence, we may use the wave equation given by Eq. (1.9) identically for $A^\mu \equiv A^\mu_{\text{tot}}$ and its corresponding source $j_{\text{tot}} = j_{\text{ind}} + j_{\text{ext}}$,

$$\left(\eta^\mu{}_\nu \,\Box + \partial^\mu \partial_\nu\right) A^\nu = \mu_0 \left(j^\mu_{\text{ind}} + j^\mu_{\text{ext}}\right) . \tag{2.116}$$

The essential strategy to transform this equation into a (homogeneous) wave equation in a medium is as follows: After separating external and induced four-current, the latter has to be reinterpreted as linear response of the material in the sense of Eq. (2.48) and eliminated in favour of the corresponding response tensor. By setting the external sources to zero, this would yield indeed a homogeneous wave equation for the (total) four-potential. However, we will retain the external current for now and continue with

$$\left(\eta^\mu{}_\nu \,\Box + \partial^\mu \partial_\nu - \mu_0 \widetilde{\chi}^\mu{}_\nu\right) A^\nu = \mu_0 j^\mu_{\text{ext}} \,, \tag{2.117}$$

which is the general microscopic and manifestly Lorentz-covariant wave equation for the electromagnetic four-potential in materials and well-known in plasma physics (cf. [22, §2.1.1]). The concrete material is then considered exclusively through the (proper) fundamental response tensor $\widetilde{\chi}$, whereas the external current could in principle be prescribed arbitrarily. At this point it is already obvious that a vanishing fundamental response tensor, $\chi^\mu{}_\nu \equiv \widetilde{\chi}^\mu{}_\nu \equiv 0$, exactly corresponds to the vacuum case described by the inhomogeneous wave equation (1.9). Correspondingly, induced fields cannot be vacuum fields because they are generated by the induced charge and current densities.

In temporal gauge, Eq. (2.117) can again be "reduced" to an equation only in terms of the vector potential,

$$\left(-\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2 - \mu_0 \left(\overleftrightarrow{\mathbb{1}} - \frac{c^2|\boldsymbol{k}|^2}{\omega^2} \overleftrightarrow{P}_{\text{L}}(\boldsymbol{k})\right) \overleftrightarrow{\widetilde{\chi}}(\boldsymbol{k},\omega)\right) \boldsymbol{A}(\boldsymbol{k},\omega) = \mu_0 \left(\overleftrightarrow{\mathbb{1}} - \frac{c^2|\boldsymbol{k}|^2}{\omega^2} \overleftrightarrow{P}_{\text{L}}(\boldsymbol{k})\right) \boldsymbol{j}_{\text{ext}}(\boldsymbol{k},\omega) \,, \tag{2.118}$$

following the same argumentation as in Sct. 1.2. For other gauges see Ref. [10, §4.1.2]. Alternatively, one could have started from Eq. (1.34), which is equivalently to

$$\left(-\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2\right) \boldsymbol{A}(\boldsymbol{k},\omega) = \mu_0 \left(\overleftrightarrow{\mathbb{1}} - \frac{c^2|\boldsymbol{k}|^2}{\omega^2} \overleftrightarrow{P}_{\text{L}}(\boldsymbol{k})\right) \boldsymbol{j}(\boldsymbol{k},\omega) \,, \tag{2.119}$$

and eliminate the spatial current with

$$\boldsymbol{j}(\boldsymbol{k},\omega) \equiv \boldsymbol{j}_{\mathrm{tot}}(\boldsymbol{k},\omega) = \overleftrightarrow{\widetilde{\chi}}(\boldsymbol{k},\omega)\,\boldsymbol{A}(\boldsymbol{k},\omega) + \boldsymbol{j}_{\mathrm{ext}}(\boldsymbol{k},\omega)\,, \tag{2.120}$$

to obtain the same result. Factoring out $-\omega^2/c^2 + |\boldsymbol{k}|^2$ in Eq. (2.118) and multiplying with its inverse yields

$$\left(\overleftrightarrow{\mathbb{1}} - \overleftrightarrow{D}_0(\boldsymbol{k},\omega)\,\overleftrightarrow{\widetilde{\chi}}(\boldsymbol{k},\omega)\right)\boldsymbol{A}(\boldsymbol{k},\omega) = \overleftrightarrow{D}_0(\boldsymbol{k},\omega)\,\boldsymbol{j}_{\mathrm{ext}}(\boldsymbol{k},\omega)\,, \tag{2.121}$$

and by Eq. (2.79) reduces even further to

$$(\overleftrightarrow{D}_0)^{-1}(\boldsymbol{k},\omega)\,\overleftrightarrow{\varepsilon}_{\mathrm{r}}(\boldsymbol{k},\omega)\,\boldsymbol{A}(\boldsymbol{k},\omega) = \boldsymbol{j}_{\mathrm{ext}}(\boldsymbol{k},\omega)\,. \tag{2.122}$$

Analogously to the (Cartesian) free Green function in Eq. (1.45), we now define the *full electromagnetic Green function* by

$$\overleftrightarrow{D}(\boldsymbol{k},\omega) \overset{\mathrm{def}}{=} \frac{\mathrm{d}\boldsymbol{A}_{\mathrm{tot}}(\boldsymbol{k},\omega)}{\mathrm{d}\boldsymbol{j}_{\mathrm{ext}}(\boldsymbol{k},\omega)}\,. \tag{2.123}$$

This response function is also known as "photon propagator" in plasma physics [22, §2.1.3]. Applying the functional chain rule multiple times to the right hand side of Eq. (2.123),

$$\frac{\mathrm{d}\boldsymbol{A}_{\mathrm{tot}}}{\mathrm{d}\boldsymbol{j}_{\mathrm{ext}}} = \frac{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{j}_{\mathrm{ext}}} + \frac{\mathrm{d}\boldsymbol{A}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}\left(\frac{\mathrm{d}\boldsymbol{j}_{\mathrm{ind}}}{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}}\right)\frac{\mathrm{d}\boldsymbol{A}_{\mathrm{ext}}}{\mathrm{d}\boldsymbol{j}_{\mathrm{ext}}}\,, \tag{2.124}$$

reveals that the free Green function is related to its direct counterpart via the Dyson equation

$$\overleftrightarrow{D} = \overleftrightarrow{D}_0 + \overleftrightarrow{D}_0\overleftrightarrow{\chi}\overleftrightarrow{D}_0\,, \tag{2.125}$$

Similarly, by replacing $\boldsymbol{A}_{\mathrm{ext}}$ with $\boldsymbol{A}_{\mathrm{tot}}$ in the last two fractions of Eq. (2.124), an equivalent Dyson equation in terms of the proper fundamental response tensor can be derived,

$$\overleftrightarrow{D} = \overleftrightarrow{D}_0 + \overleftrightarrow{D}_0\overleftrightarrow{\widetilde{\chi}}\overleftrightarrow{D}\,. \tag{2.126}$$

This is consistent to inserting Eq. (2.53) into Eq. (2.125) and alternatively can be directly read-off from Eq. (2.121). The relation between the free and full Green function can further be expressed in the "kernel form" (2.95),

$$(\overleftrightarrow{D})^{-1} = (\overleftrightarrow{D}_0)^{-1} - \overleftrightarrow{\widetilde{\chi}}\,, \tag{2.127}$$

or in terms of the dielectric tensor,

$$(\overleftrightarrow{D})^{-1} = (\overleftrightarrow{D}_0)^{-1}\overleftrightarrow{\varepsilon}_{\mathrm{r}}\,, \tag{2.128}$$

where Eq. (2.79) has been used. The concise latter equation impressively proves once more the conceptual importance of the permittivity tensor, even on the level of electromagnetic

Green function. Similar Dyson relations hold for the full Minkowskian Green function, which in temporal gauge is given by

$$D^{\mu}{}_{\nu}(\boldsymbol{k},\omega) = \begin{pmatrix} 0 & \mathbf{0}^{\mathsf{T}} \\ \mathbf{0} & \overleftrightarrow{D}(\boldsymbol{k},\omega) \end{pmatrix} , \tag{2.129}$$

as shown in [11, Eq. (3.28)].

2.7. Wave equations in media and dispersion relations

When discussing linear response as a theoretical description of spectroscopic experiments, i. e. experiments where externally controlled *transverse* light waves are radiated in direction of a material sample, then it is only natural to set the external sources to zero. Yet there are situations where it is desirable to include such sources, for example when the effect of impurities should be described or charged particles are moving through the medium. Such cases are however not considered in this thesis.

Following the assumption of vanishing external sources, $j^{\mu}_{\mathrm{ext}} \equiv 0$, the general wave equation in media for the electric field can now be obtained by multiplying Eq. (2.122) with the factor $\mathrm{i}\omega$,

$$(\overleftrightarrow{D}_0)^{-1}(\boldsymbol{k},\omega)\,\overleftrightarrow{\varepsilon}_{\mathrm{r}}(\boldsymbol{k},\omega)\,\boldsymbol{E}(\boldsymbol{k},\omega) = 0\,. \tag{2.130}$$

Setting $\varepsilon_{\mathrm{r}} \equiv 1$, the free wave equation for the vacuum case is recovered,

$$\left(-\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2\right) \boldsymbol{E}_{\mathrm{T}}(\boldsymbol{k},\omega) = 0\,, \tag{2.131}$$

$$\boldsymbol{E}_{\mathrm{L}}(\boldsymbol{k},\omega) \equiv 0\,. \tag{2.132}$$

In this case, there are no longitudinal oscillations because there simply is no material which could oscillate. The transverse field, on the other hand, unsurprisingly reproduces the free dispersion relation for light waves,

$$\omega_{\boldsymbol{k},\mathrm{T}} = c|\boldsymbol{k}|\,. \tag{2.133}$$

By identity (2.79), this case corresponds to a completely vanishing current response, which in turn agrees with what we discovered earlier in context of the covariant wave equation (2.117). After excluding the free dispersion on the other hand, Eq. (2.134) results in the concise condition

$$\overleftrightarrow{\varepsilon}_{\mathrm{r}}(\boldsymbol{k},\omega)\boldsymbol{E}(\boldsymbol{k},\omega) = 0\,, \tag{2.134}$$

which alternatively could have been derived from Eq. (1.52) by eliminating the induced current via proper Ohm's law. From a fundamental point of view, this is one of the most important relations in linear response theory, because it states that every non-trivial (total) electric field in a material is restricted to the null space of the dielectric tensor. As wave equation, this insight can at least be traced back to Dolgov and Maksimov [26, Eq. (2.34)]. A more paradigmatical discussion is given in Ref. [10]. There, especially the equivalent statement that the external field does not penetrate the material is elaborated. Put differently, electromagnetic

waves in a material correspond to the latter's *proper oscillation*. Remarkably, Eq. (2.134) already includes all effects of anisotropy, relativistic retardation and magneto-electric cross coupling, at least in the context of linear response theory. Moreover, it is not only valid for dielectrics but, quite to the contrary, applies to all materials from metal to insulator while all material-specific information is fully contained within the dielectric tensor or, equivalently by Eq. (2.79), within the current response and thus the fundamental response tensor.

A reasonable question is now how this apparently different but alleged fundamental wave equation can be justified next to the regularly employed wave equations from e. g. theoretical optics. In order to answer this, we first recall that the dielectric tensor is related to the conductivity tensor via Eq. (2.76), which reads in its full form

$$\overleftrightarrow{\varepsilon}_{\mathrm{r}}(\boldsymbol{k},\omega) = \overleftrightarrow{\mathbb{1}} - \frac{1}{\mathrm{i}\omega\varepsilon_0}\,\overleftrightarrow{\mathbb{E}}(\boldsymbol{k},\omega)\,\overleftrightarrow{\widetilde{\sigma}}(\boldsymbol{k},\omega)\,. \tag{2.135}$$

Inserting this into Eq. (2.134) yields

$$(\overleftrightarrow{\mathbb{E}})^{-1}(\boldsymbol{k},\omega)\boldsymbol{E}(\boldsymbol{k},\omega) = \frac{1}{\mathrm{i}\omega\varepsilon_0}\,\overleftrightarrow{\widetilde{\sigma}}(\boldsymbol{k},\omega)\,\boldsymbol{E}(\boldsymbol{k},\omega)\,, \tag{2.136}$$

and by inverting the electric solution generator in form of Eq. (1.54) using the inversion rule from Eq. (B.177), the latter equation can further be recast into

$$\left(-\frac{\omega^2}{c^2}\left(\overleftrightarrow{\mathbb{1}} - \frac{\overleftrightarrow{\widetilde{\sigma}}(\boldsymbol{k},\omega)}{\mathrm{i}\omega\varepsilon_0}\right) + |\boldsymbol{k}|^2\,\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\right)\boldsymbol{E}(\boldsymbol{k},\omega) = 0\,. \tag{2.137}$$

In Sct. 1.3 we already showed that the electric solution generator approaches identity in the optical limit. The general relation between dielectric tensor and (optical) conductivity is then approximately given by Eqs. (2.107) and (2.108). Apparently, the term within the inner parentheses in Eq. (2.137) can be identified with the latter. Therefore and for later reference, we *define* the *effective dielectric tensor* accordingly as

$$\overleftrightarrow{\varepsilon}_{\mathrm{eff}}(\boldsymbol{k},\omega) \overset{\mathrm{def}}{=} \overleftrightarrow{\mathbb{1}} - \frac{1}{\mathrm{i}\omega\varepsilon_0}\,\overleftrightarrow{\widetilde{\sigma}}(\boldsymbol{k},\omega)\,. \tag{2.138}$$

Additionally inserting the following vector identity (cf. App. B.5),

$$\boldsymbol{k}\times(\boldsymbol{k}\times\boldsymbol{E}(\boldsymbol{k},\omega)) = -|\boldsymbol{k}|^2\,\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\,\boldsymbol{E}(\boldsymbol{k},\omega)\,, \tag{2.139}$$

Eq. (2.137) can be brought into the equivalent form

$$-\frac{\omega^2}{c^2}\,\overleftrightarrow{\varepsilon}_{\mathrm{eff}}(\boldsymbol{k},\omega)\,\boldsymbol{E}(\boldsymbol{k},\omega) = \boldsymbol{k}\times(\boldsymbol{k}\times\boldsymbol{E}(\boldsymbol{k},\omega))\,, \tag{2.140}$$

which formally agrees with the standard wave equation used in theoretical optics and solid state physics (cf. [37, Eq. (4.11)] and [38, Eq. (1)]). As discussed in more detail in our publication [1], we observe here an astonishing "error cancellation": The *phenomenological* wave equation (2.140) combined with the *approximate* dielectric tensor in the optical limit

leads *precisely* to the *fundamental* wave equation (2.134).

In the isotropic limit where longitudinal and transverse degrees of freedom fully decouple, Eq. (2.134) splits into

$$\varepsilon_{\mathrm{r,L}}(\boldsymbol{k},\omega)\,\boldsymbol{E}_{\mathrm{L}}(\boldsymbol{k},\omega) = 0\,, \tag{2.141}$$

$$\varepsilon_{\mathrm{r,T}}(\boldsymbol{k},\omega)\,\boldsymbol{E}_{\mathrm{T}}(\boldsymbol{k},\omega) = 0\,. \tag{2.142}$$

The respective dielectric functions are given by Eqs. (2.97) and (2.99) and can be further recast in terms of the longitudinal and transverse conductivity to

$$\varepsilon_{\mathrm{r,L}}(\boldsymbol{k},\omega) = 1 - \frac{1}{\mathrm{i}\omega\,\varepsilon_0}\,\widetilde{\sigma}_{\mathrm{L}}(\boldsymbol{k},\omega)\,, \tag{2.143}$$

$$\varepsilon_{\mathrm{r,T}}(\boldsymbol{k},\omega) = 1 - \frac{\omega^2}{\omega^2 - c^2|\boldsymbol{k}|^2}\,\frac{1}{\mathrm{i}\omega\,\varepsilon_0}\,\widetilde{\sigma}_{\mathrm{T}}(\boldsymbol{k},\omega)\,. \tag{2.144}$$

Apparently, the longitudinal part *exactly* agrees with the definition of $\varepsilon_{\mathrm{eff,L}}$ which can be directly read-off from Eq. (2.135). In the transverse part by contrast, we first have to multiply both sides with the free dispersion relation (which we already excluded as non-zero) factor out the large fraction in front of the conductivity to find

$$\left(\omega^2 - c^2|\boldsymbol{k}|^2\right)\varepsilon_{\mathrm{T}}(\boldsymbol{k},\omega) = \omega^2\left(1 - \frac{1}{\mathrm{i}\omega\,\varepsilon_0}\,\widetilde{\sigma}_{\mathrm{T}}(\boldsymbol{k},\omega)\right) - c^2|\boldsymbol{k}|^2\,. \tag{2.145}$$

Because we excluded the free dispersion relation already in the beginning, inserting this relation into the condition Eq. (2.142) yields together with the longitudinal part the alternative two wave equations in terms of the *effective* dielectric functions,

$$\varepsilon_{\mathrm{eff,L}}(\boldsymbol{k},\omega)\,\boldsymbol{E}_{\mathrm{L}}(\boldsymbol{k},\omega) = 0\,, \tag{2.146}$$

$$\left(-\frac{\omega^2}{c^2}\,\varepsilon_{\mathrm{eff,T}}(\boldsymbol{k},\omega) + |\boldsymbol{k}|^2\right)\boldsymbol{E}_{\mathrm{T}}(\boldsymbol{k},\omega) = 0\,, \tag{2.147}$$

Both are fully equivalent to Eqs. (2.141) and (2.142) provided the correct identifications from Eqs. (2.143) and (2.144) are used.

This discussion in particular shows how it is possible that in ab initio physics two entirely different wave equations are used, both formulated in terms of the dielectric function: Firstly the phenomenological wave equation Eq. (2.140), which is used for optical properties and secondly the plasmon equation (2.141), which is used for longitudinal normal oscillations. Eventually, both wave equations turn out to be of the same type and especially if the isotropic limit cannot be applied, one has to work with the coupled fundamental equation (2.134).

In fact, there is yet another wave equation frequently found in textbooks on transport phenomena (e. g. [39, Eq. (2.206)]),

$$\left(-\frac{\omega^2}{c^2}\,\varepsilon_{\mathrm{L}}(\boldsymbol{k},\omega) + |\boldsymbol{k}|^2\right)\boldsymbol{E}_{\mathrm{T}}(\boldsymbol{k},\omega) = 0\,, \tag{2.148}$$

which clearly differs from Eq. (2.147). This *transverse* wave equation is in so far special, as it is formulated in terms of the *longitudinal* dielectric function. Nevertheless, also this wave equation is compatible with Eq. (2.142), provided one assumes that $\widetilde{\sigma}_{\mathrm{L}}(\boldsymbol{k},\omega) = \widetilde{\sigma}_{\mathrm{T}}(\boldsymbol{k},\omega)$, which empirically seems to be the case for many materials at optical wavelengths. Then by Eqs. (2.143) and (2.144) one can show the identity

$$\left(-\frac{\omega^2}{c^2}\,\varepsilon_{\mathrm{L}}(\boldsymbol{k},\omega) + |\boldsymbol{k}|^2\right) = \left(-\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2\right)\varepsilon_{\mathrm{T}}(\boldsymbol{k},\omega)\,, \tag{2.149}$$

and by excluding the free dispersion relation again, Eq. (2.142) is retained eventually.

From Eqs. (2.141) and (2.142) it is now clear, that non-trivial solutions for the electric field have to be obtained by setting the respective dielectric functions to zero,

$$\varepsilon_{\mathrm{r,L}}(\boldsymbol{k},\omega_{\boldsymbol{k},\mathrm{L}}) = 0\,, \tag{2.150}$$

$$\varepsilon_{\mathrm{r,T}}(\boldsymbol{k},\omega_{\boldsymbol{k},\mathrm{T}}) = 0\,. \tag{2.151}$$

In the transverse case, this translates to the implicit expression

$$\omega_{\boldsymbol{k},\mathrm{T}}^2 = c^2|\boldsymbol{k}|^2 - \frac{1}{\varepsilon_0}\,\widetilde{\chi}_{\mathrm{T}}(\boldsymbol{k},\omega_{\boldsymbol{k},\mathrm{T}})\,, \tag{2.152}$$

or in terms of the effective dielectric function,

$$\omega_{\boldsymbol{k},\mathrm{T}}^2 = \frac{c^2|\boldsymbol{k}|^2}{\varepsilon_{\mathrm{eff,T}}(\boldsymbol{k},\omega_{\boldsymbol{k},\mathrm{T}})}\,. \tag{2.153}$$

By contrast, condition (2.150) for the longitudinal field is equivalent to

$$\omega_{\boldsymbol{k},\mathrm{L}}^2 = -\frac{1}{\varepsilon_0}\,\widetilde{\chi}_{\mathrm{L}}(\boldsymbol{k},\omega_{\boldsymbol{k},\mathrm{L}})\,. \tag{2.154}$$

Apparently, inserting conditions (2.150) and (2.151) into the full electromagnetic Green function in the isotropic limit,

$$\mu_0\,\overleftrightarrow{D}^{-1}(\boldsymbol{k},\omega_{\boldsymbol{k}}) = -c^2\omega_{\boldsymbol{k}}^2\,\varepsilon_{\mathrm{r,L}}(\boldsymbol{k},\omega_{\boldsymbol{k}})\,\overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{k}) + \left(-\frac{\omega_{\boldsymbol{k}}^2}{c^2} + |\boldsymbol{k}|^2\right)\varepsilon_{\mathrm{r,T}}(\boldsymbol{k},\omega_{\boldsymbol{k}})\,\overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{2.155}$$

pinpoint the latter's singularities, just like the poles of the free Green function are given by the free dispersion relation. This connection between poles of Green functions and excitation energies is in fact a fundamental concept in field theory.

Part II.

Application to the free electron gas

3. Fundamental response tensor

In Chpt. 2 our Central Claim has been shown, which states that, as a matter of principle, all linear electromagnetic response functions can be derived from the wavevector- and frequency-dependent current response $\chi_{ij}(\boldsymbol{k},\omega)$. The latter simultaneously is the spatial part of the Lorentz-covariant fundamental response tensor $\chi^{\mu}{}_{\nu}(k)$ connecting the four-potential A^{μ} to the four-current j^{μ} by means of the linear functional $j^{\mu}_{\text{int}}[A^{\nu}_{\text{ext}}]$ (see Eq. (2.37)).

In the following sections, this concept is applied to the free, non-relativistic, homogeneous electron gas, which represents one of the essential models in theoretical materials physics, in particular for conductors. In Sct. 3.1, we will first develop an expression for the most general, genuinely non-relativistic current density based on the Pauli equation. In contrast to common textbook literature, this current will consist of three instead of only two parts, namely the diamagnetic and orbital part as well as a spinorial one which presents an essentially new contribution. This fact leads to a paradigmatically new perception of spin-generated magnetism which is also one of the central points of this thesis: Instead of regarding spin-based magnetism as an effect separated from classical electrodynamics, it should should—at least on the most fundamental level—be treated in the framework of classical Yang-Mills theory by including a corresponding spinorial contribution in the expression for the current density. This hypothesis will then be successively backed by the results from subsequent sections, while in Sct. 3.4 this new approach is put into contrast to the standard approach.

In Sct. 3.2, the Kubo formalism is shortly explained, which provides the connection between quantum field theory and linear response theory and is used in order to find definite expressions for response functions of the free electron gas. En passant, we reproduce the famous Lindhard density-density response, which will pose an important reference quantity especially in Chpt. 5.

By inserting the operator counterpart of the anticipated *full* current into the so-called spectral form of the Kubo-Greenwood formula, the retarded current-current response tensor for the free electron gas is then derived in Sct. 3.3. Including the spinorial part into the current operator then yields, besides the well-known diamagnetic and orbital parts, χ^{dia}_{ij} and χ^{orb}_{ij}, two entirely new contributions, χ^{spin}_{ij} and χ^{cross}_{ij}. While it can be shown that the spin-orbit cross-correlations vanish for spin-unpolarized systems like the free electron gas, the purely spinorial contribution eventually reproduces standard results like the Pauli paramagnetism, which is otherwise extracted from a genuine spin-spin response. On the other hand, the Landau diamagnetism can be derived from the diamagnetic part in combination with the orbital contribution. This corroborates again the Central Claim of the Functional Approach to electrodynamics in media, that *all* electromagnetic material properties have to be accessed from the microscopic charge and current densities.

3.1. Electromagnetic current density

In this section, we will motivate the expression for the *full microscopic* electromagnetic current density,

$$\boldsymbol{j}(\boldsymbol{x},t) = \boldsymbol{j}_{\mathrm{dia}}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{orb}}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{spin}}(\boldsymbol{x},t)\,, \tag{3.1}$$

which is later used to derive a Kubo formula in order to compute the electromagnetic responses of the free electron gas. Because of the many different concepts required to understand why the naïve current density from basic quantum mechanics is not suitable for this task, this aspect deserves a careful discussion.

We start with the general form of a Schrödinger-type equation,

$$\mathrm{i}\hbar\,\partial_t \Psi(\boldsymbol{x},t) = \hat{H}\,\Psi(\boldsymbol{x},t)\,. \tag{3.2}$$

For a spin-0 particle in an external electromagnetic field, $\Psi \equiv \psi$ is just the scalar single-electron wave function and the corresponding Hamiltonian is given by[1]

$$\hat{H} = \frac{|\hat{\boldsymbol{p}} + e\boldsymbol{A}(\boldsymbol{x},t)|^2}{2m} - e\,\varphi(\boldsymbol{x},t)\,, \tag{3.3}$$

where $(-e)$ is the electron charge, m its mass and $\hat{\boldsymbol{p}}$ denotes the vectorial momentum operator which in position space reads

$$\hat{\boldsymbol{p}} = -\mathrm{i}\hbar\nabla\,. \tag{3.4}$$

Although the potentials actually represent *external* fields in the framework of quantum mechanics, we will leave out the explicit index and imply $\boldsymbol{A} \equiv \boldsymbol{A}_{\mathrm{ext}}$ and $\varphi \equiv \varphi_{\mathrm{ext}}$.

In basic quantum mechanics, the naïve current and charge densities are given by

$$\rho(\boldsymbol{x},t) = (-e)\sum_{s=\uparrow,\downarrow} \psi_s^*(\boldsymbol{x},t)\,\psi_s(\boldsymbol{x},t)\,, \tag{3.5}$$

and

$$\boldsymbol{j}_{\mathrm{orb}}(\boldsymbol{x},t) = \frac{(-e)\hbar}{2m\mathrm{i}} \sum_{s=\uparrow,\downarrow} \Big(\psi_s^*(\boldsymbol{x},t)\,(\nabla\psi_s)(\boldsymbol{x},t) - (\nabla\psi_s)^*(\boldsymbol{x},t)\,\psi_s(\boldsymbol{x},t)\Big)\,, \tag{3.6}$$

where the spin summation has been introduced for later purpose and can be ignored for now. With these two expressions given, it is easy to verify that the continuity equation,

$$\partial_t \rho(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{j}(\boldsymbol{x},t) = 0\,, \tag{3.7}$$

is not fulfilled for the said Hamiltonian. In order to have it fulfilled anyway, the current has

[1] It is important to note that the potentials do not represent operators but purely classical field quantities. This "restriction" is lifted in quantum electrodynamics, where potentials (and thus the electromagnetic fields) undergo a quantization procedure themselves leading, for instance, to a Hamiltonian by which the famous black-body radiation can be explained on a fundamental basis.

to be of the following form instead:

$$\boldsymbol{j} = \frac{(-e)}{2m}\left[\psi^*\left(\frac{\hbar}{\mathrm{i}}\nabla + e\boldsymbol{A}\right)\psi + \psi\left(-\frac{\hbar}{\mathrm{i}}\nabla + e\boldsymbol{A}\right)\psi^*\right]. \tag{3.8}$$

Apparently, this adapted current can be split into two contributions,

$$\boldsymbol{j}(\boldsymbol{x},t) = \boldsymbol{j}_{\mathrm{orb}}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{dia}}(\boldsymbol{x},t)\,, \tag{3.9}$$

where the *orbital* part is just the standard expression (3.6) and the new *diamagnetic* term is defined by

$$\boldsymbol{j}_{\mathrm{dia}}(\boldsymbol{x},t) = -\frac{(-e)}{m}\,\rho(\boldsymbol{x},t)\,\boldsymbol{A}(\boldsymbol{x},t)\,. \tag{3.10}$$

This adapted current is not only gauge invariant itself as shown in App. C.3 but also leads in combination with the minimal coupling Hamiltonian from Eq. (3.3) to a gauge invariant continuity equation and therefore to a gauge invariant local and global charge conservation.

Heuristically, Eq. (3.8) could have been found as well by replacing the derivatives in Eq. (3.6) by (gauge) covariant derivatives as explained in App. C. In fact, replacing all partial derivatives in the free Schrödinger equation according to

$$\partial_t \mapsto \partial_t - \frac{\mathrm{i}}{\hbar}\,e\varphi\,, \tag{3.11}$$

$$\nabla \mapsto \nabla + \frac{\mathrm{i}}{\hbar}\,e\boldsymbol{A}\,, \tag{3.12}$$

also converts the corresponding free Hamiltonian

$$\hat{H}_0 = \frac{|\hat{\boldsymbol{p}}|^2}{2m}\,, \tag{3.13}$$

into the minimal coupling Hamiltonian given by Eq. (3.3). The associated Schrödinger equation then is again invariant under gauge transformation (cf. App. C.3).

The remaining *spinorial* contribution to the current density in Eq. (3.1) cannot be justified by the standard Schrödinger equation. Instead we have to invoke the (non-relativistic) Pauli equation, which is again of the Schrödinger type (3.2) but includes spinorial contributions as well. The corresponding (matrix-valued) Hamiltonian reads

$$\hat{H}_{\mathrm{Pauli}} = \frac{1}{2m}\left(\boldsymbol{\sigma}\cdot(\hat{\boldsymbol{p}} + e\boldsymbol{A}(\boldsymbol{x},t))\right)^2 - e\varphi(\boldsymbol{x},t)\,, \tag{3.14}$$

which in contrast to the previous ones does not act on a simple wave function but an entire two-spinor[2],

$$\Psi(\boldsymbol{x},t) = \begin{pmatrix}\psi_\uparrow(\boldsymbol{x},t)\\ \psi_\downarrow(\boldsymbol{x},t)\end{pmatrix}, \tag{3.15}$$

consisting of one scalar wave functions for each spin channel. The elements of the vector

[2] Altough spinors look like vectors they are in fact not because they transform according to their very own rules. Thus, in context of vector analysis they may be regarded as scalars.

$\boldsymbol{\sigma} = (\sigma_1, \sigma_2, \sigma_3)^\mathsf{T}$ in Eq. (3.14) are the Pauli matrices,

$$\sigma_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_2 = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix}, \quad \sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{3.16}$$

which form (together with $\sigma_0 = \mathbb{1}_{2\times2}$) a basis of the real vector space of 2x2 hermitian matrices and fulfill the identity

$$(\boldsymbol{\sigma} \cdot \boldsymbol{C})(\boldsymbol{\sigma} \cdot \boldsymbol{D}) = (\boldsymbol{C} \cdot \boldsymbol{D})\mathbb{1}_{2\times2} + \mathrm{i}\boldsymbol{\sigma} \cdot (\boldsymbol{C} \times \boldsymbol{D}), \tag{3.17}$$

for two arbitrary vectors $\boldsymbol{C}$ and $\boldsymbol{D}$. Using this relation, the Pauli Hamiltonian can be expanded into a sum of the free Hamiltonian (3.13) and an interaction Hamiltonian

$$\hat{H}_\mathrm{Pauli} = \hat{H}_0 + \hat{H}_\mathrm{int}, \tag{3.18}$$

where the latter reads

$$\hat{H}_\mathrm{int} = \left(\frac{e}{2m} (\boldsymbol{A} \cdot \hat{\boldsymbol{p}} + \hat{\boldsymbol{p}} \cdot \boldsymbol{A}) + \frac{e^2}{2m} |\boldsymbol{A}|^2 - e\varphi \right) \mathbb{1}_{2\times2} + \frac{e\hbar}{2m} \boldsymbol{\sigma} \cdot (\nabla \times \boldsymbol{A}), \tag{3.19}$$

and is obviously again a sum of the minimal coupling Hamiltonian (3.3) and a new spinorial term.

A fundamental principle from analytical mechanics states that the charge and current density can be obtained from a (classical) Hamiltonian by taking the functional derivatives w. r. t. the (external) potentials φ and $\boldsymbol{A}$,

$$\rho(\boldsymbol{x}, t) = \frac{\delta H_\mathrm{int}(t)}{\delta\varphi(\boldsymbol{x}, t)}, \qquad \boldsymbol{j}(\boldsymbol{x}, t) = -\frac{\delta H_\mathrm{int}(t)}{\delta\boldsymbol{A}(\boldsymbol{x}, t)}, \tag{3.20}$$

which couple precisely via the said densities to a free Hamiltonian (cf. [21, §1.4.3]). The classical equivalent corresponding to the operator-valued Hamiltonian in Eq. (3.19) is obtained by taking the expectation value of the latter, i. e.

$$H(t) = H_0 + H_\mathrm{int}(t) = \langle\Psi|\hat{H}_0|\Psi\rangle + \langle\Psi|\hat{H}_\mathrm{int}(t)|\Psi\rangle, \tag{3.21}$$

where the interaction part is given by the integral[3]

$$\begin{aligned} H_\mathrm{int}(t) = \int \mathrm{d}^3\boldsymbol{x}\, \Psi^\dagger(\boldsymbol{x}, t) \Bigg(& \frac{e\hbar}{m\mathrm{i}} \boldsymbol{A}(\boldsymbol{x}, t) \cdot \nabla + \frac{e\hbar}{2m\mathrm{i}} (\nabla \cdot \boldsymbol{A})(\boldsymbol{x}, t) + \frac{e\hbar}{2m} \boldsymbol{\sigma} \cdot \big(\nabla \times \boldsymbol{A}(\boldsymbol{x}, t)\big) \\ & + \frac{e\hbar}{2m} |\boldsymbol{A}(\boldsymbol{x}, t)|^2 - e\,\varphi(\boldsymbol{x}, t) \Bigg) \Psi(\boldsymbol{x}, t). \end{aligned} \tag{3.22}$$

By partially integrating some of the terms in the integrand and rearraging factors in a more

[3] A similar calculation can be found in [9, App. C2], where the Coulomb gauge, $\nabla \cdot \boldsymbol{A} = 0$, has been used. This is in fact not necessary since the divergence term cancels with one of the terms obtained by partially integrating the $\boldsymbol{A} \cdot \hat{\boldsymbol{p}}$ contribution wich then simplifies to $\boldsymbol{A} \cdot \boldsymbol{j}_\mathrm{orb}$ in the first line of Eq. (3.23). The full current density can thus be derived entirely free of any gauge.

suggestive way, this integral can be expressed equivalently by

$$\begin{aligned} H_{\text{int}}(t) = &\int \mathrm{d}^3\boldsymbol{x}\; \boldsymbol{A}(\boldsymbol{x},t) \cdot \frac{e\hbar}{2mi} \Big(\Psi^\dagger(\boldsymbol{x},t)(\nabla\Psi)(\boldsymbol{x},t) - (\nabla\Psi)^\dagger(\boldsymbol{x},t)\,\Psi(\boldsymbol{x},t) \Big) \\ &+ \int \mathrm{d}^3\boldsymbol{x}\; \boldsymbol{A}(\boldsymbol{x},t) \cdot \frac{e\hbar}{2m} \nabla \times \Big(\Psi^\dagger(\boldsymbol{x},t)\,\boldsymbol{\sigma}\,\Psi(\boldsymbol{x},t) \Big) \\ &+ \int \mathrm{d}^3\boldsymbol{x}\; \boldsymbol{A}(\boldsymbol{x},t) \cdot \frac{e^2}{2m}\, \Psi^\dagger(\boldsymbol{x},t)\,\boldsymbol{A}(\boldsymbol{x},t)\,\Psi(\boldsymbol{x},t) \\ &+ \int \mathrm{d}^3\boldsymbol{x}\; \varphi(\boldsymbol{x},t)\,(-e)\,\Psi^\dagger(\boldsymbol{x},t)\,\Psi(\boldsymbol{x},t)\,, \end{aligned} \tag{3.23}$$

which allows to directly read off the searched-for densities according to the functional derivatives in Eq. (3.20). The derivative w. r. t. the scalar potential obviously recovers the naïve charge density (3.5), whereas the (negative) derivative w. r. t. the vector potential yields a more complicated expression consisting of three terms: The first line in Eq. (3.23) apparently reverts to the orbital current (3.6) and the third line reproduces the diamagnetic part Eq. (3.10). The second line, however, reveals an entirely new third contribution which mixes different spin channels via the Pauli matrices. This spinorial current reads

$$\boldsymbol{j}_{\text{spin}}(\boldsymbol{x},t) = \frac{(-e)\hbar}{2m} \nabla \times \left(\sum_{s,s'=\uparrow,\downarrow} \psi_s^*(\boldsymbol{x},t)\,\boldsymbol{\sigma}_{ss'}\,\psi_{s'}(\boldsymbol{x},t) \right), \tag{3.24}$$

where $\boldsymbol{\sigma}_{ss'}$ denotes the spatial vector build from the (s,s') element (in the spinorial basis) of every Pauli matrix $\sigma_i = ((\sigma_i)_{ss'})$. Although this spinorial current contribution has to be regarded as non-standard and is (at least in this form) absent in the usual literature on response theory, other books like Schwabl's "Statistical Mechanics" (Ref. [40, §6.1.1]) have this covered.

Combining the orbital, diamagnetic and spinorial current densities leads to the *full* current anticipated in Eq. (3.1). This result can be motivated in an even more natural way from the Dirac current in its non-relativistic limit (see [41, Chap. XX, §29] or [42]). This derivation especially proves, that Eq. (3.1) already contains the most general non-relativistic microscopic current. Further, this current density can be regarded as a functional of the external vector potential. In some textbooks ([32, App. 2], [21, §1.4.3] and especially [30, p. 392 fn. 2]), the current is instead divided in a diamagnetic and paramagnetic part,

$$\boldsymbol{j}(\boldsymbol{x}) = \boldsymbol{j}_{\text{para}}(\boldsymbol{x}) + \boldsymbol{j}_{\text{dia}}(\boldsymbol{x})\,. \tag{3.25}$$

with

$$\boldsymbol{j}_{\text{para}}(\boldsymbol{x}) = \boldsymbol{j}(\boldsymbol{x})[\boldsymbol{A} \equiv 0]\,. \tag{3.26}$$

In other words, the current is split into a part which explicitly depends on the vector potential and a part that does not. However, in the said textbooks this paramagnetic term is usually identified with the orbital current, whereas in our case there is also the spinorial part. This way, we see that the spinorial current ranks among the paramagnetic contributions.

3.2. Kubo-Greenwood formulae

In this section, the previously found full current density will be used in order to find an explicit expression for the retarded current response χ_{ij} for the free, homogeneous and non-relativistic electron gas. This is achieved by exploiting the famous Kubo formalism[4], which provides a link between quantum field theory and classical electrodynamics.

Assuming we have a quantum system with a Hamiltonian composed of an unperturbed part $\hat{H}_0$, and an external perturbation $\hat{H}_{\text{int}}$, which depends on a parameter X such that $\hat{H}_{\text{int}}(X \equiv 0) = 0$. The simplest way this externally controlled time-dependent field $X(t)$ can enter the Hamiltonian is by linearly coupling to an operator $\hat{B}$,

$$\hat{H}_{\text{int}}(t) = X(t)\,\hat{B}\,. \tag{3.27}$$

Further, the parameter $X = X(t)$ should depend on the time in a way that for a specific start time t_0 the perturbation is "turned on", i. e.

$$\hat{H}(t) = \begin{cases} \hat{H}_0 & t < t_0 \\ \hat{H}_0 + \hat{H}_{\text{int}}(X(t)) & t \geq t_0\,. \end{cases} \tag{3.28}$$

This is why $X(t)$ is also known as "switch-on"-function and in the simplest case is just a Heaviside step function $X(t) = \Theta(t - t_0)$.

Let $|\Psi_0\rangle$ now be the ground state of the unperturbed system and $|\Psi(t)\rangle$ the state determined by the Schrödinger-type time evolution

$$\mathrm{i}\hbar \frac{\mathrm{d}}{\mathrm{d}t} |\Psi(t)\rangle = \hat{H}(t)\,|\Psi(t)\rangle\ , \quad |\Psi(t_0)\rangle = \Psi_0\,. \tag{3.29}$$

The expectation value of an observable represented by the hermetian operator $\hat{A}$ is then given by

$$A(t)[X] = \langle \Psi(t)|\hat{A}|\Psi(t)\rangle\ , \tag{3.30}$$

which implicitly is a functional of the external perturbation $X(t)$ via the wave function. The Kubo formula then states that the retarded linear response of the observable $\hat{A}$ w. r. t. a weak external perturbation that couples to $\hat{B}$ can be calculated as the expectation value in the unperturbed ground state

$$\chi^{\mathrm{R}}_{AB}(t - t') \overset{\text{def}}{=} \left.\frac{\delta A(t)}{\delta X(t')}\right|_{X=0} = -\frac{\mathrm{i}}{\hbar}\,\Theta(t - t')\,\langle \Psi_0|[\hat{A}_{\mathrm{I}}(t), \hat{B}_{\mathrm{I}}(t')]|\Psi_0\rangle\ , \tag{3.31}$$

where the time dependence of both operators in the commutator is given by the interaction picture

$$\hat{A}_{\mathrm{I}}(t) = \mathrm{e}^{\mathrm{i}\hat{H}_0 t/\hbar}\,\hat{A}\,\mathrm{e}^{-\mathrm{i}\hat{H}_0 t/\hbar}\,, \tag{3.32}$$

and likewise for $\hat{B}(t)$.

In case of a non-interacting system, i. e. systems with Hamiltonians containing only one-

[4] Named after the Japanese mathematical physicist Ryogo Kubo (1920-1995).

particle operators, the associated many-body states in fermionic Fock space can be chosen as Slater determinants,

$$|\Psi^N\rangle = |\mathrm{SL}(\varphi_1, \ldots, \varphi_N)\rangle \ , \tag{3.33}$$

build from single-particle wave functions $\varphi_i(\boldsymbol{x})$ with corresponding eigenenergies ε_i. This is in particular true for the ground state $|\Psi_0^N\rangle$ which is constructed from the energetically lowest orbitals. Higher-energy states are then generated by successively exciting one-particle levels. The corresponding many-body Hamiltonian $\hat{H}^N$ then decomposes into a sum of one-particle Hamiltonians $\hat{H}^1$, which act on the said orbitals according to

$$\hat{H}^1 \, |\varphi_i\rangle = \varepsilon_i \, |\varphi_i\rangle \ . \tag{3.34}$$

This is an extremely important result since it allows us to exclusively work with single-particle equations instead of their much more complicated many-body equivalents, which in general also contain two-particle operators. For the abstract operators it is convenient to introduce the concept of second quantization (see e. g. [21]), which simplifies calculations in many-body theory tremendously. There, an important object is the Schrödinger field operator and its hermitean adjoint,

$$\hat{\psi}^{(\dagger)}(\boldsymbol{x}) \stackrel{\mathrm{def}}{=} \hat{a}^{(\dagger)}(|\boldsymbol{x}\rangle) \, , \tag{3.35}$$

i. e. the creator and annihilator of a position state in Fock space. Specifically applying this annihilator to a single-particle state projects it onto position space and leaves only the Fock vacuum with $\langle 0|0\rangle = 1$,

$$\hat{\psi}(\boldsymbol{x}) \, |\varphi\rangle = \langle \boldsymbol{x}|\varphi\rangle \, |0\rangle = \varphi(\boldsymbol{x}) \, |0\rangle \ . \tag{3.36}$$

These two field operators given, the *number density* operator becomes

$$\hat{n}(\boldsymbol{x}) = \hat{\psi}^\dagger(\boldsymbol{x}) \hat{\psi}(\boldsymbol{x}) \, , \tag{3.37}$$

which differs from the *charge density* operator only by a constant, $\hat{\rho}(\boldsymbol{x}) = (-e) \, \hat{n}(\boldsymbol{x})$. Further, it can be shown that in general, any one-particle operators with a time evolution given by the interaction picture can be expanded as

$$\hat{A}(t) = \sum_{i,j=0}^{\infty} A_{ij} \, \mathrm{e}^{\mathrm{i}(\varepsilon_i - \varepsilon_j)t/\hbar} \, \hat{a}_i^\dagger \, \hat{a}_j \, , \tag{3.38}$$

where the matrix elements are given in the orbital basis by

$$A_{ij} = \langle \varphi_i | \hat{A} | \varphi_j \rangle \ . \tag{3.39}$$

In the grand canonical ensemble (which corresponds to the case of infinite particle number and is required for the application of the thermodynamic limit as we shall see in a moment),

the Kubo formula (3.32) then reverts to

$$\chi^{\mathrm{R}}_{\mathrm{AB}}(t-t') = -\frac{\mathrm{i}}{\hbar}\,\Theta(t-t')\sum_{i,j=0}^{\infty}\big(f_{\beta,\mu}(\varepsilon_i) - f_{\beta,\mu}(\varepsilon_j)\big)\,A_{ij}\,B_{ij}\,\mathrm{e}^{\mathrm{i}(\varepsilon_i-\varepsilon_j)t/\hbar}\,, \tag{3.40}$$

or equivalently in Fourier space under usage of Eq. (B.114),

$$\chi^{\mathrm{R}}_{\mathrm{AB}}(\omega) = \sum_{i,j=0}^{\infty}\frac{\big(f_{\beta,\mu}(\varepsilon_i) - f_{\beta,\mu}(\varepsilon_j)\big)\,A_{ij}\,B_{ij}}{\hbar(\omega+\mathrm{i}\eta) - (\varepsilon_j-\varepsilon_i)}\,, \tag{3.41}$$

where η is a positive infinitesimal regularization factor and

$$f_{\beta,\mu}(\varepsilon_i) = \frac{1}{1+\mathrm{e}^{\beta(\varepsilon_i-\mu)}} \tag{3.42}$$

denotes the Fermi-Dirac distribution at inverse temperature $\beta = 1/k_{\mathrm{B}}T$ and for chemical potential μ. The latter equation will be the starting point for the following discussion of the density and current response functions. A more detailed derivation of these so-called spectral representations (3.40) and (3.41) is given in §2 of our preprint [2], which simultaneously is the basis for this and the following sections. Alternatively, the essentials of second quantized linear response formalism are also condensed in Sec. 4.2 of Ref. [32], where the spectral representations of the Kubo formula are given by Eqs. (4.8) and (4.9).

Coming back to the free electron gas, we now have to find an appropriate single-particle basis in order to evaluate Eq. (3.39) for the current operator. In contrast to the model of a perfect crystalline bulk, there is no periodic potential generated by atomic cores in case of the Fermi gas. Instead, the system is strictly homogeneous such that response functions only depend on the difference of their two space-time arguments. This is a fundamental result which follows directly from translation invariance as shown in App. D.2. On the contrary, real probes are unlikely to fill entire space (homogeneously or not). Therefore, many-body systems are restricted to a finite volume V and Born-von Kármán periodic boundaries are postulated. This is especially relevant since quantum mechanics requires a finite particle number N (although not a constant one) in order to be able to formally operate in Fock space. The initially assumed homogeneity is then regained afterwards by performing the *thermodynamic limit*, i. e. $V, N \to \infty$, in a way that the overall particle density $n = N/V$ remains constant. As a consequence of this treatment, momentum eigenfunctions qualify as single particle states as shown in App. D.1. In particular, the resulting partitioning of reciprocal space applies with one major difference: Because of the lack of a lattice periodicity, there are no $\boldsymbol{G}$-vectors which could form a Fourier grid. Instead, the entire reciprocal space may be regarded as a single large first Brillouin zone. In the thermodynamic limit, the latter then even becomes *continuous* such that discrete sums over allowed Brillouin zone vectors,

$$\sum_{\boldsymbol{k}} \mapsto \frac{V}{(2\pi)^3}\int \mathrm{d}^3\boldsymbol{k} \tag{3.43}$$

may be replaced by continuous integrals. In the following, we will work directly in the

thermodynamic limit. Analogous calculations for a discrete set of wave vectors can be found in [2, §3].

Following the previous discussion, we now introduce the (continuous) set of spin-dependent momentum eigenstates[5] on the one-particle Hilbert space $\mathscr{H} = \mathbb{C} \otimes L^2(\mathbb{R}^3, \mathbb{C})$ by the wave functions

$$\varphi_{\boldsymbol{k},s}(\boldsymbol{x}) = \langle \boldsymbol{x} | \boldsymbol{k}, s \rangle = \frac{1}{(2\pi)^{3/2}} \, \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}} \begin{pmatrix} \delta_{\uparrow,s} \\ \delta_{\downarrow,s} \end{pmatrix} , \tag{3.44}$$

with orthonormality

$$\langle \boldsymbol{k}, s | \boldsymbol{k}', s' \rangle = \delta_{ss'} \, \delta^3(\boldsymbol{k} - \boldsymbol{k}') \, , \tag{3.45}$$

and completeness condition

$$\sum_{s=\uparrow,\downarrow} \int \mathrm{d}^3\boldsymbol{k} \; |\boldsymbol{k}, s\rangle\langle \boldsymbol{k}, s| = \mathbb{1} \, . \tag{3.46}$$

The states in Eq. (3.39) which enter the spectral representation of the Kubo formula (3.41) then have to be replaced according to

$$\varphi_i(\boldsymbol{x}) \mapsto \varphi_{\boldsymbol{k},s}(\boldsymbol{x}) \, , \tag{3.47}$$

and now denote entire Pauli spinors. Inserting this into relation (3.36) for spin-dependent Schrödinger field operators we find

$$\hat{\psi}_r(\boldsymbol{x}) \, |\boldsymbol{k}, s\rangle = (\varphi_{\boldsymbol{k},s})_r(\boldsymbol{x}) \, |0\rangle = \frac{\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}}}{(2\pi)^{2/3}} \, \delta_{rs} \, |0\rangle \; . \tag{3.48}$$

Here, φ_r now denotes the r-th component of the spinor in contrast to $\varphi_{\boldsymbol{k},s}$, which is the $(\boldsymbol{k}, s)$-th base vector of the previously mentioned one-particle Hilbert space.

The contructors $\hat{a}^\dagger_{\boldsymbol{k},s}$ and annihilators $\hat{a}_{\boldsymbol{k},s}$ corresponding to the momentum eigenstates (3.44) which act on the Fock vacuum according to

$$\hat{a}^\dagger_{\boldsymbol{k},s} \, |0\rangle = |\boldsymbol{k}, s\rangle \; , \quad \hat{a}_{\boldsymbol{k},s} \, |0\rangle = 0 \, , \tag{3.49}$$

can be regarded as Fourier transforms of the respective Schrödinger field operators. They follow the standard transformation rules (see e. g. [21, §1.3.6])

$$\hat{a}^\dagger_{\boldsymbol{k},s} = \int \frac{\mathrm{d}^3\boldsymbol{x}}{(2\pi)^{3/2}} \, \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}} \, \hat{\psi}^\dagger_s(\boldsymbol{x}) \, , \qquad \hat{a}_{\boldsymbol{k},s} = \int \frac{\mathrm{d}^3\boldsymbol{x}}{(2\pi)^{3/2}} \, \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}} \, \hat{\psi}_s(\boldsymbol{x}) \, , \tag{3.50}$$

[5] We use a modified version of the momentum eigenvectors in terms of the wavevector $\boldsymbol{k}$, which is frequently employed in theoretical solid state physics and should not be confused with the standard definition from basic quantum mechanics in terms of $\boldsymbol{p} = \hbar\boldsymbol{k}$:

$$\langle \boldsymbol{p}' | \boldsymbol{p} \rangle = \delta^3(\boldsymbol{p} - \boldsymbol{p}') = \delta^3(\hbar(\boldsymbol{k} - \boldsymbol{k}')) = \hbar^{-3}\delta^3(\boldsymbol{k} - \boldsymbol{k}') \, .$$

Together with the postulation (3.45) this gives $\hbar^{3/2} \, |\boldsymbol{p}\rangle = |\boldsymbol{k}\rangle$ for the abstract standard momentum eigenstate and $\langle \boldsymbol{x} | \boldsymbol{p} \rangle = (2\pi\hbar)^{-3/2} \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}}$ for its represenation in real space.

with the respective inverses,

$$\hat{\psi}_s^\dagger(\boldsymbol{x}) = \int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^{3/2}}\, \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}}\, \hat{a}_{\boldsymbol{k},s}^\dagger\,, \qquad \hat{\psi}_s(\boldsymbol{x}) = \int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^{3/2}}\, \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}}\, \hat{a}_{\boldsymbol{k},s}\,, \tag{3.51}$$

similar to regular (classical) field quantities (cf. App. B.3). In addition, they fulfill the canonical anti-commutation relations,

$$\{\hat{a}_i, \hat{a}_j\} = 0\,, \quad \{\hat{a}_i^\dagger, \hat{a}_j^\dagger\} = 0\,, \quad \{\hat{a}_i, \hat{a}_j^\dagger\} = \delta_{ij}\,, \tag{3.52}$$

where in our case i and j have to be replaced with $(\boldsymbol{k}, s)$ and $(\boldsymbol{k}', s')$, respectively. Using these, the second quantized expansion (3.38) of the free Hamiltonian (3.13) in the momentum basis reads

$$\hat{H}_0 = \sum_{s=\uparrow,\downarrow} \int \mathrm{d}^3\boldsymbol{k}\; \varepsilon_0(\boldsymbol{k})\, \hat{a}_{\boldsymbol{k},s}^\dagger \hat{a}_{\boldsymbol{k},s}\,, \tag{3.53}$$

where the quantum mechanical dispersion relation of a free electron is given by

$$\varepsilon_0(\boldsymbol{k}) = \hbar\omega_{\boldsymbol{k}} = \frac{\hbar^2|\boldsymbol{k}|^2}{2m}\,. \tag{3.54}$$

Obviously, this is a generalized version of the three-dimensional quantum mechanical harmonic oscillator.[6]

Before we now come to the Kubo formula for the current-current response, we first will reproduce another highly important result of the free electron gas, namely the famous Lindhard density response function. This function has not only great historical and conceptional value but is also relevant for the subsequent sections where we will prove the "Lindhard integral theorem", which apparently refers to this very response function (or at least to its associated fundamental integral).

In order to find the density-density response for the free electron gas both, the operator to which the external perturbation couples, and the one corresponding to the observable where the induced changes are to be analyzed, have to be identified with the density operator,

$$\hat{A} \;\mapsto\; \hat{\rho}(\boldsymbol{x}) \;= (-e)\,\hat{\psi}^\dagger(\boldsymbol{x})\,\hat{\psi}(\boldsymbol{x})\,, \tag{3.55}$$

$$\hat{B} \;\mapsto\; \hat{\rho}(\boldsymbol{x}') = (-e)\,\hat{\psi}^\dagger(\boldsymbol{x}')\,\hat{\psi}(\boldsymbol{x}')\,, \tag{3.56}$$

which by Eqs. (3.37) and (3.48) has the matrix elements

$$\langle \boldsymbol{k}, s|\, \hat{\rho}(\boldsymbol{x})\, |\boldsymbol{k}', s'\rangle = \frac{(-e)}{(2\pi)^3}\, \delta_{ss'}\, \mathrm{e}^{\mathrm{i}(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{x}}\,. \tag{3.57}$$

[6] Indeed, one of the essential statements of quantum field theory is that it is the field itself which is "oscillating". More precisely, there is one complex harmonic oscillator for each Fourier mode. This is best seen on the example of the electromagnetic field in Eq. (1.28). By no means do these abstract oscillators correspond to real particles in space.

Plugging this into the master formula (3.41) yields

$$\chi(\boldsymbol{x},\boldsymbol{x}';\omega) = \frac{(-e)^2}{(2\pi)^3}\sum_{ss'}\int \mathrm{d}^3\boldsymbol{k}\int \mathrm{d}^3\boldsymbol{k}'\;\delta_{ss'}\mathrm{e}^{\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\cdot(\boldsymbol{x}-\boldsymbol{x}')}\,\frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}))-f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}'))}{\hbar(\omega+\mathrm{i}\eta)-(\varepsilon_0(\boldsymbol{k}')-\varepsilon_0(\boldsymbol{k}))} \tag{3.58}$$

which, under the substitutions $\boldsymbol{q}=\boldsymbol{k}'-\boldsymbol{k}$ with $\mathrm{d}^3\boldsymbol{q}=\mathrm{d}^3\boldsymbol{k}'$ and $\boldsymbol{r}=\boldsymbol{x}-\boldsymbol{x}'$, becomes

$$\chi(\boldsymbol{r},\omega) = \int\frac{\mathrm{d}^3\boldsymbol{q}}{(2\pi)^3}\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}}\left((-e)^2\sum_{ss'}\delta_{ss'}\int\mathrm{d}^3\boldsymbol{k}\;\frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}))-f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}')}{\hbar(\omega+\mathrm{i}\eta)-(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q})-\varepsilon_0(\boldsymbol{k}))}\right). \tag{3.59}$$

By comparing this equation with the (spatial) Fourier transform of a homogeneous response function (see Eq. (B.106) in App. B.3), the expression within the large parentheses can be identified with the response function in reciprocal space,

$$\chi(\boldsymbol{q},\omega) = 2\,(-e)^2\int\mathrm{d}^3\boldsymbol{k}\;\frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}))-f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q})}{\hbar(\omega+\mathrm{i}\eta)-(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q})-\varepsilon_0(\boldsymbol{k}))}\,, \tag{3.60}$$

which is none other than the Lindhard density response [43]. Note, that it does not explicitly depend on the spin because neither the Fermi-Dirac distribution nor the orbital energy do such that the spin can be taken into account by an overall factor of two.

3.3. Diamagnetic, orbital and spinorial contribution

Turning to the current-current response, we could in principle proceed similarly to the last section. However, this case is more complicated in different respects and thus requires a more careful treatment. Firstly, the interaction Hamiltonian in which the external perturbation $\boldsymbol{A}_{\mathrm{ext}}$ couples to the current $\boldsymbol{j}$ is not of the linear form usually assumed in classical field theory, i. e.

$$H_{\mathrm{int}} \neq \int\mathrm{d}^3\boldsymbol{x}\,\big(\rho(\boldsymbol{x},t)\,\varphi_{\mathrm{ext}}(\boldsymbol{x},t)-\boldsymbol{j}(\boldsymbol{x},t)\cdot\boldsymbol{A}_{\mathrm{ext}}(\boldsymbol{x},t)\big)\,, \tag{3.61}$$

but quadratic in the vector potential as can easily be attested by Eq. (3.23). Secondly, the observable under consideration depends itself on the perturbation. In this case, the expectation value (3.30) is not only an implicit functional of the perturbation X via the time evolution of the state, but also explicitly via the abstract operator itself, $\hat{A}=\hat{A}[X]$. Consequently, the Kubo formula in Eq. (3.32) has to be extended via Leibniz' rule in order to account for this explicit dependence.[7] Following Ref. [9, App. C.1], the resulting *generalized Kubo formula* is then

$$\frac{\delta A(t)}{\delta X(t')}\Big|_{X=0} = \delta(t-t')\left\langle\frac{\partial\hat{A}}{\partial X}\Big|_{X=0}\right\rangle_{\Psi_0} - \frac{\mathrm{i}}{\hbar}\,\Theta(t-t')\left\langle\left[\left(\hat{A}(t)\big|_{X=0}\right),\left(\frac{\partial\hat{H}_{\mathrm{int}}(t)}{\partial X}\Big|_{X=0}\right)\right]\right\rangle_{\Psi_0}. \tag{3.62}$$

[7] This procedure is at variance with most text books on this topic like [32, §3.4] which prefer to stick to the standard Kubo formula, i. e. consider only the orbital (or paramagnetic) current operator, and add the diamagnetic contribution afterwards for consistency. By contrast, the spirit of the procedure followed in this thesis is based on fundamental functional derivatives Eq. (3.20) which lead to a Kubo formula where the diamagnetic term is already included in a more natural way. In any case, both versions lead to the same result, cf. e. g. [32, Eq. 3.172].

For the current-current response, we have to identify $\hat{A} \mapsto \hat{\boldsymbol{j}}(\boldsymbol{x})$ as well as $X \mapsto \boldsymbol{A}_{\text{ext}} \equiv \boldsymbol{A}$, where $\hat{\boldsymbol{j}}$ refers to the operator version of the full current in form of Eq. (3.1). In second quantized formalism, one-particle operators like $\hat{H}_{\text{int}}$ and $\hat{\boldsymbol{j}}$ can be easily obtained by simply replacing the orbitals in the classical expectation values with the corresponding Schrödinger field operators,

$$\psi(\boldsymbol{x}) \mapsto \hat{\psi}(\boldsymbol{x})\,, \tag{3.63}$$

$$\psi^*(\boldsymbol{x}) \mapsto \hat{\psi}^\dagger(\boldsymbol{x})\,. \tag{3.64}$$

The relations in Eq. (3.20) thus stay valid even for operator fields. Hence, the (negative) derivative of the interaction Hamiltonian (3.23) w. r. t. the vector potential yields by definition the current operator. Evaluating the latter at vanishing vector potential produces the paramagnetic current (operator),

$$-\left.\frac{\delta \hat{H}_{\text{int}}}{\delta \boldsymbol{A}(\boldsymbol{x})}\right|_{\boldsymbol{A}\equiv 0} = \hat{\boldsymbol{j}}_{\text{para}}(\boldsymbol{x}) = \hat{\boldsymbol{j}}_{\text{orb}}(\boldsymbol{x}) + \hat{\boldsymbol{j}}_{\text{spin}}(\boldsymbol{x})\,, \tag{3.65}$$

with

$$\hat{\boldsymbol{j}}_{\text{orb}}(\boldsymbol{x}) = \frac{(-e)\hbar}{2m\mathrm{i}} \sum_{s=\uparrow,\downarrow} \Big(\hat{\psi}_s^\dagger(\boldsymbol{x})\,(\nabla\hat{\psi}_s)(\boldsymbol{x}) - (\nabla\hat{\psi}_s^\dagger)(\boldsymbol{x})\,\hat{\psi}_s(\boldsymbol{x}) \Big)\,, \tag{3.66}$$

and

$$\hat{\boldsymbol{j}}_{\text{spin}}(\boldsymbol{x}) = \frac{(-e)\hbar}{2m} \nabla \times \left(\sum_{s,s'=\uparrow,\downarrow} \hat{\psi}_s^\dagger(\boldsymbol{x})\,\boldsymbol{\sigma}_{ss'}\,\hat{\psi}_{s'}(\boldsymbol{x}) \right)\,. \tag{3.67}$$

On the other hand, the vector potential enters the current only via the diamagnetic part (3.10). Since both fields explicitly depend on a spatial argument, the derivative in the first term of Eq. (3.62) turns again into a functional derivative which in this case explicitly reads

$$\frac{\delta \boldsymbol{j}(\boldsymbol{x})}{\delta \boldsymbol{A}(\boldsymbol{x}')} \equiv \frac{\delta \boldsymbol{j}_{\text{dia}}(\boldsymbol{x})}{\delta \boldsymbol{A}(\boldsymbol{x}')} = \frac{e}{m}\,\delta^3(\boldsymbol{x}-\boldsymbol{x}')\,\hat{\rho}(\boldsymbol{x})\overleftrightarrow{\mathbb{1}}\,. \tag{3.68}$$

The general Kubo formula for the full current-current response therefore becomes

$$\chi_{kl}(\boldsymbol{x},t;\boldsymbol{x}',t') = \frac{e}{m}\,\delta_{ij}\,\rho(\boldsymbol{x})\,\delta^3(\boldsymbol{x}-\boldsymbol{x}')\,\delta(t-t') + \frac{\mathrm{i}}{\hbar}\,\Theta(t-t')\left\langle [\hat{j}_k(\boldsymbol{x},t),\,\hat{j}_l(\boldsymbol{x}',t')] \right\rangle\,, \tag{3.69}$$

where the symbol $\hat{j}_i$ in the expectation value denotes the paramagnetic part of the current while the time-dependency of the latter is again covered by the interaction picture. Because of the bilinearity of commutators, Eq. (3.69) may be decomposed into four contributions,

$$\chi_{kl} = \chi_{kl}^{\text{dia}} + \chi_{kl}^{\text{orb}} + \chi_{kl}^{\text{spin}} + \chi_{kl}^{\text{cross}}\,, \tag{3.70}$$

where χ^{dia}_{kl} is the local contribution defined by the first term in Eq. (3.69). The remaining three contributions on the other hand read

$$\overleftrightarrow{\chi}^{\mathrm{spin}}(\boldsymbol{x},t;\boldsymbol{x}',t') = \frac{\mathrm{i}}{\hbar}\,\Theta(t-t')\left\langle[\hat{\boldsymbol{j}}_{\mathrm{spin}}(\boldsymbol{x},t),\hat{\boldsymbol{j}}_{\mathrm{spin}}(\boldsymbol{x}',t')]\right\rangle\,, \tag{3.71}$$

$$\overleftrightarrow{\chi}^{\mathrm{orb}}(\boldsymbol{x},t;\boldsymbol{x}',t') = \frac{\mathrm{i}}{\hbar}\,\Theta(t-t')\left\langle[\hat{\boldsymbol{j}}_{\mathrm{orb}}(\boldsymbol{x},t),\hat{\boldsymbol{j}}_{\mathrm{orb}}(\boldsymbol{x}',t')]\right\rangle\,, \tag{3.72}$$

$$\overleftrightarrow{\chi}^{\mathrm{cross}}(\boldsymbol{x},t;\boldsymbol{x}',t') = \frac{\mathrm{i}}{\hbar}\,\Theta(t-t')\left\langle[\hat{\boldsymbol{j}}_{\mathrm{orb}}(\boldsymbol{x},t),\hat{\boldsymbol{j}}_{\mathrm{spin}}(\boldsymbol{x}',t')]+[\hat{\boldsymbol{j}}_{\mathrm{spin}}(\boldsymbol{x},t),\hat{\boldsymbol{j}}_{\mathrm{orb}}(\boldsymbol{x}',t')]\right\rangle\,, \tag{3.73}$$

respectively. Regarding the last relation it is noteworthy that only spin and orbital parts do cross-correlate but not the diamagnetic one. Further, the two contributions within expectation value (3.73) cannot be combined because the operator fields depend on different (primed and unprimed) arguments. Lastly we emphasize that the spin-orbit cross-correlation should not be confused with spin-orbit coupling in the sense of the relativistic correction term derived from the Dirac equation by a Foldy-Wouthuysen transformation (cf. [44, §2.4.2]).

In order to evaluate the spectral representations of Eqs. (3.71) to (3.73) in the momentum basis, we need to find the matrix elements of the respective current operators. Using again relation Eq. (3.48), the expectation value of the orbital current becomes

$$\begin{aligned}
&\langle\boldsymbol{k},s|\hat{\boldsymbol{j}}_{\mathrm{orb}}(\boldsymbol{x})|\boldsymbol{k}',s'\rangle \\
&\quad= \frac{(-e)\hbar}{2m\mathrm{i}}\sum_{r=\uparrow,\downarrow}\left(\left\langle\boldsymbol{k},s\,\middle|\,\hat{\psi}^{\dagger}_{r}(\boldsymbol{x})\,(\nabla\hat{\psi}_{r})(\boldsymbol{x})\,\middle|\,\boldsymbol{k}',s'\right\rangle-\left\langle\boldsymbol{k},s\,\middle|\,(\nabla\hat{\psi}^{\dagger}_{r})(\boldsymbol{x})\,\hat{\psi}_{r}(\boldsymbol{x})\,\middle|\,\boldsymbol{k}',s'\right\rangle\right) && (3.74)\\
&\quad= \frac{(-e)\hbar}{2m\mathrm{i}}\sum_{r=\uparrow,\downarrow}\frac{\delta_{rs}\,\delta_{rs'}}{(2\pi)^3}\left(\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}}\left(\mathrm{i}\boldsymbol{k}'\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}'\cdot\boldsymbol{x}}\right)+\left(\mathrm{i}\boldsymbol{k}\,\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}}\right)\mathrm{e}^{\mathrm{i}\boldsymbol{k}'\cdot\boldsymbol{x}}\right) && (3.75)\\
&\quad= \frac{(-e)\hbar}{2m}\,\frac{\delta_{ss'}}{(2\pi)^3}\,(\boldsymbol{k}+\boldsymbol{k}')\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{x}}\,. && (3.76)
\end{aligned}$$

Because of possibly confusing notation conflicts, we follow a different way for the spinorial contribution. Instead of using again Eq. (3.48) after inserting the explicit expression of the spinorial current operator,

$$\langle\boldsymbol{k},s|\hat{\boldsymbol{j}}_{\mathrm{spin}}(\boldsymbol{x})|\boldsymbol{k}',s'\rangle = \frac{(-e)\hbar}{2m}\,\nabla\times\left(\sum_{r,r'=\uparrow,\downarrow}\left\langle\boldsymbol{k},s\,\middle|\,\hat{\psi}^{\dagger}_{r}(\boldsymbol{x})\,\boldsymbol{\sigma}_{rr'}\,\hat{\psi}_{r'}(\boldsymbol{x})\,\middle|\,\boldsymbol{k}',s'\right\rangle\right) \tag{3.77}$$

we replace the momentum states with Fock vaccum according to Eq. (3.49) and the Schrödinger field operators by their Fourier representation (see Eq. (3.51)),

$$\begin{aligned}
&\left\langle\boldsymbol{k},s\,\middle|\,\hat{\psi}^{\dagger}_{r}(\boldsymbol{x})\,\boldsymbol{\sigma}_{rr'}\,\hat{\psi}_{r'}(\boldsymbol{x})\,\middle|\,\boldsymbol{k}',s'\right\rangle \\
&\quad= \left\langle 0\,\middle|\,\hat{a}_{\boldsymbol{k},s}\left(\int\frac{\mathrm{d}^3\boldsymbol{k}_1}{(2\pi)^{3/2}}\,\mathrm{e}^{-\mathrm{i}\boldsymbol{k}_1\cdot\boldsymbol{x}}\,\hat{a}^{\dagger}_{\boldsymbol{k}_1,r}\right)\boldsymbol{\sigma}_{rr'}\left(\int\frac{\mathrm{d}^3\boldsymbol{k}_2}{(2\pi)^{3/2}}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}_2\cdot\boldsymbol{x}}\,\hat{a}_{\boldsymbol{k}_2,r'}\right)\hat{a}^{\dagger}_{\boldsymbol{k}'s'}\,\middle|\,0\right\rangle && (3.78)\\
&\quad= \boldsymbol{\sigma}_{rr'}\int\frac{\mathrm{d}^3\boldsymbol{k}_1}{(2\pi)^{3/2}}\int\frac{\mathrm{d}^3\boldsymbol{k}_2}{(2\pi)^{3/2}}\,\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_2-\boldsymbol{k}_1)\cdot\boldsymbol{x}}\left\langle 0\,\middle|\,\hat{a}_{\boldsymbol{k},s}\,\hat{a}^{\dagger}_{\boldsymbol{k}_1,r}\,\hat{a}_{\boldsymbol{k}_2,r'}\,\hat{a}^{\dagger}_{\boldsymbol{k}',s'}\,\middle|\,0\right\rangle\,. && (3.79)
\end{aligned}$$

The remaining expectation value can now be refactored by commutating the annihilators to the right via Eq. (3.52), such that

$$\left\langle 0 \,\middle|\, \hat{a}_{\boldsymbol{k},s}\, \hat{a}^{\dagger}_{\boldsymbol{k}_1,r}\, \hat{a}_{\boldsymbol{k}_2,r'}\, \hat{a}^{\dagger}_{\boldsymbol{k}',s'} \,\middle|\, 0 \right\rangle = \delta^3(\boldsymbol{k}_2 - \boldsymbol{k}')\, \delta^3(\boldsymbol{k}_1 - \boldsymbol{k})\, \delta_{r's'}\, \delta_{rs}\,. \tag{3.80}$$

Now inserting Eqs. (3.79) and (3.80) back into Eq. (3.77) finally yields the following matrix elements for the spinorial current operator,

$$\langle \boldsymbol{k}, s | \hat{\boldsymbol{j}}_{\text{spin}}(\boldsymbol{x}) | \boldsymbol{k}', s' \rangle = \frac{(-e)\hbar}{2m\,(2\pi)^3} \left(\mathrm{i}(\boldsymbol{k}' - \boldsymbol{k}) \times \boldsymbol{\sigma}_{ss'} \right) \mathrm{e}^{\mathrm{i}(\boldsymbol{k}' - \boldsymbol{k})\cdot\boldsymbol{x}}\,. \tag{3.81}$$

Hence, the i-th spatial component of the total current can be equivalently expressed by

$$(j_i)_{\boldsymbol{k},s;\boldsymbol{k}'s'}(\boldsymbol{x}) = \frac{(-e)\hbar}{2m\,(2\pi)^3} \left((2k_i + q_i)\, \delta_{ss'} + \mathrm{i}\epsilon_{ikl}\, q_k\, (\sigma_l)_{ss'} \right) \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{x}}\,, \tag{3.82}$$

where the replacement $\boldsymbol{q} = \boldsymbol{k}' - \boldsymbol{k}$ has been applied again.

By inserting the results for the matrix elements of the orbital (Eq. (3.74)) and spinorial current (Eq. (3.82)) into the master formula for the spectral representation (3.40), the Kubo formula (3.69) for the current-current response can now be re-expressed in real space as

$$\begin{aligned} \chi_{ij}(\boldsymbol{x}, \boldsymbol{x}'; \omega) = &- \frac{(-e)^2 n}{m}\, \delta_{ij}\, \delta^3(\boldsymbol{x} - \boldsymbol{x}') \\ &- \frac{(-e)^2 \hbar^2}{4m^2} \sum_{s,s'=\uparrow,\downarrow} \int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3} \int \frac{\mathrm{d}^3\boldsymbol{k}'}{(2\pi)^3}\, \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{x} - \boldsymbol{x}')}\, \frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}'))}{\hbar(\omega + \mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k}') - \varepsilon_0(\boldsymbol{k}))} \\ &\times \left((2k_i + q_i)\, \delta_{ss'} + \mathrm{i}\epsilon_{ikl}\, q_k (\sigma_l)_{ss'} \right) \left((2k_j + q_j)\, \delta_{ss'} - \mathrm{i}\epsilon_{jmn}\, q_m (\sigma_n^*)_{ss'} \right). \end{aligned} \tag{3.83}$$

Here, n has been inserted for the charge density $\rho(\boldsymbol{x}) \equiv (-e)n$, which for the free electron gas is constant by definition and explicitly given by

$$n = 2 \int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\, f(\varepsilon_0(\boldsymbol{k}))\,. \tag{3.84}$$

Its Fourier transform can then be extracted following the same steps as in the last section for the density response and reads

$$\begin{aligned} \chi_{ij}(\boldsymbol{q}, \omega) = &- \frac{e^2 n}{m}\, \delta_{ij} - \frac{e^2 \hbar^2}{4m} \sum_{s,s'=\uparrow,\downarrow} \int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\, \frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k} + \boldsymbol{q}))}{\hbar(\omega + \mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k} + \boldsymbol{q}) - \varepsilon_0(\boldsymbol{k}))} \\ &\times \left((2k_i + q_i)\, \delta_{ss'} + \mathrm{i}\epsilon_{ikl}\, q_k (\sigma_l)_{ss'} \right) \left((2k_j + q_j)\, \delta_{ss'} - \mathrm{i}\epsilon_{jmn}\, q_m (\sigma_n^*)_{ss'} \right). \end{aligned} \tag{3.85}$$

This is the searched for general expression for the current-current response tensor in Fourier space of the free, non-relativistic, homogeneous electron gas for the most general non-relativistic current density Eq. (3.1). As proved in [2, §3.2], this response is not only homogeneous but also strictly isotropic, i. e. can be decomposed by means of scalar longitudinal and transverse functions which additionally only depend on the modulus of the wavevector and not on

its direction. Algebraically this is reflected in the invariance under arbitrary rotations (see e. g. [8, App. D1] for a proof). Before preparing the current response tensor for numerical applications, we will shortly discuss each contribution separately in the following.

Diamagnetic part.—As a result of the explicit dependence of the current operator on the external vector potential, the diamagnetic part emerges as diagonal, frequency- and wavevector-independent, purely real-valued contribution,

$$\chi_{ij}^{\mathrm{dia}}(\boldsymbol{q},\omega) = -\frac{e^2 n}{m}\,\delta_{ij}\,. \tag{3.86}$$

By interpreting this non-interacting response function in context of the random phase approximation as a proper response and using the respective response relation (2.57), the corresponding conductivity tensor can be obtained,

$$\widetilde{\sigma}_{ij}(\boldsymbol{k},\omega) = -\frac{en/m}{\mathrm{i}(\omega+\mathrm{i}\eta)}\,\delta_{ij}\,, \tag{3.87}$$

where the constant density n should be regarded as a material specific parameter. In the conversion formula, the singularity has been regularized to make the retardation explicit here as well. Eq. (3.88) is a version of the famous London model which is used to describe superconductors. By replacing the infinitesimal regularization factor η by a finite so-called (inverse) relaxation time $1/\tau$, the following expression is obtained

$$\widetilde{\sigma}_{ij}(\boldsymbol{k},\omega) = -\frac{e^2 n\tau/m}{1-\mathrm{i}\omega\tau}\,\delta_{ij}\,, \tag{3.88}$$

This is known as the Drude conductivity for "electrical AC fields" and is frequently applied for metallic systems (cf. [45, §1]) as one of the simplest models that is still able to account for a variety of quantitative and qualitative feaures of this class of materials. Both models will be discussed in more detailed and from a linear response point of view in Chpt. 4 together with a proposition for a spinorial correction term.

Orbital part.—Instead of extracting the orbital contribution from Eq. (3.85), it can also be obtained by dropping the spinorial current in Eq. (3.65) as well as the diamagnetic part in the general Kubo formula, i. e. by calculating the current-current response the exact same way we did for the density-density response, using the standard Kubo expression. Since there are no spin-dependent terms left, the spin summation can be performed explicitly and yields again an overall factor of two in the resulting formula,

$$\chi_{ij}^{\mathrm{orb}}(\boldsymbol{q},\omega) = -\frac{e^2\hbar^2}{2m^2}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\,(2k_i+q_i)(2k_j+q_j)\,\frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}))}{\hbar(\omega+\mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}) - \varepsilon_0(\boldsymbol{k}))}\,. \tag{3.89}$$

By introducing the two auxiliary functions

$$\alpha_{ij}(\boldsymbol{q},\omega) = -\frac{e^2\hbar^2}{2m^2}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\, 4k_i k_j\, \frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}))}{\hbar(\omega+\mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}) - \varepsilon_0(\boldsymbol{k}))}\,, \tag{3.90}$$

$$\beta_i(\boldsymbol{q},\omega) = -\frac{e^2\hbar^2}{2m^2}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\, 2k_i\, \frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}))}{\hbar(\omega+\mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}) - \varepsilon_0(\boldsymbol{k}))}\,, \tag{3.91}$$

and with help of the Lindhard density response (3.60), the orbital contribution can be expressed by the concise formula

$$\chi^{\mathrm{orb}}_{ij}(\boldsymbol{q},\omega) = \alpha_{ij}(\boldsymbol{q},\omega) + q_i\,\beta_j(\boldsymbol{q},\omega) + \beta_i(\boldsymbol{q},\omega)\,q_j - \frac{\hbar^2}{4m^2}\,q_i\,q_j\,\chi(\boldsymbol{q},\omega)\,. \tag{3.92}$$

This is the general form of the orbital current response. The not explicitly known tensorial functions α_{ij} and β_{ij} are as characteristic for the homogeneous electron gas as is the Lindhard density response function χ. Later, we will show that these 12 scalar functions boil down to only 3 dimensionless numerical integrals, which can even be solved analytically for $T = 0\,\mathrm{K}$. The last term of this contribution can also be written more suggestively, such that

$$\overleftrightarrow{\chi}^{\mathrm{orb}}(\boldsymbol{q},\omega) = \overleftrightarrow{\alpha}(\boldsymbol{q},\omega) + \boldsymbol{q}\boldsymbol{\beta}^{\mathsf{T}}(\boldsymbol{q},\omega) + \boldsymbol{\beta}(\boldsymbol{q},\omega)\,\boldsymbol{q}^{\mathsf{T}} - \frac{\hbar^2|\boldsymbol{q}|^2}{4m^2}\,\chi(\boldsymbol{q},\omega)\,\overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{q}) \tag{3.93}$$

is obtained.

For the transverse part of this current response, a related calculation can be found in Ref. [32, §4.5], where it is also shown that the orbital and diamagnetic contribution together reproduce the Landau diamagnetism in the limit $|\boldsymbol{q}| \to 0$. The textbook by Dressel and Grüner [46] even analyze this transverse part at zero temperature to full extend. We will refer to their results later when discussing the spinorial contribution in detail.

Spinorial contribution.—We now turn to the actual "new" parts which cannot be found in standard textbooks, at least not in this form. The purely spinorial contribution to the current-current response results from those terms in Eq. (3.85) which exclusively involve the spinorial terms of the current operator matrix elements. As for the orbital contribution, the explicit expression can again be derived separately by dropping the orbital current in Eq. (3.65) or by simply reading it off the central result (3.85). Concretely, the spinorial current response is given by

$$\chi^{\mathrm{spin}}_{ij}(\boldsymbol{q},\omega) = \tag{3.94}$$
$$-\frac{e^2\hbar^2}{4m^2}\sum_{s,s'=\uparrow,\downarrow}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\,\epsilon_{ikl}\,\epsilon_{jmn}\,q_k\,q_m\,(\sigma_l)_{ss'}\,(\sigma^*_n)_{ss'}\,\frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}))}{\hbar(\omega+\mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}) - \varepsilon_0(\boldsymbol{k}))}\,.$$

In contrast to Eq. (3.89), the spin summation does not simply lead to an overall factor. Due to the spin-independence of both, the occupation number Eq. (3.42) and the dispersion relation (3.54), the spin summation only acts on the Pauli matrices. By exploiting the latter's

hermitizity and drawing on the algebraic identity

$$\sigma_i \sigma_j = \delta_{ij} \sigma_0 + \mathrm{i} \epsilon_{ijk} \sigma_k \,, \tag{3.95}$$

the factor containing the matrix product in spin basis boils down to a simple Kronecker delta,

$$\sum_{ss'} (\sigma_l)_{ss'} (\sigma_n^*)_{ss'} = \sum_{ss'} (\sigma_l)_{ss'} (\sigma_n)_{s's} = \sum_{s} (\sigma_l \sigma_n)_{ss} = \mathrm{Tr}_{\mathbb{C}^2} (\delta_{ln} \sigma_0 + \mathrm{i} \epsilon_{lnp} \sigma_p) = 2 \delta_{ln} \,, \tag{3.96}$$

where additionally the linearity of the trace operator and the trace freeness of the three Pauli matrices, $\mathrm{Tr}_{\mathbb{C}^2} \sigma_i = 0$ for $i = 1, 2, 3$, has been used. With this, Eq. (3.94) turns into

$$\chi_{ij}^{\mathrm{spin}}(\boldsymbol{q}, \omega) = -\frac{e^2 \hbar^2}{2m^2} \int \frac{\mathrm{d}^3 \boldsymbol{k}}{(2\pi)^3} \, (\delta_{ij} \delta_{km} - \delta_{im} \delta_{kj}) \, q_k \, q_m \, \frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k} + \boldsymbol{q}))}{\hbar(\omega + \mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k} + \boldsymbol{q}) - \varepsilon_0(\boldsymbol{k}))} \,, \tag{3.97}$$

which apparently can again be expressed in terms of the Lindhard response (3.60),

$$\chi_{ij}^{\mathrm{spin}}(\boldsymbol{q}, \omega) = -\frac{\hbar^2}{4m^2} \left(\delta_{ij} \, |\boldsymbol{q}|^2 - q_i \, q_j \right) \chi(\boldsymbol{q}, \omega) \,. \tag{3.98}$$

From this, it follows in particular that the spinorial current response is purely transverse, i. e.

$$q_i \, \chi_{ij}^{\mathrm{spin}}(\boldsymbol{q}, \omega) \, q_j = 0 \,, \tag{3.99}$$

which is consistent with the definition of the spin contribution to the current. This becomes even more clear when rewriting Eq. (3.98) in isotropic form,

$$\overleftrightarrow{\chi}^{\mathrm{spin}}(\boldsymbol{q}, \omega) = \chi^{\mathrm{spin}}(\boldsymbol{q}, \omega) \overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{q}) \,, \tag{3.100}$$

with

$$\chi^{\mathrm{spin}}(\boldsymbol{q}, \omega) = -\frac{\hbar^2 |\boldsymbol{q}|^2}{4m^2} \chi(\boldsymbol{q}, \omega) \,. \tag{3.101}$$

Interestingly, although the spin contribution adds only to the transverse part of the current response, its scalar part is proportional to the density response, which is by Eq. (2.93) connected to the longitudinal current response.

Using the standard relation Eq. (2.112), it can now be shown that from the spin contribution in the static limit,

$$\chi_{\mathrm{m}}^{\mathrm{spin}}(\boldsymbol{q}) = \frac{\mu_0}{|\boldsymbol{q}|^2} \lim_{\omega \to 0} \chi_{\mathrm{T}}^{\mathrm{spin}}(\boldsymbol{q}, \omega) = -\frac{\mu_0 \hbar^2}{4m^2} \chi(\boldsymbol{q}) \tag{3.102}$$

with

$$\chi(\boldsymbol{q}) \coloneqq \lim_{\omega \to 0} \chi(\boldsymbol{q}, \omega) = 2e^2 \int \frac{\mathrm{d}^3 \boldsymbol{k}}{(2\pi)^3} \, \frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k} + \boldsymbol{q}))}{\varepsilon_0(\boldsymbol{k}) - \varepsilon_0(\boldsymbol{k} + \boldsymbol{q})} \,. \tag{3.103}$$

the Pauli paramagnetism can be derived. For that reason, we additionally apply the long-wavelength limit to Eq. (3.102),

$$\lim_{|\boldsymbol{q}| \to 0} \chi(\boldsymbol{q}) = 2e^2 \int \frac{\mathrm{d}^3 \boldsymbol{k}}{(2\pi)^3} \, \frac{\mathrm{d} f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}))}{\mathrm{d} \varepsilon_0(\boldsymbol{k})} \tag{3.104}$$

which transforms the differences in the integrand into a differential quotient. By introducing the fundamental definition of the density of states *per volume* together with its counterpart in the thermodynamic limit (factor 2 arises because of spin degeneracy),

$$g(\omega) = \frac{1}{V}\sum_i \delta(\omega-\omega_i) \quad \to \quad g(\omega) = 2\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\,\delta(\omega-\omega_{\boldsymbol{k}})\,, \tag{3.105}$$

and with help of the general relation for any dispersion relation $f(\omega_{\boldsymbol{k}})$ (see e. g. [45, Eq. (2.60)]),

$$2\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\, f(\omega_{\boldsymbol{k}}) = \int \mathrm{d}\omega\; g(\omega)\, f(\omega)\,, \tag{3.106}$$

in combination with $\varepsilon_0(\boldsymbol{k}) = \hbar\omega_{\boldsymbol{k}}$, we ultimately find

$$\chi_\mathrm{m} \coloneqq \lim_{|\boldsymbol{q}|\to 0}\chi_\mathrm{m}(\boldsymbol{q}) = \frac{\mu_0\mu_\mathrm{B}^2}{\hbar}\int \mathrm{d}\omega\; g(\omega)\left(-\frac{\mathrm{d}f_{\beta,\mu}(\omega)}{\mathrm{d}\omega}\right)\,, \tag{3.107}$$

where the Bohr magneton is defined in Eq. (3.112). This is exactly the general (i. e. $T>0$) formula for the Pauli spin susceptibility (cf. e. g. [45, Eq. (31.67)]).

An alternative derivation starting from the spin-density response function can be found in [32, §4.4.1]. This simultaneously shows the crucial difference between the approach developed during the last sections and the one used in common literature. Although it is known that the Pauli paramagnetism follows from the Lindhard response, this is a result obtained from a properly defined spin-spin response function. This is in stark contrast to our approach, which relies on the inclusion of the *full*, i. e. most general non-relativistic current in *all* fundamental equations like the Kubo formula. This way, spin does not present some enigmatic "quantum source" of magnetism but instead is well covered by the Maxwell equations, where it enters as spinorial contribution to the current density. Consequently, these spin contributions should in prininciple be included in every derived fundamental formulae as well. This is in particular true for ab initio materials physics, where it could lead to presumably very small but nevertheless entirely new effects.

Spin-orbit cross correlation.—In contrast to the just discussed purely spinorial contribution, the causal correlator between spinorial and orbital contributions is—to best of our knowledge—indeed a paradigmatically new term that cannot be found yet in literature specialized on this topic. This spin-orbit cross correlation follows from the mixed terms in Eq. (3.85) and is concretely given by

$$\begin{aligned}\chi_{ij}(\boldsymbol{q},\omega) = \mathrm{i}\,\frac{e^2\hbar^2}{4m^2}\sum_{s=\uparrow,\downarrow}\int\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\Big((2k_i+q_i)\,\epsilon_{jmn}\,q_m(\sigma_n^*)_{ss} - \epsilon_{ikl}\,q_k\,(\sigma_l)_{ss}\,(2k_j+q_j)\Big)\\ \times\frac{f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) - f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}))}{\hbar(\omega+\mathrm{i}\eta) - (\varepsilon_0(\boldsymbol{k}+\boldsymbol{q}) - \varepsilon_0(\boldsymbol{k}))}\,.\end{aligned} \tag{3.108}$$

Since in the model of the free electron gas the occupation number does not depend on the spin, the summation over the spinorial index s again acts exclusively on the respective Pauli matrix, where it simply procuces its trace. The latter, however, vanishes identically as shown

before. Consequently, the spin-orbit cross correlation vanishes as well for the free electron gas, wheras this is certainly not the case for a *real* material, where occupation number and energy levels themselves are in general not identical for both spin channels.

Full current-current response tensor.—Using the results from the last paragraphs it is evident that the general current response as given by Eq. (3.85) can be simplifies further. In particular, the projector parts of the two terms involving the Lindhard response, one as part of the orbital contribution (3.93) and the other one provided by the spinorial part (3.98), surprisingly combine to identity. Hence, the full current response tensor can be expressed conveniently by the following concise formula,

$$\chi_{ij}(\boldsymbol{q},\omega) = -\frac{e^2 n}{m}\,\delta_{ij} + \alpha_{ij}(\boldsymbol{q},\omega) + q_i\,\beta_j(\boldsymbol{q},\omega) + \beta_i(\boldsymbol{q},\omega)\,q_j - \delta_{ij}\,\frac{\hbar^2|\boldsymbol{q}|^2}{4m^2}\,\chi(\boldsymbol{q},\omega)\,. \tag{3.109}$$

By following the same steps as for the mere spinorial part, i. e. by performing the limits $\omega \to 0$ and $|\boldsymbol{q}| \to 0$ consecutively, the corresponding *full* magnetic permeability constant turns out to be the sum of the Landau diamagnetism and the Pauli paramagnetism as result for the free, non-relativistic, homogeneous electron gas. This corroborates the central tenet of the Functional Approach to electrodynamics in media that all electromagnetic material properties have to be accessed from the microscopic charge and current densities and, in particular, that spin-generated magnetism should be treated—at least on the most fundamental level—by including a spinorial contribution to the electric current density. Moreover, we note that in the long wavelength limit $\boldsymbol{q} \to 0$ all $\boldsymbol{q}$-dependent terms tend to zero and thus only the diamagnetic term remains,

$$\chi_{ij}(\omega) = -\frac{e^2 n}{m}\,\delta_{ij}\,, \tag{3.110}$$

Hence, the electron gas is exclusively described by the Drude conductivity or (for vanishing dissipation) by the London conductivity (see Eqs. (3.87) and (3.88)).

In general, the characteristic functions of the free electron gas are α_{ij}, β_i and χ. As we will show in Sct. 5.1, the 12 scalar functions corresponding to the first two contribution can actually be reduced to only three parameter integrals. Although they cannot be expressed in terms of the Lindhard response χ, we will prove the "Lindhard Integral Theorem" stating that for $T = 0\,\mathrm{K}$, their associated fundamental integrals can be expressed through the one inherent to the Lindhard response.

In spite of the fact that the longitudinal current response can be obtained—at least in principle—from the density response by virtue of the response relation Eq. (2.93), the entire discussion is actually only relevant for the decoupled transverse part. However, as shown in our preprint [2, §3.2, p.28], Eq. (2.93) is in fact not valid anymore for the Lindhard response (3.60) and the general current response (3.85). Instead one has to revert to the respective Fourier pre-images w. r. t. the frequency variable. The reason for this seemingly contradiction lies in the subtle difficulties arising when working algebraically in the Fourier space. Simply speaking, in Fourier space one can always add a term proportional to Dirac delta without altering the real space result. This is because terms involving such Dirac delta distributions vanish identically under inverse Fourier trasformation (see also remarks in Sct. 4.1). Hence,

we will mostly restrict to the discussion of the transverse current response, in particular because the spinorial part is a purely transverse contribution.

3.4. Spin susceptibility vs. spinorial current response

An immediate result of the microscopic Maxwell equations in media as introduced in Sct. 2.1 is that the magnetization is always generated by a microscopic current as a matter of principle. Nevertheless, magnetism originating in spinorial degrees of freedom is usually treated entirely offside any electrodynamic field theory. Instead, for the Pauli paramagnetism of metals, for instance, the magnetization is introduced as the difference of the two spin-polarized charge densities (see e. g. [45, Eq. (31.55)], [47, §12.4.1] or [48, §15.3]),

$$M = -\mu_{\mathrm{B}} \left(n_{\uparrow} - n_{\downarrow} \right) , \tag{3.111}$$

where

$$\mu_{\mathrm{B}} = \frac{e\hbar}{2m} \tag{3.112}$$

denotes the Bohr magneton. As can be found in standard literature, this is a result of setting $\boldsymbol{B}_{\mathrm{ext}} = B_3 \, \boldsymbol{e}_3$ in the spinorial part of the matrix-valued Pauli interaction Hamiltonian (3.19), such that

$$\hat{H}_{\mathrm{spin}} = -\boldsymbol{\mu} \cdot \boldsymbol{B}_{\mathrm{ext}} \tag{3.113}$$

is replaced by the so-called "longitudinal" spin Hamiltonian

$$\hat{H}_{\mathrm{spin,L}} = -\mu_3 B_3 = \left(\mu_{\mathrm{B}} B_3 \right) \sigma_3 \, . \tag{3.114}$$

In the latter equations, the magnetic moment $\boldsymbol{\mu}$ is defined by (cf. [49, Eq. (36.62)])

$$\boldsymbol{\mu} = \frac{g_{\mathrm{e}}}{2} \frac{(-e)}{m} \boldsymbol{S} = -\frac{g_{\mathrm{e}}}{2} \, \mu_{\mathrm{B}} \boldsymbol{\sigma} \, , \tag{3.115}$$

with spin density

$$\boldsymbol{S} = \frac{\hbar}{2} \boldsymbol{\sigma} \, , \tag{3.116}$$

and Landé factor for electrons $g_{\mathrm{e}} \equiv 2$ (according to Dirac theory)[8]. As can best be seen from the useful identity

$$\boldsymbol{\sigma} \cdot \boldsymbol{B} = \begin{pmatrix} B_3 & B_1 - \mathrm{i} B_2 \\ B_1 + \mathrm{i} B_2 & -B_3 \end{pmatrix} , \tag{3.117}$$

fixing the magnetic field in z-direction decouples the Pauli Hamiltonian and leads, depending on the direction of the spin, to an energy shift $\Delta E = \pm \mu_{\mathrm{B}} B_3$ compared to the unperturbed system, which is known as Zeeman splitting[9]. Explicitly computing the expectation value of

[8] In fact, renormalized quantum electrodynamics shows that this Landé factor (a. k. a. gyromagnetic factor) for the electron is not exactly 2 as predicted by the Dirac equation. Instead, the famous US-american physicist Julian Seymour Schwinger (1918-1994) proved that including the electron-photon vertex in the calculation of the magnetic moment operator corrects this factor in first order to $g_{\mathrm{e}} = 2(1 + \alpha/\pi)$, where α is the finestructure constant (see e. g. [49, §41]).

the simplified spin Hamiltonian (3.114) in an N-electron state,

$$E = \langle \Psi^N | \hat{H}_{\mathrm{spin,L}} | \Psi^N \rangle = \mu_{\mathrm{B}} B_3 \sum_{i=1}^{N} \begin{pmatrix} \psi_\uparrow \\ \psi_\downarrow \end{pmatrix}^\dagger \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} \psi_\uparrow \\ \psi_\downarrow \end{pmatrix} = \mu_{\mathrm{B}} (N_\uparrow - N_\downarrow) B_3 \,, \tag{3.118}$$

and inserting the result into the frequently employed defining equation for the magnetization density (cf. [45, Eq. (31.1)])

$$M(B) = -\frac{1}{V} \frac{\partial E(B)}{\partial B} \,, \tag{3.119}$$

directly leads back to the initial expression (3.111). This calculation is usually not carried out explicitly in most textbooks. Instead they simply argue that every electron contributes to the magnetization density with $\pm\mu_{\mathrm{B}}$ depending on its spin direction, which then leads to the same result except for a possibly varying overall sign. After some elementary manipulations of the spin-polarized density of states and under usage of Eq. (3.84) in conjunction with Eq. (3.106) for the integral against the Fermi-Dirac distribution, the Pauli spin susceptibility as given by Eq. (3.107) is retained (cf. [45, p. 661f]).

A more general way to treat the spinorial magnetism is by introducing a spin-resolved number density operator,

$$\hat{n}_{ss'}(\boldsymbol{x}) = \hat{\psi}_s^\dagger(\boldsymbol{x}) \hat{\psi}_{s'}(\boldsymbol{x}) \,, \tag{3.120}$$

and a corresponding spin response function (see e. g. [32, §3.5] and especially [50] as reference for the numerical implementation as realized in the ELK code [31]),

$$n_{ss'}(\boldsymbol{x}, t) = \int \mathrm{d}t' \int \mathrm{d}^3\boldsymbol{x}' \sum_{rr'} \chi_{ss'rr'}(\boldsymbol{x}, \boldsymbol{x}'; t - t') V^{\mathrm{ext}}_{rr'}(\boldsymbol{x}', t') \,, \tag{3.121}$$

where the external potential energy is defined by

$$V^{\mathrm{ext}} = (-e\varphi)\, \mathbb{1}_{2\times 2} + \mu_{\mathrm{B}} \boldsymbol{\sigma} \cdot \boldsymbol{B} = \begin{pmatrix} -e\varphi + \mu_{\mathrm{B}} B_3 & \mu_{\mathrm{B}}(B_1 - \mathrm{i}B_2) \\ \mu_{\mathrm{B}}(B_1 + \mathrm{i}B_2) & -e\varphi - \mu_{\mathrm{B}} B_3 \end{pmatrix} . \tag{3.122}$$

With the latter, part of the second-quantized version of the interaction Hamiltonian (3.19) can be equivalently expressed as

$$\hat{H}_{\mathrm{ext}} = \int \mathrm{d}^3\boldsymbol{x} \sum_{s,s'=\uparrow,\downarrow} V^{\mathrm{ext}}_{ss'}(\boldsymbol{x}) \hat{n}_{ss'}(\boldsymbol{x}) \,. \tag{3.123}$$

The Kubo formula for the retarded spin-spin response then states

$$\chi_{ss'rr'}(\boldsymbol{x}, \boldsymbol{x}', t - t') = -\frac{\mathrm{i}}{\hbar} \Theta(t - t') \left\langle [\hat{n}_{ss'}(\boldsymbol{x}, t), \hat{n}_{rr'}(\boldsymbol{x}', t')] \right\rangle \,. \tag{3.124}$$

[9] Named after the Dutch physicist Pieter Zeeman (1865-1943), who shared the 1902 Nobel Prize in Physics with Hendrik Lorentz for the discovery of this effect.

By re-identifying Eq. (3.37) with

$$\hat{n}(\boldsymbol{x}) = \sum_{s=\uparrow,\downarrow} \hat{\psi}_s^\dagger(\boldsymbol{x})\,\hat{\psi}_s(\boldsymbol{x}) = \sum_{s=\uparrow,\downarrow} \hat{n}_{ss}(\boldsymbol{x}) = \hat{n}_{\uparrow\uparrow}(\boldsymbol{x}) + \hat{n}_{\downarrow\downarrow}(\boldsymbol{x})\,, \tag{3.125}$$

and using $\hat{n}_s \equiv \hat{n}_{ss}$ and $\chi_{rs} \equiv \chi_{rrss}$ as abbreviations, it is clear that because of the expansion

$$[\hat{n}(\boldsymbol{x}), \hat{n}(\boldsymbol{x}')] = [\hat{n}_\uparrow(\boldsymbol{x}), \hat{n}_\uparrow(\boldsymbol{x}')] + [\hat{n}_\downarrow(\boldsymbol{x}), \hat{n}_\downarrow(\boldsymbol{x}')] + [\hat{n}_\uparrow(\boldsymbol{x}), \hat{n}_\downarrow(\boldsymbol{x}')] + [\hat{n}_\downarrow(\boldsymbol{x}), \hat{n}_\uparrow(\boldsymbol{x}')]\,, \tag{3.126}$$

a summation over all spin pairs exactly reproduces the already familiar density-density response (cf. [32, Eq. (3.201)]),

$$\chi_{nn}(\boldsymbol{x}, \boldsymbol{x}', t - t') = \sum_{s,s'=\uparrow,\downarrow} \chi_{ss'}(\boldsymbol{x}, \boldsymbol{x}', t - t')\,, \tag{3.127}$$

where χ_{nn} refers to the *number density* response.[10]

On the other hand, the so-called *longitudinal* spin-spin response with the associated Kubo formula

$$\chi_{\mu_3,\mu_3}(\boldsymbol{x}, \boldsymbol{x}', t - t') = -\frac{\mathrm{i}}{\hbar}\,\Theta(t - t')\,\big\langle[\hat{\mu}_3(\boldsymbol{x}, t), \hat{\mu}_3(\boldsymbol{x}', t')]\big\rangle\,, \tag{3.128}$$

corresponds to the simplified paramagnetic case from the beginning of this section. This can best be seen from the electromagnetic spin density operator in second quantization,

$$\hat{\boldsymbol{\mu}}(\boldsymbol{x}, t) = \frac{(-e)}{m}\,\hat{\boldsymbol{S}}(\boldsymbol{x}, t) = \frac{(-e)}{m}\,\hat{\Psi}^\dagger(\boldsymbol{x}, t)\left(\frac{\hbar}{2}\,\boldsymbol{\sigma}\right)\hat{\Psi}(\boldsymbol{x}, t)\,, \tag{3.129}$$

with z-component

$$\hat{\mu}_3 = \frac{(-e)\hbar}{2m}\begin{pmatrix}\hat{\psi}_\uparrow\\ \hat{\psi}_\downarrow\end{pmatrix}^\dagger\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}\begin{pmatrix}\hat{\psi}_\uparrow\\ \hat{\psi}_\downarrow\end{pmatrix} = -\mu_\mathrm{B}\,(\hat{n}_\uparrow - \hat{n}_\downarrow)\,. \tag{3.130}$$

Expanding the corresponding commutator as before yields

$$\frac{1}{\mu_\mathrm{B}^2}\,[\hat{\mu}_3(\boldsymbol{x}), \hat{\mu}_3(\boldsymbol{x}')] = [\hat{n}_\uparrow(\boldsymbol{x}), \hat{n}_\uparrow(\boldsymbol{x}')] + [\hat{n}_\downarrow(\boldsymbol{x}), \hat{n}_\downarrow(\boldsymbol{x}')] - [\hat{n}_\uparrow(\boldsymbol{x}), \hat{n}_\downarrow(\boldsymbol{x}')] - [\hat{n}_\downarrow(\boldsymbol{x}), \hat{n}_\uparrow(\boldsymbol{x}')]\,, \tag{3.131}$$

such that it is easy to verify that Eq. (3.128) can again be expressed in terms of the more general spin-spin response (3.124) by (cf. [32, Eq. (3.203)])

$$\chi_{\mu_3,\mu_3}(\boldsymbol{x}, \boldsymbol{x}', t - t') = \mu_\mathrm{B}^2 \sum_{s,s'=\uparrow,\downarrow} ss'\,\chi_{ss'}(\boldsymbol{x}, \boldsymbol{x}', t - t')\,, \tag{3.132}$$

where $s = 1$ maps to spin-up ($\uparrow$) and $s = -1$ maps to spin-down ($\downarrow$) as factors outside of indices. For non-interacting systems the two spin channels are entirely decoupled, i. e. the mixed-spin density commutators in Eq. (3.131) vanish and therefore the longitudinal spin-spin

[10] The *number density* response χ_{nn} from this section differs from the *charge density* response χ defined in Eq. (2.85) exactly by the squared elementary charge, i. e. $\chi = (-e)^2\chi_{nn}$.

response is (up to a constant prefactor) identical to the density-density response,

$$\chi^{(0)}_{\mu_3,\mu_3} \equiv \mu_{\mathrm{B}}^2 \chi_{nn} \,. \tag{3.133}$$

In general, however, only the additional symmetry $\chi_{\uparrow\downarrow} \equiv \chi_{\downarrow\uparrow}$ is present and Eq. (3.133) does not apply. Additionally, one can also introduce the mixed spin-density response (cf. [32, Eq. (3.204)]),

$$\chi_{n,\mu_3} = \chi_{\mu_3,n} = -\mu_{\mathrm{B}} \sum_{s,s'=\uparrow,\downarrow} s' \, \chi_{ss'} \,, \tag{3.134}$$

which vanish as well in the so-called paramagnetic state where $\chi_{\uparrow\uparrow} \equiv \chi_{\downarrow\downarrow}$.

There is yet another so-called *transverse* spin-response which corresponds to a magnetic field perpendicular to the direction of the static spin polarization. In this case, the spinorial Hamiltonian is often written as (cf. [32, Eq. (3.209)] or [50, Eq. (12)])

$$\hat{H}_{\mathrm{spin,T}} = -\frac{1}{2} \int \mathrm{d}^3\boldsymbol{x} \left(\hat{\mu}_+(\boldsymbol{x}) \, B_-(\boldsymbol{x}) - \hat{\mu}_-(\boldsymbol{x}) \, B_+(\boldsymbol{x}) \right) , \tag{3.135}$$

where the occuring fields are defined by

$$\hat{\mu}_\pm \overset{\mathrm{def}}{=} \hat{\mu}_1 \pm \mathrm{i}\hat{\mu}_2 \,, \tag{3.136}$$

$$B_\pm \overset{\mathrm{def}}{=} B_1 \pm \mathrm{i}B_2 \,. \tag{3.137}$$

Consequently, Eq. (3.135) expands to

$$\hat{H}_{\mathrm{spin,T}} = -\int \mathrm{d}^3\boldsymbol{x} \left(\hat{\mu}_1(\boldsymbol{x}) \, B_1(\boldsymbol{x}) + \hat{\mu}_2(\boldsymbol{x}) \, B_2(\boldsymbol{x}) \right) , \tag{3.138}$$

which is by

$$\hat{\mu}_1 \, B_1 + \hat{\mu}_2 \, B_2 = -\mu_{\mathrm{B}} \begin{pmatrix} 0 & B_1 - \mathrm{i}B_2 \\ B_1 + \mathrm{i}B_2 & 0 \end{pmatrix} \tag{3.139}$$

equivalent to the off-diagonal part of the general Pauli spin Hamiltonian Eq. (3.113). The corresponding retarded transverse spin response defined by

$$\chi_{\pm\mp}(\boldsymbol{x},\boldsymbol{x}',t-t') = -\frac{\mathrm{i}}{2\hbar} \, \Theta(t-t') \left\langle [\hat{\mu}_\pm(\boldsymbol{x},t), \hat{\mu}_\mp(\boldsymbol{x}',t')] \right\rangle \,, \tag{3.140}$$

with

$$\hat{\mu}_+(\boldsymbol{x}) = \frac{(-e)\hbar}{m} \, \hat{n}_{\uparrow\downarrow}(\boldsymbol{x}) \,, \tag{3.141}$$

$$\hat{\mu}_-(\boldsymbol{x}) = \frac{(-e)\hbar}{m} \, \hat{n}_{\downarrow\uparrow}(\boldsymbol{x}) \,, \tag{3.142}$$

can then be converted to the spin-spin response by virtue of

$$\chi_{+-}(\boldsymbol{x},\boldsymbol{x}',t-t') = 2\mu_{\mathrm{B}}^2 \, \chi_{\uparrow\downarrow\downarrow\uparrow}(\boldsymbol{x},\boldsymbol{x}',t-t') \,, \tag{3.143}$$

and

$$\chi_{-+}(\boldsymbol{x},\boldsymbol{x}',t-t') = 2\mu_{\mathrm{B}}^2\,\chi_{\downarrow\uparrow\uparrow\downarrow}(\boldsymbol{x},\boldsymbol{x}',t-t')\,. \tag{3.144}$$

Simultaneously, these two are the only remaining non-zero spin response functions since $\chi_{\mu_1\mu_2} = -\chi_{\mu_2\mu_1}$ and therefore $\chi_{++} \equiv \chi_{--} \equiv 0$.

Combining the discussion for longitudinal and transverse spin-spin response proves, that the entire magnetic response is covered by Pauli spin Hamiltonian Eq. (3.113). Comparing this to the approach followed in the preceding sections which eventuallly led to the spinorial current contribution (3.24), we immediately find the connection

$$\hat{\boldsymbol{j}}_{\mathrm{s}}(\boldsymbol{x},t) = -\mu_{\mathrm{B}}\nabla\times\left(\hat{\Psi}^{\dagger}(\boldsymbol{x},t)\,\boldsymbol{\sigma}\,\hat{\Psi}(\boldsymbol{x},t)\right) = (\nabla\times\hat{\boldsymbol{\mu}})(\boldsymbol{x},t)\,. \tag{3.145}$$

This current can now be treated equivalently to any other one and consequently leads to a magnetization in the very same manner. More precisely, the spinorial current contribution induced by an externally applied magnetic field leads to a spinorial magnetization $\boldsymbol{M}_s$, just like the orbital current does for the so-called orbital magnetization (see Sct. 3.1). Hence, instead of regarding spin as a separate source of magnetism, it can be included into the common field theoretical framework by virtue of the above identification. Even a potentially constant spin density allegedly violating $\nabla\cdot\boldsymbol{M}_s = 0$ (see Sct. 2.1) would not pose an inherent problem of this approach since $j_s = \langle\hat{j}_s\rangle$ is a purely transverse field and therefore produces a purely transverse magnetization. We show this, for the sake of simplicitly, on the example of a *static* spin density $\boldsymbol{S}(\boldsymbol{x})$ (which is in fact already more general than a merely static one). The vector potential generated by the corresponding spinorial current reads

$$\boldsymbol{A}_{\mathrm{ind}}(\boldsymbol{x}) = \frac{\mu_0}{4\pi}\int \mathrm{d}^3\boldsymbol{x}'\,\frac{\boldsymbol{j}_{\mathrm{s}}(\boldsymbol{x}')}{|\boldsymbol{x}-\boldsymbol{x}'|}\,, \tag{3.146}$$

which by $\boldsymbol{B} = \nabla\times\boldsymbol{A}$ translates to the following magnetic field,

$$\boldsymbol{B}_{\mathrm{ind}}(\boldsymbol{x}) = \mu_0\nabla\times\left(\frac{1}{4\pi}\int \mathrm{d}^3\boldsymbol{x}\,'\frac{\nabla\times\boldsymbol{\mu}(\boldsymbol{x}')}{|\boldsymbol{x}-\boldsymbol{x}'|}\right)\,. \tag{3.147}$$

Comparing this result with the partitioning of fields according to the Helmholtz vector theorem (see App. B.5) reveals

$$\boldsymbol{M}_{\mathrm{s}}(\boldsymbol{x}) = \boldsymbol{\mu}_{\mathrm{T}}(\boldsymbol{x}) \tag{3.148}$$

and therefore

$$\nabla\cdot\boldsymbol{M}_{\mathrm{s}}(\boldsymbol{x}) = 0\,. \tag{3.149}$$

For time-dependent fields, an equivalent result can be shown using the formalism introduced in Sct. 1.2. Moreover, we note that the spinorial current does not lead to a charge transport. This can easily be verified with the continuity equation,

$$\partial_t\rho_{\mathrm{s}}(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{j}_{\mathrm{s}}(\boldsymbol{x},t) = 0\,, \tag{3.150}$$

where the identity $\nabla \cdot (\nabla \times \boldsymbol{C})(\boldsymbol{x}, t) \equiv 0$ for arbitrary vector fields $\boldsymbol{C}(\boldsymbol{x}, t)$ has been used.

Coming back to the difference between a genuine spin-susceptibility $\chi_{ss'rr'}$ and the spinorial part of the current response $\chi_{ij}^{\mathrm{spin}}$, we can say the following: The combined longitudinal and transverse spin-response emerges from the *full* Pauli spin term in the interaction Hamiltonian Eq. (3.113). Simultaneously, this is the starting point from which the (paramagnetic) spinorial current operator (3.67) is derived in Sct. 3.3 via Eq. (3.65), which eventually enters the Kubo formula (3.69) for the current-current response. The resulting tensor can then be expanded into different contributions, one of which is the purely transverse spinorial contribution $\chi_{ij}^{\mathrm{spin}}$. In the static limit, the latter then reverts to the Pauli spin susceptibility, which is also connected to the longitudinal spin-spin response. For non-interacting systems (and only in this particular case), both response functions are then related by

$$\chi^{\mathrm{spin}}(\boldsymbol{q}, \omega) = -|\boldsymbol{q}|^2 \chi^{(0)}_{\mu_3\mu_3}(\boldsymbol{q}, \omega)\,. \tag{3.151}$$

This can be read-off directly from Eq. (3.101), and with Eq. (3.102) we further find

$$\chi_{\mathrm{m}}^{\mathrm{spin}}(\boldsymbol{q}) = -\mu_0 \chi^{(0)}_{\mu_3\mu_3}(\boldsymbol{q})\,, \tag{3.152}$$

in the static limit.

Recapitulating all response relations from this section we can with certainty that χ^{spin} and $\chi_{\mu_3\mu_3}$ do indeed present different response functions and must not be confused with each other.

4. London model and diamagnetic response

In this chapter, the famous phenomenological London model of superconductivity (and by association the Drude model for metals), is interpreted in the framework of linear response theory. This way it becomes apparent that the London model with its completely isotropic, instantaneous and local current response tensor represents the simplest possible approximation for any material w. r. t. electrodynamics apart from the vacuum case (i. e. no material at all). In particular, the current response of the London model can be identified with the diamagnetic contribution to the response tensor of the non-relativistic, free and homogeneous electron gas.

Sct. 4.1 starts with a short discussion of the main features and some mathematical intricats inherent to this model and in particular includes the standard derivation of the Meißner-Ochsenfeld effect. In Sct. 4.2, an alternative derivation is presented based on the universal response relations from Sct. 2.3. An important aspect there is the distinction between proper and direct response functions: While the direct magnetic susceptibility predict a perfect diamagnet in the combined static and long-wavelength limit, its proper counterpart tends to negative infinity under identical conditions. Optical properties in form of longitudinal and transverse electromagnetic dispersion relations are discussed as well in this section.

Finally, in Sct. 4.3 the London response is used as basis for a spin-corrected and self-consistent toy model in order to anticipate the central result of this thesis regarding the impact of the spin-contribution on optical and magnetic properties.

4.1. Interpretation as response function

According to the general microscopic wave equation (2.117), a current response equal to zero (and consequently a vanishing fundamental response tensor) corresponds to the vacuum case, where no material is present at all. Following the spirit of the Lindhard theory (i. e. reinterpreting the direct response function of a free system as an approximation for the proper response function of the interacting system), the most simple "material toy model" is hence obtained by setting the Fourier representation of the *proper* current response to a constant:

$$\overleftrightarrow{\widetilde{\chi}}(\boldsymbol{k},\omega) = \widetilde{\chi}_0 \overleftrightarrow{\mathbb{1}} \,. \tag{4.1}$$

As a tensor, $\widetilde{\chi}_{ij}$ is then proportional to Kronecker delta. Further, this simplification does not only lead to a strictly isotropic (see Sct. 2.5), but even to a "completely" isotropic tensor, i. e. its longitudinal and transverse parts fulfill the condition

$$\widetilde{\chi}_{\mathrm{T}}(\boldsymbol{k},\omega) = \widetilde{\chi}_{\mathrm{L}}(\boldsymbol{k},\omega) \equiv \widetilde{\chi}_0 \,. \tag{4.2}$$

According to the stiffness theorem, static response functions $\chi_{AB}(\boldsymbol{k}, \omega = 0)$ are always required to be negative [32, p. 174]. Therefore, the constant $\widetilde{\chi}_0$ must not only be purely real because of the reality condition for Fourier transforms, $\chi_{AB}(-\boldsymbol{k}) = \chi^*_{AB}(\boldsymbol{k})$ (see App. B.3), but must be negative as well.

Although this kind of approximation almost seems bizarre in regard of the extensive preceding discussion in Part I, where the current reponse was introduced as the most fundamental of all response functions, this drastic simplification leads to a model which is astonishingly well capable of reproducing phenomenological materials behavior as we will see throughout this section.

First we notice that this very approximation already appears in the famous London model, which is, next to the Landau-Ginsburg phase transition and the Nobel Prize-winning BCS theory, one of the three models typically employed to describe the characteristic phenomena of superconductivity. The essential statement proposed by the London brothers[1] is that for superconducters, the response law

$$\boldsymbol{j} = -\frac{ne^2}{m}\,\boldsymbol{A}\,, \tag{4.3}$$

relating the current density to the vector potential (cf. [53, Eq. (12.99)]), should *replace* Ohm's law, which by contrast relates the current density to the electric field. In this particular case, "Ohm's law" refers to the phenomenological constitutive law $\boldsymbol{j} = \sigma \boldsymbol{E}$, with scalar and constant conductivity σ, which has been successfully used for decades to describe ordinary conductors at constant temperature. As of today, it is still employed as necessary condition for an electric resistor to qualify as "ohmic" (see e. g. [54, V3/§1.2.1]). Being only a very simplistic limiting case, this should not be confused with the fundamental version of Ohm's law given by Eq. (2.56).

Obviously, Eq. (4.3) coincides with our initial suggestion (4.1) for a toy model under the identification of $\widetilde{\chi}_0$ with the negative inverse of the so-called London coefficient Λ,

$$\widetilde{\chi}_0 = -\frac{1}{\Lambda} \overset{\text{def}}{=} -\frac{nq^2}{m}\,, \tag{4.4}$$

where m is the electron mass, $q = (-e)$ the charge of an electron and n the (constant) electronic number density.[2] The latter expression, in turn, matches exactly the diamagnetic contribution to the current response tensor of the free electron gas, Eq. (3.86). Hence, it is not surprising that the London model despite its simplicity is perfectly capable of describing all substantial characteristics of a so-called "ideal diamagnet".

Aside from that, Eq. (4.3) can also be recast into a form that fits better into the context

[1] The London model is named after the German brothers and Fritz (1900-1954) and Heinz (1907-1970) London, who developed the constitutive relations (4.7) and (4.8) in 1935 as phenomenological explanation of the Meißner-Ochsenfeld effect in superconducters [51]. The latter, in turn, is named after the German experimentalists W. Meißner (1882-1974) and R. Ochsenfeld (1901-1993), who discovered the said effect only two years earlier in 1933 [52].

[2] In fact, BCS theory shows that (at least for conventional superconducters) the "superconducting charge carriers" are not mere electrons but so called "Cooper pairs", i. e. bound pairs of two electrons (fermions with spin ½) that form a composite boson (integer spin 0 $\hat{=}$ singlet state or 1 $\hat{=}$ triplet state)—an effect attributed to electron-phonon interaction. This leads to a different interpretation of the quantities involved in the Landau coefficient, each differing exactly by a factor two in such a way, that these additional factors just cancel each other. [55, §13.3.1]

of linear response theory. More concretely, it should mimic Eq. (2.36), where the current at a specific point in spacetime depends on the entire external vector potential. For the London conductivity, this translates to

$$\boldsymbol{j}(\boldsymbol{x},t) = c\int \mathrm{d}t' \int \mathrm{d}^3\boldsymbol{x}' \left(-\frac{ne^2}{m}\right) \delta^3(\boldsymbol{x}-\boldsymbol{x}')\,\delta(ct-ct')\,\boldsymbol{A}(\boldsymbol{x}',t')\,. \tag{4.5}$$

The corresponding response tensor in real space can now easily be read-off as

$$\overleftrightarrow{\widetilde{\chi}}(\boldsymbol{x},t;\boldsymbol{x}',t') = -\frac{ne^2}{m}\,\delta^3(\boldsymbol{x}-\boldsymbol{x}')\,\delta(ct-ct')\,, \tag{4.6}$$

which is according to Eq. (B.106) consistent with the inverse Fourier transform of our initial proposition (4.1). Physically, Eq. (4.6) implies the following crucial properties of the London current response:

(i) Locality, i. e. the induced effect generated by a perturbative field at a specific spatial point is limited to this very point.

(ii) Instantaneousness, i. e. there is no delay in the material's response to an external perturbation.

Compared to the general form of a homogeneous response function in real space, these two conditions again constitute the most primitive approximation.

By respectively applying the curl or the time derivative to the response law in Eq. (4.3) and inserting the matching connection between field and (vector) potential (Eqs. (1.5) and (1.6) with $\varphi \equiv 0$), the following alternative statements can be deduced,

$$\partial_t \boldsymbol{j}(\boldsymbol{x},t) = +\frac{ne^2}{m}\,\boldsymbol{E}(\boldsymbol{x},t)\,, \tag{4.7}$$

$$\nabla\times\boldsymbol{j}(\boldsymbol{x},t) = -\frac{ne^2}{m}\,\boldsymbol{B}(\boldsymbol{x},t)\,, \tag{4.8}$$

which relate the current density to the electric and magnetic fields. In standard textbooks, the latter two constitutive laws are usually introduced as first and second London equation (cf. e. g. [47, Eqs. (13.3.2) and (13.3.4)]). Fourier transforming both of them yields

$$\mathrm{i}\omega\,\boldsymbol{j}(\boldsymbol{k},\omega) = \left(-\frac{ne^2}{m}\right)\boldsymbol{E}(\boldsymbol{k},\omega)\,, \tag{4.9}$$

$$\mathrm{i}\boldsymbol{k}\times\boldsymbol{j}(\boldsymbol{k},\omega) = \left(-\frac{ne^2}{m}\right)\boldsymbol{B}(\boldsymbol{k},\omega)\,, \tag{4.10}$$

which directly shows that the two London equations are not independent but consistent with Faraday's law, Eq. (1.66). Further, by inserting once more the relation between the vector potential and the electric field, Eq. (1.35), the Fourier space equivalent of the initial London equation (4.3) is retained. This confirms again our Gauge Claim, that the temporal gauge has to be considered as the preferred one for electrodynamics in media and especially for the London model, contrary to what is stated in Ref. [56], fn. 9 on p. 6.

As mentioned earlier, the basic idea for superconducters was to replace the *phenomenological* Ohm law with the London equation(s). Fundamentally, however, the *microscopic* version of Ohm's law as given by Eq. (2.56) is always valid (at least within linear response theory) and thus leads to an associated London conductivity tensor that can be read-off directly from Eq. (4.9) and reads in Fourier space

$$\overleftrightarrow{\widetilde{\sigma}}(\boldsymbol{k},\omega) = \widetilde{\sigma}(\omega)\overleftrightarrow{\mathbb{1}}\,, \tag{4.11}$$

with the scalar function

$$\widetilde{\sigma}(\omega) = \frac{\widetilde{\chi}_0}{\mathrm{i}\omega} = -\frac{ne^2}{\mathrm{i}\omega m}\,. \tag{4.12}$$

The same result is obtained when transforming the current response tensor directly into the conductivity with help of the universal response relation (2.57), which is also valid for the proper versions of the respective tensors.

At this point, one can draw at least three remarkable conclusions about the proper London conductivity:

(i) It does not depend on the wavevector $\boldsymbol{k}$, but only on the frequency ω,

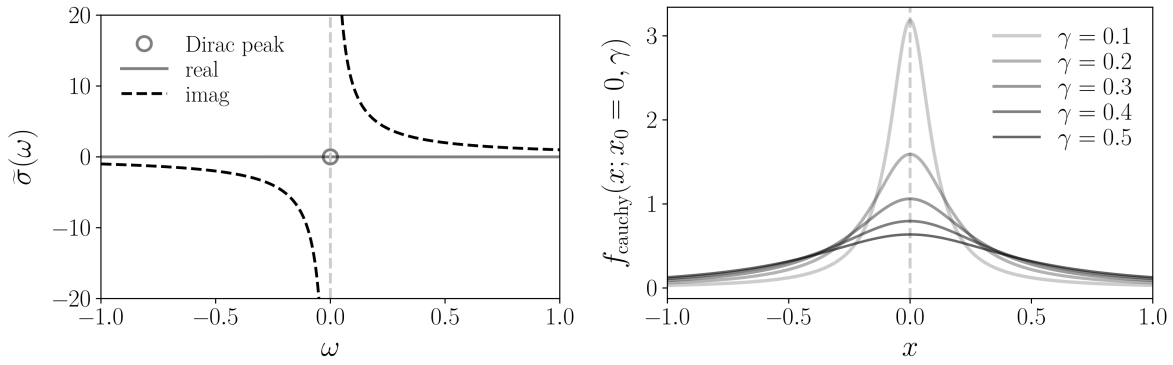
(ii) As a tensor, it is still proportional to identity and thus again "completely" isotropic with $\widetilde{\sigma}(\omega) \equiv \widetilde{\sigma}_{\mathrm{L}}(\omega) = \widetilde{\sigma}_{\mathrm{T}}(\omega)$,

(iii) It is purely imaginary.

The success of the London model can already be surmised from these observations, because they reflect some essential materials characteristics of superconducters: The first point seems to be a general bulk property at least for optical wavelengths as can be shown from ab initio calculations. Hence, this hypothesis is also one of the central conjectures in our recent publication [1], where we derive wavevector-dependent optical materials properties from wavevector-independent, ab initio calculated conductivity tensors. The key concept which makes this post-processing possible in the first place is based on the assumption that, although a specific response function may be independent of the wavelength, this property can—according to the universal response relations—not be upheld for all response functions simultaneously. This applies to the London conductivity as well as we will notice in the discussion of the magnetic susceptibility in the next section.

Since we did not assume a specific macroscopic structure like thin films or other complicated surfaces, but rather want to describe *bulk effects* of superconducters like the zero resistance which this material class shows independently of the direction, it is reasonable that this isotropy is also reflected in the conductivity tensor. As discussed in Sct. 2.5, being isotropic is, however, not equal to being completely independent from the wavevector argument. In general, there should still remain a dependence on the absolute value of $\boldsymbol{k}$, at least outside the optical regime. This is a feature which lies beyond the capability of the London model but has already been studied by other means [57, 58].

Regarding the last statement in the above list, one has to take special care w. r. t. the distributional character of response functions in Fourier space. As discussed in App. B.3, for



(a) London conductivity **(b)** Cauchy probability density

Fig. 4.1.: In (a), the London conductivity as given by Eq. (4.12) and normalized to $|\widetilde{\chi}_0| = \frac{ne^2}{m}$ is shown. The imaginary part tends to $\pm\infty$ for $\omega \to \pm 0$, whereas the real part is zero except for $\omega = 0$, where it shows a Dirac peak as analyzed in Eq. (4.17). The latter results from the limiting case $\eta \to 0$ in Eq. (4.14) and has the form of a (renormalized) Cauchy distribution (4.15), which in turn is plotted in (b) for different scaling parameters γ. Furthermore, the London conductivity satisfies the reality conditions stated in App. B.3, i. e. its real part is an even function of ω, whereas its imaginary part is odd.

retarded response functions the frequency has to be replaced according to $\omega \mapsto \omega + \mathrm{i}\eta$, and afterwards the limit $\eta \to 0$ has to be taken. Applying the same regularization procedure to the scalar conductivity (4.12) leads to the equivalent form

$$\widetilde{\sigma}(\omega) = \widetilde{\chi}_0 \lim_{\eta \to 0} \frac{\mathrm{i}}{\omega + \mathrm{i}\eta} \tag{4.13}$$

$$= \widetilde{\chi}_0 \lim_{\eta \to 0} \left(\frac{\eta}{\omega^2 + \eta^2} + \mathrm{i} \frac{\omega}{\omega^2 + \eta^2} \right) , \tag{4.14}$$

where in the last step, real and imaginary parts have been separated by complex expansion. Now it is obvious that conclusion (iii) is not true until the limit $\eta \to 0$ is actually executed. By analyzing the complex fraction in Eq. (4.13) further, one finds that its real part represents a (renormalized) Cauchy distribution, i. e. a function of the form

$$f(x; x_0, \gamma) = \frac{1}{\pi\gamma} \frac{\gamma^2}{(x - x_0)^2 + \gamma^2} = -\frac{1}{\pi} \operatorname{Re} \left(\frac{i}{x - x_0 + \mathrm{i}\gamma} \right) . \tag{4.15}$$

Here, γ is a scaling parameter specifying the half-width at half-maximum (HWHM) and x_0 is the location parameter that determines the point where the curve is centered at. Continuously decreasing γ for such a Lorentzian while retaining a constant integration value yields a so-called Dirac series, which eventually culminates in the Dirac delta distribution (cf. [59, §10.1]),

$$\lim_{\gamma \to 0} f(x, x_0, \gamma) = \delta(x - x_0) , \tag{4.16}$$

as visualized in Fig. 4.1b. Hence, performing the limit $\eta \to 0$ in Eq. (4.14) explicitly leads to

a distributional though non-vanishing real part for the conductivity,[3]

$$\widetilde{\sigma}(\omega) = \frac{\pi n e^2}{m}\,\delta(\omega) + \mathrm{i}\,\frac{n e^2}{m\omega}\,, \tag{4.17}$$

which apparently seems to be in conflict with what we found earlier in Eq. (4.12) by simply dividing the current response by a factor $\mathrm{i}\omega$. This supposed contradiction is due to the fact that one cannot simply apply all conventional algebraic transformations when working with distributions in Fourier space. Instead, one rather has to understand that e. g. dividing by $\mathrm{i}\omega$ is the inverse of multiplying with this factor. While the latter maps to a time derivative in real space, the said division consequently corresponds to an integration in real space and can thus always be complemented by an addition of a constant term times Dirac delta. Multiplying again with $\mathrm{i}\omega$ yields exactly the original expression because of the algebraic identity $\omega\,\delta(\omega) = 0$. The same result could have been shown using the distributional Sokhotski-Plemelj identity (B.109).

On physical grounds, this seemingly purely mathematical fact leads to an important qualitative change: For static electric fields, i. e. fields with a frequency equal to zero, we now find an infinitely large conductivity, or in other words, a so-called zero electrical "DC resistance" as expected for a perfect conductor. By contrast, for real AC fields (i. e. $\omega \neq 0$), the conductivity is indeed purely imaginary as stated in conclusion (iii) on p. (iii).

Below the critical temperature and for sufficiently small currents and fields (as assumed in linear response), the second important characteristic of a superconducter besides the zero DC resistance (which simultaneously aparts them from mere perfect conductors)[4] is the famous Meißner-Ochsenfeld effect. In the so-called Meißner phase, superconducting materials are said to expel any (external) magnetic field from their interior, i. e. act like perfect diamagnets [45, p. 727]. In fact, reproducing the Meißner-Ochsenfeld effect with the help of a phenomenological model was the original motivation behind the London conductivity. The canonical proof of this assertion works as follows: First, the curl is taken on both sides of Ampère's law (Eq. (1.2)). The left-hand side then reduces to

$$\nabla \times (\nabla \times \boldsymbol{B}(\boldsymbol{x}, t)) = -\,\Delta\,\boldsymbol{B}(\boldsymbol{x}, t)\,, \tag{4.18}$$

because of Gauß's law for magnetic fields (Eq. (1.3)). In absence of electric fields, the current on the right-hand side of Ampère's law can further be eliminated using the second London equation (4.8). Eventually, this leads to a Helmholtz equation for the magnetic field,

$$\Delta\,\boldsymbol{B}(\boldsymbol{x}, t) = \frac{1}{\lambda_{\mathrm{L}}^2}\,\boldsymbol{B}(\boldsymbol{x}, t)\,, \tag{4.19}$$

[3] A similar expression can be found in [56, Eq. 2.44a] in context of the "two-fluid approximation", although with a prefactor differing by two. This is because the author insists on the misconception that negative frequencies would be "unphysical" while they in fact represent a mathematical necessity in context of Fourier transformation (see App. B.3). Without equally considering the negative frequency axis during the inverse transformation, the resulting function or kernel would simply not agree with the initial function anymore. This can be checked most easily using the example of sine and cosine.

[4] A ficticious perfect conductor with zero resistance would not necessarily expel externally applied magnetic fields as superconducters do below their (first) critical temperature, but may also comprise a nonzero constant magnetic field. [60]

where the laplacian eigenvalue is given by the so-called London penetration depth,

$$\lambda_{\mathrm{L}} = \sqrt{\frac{m}{\mu_0 n e^2}}\,, \tag{4.20}$$

which is regarded as a characteristic length scale for superconducters and lies in the range of 10 nm to 100 nm [45, p. 739]. The reason for this naming originates in the solution of Eq. (4.19) for a specific geometry[5] in form of an exponentially decaying field,

$$B_z(x) = B_0\, \mathrm{e}^{-x/\lambda_{\mathrm{L}}}\,. \tag{4.21}$$

The London penetration depth is then simply the distance from the surface boundary ($x = 0$) to the plane within the material ($x > 0$), where the external magnetic field is reduced by the factor e^{-1}.

From a response theoretical point of view, this tinkered explanation is not very satisfying. Fortunately, the universal response relation framework provides an even better way to show the Meißner-Ochsenfeld effect analytically, as we will see in the next section.

4.2. Application of universal response relations

Having reinterpreted the London conductivity as a proper response tensor, we first want to revisit magnetic properties from a response theoretical point of view again. For that reason, we recall that direct and proper response functions are related via the Dysonian equation (2.53) which, in case of the London model, implies an isotropic structure for the direct response tensor as well. More explicitly, the latter's longitudinal and transverse parts can be calculated from Eqs. (2.91) and (2.92) and read

$$\chi_{\mathrm{L}}(\boldsymbol{k},\omega) = \left(-\frac{ne^2}{m}\right)\frac{\omega^2}{\omega^2 - \omega_{\mathrm{p}}^2}\,, \tag{4.22}$$

$$\chi_{\mathrm{T}}(\boldsymbol{k},\omega) = \left(-\frac{ne^2}{m}\right)\frac{\omega^2 - c^2|\boldsymbol{k}|^2}{\omega^2 - c^2|\boldsymbol{k}|^2 - \omega_{\mathrm{p}}^2}\,, \tag{4.23}$$

where

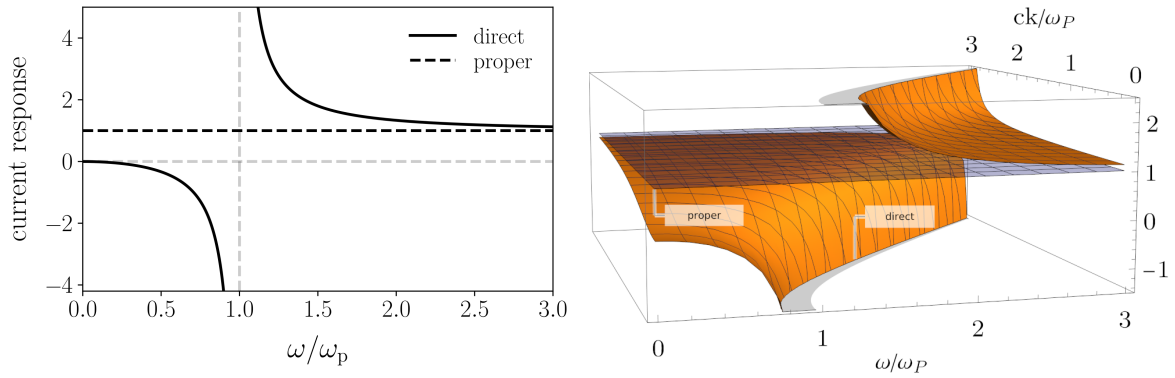
$$\omega_{\mathrm{p}}^2 \overset{\mathrm{def}}{=} \frac{ne^2}{\varepsilon_0 m}\,, \tag{4.24}$$

has been introduced as the plasma frequency (cf. [45, Eq. (1.38)]). In the long-wavelength limit both parts obviously coincide,

$$\lim_{|\boldsymbol{k}|\to 0}\chi_{\mathrm{T}}(\boldsymbol{k},\omega) = \lim_{|\boldsymbol{k}|\to 0}\chi_{\mathrm{L}}(\boldsymbol{k},\omega) = \chi_{\mathrm{L}}(\omega)\,, \tag{4.25}$$

such that the tensor becomes "completely" isotropic again. By comparing Eqs. (4.22) and (4.23) with Eq. (4.2), it becomes immediately clear that the distinction between direct and proper

[5] Here, the superconducter is occupying the positive real half-space $x \geq 0$ and a *constant* magnetic vector field is applied outside pointing to the material's boundary plane in z-direction.



(a) longitudinal parts **(b)** transverse parts

Fig. 4.2.: Proper vs. direct London current response tensor, $\widetilde{\chi}_{ij} = \widetilde{\chi}_0 \delta_{ij}$ and $\chi_{ij}(k,\omega)$ with $k = |\boldsymbol{k}|$, separated by longitudinal and transverse parts. Function values are normalized to $\widetilde{\chi}_0$, such that the proper versions represented by the straight dashed line in (a) and the blue plane in (b) are both located at 1. Direct versions tend to their proper counterparts only in regions where the difference $\omega^2 - c^2|\boldsymbol{k}|$ is large compared to the plasma frequency ω_p. Both plots show a pole at the respective diamagnetic dispersion relation (Eqs. (4.34) and (4.35)), indicated by the vertical dashed grey line in (a) and the grey areas on top and bottom of the box in (b). The function values of the transverse part at $ck/\omega_\mathrm{p} = 0$ in (b) match in particular the plot in (a) as shown in Eq. (4.25).

response functions is most essential. While longitudinal and transverse parts of one type of response function strive towards each other in the long-wavelength limit or may even coincide right from the beginning as in the case of Eq. (4.2), direct and proper versions of the same response apparently do not show such a tendency. In case of the London model, the direct longitudinal and transverse parts can nevertheless be forced to resemble their proper counterparts in the limit $\boldsymbol{k} \to 0$, although this requires to set the plasma frequency to zero as well. The latter, however, is directly connected to the electronic charge density n, such that $\omega_\mathrm{p} \equiv 0$ implies $n \equiv 0$ and thus precisely recovers the vacuum case where the current response entirely vanishes anyway. On the other hand, as long as the difference $\omega^2 - c^2|\boldsymbol{k}|^2$ is large compared to the plasma frequency, the fraction in Eq. (4.23) is approximately one and thus the transverse direct response indeed tends to its proper counterpart as can easily be verified visually in Fig. 4.2b. The same holds for $\omega \gg \omega_\mathrm{p}$ in the transverse case. All these considerations, however, are only true for the London model and do not apply in general.

In any case, the plasma frequency seems to play a special rôle in the London model since it determines the location of the poles visualized in Figs. 4.2a and 4.2b. There, the evident deviations between direct and proper response functions show in particular that both types are substantially different and must not be confused with each other. All the more astonishing in this respect is the circumstance that magnetic materials properties like the magnetic susceptibility χ_m or the permeability μ_r are usually accessed via direct response functions, whereas optical properties are typically derived from proper tensors like the optical conductivity $\widetilde{\sigma}_{ij}$ or the closely connected the dielectric tensor $(\varepsilon_\mathrm{r})_{ij}$ (see Sct. 2.4 for details). In the following, we will take a closer look at these two types of materials properties and which effects can already be inferred from the simple London model, or equivalently, the mere diamagnetic part of the current response of the free electron gas.

We begin with the (purely transverse) magnetic susceptibility, which can be derived explic-

itly via the universal response relation Eq. (2.102) and reads

$$\chi_{\mathrm{m}}(\boldsymbol{k},\omega) = \frac{\omega_{\mathrm{p}}^2}{\omega^2 - c^2|\boldsymbol{k}|^2 - \omega_{\mathrm{p}}^2}\,. \tag{4.26}$$

In the combined static and optical limit this simplifies to

$$\lim_{\omega,|\boldsymbol{k}|\to 0} \chi_{\mathrm{m}}(\boldsymbol{k},\omega) = -1\,, \tag{4.27}$$

independently of the order these limits are taken. By its definition (2.61), χ_m or equivalently χ_{BB} contains the information how the induced magnetic field varies with changes in the externally applied one. In case of Eq. (4.27), the relation between external and induced fields becomes particularly intuitive even in real space, where

$$\boldsymbol{B}_{\mathrm{ind}}(\boldsymbol{x},t) = \int \mathrm{d}t' \int \mathrm{d}^3\boldsymbol{x}'\, (-1)\,\delta^3(\boldsymbol{x}-\boldsymbol{x}')\,\delta(t-t')\,\boldsymbol{B}_{\mathrm{ext}}(\boldsymbol{x}',t') = -\boldsymbol{B}_{\mathrm{ext}}(\boldsymbol{x},t)\,. \tag{4.28}$$

Inserting this with the fundamental splitting of fields, $\boldsymbol{B}_{\mathrm{tot}} = \boldsymbol{B}_{\mathrm{ind}} + \boldsymbol{B}_{\mathrm{ext}}$ (see Sct. 2.1), immediately shows that the resulting total field vanishes in this case. In other words, the induced field completely cancels the external one, viz. locally ($\boldsymbol{k} = 0$) and instantaneously ($\omega = 0$), which makes the material a perfect diamagnet. We especially note, that this result does not contradict the canonical proof of the Meißner-Ochsenfeld effect, where the external magnetic field enters the material's surface by a few nanometers before vanishing completely because of the induced surface currents. Quite to the contrary, in the framework of linear response for homogeneous media there simply is no surface in the first place (see App. D.2), such that the statement in Eq. (4.28) is perfectly in line with the traditional proof and in addition does not require a cobbled geometry.

Next we want to consider optical properties for which the dielectric tensor is required as discussed in Sct. 2.7. By applying the relation between the density response and the transverse current response (2.93) to Eq. (4.23), an explicit expression for the (direct) London density response can be obtained,

$$\chi(\boldsymbol{k},\omega) = \left(-\frac{ne^2}{m}\right)\frac{-|\boldsymbol{k}|^2}{\omega^2 - \omega_{\mathrm{p}}^2}\,. \tag{4.29}$$

This is consistent with the analogous relation between the two proper versions in combination with the Dysonian equation for the density response (2.94). The longitudinal and transverse parts of the dielectric tensor can then be obtained via Eqs. (2.96) and (2.98), or alternatively using Eqs. (2.97) and (2.99), and read

$$\varepsilon_{\mathrm{L}}(\boldsymbol{k},\omega) = 1 - \frac{\omega_{\mathrm{p}}^2}{\omega^2}\,, \tag{4.30}$$

$$\varepsilon_{\mathrm{T}}(\boldsymbol{k},\omega) = 1 - \frac{\omega_{\mathrm{p}}^2}{\omega^2 - c^2|\boldsymbol{k}|^2}\,. \tag{4.31}$$

Obviously, the transverse dielectric function tends to its longitudinal counterpart in the op-

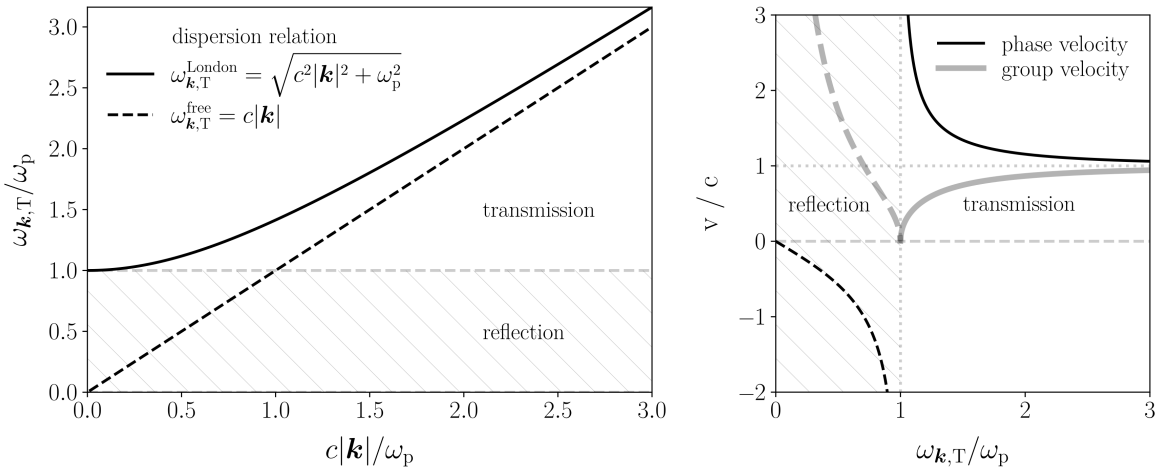


Fig. 4.3.: Transverse dispersion relation obtained from the London model (solid curve) compared to the free electromagnetic dispersion (dashed line). Transmission is only possible for frequencies higher than the plasma frequency. Below ω_p, light cannot penetrate the surface and is completely reflected (hatched area). For this region, phase and group velocities take purely imaginary values (dashed continuations in right plot). Normalizations are chosen such that both plots are compatible, i. e. the derivative of the London dispersion from the left plot yields the group velocity from the right one, $\frac{\partial(\omega_{\boldsymbol{k}}/\omega_\mathrm{p})}{\partial(c|\boldsymbol{k}|/\omega_\mathrm{p})} = \frac{1}{c}\frac{\partial\omega_{\boldsymbol{k}}}{\partial|\boldsymbol{k}|} = \frac{v_\mathrm{g}}{c}$. Note the different abscissae.

tical limit, i. e.

$$\lim_{|k|\to 0} \varepsilon_\mathrm{L}(\boldsymbol{k},\omega) = \lim_{|k|\to 0} \varepsilon_\mathrm{L}(\boldsymbol{k},\omega) = \varepsilon_\mathrm{L}(\omega)\,. \tag{4.32}$$

Now given these two functions, the London model is perfectly suited to prove the astonishing equivalence between the two wave operators in Fourier space from Sct. 2.7,

$$\left(-\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2\right)\varepsilon_\mathrm{T}(\boldsymbol{k},\omega) = -\frac{\omega^2}{c^2}\,\varepsilon_\mathrm{L}(\boldsymbol{k},\omega) + |\boldsymbol{k}|^2 = -\frac{\omega^2}{c^2} + |\boldsymbol{k}|^2 + \frac{\omega_\mathrm{p}^2}{c^2} \tag{4.33}$$

According to the fundamental wave equation in media (2.134), or more precisely Eqs. (2.150) and (2.151) for the isotropic limit, non-trivial solutions are determined by setting the transverse and longitudinal dielectric functions to zero. In case of the London model, this yields the following dispersion relations,

$$\omega_{\boldsymbol{k},\mathrm{L}}^2 = \omega_\mathrm{p}^2\,, \tag{4.34}$$

$$\omega_{\boldsymbol{k},\mathrm{T}}^2 = c^2|\boldsymbol{k}|^2 + \omega_\mathrm{p}^2\,. \tag{4.35}$$

The first equation unsurprisingly recovers the plasma frequency for oscillations of the electronic charge density (a. k. a. plasma oscillations), whereas the second equation, on the other hand, reproduces the left graph plotted in Fig. 4.3. Conceptually important quantities in this repect are the group velocity,

$$v_\mathrm{G} \overset{\mathrm{def}}{=} \frac{\partial\omega_{\boldsymbol{k},\mathrm{T}}}{\partial|\boldsymbol{k}|} = c\sqrt{1 - \frac{\omega_\mathrm{p}^2}{\omega^2}}\,, \tag{4.36}$$

the phase velocity,

$$v_\mathrm{p} \overset{\mathrm{def}}{=} \frac{\omega_{\boldsymbol{k},\mathrm{T}}}{|\boldsymbol{k}|} = c\sqrt{1+\frac{\omega_\mathrm{p}^2}{c^2|\boldsymbol{k}|^2}}\,, \tag{4.37}$$

and the index of refraction.

$$n \overset{\mathrm{def}}{=} \frac{c|\boldsymbol{k}|}{\omega_{\boldsymbol{k}}} = \sqrt{1-\frac{\omega_\mathrm{p}^2}{\omega_{\boldsymbol{k}}^2}} \tag{4.38}$$

For the London model, phase and group velocities are further related by

$$n \equiv \frac{c}{v_\mathrm{p}} = \frac{c}{v_\mathrm{p}}\,. \tag{4.39}$$

and thus

$$\frac{v_\mathrm{g}}{c} = \frac{c}{v_\mathrm{p}}\,. \tag{4.40}$$

With these, the following regions in Fig. 4.3 can now be distinguished (see e. g. [47, §11.6.3]):

(i) $\omega < \omega_\mathrm{p}$: In this region there is no solution for the (bulk) wave equation. Transverse electromagnetic waves cannot propagate through the medium. Instead, any radiation would be completely reflected at a supposed surface due to the so-called "inifite polarizability" of the electron gas. Group velocity v_g, phase velocity v_p and index of refraction n take purely imaginary values here.

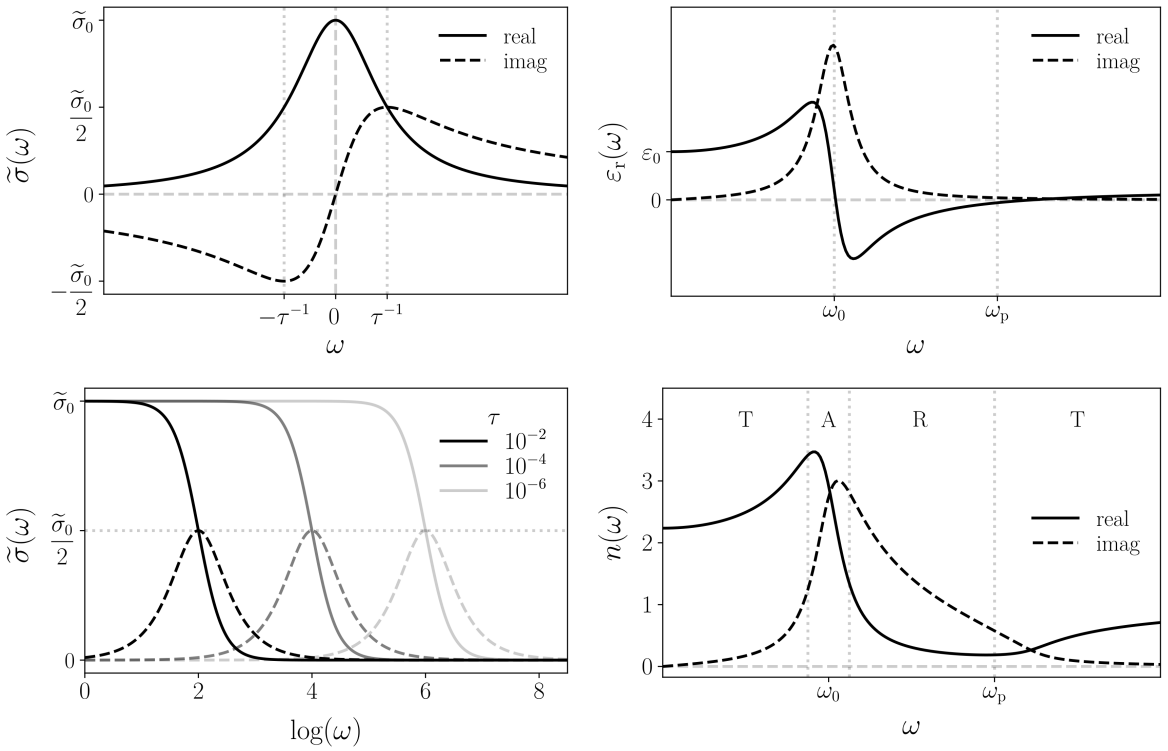
(ii) $\omega = \omega_\mathrm{p}$: Here, the group velocity is equal to zero, hence still no transmission. The phase velocity, by contrast, is infinite, which corresponds to a refractive index of zero.[6] This means that the medium oscillates as a whole, a behavior which is sometimes referred to as "plasmon" [1, p. 8]

(iii) $\omega > \omega_\mathrm{p}$: External fields can now cause proper oscillations within the medium. External fields are not cancelled completely, such that the material becomes transparent. While the group velocity stays subluminal, the phase velocity is still superluminal, but drops quickly.

(iv) $\omega \gg \omega_\mathrm{p}$: In this limit, the charge density of the material (or in this case the plasma) cannot follow the external perturbation anymore. The medium behaves completely transparent for electromagnetic radiation. Group and phase velocities tend to the vacuum speed of light c.

For metals whose plasma frequencies usually lie in the visible or ultraviolet range, a mechanism similar to the one in the $\omega < \omega_\mathrm{p}$ region is in fact (part of) the reason why metals appear shiny. Due to the rather low charge carrier concentration in semi- and superconductors, ω_p is shifted to the infrared spectrum for these material classes (see e. g. [64, p. 6.5.1]). Another

[6] This existence of phase velocities larger than the speed of light is not in contradiction with special relativity. For example, superposing two plane waves which propagate in not quite exactly opposite directions leads to humps in the interference pattern which propagate faster than c. In fact, this is even a desired property in the research field of photonics. There, zero or near-to-zero refractive indices have already been observed experimentally for some so-called metamaterials (see e. g. [61, 62]). The relevant quantity w. r. t. relativistic constraints is the group velocity, or more precisely the wave front velocity $v_\mathrm{f} = \lim_{\boldsymbol{k}\to\infty} v_\mathrm{p}$, which replaces v_g in lossy materials in its rôle as signal velocity. The latter must not exceed the speed of light. [63].



(a) Drude model **(b)** Lorentz oscillator model

Fig. 4.4.: Drude model for metals and Lorentz oscillator model for semiconductors. Both phenomenological models are used to describe optical effects for the respective material types. The Drude model can be derived from the London model in the relaxation time approximation, where the infinitesimal factor η is replaced by a finite relaxation rate $1/\tau$. The Lorentz oscillator model for semiconductors by contrast covers more optical effects as indicated by T-A-R-T (transmittion—absorption—reflection—transmission) in the bottom (b) plot. Formally it reverts to the London or Drude model for $\omega_0 = 0$ and suitable choices for the damping constant γ.

relevant property in this respect is the surface condition. Since metals are relatively hard without being brittle, they are easy to polish. This leads to a strongly directed (specular) reflection compared to the diffuse light scattered at rather rough surfaces and thus enhances the perceptible effect of reflection.

Speaking of metals and semiconductors as two further types of materials besides superconductors, it should also be noted that the respective phenomenological models used to describe optical properties are highly related to the London model. For metals, the famous Drude model can be derived from the London conductivity in the so-called relaxation time approximation by replacing the infinitesimal regularization factor η in Eq. (4.13) with a finite relaxation time τ,

$$\frac{\mathrm{i}}{\omega + \mathrm{i}\eta} \overset{\eta \leftrightarrow \frac{1}{\tau}}{=} \frac{\tau}{1 - \mathrm{i}\omega\tau}\,, \tag{4.41}$$

such that the proper response tensor becomes (visualized in Fig. 4.4a)

$$\overleftrightarrow{\widetilde{\sigma}}(\omega) = \frac{\widetilde{\sigma}_0}{1 - \mathrm{i}\omega\tau}\,\overleftrightarrow{\mathbb{1}}\,, \tag{4.42}$$

with constant “DC-conductivity”

$$\widetilde{\sigma}_0 \overset{\mathrm{def}}{=} \frac{ne^2\tau}{m}\,. \tag{4.43}$$

In this model, τ is related to the so-called "mean free path" $l = v_\mathrm{F}\tau$ (with Fermi velocity v_F, see Eq. (5.26)), which is said to be a measure for the mobility of electrons. In other words, this quantity models the ability of charged particles to move through a medium in response to externally applied electromagnetic fields. This DC-conductivity is usually of order $10^6\,\mathrm{S\,cm^{-1}}$ [46, p. 302]. Therefore, the relaxation rate is typically $1/\tau \ll \omega_\mathrm{p}$ for simple metals with charge carrier densities of about $10^{28}\,\mathrm{m^{-3}}$ and hence a plasma frequency well in the ultraviolet spectrum.

For semiconductors on the other hand, the Lorentz oscillator model is frequently employed. There, the dielectric function is modelled instead of the conductivity and takes the following form (visualized in Fig. 4.4b):

$$\varepsilon(\omega) = 1 + \frac{\omega_\mathrm{p}^2}{\omega_0^2 - \omega^2 - \mathrm{i}\gamma\omega}\,. \tag{4.44}$$

This permittivity is derived by a completely different approach (see e. g. [65, §4]), namely by Fourier transforming the following equation of motion,

$$m\ddot{\boldsymbol{x}}(t) + m\gamma\dot{\boldsymbol{x}}(t) + m\omega_0^2\boldsymbol{x}(t) = -e\boldsymbol{E}(t)\,, \tag{4.45}$$

with resonant frequency ω_0 and damping parameter γ, into

$$\left(\omega^2 + \mathrm{i}\omega\gamma - \omega_0^2\right)\boldsymbol{x}(\omega) = \frac{e}{m}\boldsymbol{E}(\omega)\,, \tag{4.46}$$

and inserting $\boldsymbol{x}(\omega)$ into the phenomenological definition of polarization in terms of electric dipole moments. This leads via the standard relation $n^2(\omega) = \varepsilon(\omega)$ to the refractive index plotted in Fig. 4.4b, which is also known as T-A-R-T plot. The latter acronym describes the following four regions:

- **T**ransmission for $\omega < \omega_0 - \gamma/2$
- **A**bsorption for $\omega_0 - \gamma/2 < \omega < \omega_0 + \gamma/2$
- **R**eflection for $\omega_0 + \gamma/2 < \omega < \omega_\mathrm{p}$
- **T**ransmission for $\omega > \omega_\mathrm{p}$ (like metals)

Evidently, this material behavior is much more complex than the one modelled by the London and Drude model. Nevertheless, for $\omega_0 = 0$, the Lorentz oscillator reverts to either Drude or London model, depending on the choice of the damping parameter γ as can be seen from Eq. (4.31) in the optical limit or by applying Eq. (2.108) to the Drude conductivity.

At this point, it is important to note that no quantum mechanical treatment is in fact required to find all *qualitative* characteristics of (super-)conductors as predicted by the London and Drude model. The complete discussion of this section could have done entirely without knowing the explicit composition of the initially introduced constant current response factor χ_0. The following identities could have equally obtained from dimensional considerations,

$$\chi_0 = \varepsilon_0\omega_\mathrm{p}^2 = \frac{\sigma_0}{\tau} \tag{4.47}$$

but then of course without fundamental theoretical support. In fact, most traditional treatments just cite London's original work (where no explanation for this factor is given but rather referred to even older publications like Ref. [66]), copy the classical motivation of the Drude model (e. g. [56, §2.1] or [45, p.737]), or follow the lines of the rather dubious "generalized London theory" based on "macroscopic wave function" (see e. g. [47, §13.3.2]). Textbooks with a more theoretical aspiration like [32] know however that the London model is part of a more general expression based on the quantum field theoretical treatment.

4.3. Spin correction

In this section, we propose a spin correction to the London model based on the results of Sct. 3.3 and study how this affects optical and magnetic properties. We start from the proper London current response given by Eq. (4.2). The corresponding density response calculated via relation (2.93) then reads

$$\widetilde{\chi}(\boldsymbol{k},\omega) = \varepsilon_0 |\boldsymbol{k}|^2 \frac{\omega_\mathrm{p}^2}{\omega^2} . \tag{4.48}$$

As shown in Sct. 3.3, the spinorial contribution to the current response tensor of the free electron gas is purely transverse and proportional to the density response at that. Thus, inserting Eq. (4.48) into the defining equation (3.100), we find the spin contribution for the London model,

$$\overleftrightarrow{\widetilde{\chi}}^{\mathrm{spin}}(\boldsymbol{k},\omega) = -\varepsilon_0 \frac{\hbar^2|\boldsymbol{k}|^4}{4m^2} \frac{\omega_\mathrm{p}^2}{\omega^2} \overleftrightarrow{P}_\mathrm{T}(\boldsymbol{k}) . \tag{4.49}$$

This can now be integrated into the initial tensor by replacing the original London current response according to

$$\overleftrightarrow{\widetilde{\chi}}^{\mathrm{London}} \quad \mapsto \quad \overleftrightarrow{\widetilde{\chi}}' = \overleftrightarrow{\widetilde{\chi}}^{\mathrm{London}} + \overleftrightarrow{\widetilde{\chi}}^{\mathrm{spin}} . \tag{4.50}$$

Since this will only affect the transverse part, nothing changes for the oscillations of the electronic charge density (i. e. "plasmons"). We will, however, observe a difference in optical and magnetic properties.

From the generally valid form of the dispersion relation in terms of the current response, Eq. (2.152), the following implicit expression for frequencies corresponding to transverse oscillations can be derived:

$$\omega_{\boldsymbol{k},\mathrm{T}}^2 = c^2|\boldsymbol{k}|^2 + \omega_\mathrm{p}^2 + \left(\frac{\hbar|\boldsymbol{k}|^2}{2m}\right)^2 \frac{\omega_\mathrm{p}^2}{\omega_{\boldsymbol{k},\mathrm{T}}^2} . \tag{4.51}$$

Solving this for the squared frequency yields

$$(\omega_{\boldsymbol{k},\mathrm{T}}^2)_\pm = \frac{1}{2}\left(c^2|\boldsymbol{k}|^2 + \omega_\mathrm{p}^2\right) \pm \frac{1}{2}\sqrt{\left(c^2|\boldsymbol{k}|^2 + \omega_\mathrm{p}^2\right)^2 + \left(\frac{\hbar\omega_\mathrm{p}}{mc^2}\right)^2 (c|\boldsymbol{k}|)^4} . \tag{4.52}$$

For simple metals, the plasma energy corresponding to ω_p lies in the range of 3 to 15 eV. This is insignificantly small compared to the rest energy of the electron with $mc^2 \approx 0.511\,\mathrm{MeV}$,

and consequently

$$\left(\frac{\hbar\omega_\mathrm{p}}{mc^2}\right)^2 \approx 0\,. \tag{4.53}$$

Hence, the second term within the square root can be neglected such that

$$(\omega^2_{\boldsymbol{k},\mathrm{T}})_+ \approx c^2|\boldsymbol{k}|^2 + \omega_\mathrm{p}^2\,, \tag{4.54}$$

$$(\omega^2_{\boldsymbol{k},\mathrm{T}})_- \approx 0\,, \tag{4.55}$$

should approximate the two branches sufficiently well. The plus-branch now obviously coincides with the standard London result (4.35), whereas the minus-branch does not contain any information and can be discarded. Therefore we can conclude that spin-correcting the London response does not lead to any significant qualitative effect for optical applications.

Turning to magnetic properties, we first have to find the direct current response of the *combined* diamagnetic and spinorial terms. This is necessary because of the non-linearity of the Dysonian response relations. For the spin-London model, response relation (2.92) yields

$$\chi'_\mathrm{T}(\boldsymbol{k},\omega) = \widetilde{\chi}_0\,\frac{\left(\omega^2 - c^2|\boldsymbol{k}|^2\right)(1+\kappa)}{\omega^2 - c^2|\boldsymbol{k}|^2 - \omega_\mathrm{p}^2(1+\kappa)}\,, \tag{4.56}$$

with

$$\kappa = \frac{\widetilde{\chi}^\mathrm{spin}(\boldsymbol{k},\omega)}{\widetilde{\chi}_0} = \frac{\hbar^2|\boldsymbol{k}|^4}{4m^2\omega^2}\,. \tag{4.57}$$

Although this only appears like a slightly altered version of the original direct current response of the London model, Eq. (4.23), it has at least one interesting effect: In the static limit, where Eq. (4.56) simplifies to

$$\lim_{\omega\to 0}\chi_\mathrm{T}(\boldsymbol{k},\omega) = -\varepsilon_0\,c^2|\boldsymbol{k}|^2\,, \tag{4.58}$$

drawing again on relation (2.112) between transverse current response and magnetic susceptibility results in

$$\chi'_\mathrm{m}(\boldsymbol{k}) = -1\,. \tag{4.59}$$

This recovers the Meißner effect even without the need to take any additional limit. The reason for this rather significant change compared to the original London susceptibility (4.26) is the singularity of κ in the static limit. Likewise, in the optical limit we first obtain

$$\lim_{|\boldsymbol{k}|\to 0}\chi'_\mathrm{m}(\boldsymbol{k},\omega) = \lim_{|\boldsymbol{k}|\to 0}\mathbb{D}(\boldsymbol{k},\omega)\chi'_\mathrm{T}(\boldsymbol{k},\omega) = \frac{\omega_\mathrm{p}^2}{\omega^2-\omega_\mathrm{p}^2}\,, \tag{4.60}$$

via the more general response relation Eq. (2.102), which is identical to the original function in this limit (cf. Eq. (4.26)) and for $\omega \to 0$ results in the Meißner effect as well. Hence, the result is again independent of the order these limits are taken.

Provided this spin-London model is suitable to anticipate the response functions and limits that will be affected more or less by including the spinorial contribution to the current response, we may state the following: Optical properties are unlikely to change in any significant manner. This should be especially true for the dispersion relation. Magnetic properties,

on the other hand, are more sensible to this correction and may show small or possibly even not so small qualitative changes.

Last but not least, we remark that the spin correction procedure is self-consistent in the sense that applying again the substitution from Eq. (4.50) on the already spin-corrected current response has no stacking effect. The spin correction is effectively idempotent. This is because the spin correction affects only the *transverse* part. The density response required for constructing the spin correction on the other hand, is derived from the *longitudinal* current response. Thus, the spin correction of the London model is always given by Eq. (4.49).

5. Full current response

In this final chapter, we analyze the full current response tensor as derived in Sct. 3.3. In particular, we show how magnetic and optical properties of the free electron gas compare to the ones derived for the London model and the ones found in standard literature for the "spinless" conductivity.

Before starting with an in-depth study of the physical properties of the current response, we first transform all material-characteristic functions, which occur in the analytical master formula (3.109), into dimensionless integrals. This especially facilitates any later numerical treatment. For zero temperature, where the Fermi-Dirac distribution reduces to a Heaviside step function, these integrals can even be solved analytically. Although this is already known in principle (see Ref. [46]), we present in Sct. 5.1 an alternative way based on our formulation of the *full* current response tensor, i. e. including the spinorial part. It can be shown that the 12 initially unknown scalar component functions in fact reduce to 3 dimensionless integrals. Hence, a material in this model is completely characterized by the said integrals, the Lindhard density response and the constant charge density. A particularly aesthetic result is the master formula Eq. (5.54), which relates all auxiliary functions to their characteristic integrals. With the Lindhard Integral Theorem, we then prove in Sct. 5.2 that all of the latter can be expressed in terms of the characteristic Lindhard integral. In Sct. 5.3, the Laurent series for the optical and static limit of the spinless and spinorial transverse current response are derived. Based on these, optical and magnetic properties are finally discussed in Scts. 5.4 and 5.5.

5.1. Dimensionless formulae

In this first section, a dimensionless variant of the current response tensor loosely based on the conventions of [32, §4.4] is introduced, which will serve as basis for all subsequent analyses in this chapter.

Isotropy.—We start by proving that the current response tensor for the free electron gas is (as one would expect) not only homogeneous but also strictly isotropic, i. e. is of the isotropic form (2.89) *and* does not depend on the direction of its wavevector argument. In Sct. 3.3 we already showed that the spinorial contribution is purely transverse anyway. Further, the diamagnetic part is a simple constant and the spin-orbit cross-correlation vanishes entirely. Hence, it is sufficient to show this property for the orbital current response. For convenience, we write the latter in the form

$$\overleftrightarrow{\chi}^{\mathrm{orb}}(\boldsymbol{q},\omega) = -\frac{e^2\hbar^2}{2m^2}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\,(2\boldsymbol{k}+\boldsymbol{q})(2\boldsymbol{k}+\boldsymbol{q})^{\mathsf{T}}\,\frac{f_{\boldsymbol{k}}-f_{\boldsymbol{k}+\boldsymbol{q}}}{\hbar\omega^{+}+\varepsilon_{\boldsymbol{k}}-\varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\,, \tag{5.1}$$

where the abbreviations $f_{\boldsymbol{k}} \equiv f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k}))$ for the Fermi-Dirac distribution, $\varepsilon_{\boldsymbol{k}} \equiv \varepsilon_0(\boldsymbol{k})$ for the single-particle energy levels and $\omega^+ \equiv \omega + \mathrm{i}\eta$ for the regularized frequency has been used. Decomposing the integration variable $\boldsymbol{k}$ into parts parallel ($\parallel$) and orthogonal ($\perp$) to the function argument $\boldsymbol{q}$,

$$\boldsymbol{k}_\parallel = \overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{q})\,\boldsymbol{k}\,, \tag{5.2}$$

$$\boldsymbol{k}_\perp = \overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{q})\,\boldsymbol{k}\,, \tag{5.3}$$

the integrand in Eq. (5.1) can be expanded such that the tensor reads

$$\overleftrightarrow{\chi}^{\mathrm{orb}}(\boldsymbol{q},\omega) = -\frac{e^2\hbar^2}{2m^2}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\,(2\boldsymbol{k}_\perp + 2\boldsymbol{k}_\parallel + \boldsymbol{q})(2\boldsymbol{k}_\perp + 2\boldsymbol{k}_\parallel + \boldsymbol{q})^{\mathsf{T}}\frac{f_{\boldsymbol{k}} - f_{\boldsymbol{k}+\boldsymbol{q}}}{\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\,. \tag{5.4}$$

From the defining equations for the Fermi-Dirac distribution (Eq. (3.42)) and the quantum mechanical dispersion relation (Eq. (3.54)) it is clear that the fraction in the integrand is invariant under the exchange $\boldsymbol{k}_\perp \mapsto -\boldsymbol{k}_\perp$. Consequently, integrating against a term which is linear in $\boldsymbol{k}_\perp$ makes the corresponding integral vanish. Alternatively, this can be seen by expanding the product in front of the fraction and using $\boldsymbol{k}_\perp \cdot \boldsymbol{k}_\parallel = \boldsymbol{k}_\perp \cdot \boldsymbol{q} = 0$. Therefore, the only non-zero contributions in Eq. (5.4) are

$$\overleftrightarrow{\chi}^{\mathrm{orb}}_{\mathrm{L}}(\boldsymbol{q},\omega) = -\frac{e^2\hbar^2}{2m^2}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\,(2\boldsymbol{k}_\parallel + \boldsymbol{q})(2\boldsymbol{k}_\parallel + \boldsymbol{q})^{\mathsf{T}}\frac{f_{\boldsymbol{k}} - f_{\boldsymbol{k}+\boldsymbol{q}}}{\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\,, \tag{5.5}$$

$$\overleftrightarrow{\chi}^{\mathrm{orb}}_{\mathrm{T}}(\boldsymbol{q},\omega) = -\frac{e^2\hbar^2}{2m^2}\int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\,4\boldsymbol{k}_\perp\boldsymbol{k}_\perp^{\mathsf{T}}\frac{f_{\boldsymbol{k}} - f_{\boldsymbol{k}+\boldsymbol{q}}}{\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\,. \tag{5.6}$$

which simultaneously represent the longitudinal and transverse parts of the full current response. Eq. (5.6) in particular coincides essentially with Eq. (4.47) in [32]. The *strict* isotropy follows now trivially from the direction-independence of the $\boldsymbol{k}$-integration. This will later become more obvious after we have transitioned to spherical coordinates. For now, it is sufficient to see that the full current response tensor is completely determined by

$$\overleftrightarrow{\chi}(\boldsymbol{q},\omega) \equiv \overleftrightarrow{\chi}(|\boldsymbol{q}|,\omega) = \chi_{\mathrm{L}}(|\boldsymbol{q}|,\omega)\overleftrightarrow{P}_{\mathrm{L}}(|\boldsymbol{q}|) + \chi_{\mathrm{T}}(|\boldsymbol{q}|,\omega)\overleftrightarrow{P}_{\mathrm{T}}(|\boldsymbol{q}|)\,. \tag{5.7}$$

The two scalar functions of this isotropic expansion are obtained as usual via

$$\chi_{\mathrm{L}}(|\boldsymbol{q}|,\omega) = \frac{\boldsymbol{q}^{\mathsf{T}}\overleftrightarrow{\chi}(\boldsymbol{q})\,\boldsymbol{q}}{|\boldsymbol{q}|^2}\,, \tag{5.8}$$

$$\chi_{\mathrm{T}}(|\boldsymbol{q}|,\omega) = \boldsymbol{e}^{\mathsf{T}}(\boldsymbol{q})\,\overleftrightarrow{\chi}(\boldsymbol{q},\omega)\,\boldsymbol{e}(\boldsymbol{q})\,, \tag{5.9}$$

where $\boldsymbol{e}(\boldsymbol{q})$ denotes an arbitrary transverse unit vector, i. e. a vector with the properties $\boldsymbol{e}(\boldsymbol{q}) \cdot \boldsymbol{q} = 0$ and $\boldsymbol{e}(\boldsymbol{q}) \cdot \boldsymbol{e}(\boldsymbol{q}) = 1$. By specifically setting

$$\boldsymbol{q} = q\,\boldsymbol{e}_z \equiv q\,(0,0,1)^{\mathsf{T}}\,, \quad \text{with} \quad q \equiv |\boldsymbol{q}|\,, \tag{5.10}$$

we may choose the other Cartesian base vectors for the two (linearly independent) transverse

unit vectors, $\boldsymbol{e}_1(\boldsymbol{q}) = \boldsymbol{e}_x \equiv (1,0,0)^\mathsf{T}$ and $\boldsymbol{e}_2(\boldsymbol{q}) = \boldsymbol{e}_y \equiv (0,1,0)^\mathsf{T}$, such that the right-hand sides of Eqs. (5.8) and (5.9) simply project out the respective diagonal matrix elements. Thus we find for the longitudinal and transverse current response

$$\chi_\mathrm{L}(q,\omega) = \chi_{zz}(q\,\boldsymbol{e}_z,\omega)\,, \tag{5.11}$$

$$\chi_\mathrm{T}(q,\omega) = \chi_{xx}(q\,\boldsymbol{e}_z,\omega) = \chi_{yy}(q\,\boldsymbol{e}_z,\omega)\,, \tag{5.12}$$

and hence it is obvious that the complete information is contained only in the diagonal entries of the tensor. The latter will be specified further in the next step.

Spherical coordinates and adapted units.—Before proceeding, we recall that the current response of the free electron gas is in general given by Eq. (3.109), which can also be written as

$$\chi_{ij}(\boldsymbol{q},\omega) = -\left(\frac{e^2 n}{m} + \frac{\hbar^2|\boldsymbol{q}|^2}{4m^2}\,\chi(\boldsymbol{q},\omega)\right)\delta_{ij} + \alpha_{ij}(\boldsymbol{q},\omega) + q_i\,\beta_j(\boldsymbol{q},\omega) + \beta_i(\boldsymbol{q},\omega)\,q_j\,. \tag{5.13}$$

Re-expressing the two auxiliary functions α_{ij} and β_i given by Eqs. (3.90) and (3.91) with the abbreviations introduced in the beginning of this section yields

$$\alpha_{ij}(\boldsymbol{q},\omega) = -\frac{\hbar^2}{4m^2}\left(2e^2\int\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\;4k_i k_j\,\frac{f_{\boldsymbol{k}} - f_{\boldsymbol{k}+\boldsymbol{q}}}{\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\right), \tag{5.14}$$

$$\beta_i(\boldsymbol{q},\omega) = -\frac{\hbar^2}{4m^2}\left(2e^2\int\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\;2k_i\,\frac{f_{\boldsymbol{k}} - f_{\boldsymbol{k}+\boldsymbol{q}}}{\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\right), \tag{5.15}$$

and likewise for the Lindhard density response,

$$\chi(\boldsymbol{q},\omega) = 2e^2\int\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\;\frac{f_{\boldsymbol{k}} - f_{\boldsymbol{k}+\boldsymbol{q}}}{\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\,. \tag{5.16}$$

Further, the general expression for the (total) density is given by

$$n = 2\int\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\;f_{\boldsymbol{k}} = \int\mathrm{d}\omega\;g(\omega)f(\omega)\,, \tag{5.17}$$

where the integral relation for the density of states (3.106) has been used.

In the following, the prodecure of finding a numerically processable form of the current response tensor is demonstrated, based on the conventions used in Ref. [32, §4.4]. Since the basic steps are similar for all parts of Eq. (5.13), the general approach is explicitly performed only on the example of the well-known Lindhard density response (5.16). The remaining parts can then be deduced using the Lindhard Integral Theorem proved in Sct. 5.2.

We begin by splitting the fraction in the integrand of the Lindhard function into two parts followed by the substitution $\boldsymbol{k} \mapsto -\boldsymbol{k} - \boldsymbol{q}$ only for the second part. Finally using $f_{-\boldsymbol{k}} \equiv f_{\boldsymbol{k}}$, both parts are merged again under one integral which reads

$$\chi(\boldsymbol{q},\omega) = 2e^2\int\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3}\;f_{\boldsymbol{k}}\left(\frac{1}{\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}} + \frac{1}{-\hbar\omega^+ + \varepsilon_{\boldsymbol{k}} - \varepsilon_{\boldsymbol{k}+\boldsymbol{q}}}\right). \tag{5.18}$$

Additionally, the symmetry $\varepsilon_0(-\boldsymbol{k}) \equiv \varepsilon_0(\boldsymbol{k})$ has been used in the right fraction to make both parts look more similar. For convenience, we introduce the shortcut $\{\omega^+ \mapsto -\omega^+\}$, symbolizing that the second term in the integrand of Eq. (5.18) differs only by this very substitution from the first one. For zero absolute temperature, the chemical potential μ (a. k. a. Fermi level) is according to the Sommerfeld expansion equal to the Fermi energy E_F (cf. [67, §30.2]). In the same limit, the Fermi-Dirac distribution (3.42) approaches a Heaviside theta-function,

$$\lim_{T\to 0} f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) \equiv \lim_{\beta\to\infty} f_{\beta,\mu}(\varepsilon_0(\boldsymbol{k})) = \Theta(E_\mathrm{F} - \varepsilon_0(\boldsymbol{k}))\,. \tag{5.19}$$

Expanding the energies in the denominators of Eq. (5.18) with the dispersion relation (3.54) and applying the zero-temperature limit then yields

$$\chi(\boldsymbol{q},\omega) = \frac{2e^2}{(2\pi)^3}\,\frac{2m}{\hbar^2}\int \mathrm{d}^3\boldsymbol{k}\;\Theta(E_\mathrm{F}-\varepsilon_{\boldsymbol{k}})\left(\frac{1}{2m\omega^+/\hbar + |\boldsymbol{k}|^2 - |\boldsymbol{k}+\boldsymbol{q}|^2} + \{\omega^+ \mapsto -\omega^+\}\right). \tag{5.20}$$

Next, we use Eq. (5.10) for the $\boldsymbol{q}$-vector and perform a transformation to spherical coordinates for the integration variable,

$$\boldsymbol{k} = k\,(\sin\theta\,\cos\varphi, \sin\theta\,\sin\varphi, \cos\theta)^\mathsf{T} \quad \text{with} \quad k \equiv |\boldsymbol{k}|\,, \tag{5.21}$$

such that $\theta = 0$ corresponds to the direction of $\boldsymbol{q} = q\,\boldsymbol{e}_z$. By inserting the identity

$$|\boldsymbol{k}+\boldsymbol{q}|^2 = (\boldsymbol{k}+\boldsymbol{q})\cdot(\boldsymbol{k}+\boldsymbol{q}) = q^2 + 2kq\cos\theta + k^2\,, \tag{5.22}$$

as well as the invariance of the Heaviside function w. r. t. to the following change of arguments,

$$\Theta(E_\mathrm{F} - \varepsilon_0(\boldsymbol{k})) = \Theta\left(\frac{\hbar^2 k_\mathrm{F}^2}{2m} - \frac{\hbar^2 k^2}{2m}\right) = \Theta(k_\mathrm{F} - k)\,, \tag{5.23}$$

the Lindhard function becomes

$$\chi(q,\omega) = \frac{me^2}{\pi^2\hbar^2}\,\frac{1}{2\pi}\int_0^{k_\mathrm{F}}\mathrm{d}k\int_0^{\pi}\mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\varphi\,k^2\sin\theta\left(\frac{1}{2m\omega^+/\hbar - 2kq\cos\theta - q^2} + \{\omega^+ \mapsto -\omega^+\}\right). \tag{5.24}$$

In view of a numerical implementation it is convenient to introduce dimensionless versions of wavenumber and frequency variables,

$$\hat{k} := \frac{k}{k_\mathrm{F}}\,, \quad \hat{q} := \frac{q}{k_\mathrm{F}}\,, \quad \hat{\omega} := \frac{\hbar\omega}{E_\mathrm{F}} = \frac{2m\omega}{\hbar k_\mathrm{F}^2} = \frac{2\omega}{v_\mathrm{F} k_\mathrm{F}}\,, \tag{5.25}$$

where the F-indexed constants are the respective Fermi momentum k_F as well as the Fermi energy and Fermi velocity,

$$E_\mathrm{F} := \frac{\hbar^2 k_\mathrm{F}^2}{2m}\,, \quad v_\mathrm{F} := \frac{\hbar k_\mathrm{F}}{m}\,. \tag{5.26}$$

For later reference we additionally define the likewise dimensionless variables

$$\nu_\pm \equiv \nu_\pm(\hat{q},\hat{\omega}) \coloneqq \frac{1}{2}\left(\frac{\hat{\omega}}{\hat{q}} \pm \hat{q}\right) = \frac{\omega}{q v_\mathrm{F}} \pm \frac{q}{2k_\mathrm{F}}\,. \tag{5.27}$$

Now, substituting the integrand in the Lindhard function (5.20) according to $\mathrm{d}k \mapsto k_\mathrm{F}\,\mathrm{d}\hat{k}$ and explicitly carrying out the integral w. r. t. the variable φ leads to

$$\chi(\hat{q},\hat{\omega}) = \frac{m k_\mathrm{F}}{\pi^2\hbar^2}\frac{e^2}{2\hat{q}}\int_0^1 \mathrm{d}\hat{k}\,\hat{k}^2 \int_0^\pi \mathrm{d}\theta\,\sin\theta\left(\frac{1}{\frac{1}{2}(\hat{\omega}^+/\hat{q}-\hat{q}) - \hat{k}\cos\theta} + \{\hat{\omega}^+ \mapsto -\hat{\omega}^+\}\right). \tag{5.28}$$

After inserting the (total) density of states per unit volume of the free electron gas (cf. [32, Eq. (4.22)])

$$g(E_\mathrm{F}) = \frac{m k_\mathrm{F}}{\pi^2\hbar^2} \tag{5.29}$$

we then finally arrive at the concise formula

$$\chi(\hat{q},\hat{\omega}) = g(E_\mathrm{F})\frac{e^2}{2\hat{q}}\left(I_\chi(\nu_-) + I_\chi(-\nu_+)\right), \tag{5.30}$$

where the dimensionless integral I_χ is given by (see Sct. 5.2 for proof)

$$I_\chi(z) \overset{\mathrm{def}}{=} \int_0^1 \mathrm{d}x\,x^2 \int_0^\pi \mathrm{d}\theta\,\frac{\sin\theta}{z - x\cos\theta} = z + \frac{1-z^2}{2}\,\mathrm{Ln}\left(\frac{z+1}{z-1}\right). \tag{5.31}$$

Here, z denotes an input variable that always have a non-zero imaginary part via the regularized frequency $\omega^+ = \omega + \mathrm{i}\eta$, such that the complex logarithm has to be decomposed into

$$\mathrm{Ln}(z) = \ln|z| + \mathrm{i}\arg(z) \tag{5.32}$$

for numerical evaluation (see App. B.6 for details). This result essentially coincides with Eqs. (4.20) and (4.21) in Ref. [32].

Auxiliary functions.—Next, we want to transfer this procedure to the other material characteristic functions α_{ij} and β_i. Following the same steps as for the Lindhard function we find

$$\alpha_{ij}(\hat{q},\hat{\omega}) = -\frac{\hbar^2 e^2}{4m^2}\frac{g(E_\mathrm{F})}{2\hat{q}2\pi}\int_0^1 \mathrm{d}k\hat{k}^2 \int_0^\pi \mathrm{d}\theta\sin\theta \int_0^{2\pi}\mathrm{d}\varphi\,4\hat{k}_i\hat{k}_j k_\mathrm{F}^2\left(\frac{1}{\nu_- - \hat{k}\cos\theta} + \{\nu_- \mapsto -\nu_+\}\right), \tag{5.33}$$

$$\beta_i(\hat{q},\hat{\omega}) = -\frac{\hbar^2 e^2}{4m^2}\frac{g(E_\mathrm{F})}{2\hat{q}2\pi}\int_0^1 \mathrm{d}k\,\hat{k}^2 \int_0^\pi \mathrm{d}\theta\sin\theta \int_0^{2\pi}\mathrm{d}\varphi\,2\hat{k}_i k_\mathrm{F}\left(\frac{1}{\nu_- - \hat{k}\cos\theta} + \{\nu_- \mapsto -\nu_+\}\right), \tag{5.34}$$

where, in contrast to the Lindhard function, the integrals w. r. t. the variable φ have no trivial integrands but ones that depend on the choice of the indices i and j and the associated mapping of k_i and k_j to the spherical coordinates defined in Eq. (5.21). Because of the identities

$$\int_0^{2\pi} \mathrm{d}\varphi \, \sin\varphi = \int_0^{2\pi} \mathrm{d}\varphi \, \cos\varphi = \int_0^{2\pi} \mathrm{d}\varphi \, \cos\varphi \sin\varphi = 0 \,, \tag{5.35}$$

and

$$\int_0^{2\pi} \mathrm{d}\varphi \, \sin^2\varphi = \int_0^{2\pi} \mathrm{d}\varphi \, \cos^2\varphi = \pi \,, \tag{5.36}$$

it is obvious that only the components $\alpha_{xx} = \alpha_{yy}, \alpha_{zz}$ and β_z integrate to non-zero functions. This matches the result we previously found for the longitudinal and transverse current response in Eqs. (5.11) and (5.12), which we can now specify further as

$$\chi_{\mathrm{L}} = -\frac{e^2}{m} \, n - \frac{\hbar^2}{4m^2} \, q^2 \chi + 2q \, \beta_z + \alpha_{zz} \,, \tag{5.37}$$

$$\chi_{\mathrm{T}} = -\frac{e^2}{m} \, n - \frac{\hbar^2}{4m^2} \, q^2 \chi + \alpha_{xx} \,. \tag{5.38}$$

From the fact that only 3 of the initial 12 unknown auxiliary component functions do not vanish we can conclude that for a complete material description it is sufficient to know the following remaining functions: α_{xx}, α_{zz}, β_z, χ and the constant n. These in turn can be expressed similarly to the Lindhard function by the concise equations

$$\alpha_{xx} = E_{\mathrm{F}} \, g(E_{\mathrm{F}}) \, \frac{e^2}{m} \, \frac{(-2)}{4\hat{q}} \left(I_{\alpha_{xx}}(\nu_-) - I_{\alpha_{xx}}(\nu_+) \right) , \tag{5.39}$$

$$\alpha_{zz} = E_{\mathrm{F}} \, g(E_{\mathrm{F}}) \, \frac{e^2}{m} \, \frac{(-4)}{4\hat{q}} \left(I_{\alpha_{zz}}(\nu_-) - I_{\alpha_{zz}}(\nu_+) \right) , \tag{5.40}$$

$$\beta_z = \frac{E_{\mathrm{F}} \, g(E_{\mathrm{F}})}{k_{\mathrm{F}}} \, \frac{e^2}{m} \, \frac{(-2)}{4\hat{q}} \left(I_{\beta_z}(\nu_-) + I_{\beta_z}(\nu_+) \right) , \tag{5.41}$$

$$\frac{\hbar^2}{4m^2} \, \chi = \frac{E_{\mathrm{F}} \, g(E_{\mathrm{F}})}{k_{\mathrm{F}}^2} \, \frac{e^2}{m} \, \frac{(+1)}{4\hat{q}} \left(I_{\chi}(\nu_-) - I_{\chi}(\nu_+) \right) , \tag{5.42}$$

where the respective dimensionless integrals are given by (see Sct. 5.2 for proofs)

$$I_{\alpha_{xx}}(z) \overset{\mathrm{def}}{=} \int_0^1 \mathrm{d}x \, x^4 \int_0^\pi \mathrm{d}\theta \, \frac{\sin^3\theta}{z - x\cos\theta} = \frac{5}{6} \, z - \frac{1}{2} \, z^3 + \frac{1}{4} \, (z^2 - 1)^2 \, \mathrm{Ln}\left(\frac{z+1}{z-1} \right) , \tag{5.43}$$

$$I_{\alpha_{zz}}(z) \overset{\mathrm{def}}{=} \int_0^1 \mathrm{d}x \, x^4 \int_0^\pi \mathrm{d}\theta \, \frac{\sin\theta \cos^2\theta}{z - x\cos\theta} = -\frac{2}{3} \, z + z^3 - \frac{1}{2} \, z^2 \left(z^2 - 1 \right) \mathrm{Ln}\left(\frac{z+1}{z-1} \right) , \tag{5.44}$$

$$I_{\beta_z}(z) \overset{\mathrm{def}}{=} \int_0^1 \mathrm{d}x \, x^3 \int_0^\pi \mathrm{d}\theta \, \frac{\sin\theta \cos\theta}{z - x\cos\theta} = \frac{1}{z} \, I_{\alpha_{zz}}(z) \,, \tag{5.45}$$

and Eq. (5.31). It is noteworthy that all but one of these integrals are odd functions in the complex variable z, i. e. $I_\gamma(-z) = -I(z)$ for $\gamma = \alpha_{xx}, \alpha_{zz}, \chi$ and $I_{\beta_z}(-z) = I_{\beta_z}(z)$.[1] With numerical implementation kept in mind, it seems easier to have all functions in Eqs. (5.39) to (5.42) use a sum instead of a difference for the respective integrals and absorb the different signs of the second summand into the integral arguments ν_+.

Dimensionless equations.—In the final step, we introduce dimensionless versions of all relevant parts of the current response. Specifically, we choose

$$\chi_{ij} = \hat{\chi}_{ij} \times E_\mathrm{F}\, g(E_\mathrm{F})\, \frac{e^2}{m}\,, \tag{5.46}$$

$$\alpha_{ij} = \hat{\alpha}_{ij} \times E_\mathrm{F}\, g(E_\mathrm{F})\, \frac{e^2}{m}\,, \tag{5.47}$$

$$\beta_i = \hat{\beta}_i \times E_\mathrm{F}\, g(E_\mathrm{F})\, \frac{e^2}{m}\, \frac{1}{k_\mathrm{F}}\,, \tag{5.48}$$

and anticipatory w. r. t. to the usage in Eqs. (5.37) and (5.38),

$$\frac{\hbar^2}{4m^2}\,\chi = \hat{\chi} \times E_\mathrm{F}\, g(E_\mathrm{F})\, \frac{e^2}{m}\, \frac{1}{k_\mathrm{F}^2}\,, \tag{5.49}$$

$$\frac{e^2}{m}\, n = \hat{n} \times E_\mathrm{F}\, g(E_\mathrm{F})\, \frac{e^2}{m}\,. \tag{5.50}$$

Alternatively, these conversion formulae can be expressed in terms of the ground state density,

$$E_\mathrm{F}\, g(E_\mathrm{F}) = \frac{3}{2}\, n = \frac{k_\mathrm{F}^3}{2\pi^2}\,, \tag{5.51}$$

or in terms of the plasma frequency,

$$E_\mathrm{F}\, g(E_\mathrm{F})\, \frac{e^2}{m} = \frac{3}{2}\, \frac{ne^2}{m} = \frac{3}{2}\, \varepsilon_0 \omega_\mathrm{p}^2\,. \tag{5.52}$$

where for the free electron gas, the constant n can explicitly determined for zero temperature Eq. (5.17) to be

$$n = 2\, \frac{4\pi}{(2\pi)^3} \int_0^\infty \mathrm{d}k\, k^2\, \Theta(k_\mathrm{F} - k) = \frac{k_\mathrm{F}^3}{3\pi^2}\,. \tag{5.53}$$

From the above relation and the conversion rules for dimensionless quantities it follows directly that the normalized density in adapted units simply becomes $\hat{n} = 2/3$.

Putting all this together, the dimensionless versions of the material specific functions in Eqs. (5.39) to (5.42) can now be expressed by the concise master formula

$$\hat{\gamma}(\hat{q}, \hat{\omega}) = \frac{\gamma_0}{4\hat{q}} \left(I_\gamma(\nu_-) + I_\gamma(-\nu_+) \right) \qquad \text{with} \qquad \begin{array}{c|cccc} \gamma & \chi & \alpha_{xx} & \alpha_{zz} & \beta_z \\ \hline \gamma_0 & +1 & -2 & -4 & -2 \end{array}\,, \tag{5.54}$$

[1] This can be shown also without knowing the explicit results by substituting $\theta' = \pi - \theta$ in $I(-z)$ and using the identities $\cos(\pi - x) = -\cos(x)$ and $\sin(\pi - x) = \sin(x)$.

where $\nu_\pm \equiv \nu_\pm(\hat{q},\hat{\omega})$ is defined in Eq. (5.27). This in turn implies the following form for the dimensionless longitudinal and transverse current response functions,

$$\hat{\chi}_{\text{L}} = -\frac{2}{3} - \hat{q}^2\hat{\chi} + 2\hat{q}\hat{\beta}_z + \hat{\alpha}_{zz}\,, \tag{5.55}$$

$$\hat{\chi}_{\text{T}} = -\frac{2}{3} - \hat{q}^2\hat{\chi} + \hat{\alpha}_{xx}\,, \tag{5.56}$$

which can be implemented numerically right away. Note, that a factor 2 for spin degeneracy has already been included in most formulae. For spin-resolved versions, one had to separate orbital and spinorial contributions again and replace

$$E_{\text{F}} \mapsto E_{\text{F}}^s\,, \quad k_{\text{F}} \mapsto k_{\text{F}}^s\,, \quad \nu_\pm \mapsto \nu_\pm^s\,, \quad g \mapsto g^s/2\,, \quad n \mapsto n^s/2\,, \tag{5.57}$$

with

$$g^s(E_{\text{F}}^s) = \frac{3}{2}\frac{n^s}{E_{\text{F}}} = \frac{mk_{\text{F}}^s}{2\pi^2\hbar^2}\,, \quad g(E_{\text{F}}) = \sum_{s=\uparrow,\downarrow} g^s(E_{\text{F}}) = \frac{mk_{\text{F}}}{\pi^2\hbar^2}\,, \tag{5.58}$$

in appropriate places.

5.2. Lindhard integral theorem

This section is dedicated to the proofs of the explicit expressions for the characteristic integrals on the right-hand sides of Eqs. (5.43) to (5.45). For preparation, we first solve the well-known Lindhard integral in order to introduce all neccesary identities that will be used for the remaining integrals. These results are then used to prove the Lindhard Integral Theorem at the end of this section.

Preparations.—We want to prove the Lindhard integral

$$I_\chi(z) \stackrel{\text{def}}{=} \int_0^1 \mathrm{d}x\, x^2 \int_0^\pi \mathrm{d}\theta\, \frac{\sin\theta}{z - x\cos\theta} = z + \frac{1-z^2}{2}\,\text{Ln}\left(\frac{z+1}{z-1}\right)\,, \tag{5.31 rev.}$$

with $z \in \mathbb{C}$.

For that reason, we first substitute $u = \cos\theta$ together with the corresponding limits $u(0) = 1$ and $u(\pi) = -1$ and obtain

$$I_\chi(z) = \int_0^1 \mathrm{d}x\, x^2 \int_{-1}^1 \mathrm{d}u\, \frac{1}{z - xu}\,. \tag{5.59}$$

The inner integral is a standard one and can be solved right-away with

$$\int_{-1}^1 \mathrm{d}u\, \frac{1}{z-xu} = -\frac{\text{Ln}(z-xu)}{x}\bigg|_{-1}^1 = \frac{1}{x}\,\text{Ln}\,\frac{z+x}{z-x}\,, \tag{5.60}$$

where Ln denotes the *complex* logarithm with its branch cut running along the negative real

axis as defined in App. B.6. Inserting this result back into Eq. (5.59) and integrating the remaining expression by parts yields

$$I_\chi(z) = \int_0^1 \mathrm{d}x\, x \,\mathrm{Ln}\,\frac{z+x}{z-x} \tag{5.61}$$

$$= \int_0^1 \mathrm{d}x\, \frac{\mathrm{d}}{\mathrm{d}x}\left(\frac{1}{2}\,x^2\right) \mathrm{Ln}\,\frac{z+x}{z-x} \tag{5.62}$$

$$= \frac{x^2}{2}\,\mathrm{Ln}\,\frac{z+x}{z-x}\bigg|_0^1 - \int_0^1 \mathrm{d}x\, \frac{1}{2}\,x^2\,\frac{z-x}{z+x}\,\frac{(z-x)+(z+x)}{(z-x)^2} \tag{5.63}$$

$$= \frac{1}{2}\,\mathrm{Ln}\,\frac{z+1}{z-1} - z\int_0^1 \mathrm{d}x\, \frac{x^2}{z^2-x^2}\,. \tag{5.64}$$

By rewriting the integrand of the second term using

$$\frac{x^2}{z^2-x^2} = \frac{(x^2-z^2)+z^2}{z^2-x^2} = -1 + \frac{z^2}{z^2-x^2}\,, \tag{5.65}$$

the above integral can be simplified further into

$$I_\chi(z) = \frac{1}{2}\,\mathrm{Ln}\,\frac{z+1}{z-1} + z - z\int_0^1 \mathrm{d}x\, \frac{z^2}{x^2-z^2}\,. \tag{5.66}$$

One way to solve the new integral in the second summand is by rewriting the expression using partial fraction decomposition, i. e.

$$\frac{z^2}{z^2-x^2} = \frac{z^2}{(z+x)(z-x)} = \frac{z}{2}\left(\frac{1}{z+x} + \frac{1}{z-x}\right)\,, \tag{5.67}$$

such that it can be integrated directly into

$$\int_0^1 \mathrm{d}x\, \frac{z^2}{x^2-z^2} = \frac{z}{2}\int_0^1 \mathrm{d}x \left(\frac{1}{z+x} + \frac{1}{z-x}\right) = \frac{z}{2}\,\mathrm{Ln}\,\frac{z+x}{z-x}\bigg|_0^1 = \frac{z}{2}\,\mathrm{Ln}\,\frac{z+1}{z-1}\,. \tag{5.68}$$

Alternatively, this could have been solved by successive u- and trigonometric substitution with

$$u = \frac{x}{z} = \tanh y\,, \tag{5.69}$$

$$\mathrm{d}u = \mathrm{d}y\,(1-\tanh^2 y)\,, \tag{5.70}$$

where

$$\tanh y = \frac{\sinh y}{\cosh y} = \frac{\mathrm{e}^y - \mathrm{e}^{-y}}{\mathrm{e}^y + \mathrm{e}^{-y}} = \frac{\mathrm{e}^{2y}-1}{\mathrm{e}^{2y}+1} \tag{5.71}$$

is the *tangens hyperbolicus*. The right-hand side of Eq. (5.68) is then retained with the

following identity for the *area tangens hyperbolicus* (i. e. the inverse of tanh),

$$\operatorname{artanh}(u) = \frac{1}{2}\,\operatorname{Ln}\left(\frac{1+u}{1-u}\right). \tag{5.72}$$

Finally, by inserting the result (5.68) into Eq. (5.66) one arrives at the assertion, Eq. (5.31).

Auxiliary functions.—Having proved the explicit form of the Lindhard integral, we can continue with the integrals corresponding to the auxiliary functions α_{ij} and β_i. Starting with

$$I_{\alpha_{xx}}(z) = \int_0^1 \mathrm{d}x\, x^4 \int_0^\pi \mathrm{d}\theta\, \frac{\sin^3\theta}{z - x\cos\theta} = \frac{5}{6}\,z - \frac{1}{2}\,z^3 + \frac{1}{4}\,(z^2-1)^2\,\operatorname{Ln}\left(\frac{z+1}{z-1}\right), \quad (5.43\text{ rev.})$$

we will use the intermediate results for I_χ to derive analytic expressions for the only three non-vanishing integrals $I_{\alpha_{xx}}$, $I_{\alpha_{zz}}$ and I_{β_z}.

First, the inner integral is transformed by using the identity $\sin^2\theta = 1 - \cos^2\theta$ and by substituting $u = \cos\theta$,

$$I_{\alpha_{xx}}(z) = \int_0^1 \mathrm{d}x\, x^3 \int_{-1}^1 \mathrm{d}u\, \frac{1-u^2}{z/x - u}. \tag{5.73}$$

Introducing the shortcut $a = z/x$ in the second integrand and performing a polynomial division yields

$$\frac{1-u^2}{a-u} = u + a + \frac{a^2-1}{u-a}. \tag{5.74}$$

Since the integration limits of the inner integral are symmetric, the contribution of the linear term in u vanishes identically. For the two remaining parts we find

$$\int_{-1}^1 \mathrm{d}u\, \frac{1-u^2}{a-u} = au + (a^2-1)\operatorname{Ln}(u-a)\Big|_{-1}^1 = 2a - (a^2-1)\operatorname{Ln}\frac{a+1}{a-1}. \tag{5.75}$$

Inserting this intermediate result back into Eq. (5.73) leads to

$$I_{\alpha_{xx}}(z) = 2z\int_0^1 \mathrm{d}x\, x^2 - z^2\int_0^1 \mathrm{d}x\, x \operatorname{Ln}\frac{z+x}{z-x} + \int_0^1 \mathrm{d}x\, x^3 \operatorname{Ln}\frac{z+x}{z-x}, \tag{5.76}$$

or equivalently,

$$I_{\alpha_{xx}}(z) = \frac{2}{3}\,z - z^2 I_\chi(z) + I_3(z). \tag{5.77}$$

In the last step, the first term has been solved immediately by elementary rules and the second part is apparently identical to the Lindhard integral in form of Eq. (5.61). This already suggests that the latter will also play an important rôle for the characteristic integrals. The remaining third part in Eq. (5.76), on the other hand, needs to be treated further but can be solved by following similar steps as performed in Eq. (5.61) and following. First, we integrate

again by parts to cast off the logarithm in the integrand,

$$I_3(z) = \int_0^1 \mathrm{d}x\, x^3 \,\mathrm{Ln}\,\frac{z+x}{z-x} \tag{5.78}$$

$$= \int_0^1 \mathrm{d}x\, \frac{\mathrm{d}}{\mathrm{d}x}\left(\frac{1}{4}\,x^4\right) \mathrm{Ln}\,\frac{z+x}{z-x} \tag{5.79}$$

$$= \frac{1}{4}\,\mathrm{Ln}\,\frac{z+1}{z-1} - \frac{z}{2}\int_0^1 \mathrm{d}x\, \frac{x^4}{z^2-x^2}\,. \tag{5.80}$$

Then, by polynomial division, the fraction in the second summand is factorized into

$$\frac{x^4}{z^2-x^2} = -x^2 - z^2 + \frac{z^4}{z^2-x^2}\,, \tag{5.81}$$

which can be handled more easily during successive integration. Carrying out the above integral explicitly gives

$$I_3(z) = \frac{1}{4}\,\mathrm{Ln}\,\frac{z+1}{z-1} + \frac{z}{2}\left(\frac{1}{3}\,x^3 + z^2 x\right)\Bigg|_0^1 - \frac{z^3}{2}\int_0^1 \mathrm{d}x\, \frac{z^2}{z^2-x^2} \tag{5.82}$$

$$= \frac{1}{4}\,\mathrm{Ln}\,\frac{z+1}{z-1} + \frac{z}{6} + \frac{z^3}{2} - \frac{z^3}{2}\left(\frac{z}{2}\,\mathrm{Ln}\,\frac{z+1}{z-1}\right)\,, \tag{5.83}$$

where in the last step the previously determined integral from Eq. (5.68) has been used. By suitably refactoring terms in Eq. (5.83), this result can again be expressed in terms of the characteristic integral I_χ, namely

$$I_3(z) = -\frac{1}{3}\,z + \frac{1+z^2}{2}\,I_\chi(z)\,, \tag{5.84}$$

and hence yields for Eq. (5.77)

$$I_{\alpha_{xx}}(z) = \frac{1}{3}\,z + \frac{1-z^2}{2}\,I_\chi(z)\,. \tag{5.85}$$

Finally, by inserting the Lindhard integral I_χ into the latter equation we arrive at the expression stated in Eq. (5.43).

The next integral to be explicitly solved is

$$I_{\alpha_{zz}}(z) = \int_0^1 \mathrm{d}x\, x^4 \int_0^\pi \mathrm{d}\theta\, \frac{\sin\theta\cos^2\theta}{z - x\cos\theta} = -\frac{2}{3}\,z + z^3 - \frac{1}{2}\,z^2\left(z^2-1\right)\mathrm{Ln}\left(\frac{z+1}{z-1}\right)\,. \quad (5.44\ \text{rev.})$$

Similar to the previous ones, the substitution $u = \cos\theta$ leads to the following simplification,

$$I_{\alpha_{zz}}(z) = \int_0^1 \mathrm{d}x\, x^3 \int_{-1}^1 \mathrm{d}u\, \frac{u^2}{a-u}\,, \tag{5.86}$$

where $a = z/x$ has been used again as a convenient shortcut. After factorizing the new inner integrand into

$$\frac{u^2}{a-u} = -u - a + \frac{a^2}{a-u}\,, \tag{5.87}$$

and integrating within the proper limits, Eq. (5.86) translates to

$$I_{\alpha_{zz}}(z) = \int_0^1 \mathrm{d}x\, x^3 \left(-2a + a^2 \operatorname{Ln}\frac{a+1}{a-1}\right) \tag{5.88}$$

$$= -2z \int_0^1 \mathrm{d}x\, x^2 + z^2 \int_0^1 \mathrm{d}x\, x \operatorname{Ln}\frac{z+x}{z-x}\,. \tag{5.89}$$

The second part can again be identified with the Lindhard integral, such that

$$I_{\alpha_{zz}}(z) = -\frac{2}{3}\, z + z^2 I_\chi(z)\,, \tag{5.90}$$

which can alternatively be written as

$$I_{\alpha_{zz}}(z) = I_3(z) - I_{\alpha_{xx}}(z)\,. \tag{5.91}$$

Inserting the explicit form of I_χ from Eq. (5.31), we arrive exactly at Eq. (5.44).

The last fundamental integral for the current response tensor is given by

$$I_{\beta_z}(z) = \int_0^1 \mathrm{d}x\, x^3 \int_0^\pi \mathrm{d}\theta\, \frac{\sin\theta\cos\theta}{z - x\cos\theta} = \frac{1}{z}\, I_{\alpha_{zz}}(z)\,. \tag{5.45 rev.}$$

The right-hand side already suggests a close resemblance to the prior characteristic integral, $I_{\alpha_{zz}}$. Performing the very same substition as in all previous cases, $u = \cos\theta$, leads here to

$$I_{\beta_z}(z) = \int_0^1 \mathrm{d}x\, x^2 \int_{-1}^1 \mathrm{d}u\, \frac{u}{a-u}\,. \tag{5.92}$$

Using the relation

$$\frac{u}{a-u} = -1 + \frac{a}{a-u}\,, \tag{5.93}$$

and integrating w. r. t. u within the proper limits, we arrive at

$$I_{\beta_z}(z) = -\frac{2}{3} + z \int_0^1 \mathrm{d}x\, x \operatorname{Ln}\frac{z+x}{z-x} = -\frac{2}{3} + z I_\chi(z)\,, \tag{5.94}$$

where we can directly read off the assertion by comparing with Eq. (5.90).

Lindhard Integral Theorem. *Let I_χ be the complex-valued Lindhard integral which by Eq. (5.54) essentially determines the Lindhard response, i. e. the frequency- and wavevector-dependent density-density response of the free homogeneous electron gas, for the zero temperature case. Then, all other electromagnetic response functions can be expressed in terms of this Lindhard integral, the frequency and the wavevector.*

Proof. Eqs. (5.85), (5.90) and (5.94) show that the theorem holds for all characteristic integrals required to build the full current response function. By the universal response relations Eqs. (2.57) to (2.61), this then applies to all electromagnetic response functions as well. □

5.3. Laurent expansions

In order to estimate the impact of the spin correction in the transverse current response function, we need to find the Laurent series for the relevant functions. From the general expression for the spinorial part of the current response, Eq. (3.100), we know already that this contribution is purely transverse itself. If we now want to find an expression analogous to Eq. (5.56) which does not include the spinorial contribution, all we have to do is remove the Lindhard term such that only the terms corresponding to the diamagnetic and orbital parts remain. The resulting dimensionless formula then reads

$$\hat{\chi}_{\mathrm{T}}^{\mathrm{ns}}(\hat{q},\hat{\omega}) = -\frac{2}{3} + \left(-\frac{1}{2\hat{q}}\right)\left(I_{\alpha_{xx}}(\nu_-) - I_{\alpha_{xx}}(\nu_+)\right), \tag{5.95}$$

where "ns" is short for "no spinorial contribution" (or "spinless" in the following) and $\nu_\pm$ is defined in Eq. (5.27). Here, we have inserted already the explicit expression for the auxiliary function α_{xx} according to the master formula (5.54). Re-introducing proper units for the response function (but not for the arguments yet) gives (retarded regularization for ω implied)

$$\chi_{\mathrm{T}}^{\mathrm{ns}}(\hat{q},\hat{\omega}) = \frac{3}{2}\frac{ne^2}{m}\,\hat{\chi}_{\mathrm{T}}^{\mathrm{ns}}(\hat{q},\hat{\omega}) \tag{5.96}$$

$$= -\frac{ne^2}{m}\left(1 + \frac{3}{4\hat{q}}\left(I_{\alpha_{xx}}(\nu_-) - I_{\alpha_{xx}}(\nu_+)\right)\right). \tag{5.97}$$

The characteristic integral of α_{xx} has already been formulated explicitly in Eq. (5.43) and reduces for zero temperature to the simple expression

$$I_{\alpha_{xx}}(z) = \frac{5}{6}\,z - \frac{1}{2}\,z^3 + \frac{1}{4}\,\left(z^2-1\right)^2 \mathrm{Ln}\left(\frac{z+1}{z-1}\right), \tag{5.98}$$

where $\mathrm{Ln}(w)$ with $w \in \mathbb{C}$ denotes the complex logarithm as defined in App. B.6. The latter equation can be expanded into the following Laurent series (cf. Eq. (B.200) in App. B.6),

$$\mathrm{Ln}\left(\frac{z+1}{z-1}\right) = 2\left(\frac{1}{z} + \frac{1}{3z^3} + \frac{1}{5z^5} + \frac{1}{7z^7} + \mathcal{O}\left(\frac{1}{z^9}\right)\right), \tag{5.99}$$

which converges to the left-hand side for $|z| > 1$. The searched for expression for the last term in Eq. (5.98) is then approximately given by

$$\begin{aligned}
&\frac{1}{4}\left(z^2-1\right)^2 \operatorname{Ln}\left(\frac{z+1}{z-1}\right) \\
&\quad = \frac{1}{2}\left[\left(z^4-2z^2+1\right)\left(\frac{1}{z}+\frac{1}{3z^3}+\frac{1}{5z^5}+\cdots\right)\right] && (5.100)\\
&\quad = \frac{1}{2}\left[\left(z^3-2z+\frac{1}{z}\right)+\left(\frac{1}{3}z-\frac{2}{3z}+\frac{1}{3z^3}\right)+\left(\frac{1}{5z}-\frac{2}{5z^3}+\frac{1}{5z^5}\right)+\ldots\right] && (5.101)\\
&\quad = \frac{1}{2}\left[z^3-\frac{5}{3}z+\frac{8}{15}\frac{1}{z}+\frac{8}{105}\frac{1}{z^3}+\ldots\right]. && (5.102)
\end{aligned}$$

By defining

$$\kappa_\pm \coloneqq \frac{1}{4}\left(\nu_\pm^2-1\right)^2 \operatorname{Ln}\left(\frac{\nu_\pm+1}{\nu_\pm-1}\right), \tag{5.103}$$

and using the following expressions for the powers of the auxiliary variable $\nu_\pm \equiv \nu_\pm(\hat{q},\hat{\omega})$,

$$(\nu_\pm)^3 = \frac{1}{8}\left(\frac{\hat{\omega}^3}{\hat{q}^3}+3\,\hat{q}\hat{\omega}\pm\hat{q}^3\pm\frac{3\hat{\omega}^2}{\hat{q}}\right), \tag{5.104}$$

$$(\nu_\pm)^{-1} = \frac{2\,\hat{q}}{\hat{\omega}\pm\hat{q}^2}, \tag{5.105}$$

$$(\nu_\pm)^{-3} = \pm\frac{8\,\hat{q}^3}{(\hat{q}^2\pm\hat{\omega})^3}, \tag{5.106}$$

together with their respective differences,

$$(\nu_- - \nu_+) = -\hat{q}, \tag{5.107}$$

$$\left(\nu_-^3-\nu_+^3\right) = -\frac{1}{4}\left(\hat{q}^3+\frac{3\hat{\omega}^2}{\hat{q}}\right), \tag{5.108}$$

$$\left(\frac{1}{\nu_-}-\frac{1}{\nu_+}\right) = 4\,\frac{\hat{q}^3}{\hat{\omega}^2-\hat{q}^4} \overset{\hat{q}\to 0}{\approx} 4\,\hat{q}\left(\frac{\hat{q}^2}{\hat{\omega}^2}+\mathcal{O}\left(\hat{q}^6\right)\right), \tag{5.109}$$

$$\left(\frac{1}{\nu_-^3}-\frac{1}{\nu_+^3}\right) = -16\,\frac{\hat{q}^5\left(\hat{q}^4+3\hat{\omega}^2\right)}{\left(\hat{q}^4-\hat{\omega}^2\right)^3} \overset{\hat{q}\to 0}{\approx} 48\,\hat{q}\left(\frac{\hat{q}^4}{\hat{\omega}^4}+\mathcal{O}\left(\hat{q}^8\right)\right), \tag{5.110}$$

we find the intermediate expression

$$\frac{3}{4\hat{q}}\left(\kappa_- - \kappa_+\right) = \frac{3}{8}\left[\frac{5}{3}-\frac{3}{4}\frac{\hat{\omega}^2}{\hat{q}^2}-\frac{1}{4}\hat{q}^2+\frac{32}{15}\frac{\hat{q}^2}{\hat{\omega}^2}+\frac{128}{35}\frac{\hat{q}^4}{\hat{\omega}^4}+\mathcal{O}\left(\hat{q}^6\right)\right]. \tag{5.111}$$

Combining this with the remaining two terms of Eq. (5.98),

$$\frac{3}{4\hat{q}}\left(\frac{5}{6}\left(\nu_- - \nu_+\right)-\frac{1}{2}\left(\nu_-^3-\nu_+^3\right)\right) = \frac{3}{8}\left(-\frac{5}{3}+\frac{3}{4}\frac{\hat{\omega}^2}{\hat{q}^2}+\frac{1}{4}\hat{q}^2\right) \tag{5.112}$$

exactly cancels the first three terms in Eq. (5.111) and hence yields for the current response

$$\chi_{\mathrm{T}}^{\mathrm{ns}}(\hat{q},\hat{\omega}) = -\frac{ne^2}{m}\left(1+\frac{4}{5}\frac{\hat{q}^2}{\hat{\omega}^2}+\frac{48}{35}\frac{\hat{q}^4}{\hat{\omega}^4}+\mathcal{O}\left(\hat{q}^6\right)\right). \tag{5.113}$$

This result is valid for $|\nu_\pm| > 1$, or equivalently, $|\hat{\omega}/\hat{q} \pm \hat{q}| > 2$, which is for small $\hat{q}$ with $\hat{q} \ll \hat{\omega}$ (i. e. in the optical limit) certainly the case.

For $|\nu_\pm| < 1$, Eq. (5.113) cannot be used however. This is in particular true in the static limit, where $\hat{\omega} \to 0$ (but still leaving an infinitesimal positive imaginary part due to regularization). Evaluating the current response for small wavevectors there requires a different series representation, which is derived in App. B.6. Performing the static limit for the analytic expression of the current response yields (retardation again implied)

$$\chi_{\mathrm{T}}^{\mathrm{ns}}(\hat{q},\hat{\omega}=0) = -\frac{3}{2}\frac{ne^2}{m}\left(1+\frac{1}{2\hat{q}}\left[I_{\alpha_{xx}}\left(-\frac{\hat{q}}{2}\right)-I_{\alpha_{xx}}\left(+\frac{\hat{q}}{2}\right)\right]\right). \tag{5.114}$$

Then inserting the Laurent expansion (B.207) of the complex logarithm for the third term in Eq. (5.98) returns

$$\frac{1}{4}\left(z^2-1\right)^2 \operatorname{Ln}\left(\frac{z+1}{z-1}\right) = \frac{1}{4}\left[\left(z^4-2z^2+1\right)\left(-\mathrm{i}\pi+z+\frac{z^3}{3}+\frac{z^5}{5}+\cdots\right)\right] \tag{5.115}$$

$$= -\frac{\mathrm{i}\pi}{4}\left(z^2-1\right)^2+\frac{z}{2}-\frac{5z^3}{6}+\frac{4z^5}{15}+\mathcal{O}(z^7), \tag{5.116}$$

in contrast to Eq. (5.100). This, in turn, leads to the following representation for the characteristic integral in this region,

$$I_{\alpha_{xx}}\left(\pm\frac{\hat{q}}{2}\right) = \frac{\mathrm{i}\pi}{64}\left(\hat{q}^2-4\right)^2 \pm \frac{2}{3}\left(\hat{q}-\frac{\hat{q}^3}{4}+\frac{\hat{q}^5}{80}+\mathcal{O}(\hat{q}^7)\right), \tag{5.117}$$

and eventually to

$$\chi_{\mathrm{T}}^{\mathrm{ns}}(\hat{q},0) = \frac{3}{2}\frac{ne^2}{m}\hat{q}^2\left(-\frac{1}{6}+\frac{\hat{q}^2}{120}+\mathcal{O}(\hat{q}^4)\right), \tag{5.118}$$

for the current response in proper units.

Having obtained two representations for the combined diamagnetic and orbital part of the current response function which cover the relevant limiting cases, we can now turn to the spinorial part. Since $\chi_{\mathrm{T}}^{\mathrm{spin}}$ is essentially given by the Lindhard density response,

$$\chi_{\mathrm{T}}^{\mathrm{spin}}(\hat{q},\hat{\omega}) = \frac{3}{2}\frac{ne^2}{m}\hat{\chi}_{\mathrm{T}}^{\mathrm{spin}}(\hat{q},\hat{\omega}) = -\frac{3}{2}\frac{ne^2}{m}\hat{q}^2\hat{\chi}(\hat{q},\hat{\omega}), \tag{5.119}$$

it suffices to find the respective series expansions for the latter. This is a standard result which can be found in many textbooks like Ref. [46] (although mostly disguised in form of the *longitudinal* dielectric function via Eq. (2.97)) and whose derivation works exactly the way $\chi_{\mathrm{T}}^{\mathrm{ns}}$ has been treated previously. Hence, we will only state the final expressions here. In

case of the optical limit, the spinorial part can be expanded into

$$\chi_{\mathrm{T}}^{\mathrm{spin}}(\hat{q},\hat{\omega}) = \frac{3}{2}\frac{ne^2}{m}\,\hat{q}^2\left(-\frac{2}{3}\frac{\hat{q}^2}{\hat{\omega}^2} - \frac{8}{5}\frac{\hat{q}^4}{\hat{\omega}^4} + \mathcal{O}(\hat{q}^6)\right) \tag{5.120}$$

$$= -\frac{ne^2}{m}\left(\frac{\hat{q}^4}{\hat{\omega}^2} + \frac{12}{5}\frac{\hat{q}^6}{\hat{\omega}^4} + \mathcal{O}(\hat{q}^8)\right), \tag{5.121}$$

whereas in the static limit, the same function becomes

$$\chi_{\mathrm{T}}^{\mathrm{spin}}(\hat{q},0) = \frac{3}{2}\frac{ne^2}{m}\,\hat{q}^2\left(\frac{1}{2} - \frac{\hat{q}^2}{24} + \mathcal{O}(\hat{q}^4)\right). \tag{5.122}$$

The crucial difference to the mere Lindhard response is, evidently, the difference in the leading q-order which has far reaching effects as we will see in the following sections.

5.4. Optical properties

Drawing on the results of the previous section, we first prove that the combination of diamagnetic and orbital contributions to the (transverse) current response derived within our formalism exactly leads to the already known expression for the (transverse) conductivity. Using the relation between current response and conductivity tensor (2.57) in the $\eta \to 0$ limit (see also Eq. (4.14)), and substituting back $\hat{q}/\hat{\omega} = {}^{q v_{\mathrm{F}}}/_{2\omega}$, the imaginary part of the transverse conductivity can be readily expressed as

$$\operatorname{Im}\sigma_{\mathrm{T}}^{\mathrm{ns}}(\hat{q},\hat{\omega}) = \frac{ne^2}{\omega m}\left(1 + \frac{1}{5}\frac{q^2 v_{\mathrm{F}}^2}{\omega^2} + \frac{3}{35}\frac{q^4 v_{\mathrm{F}}^4}{\omega^4} + \mathcal{O}\left(q^6\right)\right). \tag{5.123}$$

At this point the relation to the London model (in particular Eqs. (4.2) and (4.12)) is again evident. Incorporating the orbital current simply adds more terms proportional to even powers of q to the current response. The same is necessarily true for the Drude model after performing the relaxation time approximation ($\omega \mapsto \omega + {}^{\mathrm{i}}/_{\tau}$). Therefore, the London model can indeed be regarded as the most simple toy model w. r. t. linear response theory, which is extended first by the purely frequency-dependent Drude model to account for impurities [32, §4.6], and finally by including the orbital (and possibly also the spinorial) contribution, leading to a genuine dependence on the wavevector.

Moreover, Eq. (5.123) agrees essentially with the findings of Dressel and Grüner (cf. [46, Eq. (5.3.8)])[2]. In fact, the equivalence between the conductivity in Ref. [46] and $\sigma_{\mathrm{T}}^{\mathrm{ns}}$ can already be seen by (partially) expanding the expression in Eq. (5.97) in terms of frequency and wavenumber and dividing the result by $\mathrm{i}\omega$,

$$\sigma_{\mathrm{T}}^{\mathrm{ns}}(q,\omega) = \frac{\mathrm{i}ne^2}{\omega m}\left(\frac{3}{8}\left[1 + 3\left(\frac{\omega^2}{q v_{\mathrm{F}}}\right)^2 + \left(\frac{q}{2k_{\mathrm{F}}}\right)^2\right]\right. \tag{5.124}$$

$$\left. - \frac{3}{16}\frac{k_{\mathrm{F}}}{q}\left[(\nu_-^2-1)^2\,\mathrm{Ln}\left(\frac{-\nu_- + 1}{-\nu_- - 1}\right) + (\nu_+^2-1)^2\,\mathrm{Ln}\left(\frac{\nu_+ + 1}{\nu_+ - 1}\right)\right]\right),$$

[2] Note, that the prefactor in front of the q^4 term in [46, Eq. (5.3.8)] has been incorrectly determined there.

which *exactly* agrees with [46, Eq. (5.3.6)] and expands in the small q limit to Eq. (5.123). As already discussed in Sct. 4.1, taking the $\eta \to 0$ limit of a seemingly purely imaginary function can nevertheless lead to real contributions. In case of the current response (and likewise the conductivity), the complex logarithm contributes additional terms as can be seen from Eq. (B.203).

The real part of the conductivity in this limit has already been analized for different cases in [46, Eq. (5.3.7a)] and is not relevant any further for our purposes. For the remaining discussion, even the exact expression for the imaginary part of the conductivity is not so important. Rather, the leading power of q in the optical limit is of interest. Performing the same kind of calculation now for the isolated spinorial part given by the series in Eq. (5.120), we further find

$$\operatorname{Im} \sigma_{\mathrm{T}}^{\mathrm{spin}}(\boldsymbol{k}, \omega) = \frac{ne^2}{\omega m} \left(\frac{1}{4} \frac{v_{\mathrm{F}}^2}{k_{\mathrm{F}}^2} \frac{q^4}{\omega^2} + \mathcal{O}(q^6) \right) . \tag{5.125}$$

Hence, the full conductivity in the optical limit is identical to its spinless contribution up to the q^2 term, while the spinorial part would only contribute for very large wavevectors. For $q \to 0$, this "correction" is insignificant and therefore, it is not surprising that the missing spinorial part in the current response, or equivalently in the conductivity tensor, has not been noticed so far in the standard literature on this topic.

Next, we want to verify how the dispersion relations are affected by the spinorial contribution. For the spin-London model proposed in Sct. 4.2, we found that adding the latter had no substantial impact on the transverse diamagnetic dispersion relation and did not change the longitudinal one at all. With the current response function in form of Eqs. (5.55) and (5.56), we are now in a position to explicitly calculate the transverse and longitudinal dispersion relations, defined by the conditions (2.150) and (2.151), for the free electron gas. Since the spinorial contribution is still purely transverse and thus has no impact on the longitudinal current response, we refer to [46, §5.4.4] for the respective dispersion relation,

$$\omega_{\boldsymbol{k},\mathrm{L}}^2 \approx \omega_{\mathrm{p}}^2 \left(1 + \frac{3}{5} \frac{q^2 v_{\mathrm{F}}^2}{\omega_{\mathrm{p}}^2} \right) , \tag{5.126}$$

There, the essential calculation based on Eq. (2.150) has already been carried out analytically.

Because a completely analytic derivation for the transverse dispersion relation would be quite cumbersome, we rely on a numerical approach here instead. As a first step, we reinterpret the current response obtained by the Kubo formula again in the RPA spirit, i. e. as a *proper* response tensor. Then, by exploiting the following conversion formula for the dimensionless response,

$$\widetilde{\chi}_T = \frac{3}{2} \frac{ne^2}{m} \hat{\chi}_{\mathrm{T}} = \frac{3}{2} \varepsilon_0 \omega_{\mathrm{p}}^2 \hat{\chi}_{\mathrm{T}} , \tag{5.127}$$

the transverse dielectric function,

$$\varepsilon_{\mathrm{r,T}}(\boldsymbol{q}, \omega) = 1 + \frac{1}{\varepsilon_0} \frac{1}{\omega^2 - c^2 |\boldsymbol{q}|^2} \widetilde{\chi}_{\mathrm{T}}(\boldsymbol{q}, \omega) , \tag{5.128}$$

can be expressed using the previously introduced adapted units and reads

$$\varepsilon_{\mathrm{r,T}}(\hat{q},\hat{\omega}) = 1 + \frac{3}{2}\left[\left(\frac{\omega_\mathrm{F}}{\omega_\mathrm{p}}\right)^2\hat{\omega}^2 - \left(\frac{ck_\mathrm{F}}{\omega_\mathrm{p}}\right)^2\hat{q}^2\right]^{-1}\hat{\chi}_\mathrm{T}(\hat{q},\hat{\omega})\,. \tag{5.129}$$

Condition (2.151) is then equivalent to the implicit equation

$$\hat{\omega}_{q,\mathrm{T}} = \frac{\omega_{q,\mathrm{T}}}{\omega_\mathrm{F}} = \sqrt{\left(\frac{ck_\mathrm{F}}{\omega_\mathrm{F}}\right)^2\hat{q}^2 - \frac{3}{2}\left(\frac{\omega_\mathrm{p}}{\omega_\mathrm{F}}\right)^2\hat{\chi}_\mathrm{T}(\hat{q},\hat{\omega}_{q,\mathrm{T}})}\,, \tag{5.130}$$

or, using a second scaling which is more convenient for plotting in the lower energy region,

$$\frac{\omega_{q,\mathrm{T}}}{\omega_\mathrm{p}} = \sqrt{\left(\frac{cq}{\omega_\mathrm{p}}\right)^2 - \frac{3}{2}\hat{\chi}_\mathrm{T}(\hat{q},\hat{\omega}_{q,\mathrm{T}})}\,. \tag{5.131}$$

For the numerical implementation, two different methods to solve this dispersion relation for the transverse frequency have been used:

(i) Solve Eq. (5.131) self-consistently and use the diamagnetic (i. e. London) dispersion relation as initial guess.

(ii) Refactor the dispersion relation and use a cunning algorithm to find the roots of Eq. (5.129) directly.

Both strategies have certain advantages and drawbacks. The self-consistent approach is evidently the less complex option. Eq. (5.131), and in particular $\hat{\chi}_\mathrm{T}(\hat{q},\hat{\omega})$ therein, can be implemented right away using the framework provided by Sct. 5.1. One iteration step would then solve the following equation for each scaled wavevector x_0 ,

$$y^{(n+1)} = \sqrt{x_0^2 - \frac{3}{2}\hat{\chi}_\mathrm{T}\left(\frac{\omega_\mathrm{p}}{ck_\mathrm{F}}x_0, \frac{\omega_\mathrm{p}}{\omega_\mathrm{F}}y^{(n)}\right)}\,, \tag{5.132}$$

with

$$y = \frac{\omega_{q,\mathrm{T}}}{\omega_\mathrm{p}} \qquad \text{and} \qquad x = \frac{cq}{\omega_\mathrm{p}}\,, \tag{5.133}$$

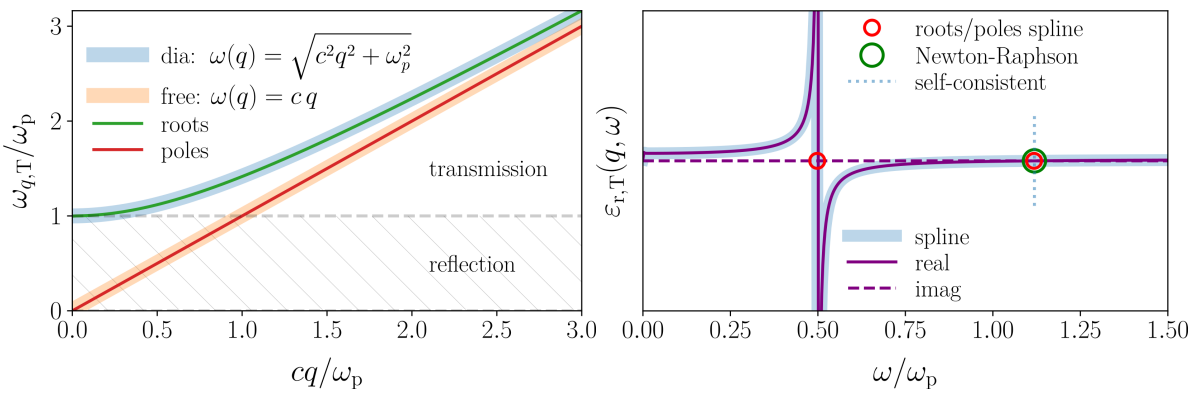
until the convergence criterion is met. The obvious choice for the latter is, that the absolute value $|y^{(n+1)} - y^{(n)}| < \epsilon_\mathrm{tol}$ should be less than a certain tolerance for consecutive iteration steps n and $(n+1)$. For the initial guess, the following form of the diamagnetic dispersion relation can be used:

$$y^{(0)} = \sqrt{x^2+1}\,. \tag{5.134}$$

By mixing scaled frequencies of successive iterations according to

$$y^{(n+1)} = \beta y^{(n+1)} + (1-\beta)\,y^{(n)}\,, \tag{5.135}$$

the convergence behavior can be improved by a certain degree. For small wavevectors, however, the numerics become increasingly unstable. This is a direct consequence of $\lim_{q\to 0}\nu_\pm = \infty$



(a) dispersion relation **(b)** root finding

Fig. 5.1.: Dispersion relation obtained from different approaches for finding the roots of the transverse dielectric function. All methods indicated in the right picture led to the diamagnetic dispersion, independently of using the full or only the spinless current response as input. By exploiting the zero-crossing of spline at poles, also the free dispersion relation could be recovered.

taking very large values such that the differences in Eq. (5.54) lie outside the floating point precision. This effect is also known as "catastrophic cancellation" in numerical analysis.

A numerically more stable, but on the other hand much more elaborate approach is to search for the roots of Eq. (5.129) directly. Since the usual optimization routines highly rely on the passed initial value, one has to find a good approximation for this first. Drawing again on the diamagnetic dispersion as reference, an interpolated univariate spline was created for $\varepsilon_{\mathrm{r,T}}(x_0, y)$ as shown in Fig. 5.1b. Function values far greater than the plot section in Fig. 5.1b were cut-off to prevent the spline from oscillating around the pole. In order to make absolutely sure that the correct root is found, the spline root was then in turn passed to a Newton-Raphson library routine (`scipy.optimize.newton`). As can be seen from the plot, all three methods basically found the same root within numerical accuracy. A particularly helpful feature of the spine plot is, that not only roots but also poles can be identified. Exploiting this circumstance proved numerically that the poles of the transverse dielectric function are given by the free dispersion relation. In fact, this is already obvious from Eq. (5.129), where the said dispersion enters through the denominator.

In any way, the resulting dispersion relation for the spinless as well as for the full current response do not deviate significantly from the diamagnetic London result. This confirms our finding from Sct. 4.3 as well as the claim, that optical properties can and should be accessed in general from the full current response tensor.

5.5. Magnetic properties

We now turn to magnetic properties, which should be—at least according to our Central Claim—fully accessible from the current response just as well as the optical properties from last section. In particular, we will show that in the static limit the two famous heuristic model types of magnetism for the free electron gas, namely the Pauli paramagnetism and the

Landau diamagnetism (cf. [68, Eqs. (7.15) and (7.61)]),

$$\chi_{\mathrm{m}}^{\mathrm{Pauli}} = \mu_0 \mu_{\mathrm{B}}^2 \, g(E_{\mathrm{F}}) \,, \quad \chi_{\mathrm{m}}^{\mathrm{Landau}} = -\frac{1}{3} \, \mu_0 \mu_{\mathrm{B}}^2 \, g(E_{\mathrm{F}}) \,, \tag{5.136}$$

can be inferred from different parts of the full frequency- and wavevector-dependent current response tensor. Both, $\chi_{\mathrm{m}}^{\mathrm{Pauli}}$ and $\chi_{\mathrm{m}}^{\mathrm{Landau}}$, are constant at zero temperature and differ only by a factor $-1/3$, although they are usually derived by quite different means (cf. e. g. [68, §7.2.1 vs. §7.6] or [69, §3]). Following the spirit of our approach, both necessarily have to combine to a total (paramagnetic) response, which is otherwise only obtained by manually summing up contributions to the magnetic susceptibility obtained from different models. Admittedly, in more theoretically oriented textbooks like Ref. [70], the different "magnetic effects" are only separated in order to discuss the origin behind the two contributions independently. However, from these books it is clear (although usually not even mentioned explicitly) that the full magnetic susceptibility is obtained from combining all relevant coupling terms into a single fundamental Hamiltonian, which eventually leads to our starting point, the Pauli equation (3.14). In fact, Nolting even states in his recent treatise:

> "The coupling of the [magnetic] field to the spin leads to paramagnetism and the coupling of the field to the orbital motion leads to diamagnetism. The two couplings cannot, however, be separated." [70, p. 90]

In view of this background, our result is truly not surprising. Quite to the contrary, it is puzzling that this reasoning seems to be completely forgotten when it comes to practically calculating the current response tensor.

Coming back to our objective of extracting magnetic properties the same way optical properties have been before, the essential difference compared to last section now lies in the specific limit, or more precisely in the specific order of limits that has to be performed. Contrary to the optical limit which, as the name suggests, fits best for optical applications, we now have to consider the static, i. e. $\omega \to 0$ limit, which is more suitable for deriving magnetic properties via the standard relation (2.112). Because we are still interested in the small q region, we cannot use Eqs. (5.113) and (5.120) as approximative representations of the transverse current response function but have to revert to the expansions (5.122) and (5.118), which are valid in the relevant argument region. Inserting the identity

$$\frac{3}{2} \frac{n e^2}{m} \frac{1}{k_{\mathrm{F}}^2} = E_{\mathrm{F}} \, g(E_{\mathrm{F}}) \frac{e^2}{m} \frac{1}{k_{\mathrm{F}}^2} = 2 \mu_{\mathrm{B}}^2 \, g(E_{\mathrm{F}}) \tag{5.137}$$

into the referenced series immediately leads to the following two relations for the respective parts of the transverse current response,

$$\chi_{\mathrm{T}}^{\mathrm{ns}}(q,0) = \mu_{\mathrm{B}}^2 \, g(E_{\mathrm{F}}) \, q^2 \left(-\frac{1}{3} + \frac{1}{60} \frac{q^2}{k_{\mathrm{F}}^2} + \mathcal{O}(q^4) \right) , \tag{5.138}$$

$$\chi_{\mathrm{T}}^{\mathrm{spin}}(q,0) = \mu_{\mathrm{B}}^2 \, g(E_{\mathrm{F}}) \, q^2 \left(1 - \frac{1}{12} \frac{q^2}{k_{\mathrm{F}}^2} + \mathcal{O}(q^4) \right) , \tag{5.139}$$

and via Eq. (2.112) eventually to

$$\chi_{\mathrm{m}}^{\mathrm{ns}}(q,0) = \mu_0\mu_{\mathrm{B}}^2\, g(E_{\mathrm{F}}) \left(-\frac{1}{3} + \frac{1}{60}\,\frac{q^2}{k_{\mathrm{F}}^2} + \mathcal{O}(q^4)\right) , \tag{5.140}$$

$$\chi_{\mathrm{m}}^{\mathrm{spin}}(q,0) = \mu_0\mu_{\mathrm{B}}^2\, g(E_{\mathrm{F}}) \left(1 - \frac{1}{12}\,\frac{q^2}{k_{\mathrm{F}}^2} + \mathcal{O}(q^4)\right) , \tag{5.141}$$

for the corresponding isolated magnetic susceptibilities. Evidently, the leading terms in the latter equations exactly reproduce the two model susceptibilities in Eq. (5.136). More specifically, the *combined* diamagnetic and orbital contributions to the current response tend in the static limit and for small wavevectors to the diamagnetic Landau susceptibility, whereas the spinorial part tends under identical conditions to the paramagnetic Pauli susceptibility. The first result matches in particular [32, Eq. (4.50)], whereas the second one is covered by [32, Eq. (4.29)] in established literature. Note, that in the latter reference the *non-interacting* spin susceptibility $\chi^{(0)}_{\mu_3\mu_3}$ (see Sct. 3.4) is again adduced. By contrast, our result is exclusively based on the Kubo formalism for the conductivity (or equivalently the current response) for the full, i. e. most general non-relativistic current operator (3.1).

Blundell also states these mere two limiting cases in his renowned monograph although without derivation (cf. [68, §7.7]). Interestingly, his result for the wavevector-dependent "diamagnetic response" ([68, Eq. (7.85)])[3] is obviously based on the spinless current response (5.97), just like the result for the optical conductivity in Dressel and Grüner's treatise is ([46, Eq. (5.3.6)]). All the more it seems astonishing that in the respective literature no connection is made between optical and magnetic materials properties. At the same time, these relations emerge only so obviously in this thesis because of the extensive preceding framework. Further, an autonomous realization by the ordinary reader is additionally impeded by the evidently erroneous key equations in popular literature.

In any case, combining spinless and spinorial parts to the full magnetic susceptibility,

$$\chi_{\mathrm{m}} \equiv \chi_{\mathrm{m}}^{\mathrm{full}} = \chi_{\mathrm{m}}^{\mathrm{ns}} + \chi_{\mathrm{m}}^{\mathrm{spin}} \tag{5.143}$$

returns a positive (i. e. paramagnetic) response in total, i. e. the magnetic response of the free electron gas reinforces the externally applied perturbation within the medium. This behavior can also be verified visually in Fig. 5.2.

So far, we did not explicitly comment on the question if the discussed magnetic susceptibilities are proper or direct. Following the RPA spirit, the current response should again be re-intepreted as a *proper* response. Since Pauli paramagnetism and Landau diamagnetism are equally derived from a non-interacting approximation, the same dogma actually holds for them as well. On the other hand, in Sct. 2.4 we showed that the commonly termed "magnetic

[3] Unfortunately, also in this reference the key equation is erroneous. Correctly, it should read

$$\chi_{\boldsymbol{q}} = \chi_{\mathrm{L}}\,\frac{3k_{\mathrm{F}}^2}{2q^2}\left[1 + \frac{q^2}{4k_{\mathrm{F}}^2} - \frac{k_{\mathrm{F}}}{q}\left(1 - \frac{q^2}{4k_{\mathrm{F}}^2}\right)^2 \ln\left|\frac{q+2k_{\mathrm{F}}}{q-2k_{\mathrm{F}}}\right|\right] . \tag{5.142}$$

This can easily be checked by plotting both versions and verifying visually that only Eq. (5.142) leads to the attached plot [68, Fig. 7.11], which in turn agrees with the orange line in Fig. 5.2.

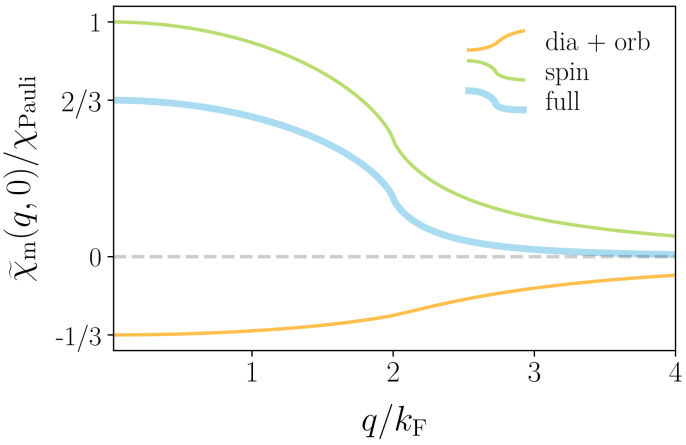


Fig. 5.2.: Spinless (i. e. diamagnetic plus orbital) and spinorial part of the *proper* magnetic susceptibility. While the first contribution reduces the magnetic field within the medium, the separated spinorial part as well as the total susceptibility lead to a reinforcement of the externally applied perturbation. All plots are normalized to the Pauli susceptibility (5.136) and do not change with varying reference susceptibility $\chi_\mathrm{m}^\mathrm{Pauli}$.

susceptibility" is in fact a *direct* response because it is defined as the functional derivative of the magnetization $\boldsymbol{M}$ (i. e. the induced magnetic field) w. r. t. the *external* magnetic field $\boldsymbol{H}$ and not w. r. t. the *total* field $\boldsymbol{B}$. Consequently, Eqs. (5.140) and (5.141) should actually not be used to compare with this quantity. This is particularly obvious on the example of the isolated diamagnetic part of the current response given by the first term in Eq. (5.95),

$$\hat{\chi}_\mathrm{T}^\mathrm{dia} = -\frac{2}{3} \quad \rightarrow \quad \chi_\mathrm{m}^\mathrm{dia} = -\frac{4}{3}\,\mu_0\mu_\mathrm{B}^2\,g(E_\mathrm{F})\left(\frac{k_\mathrm{F}}{q}\right)^2 \quad \neq \quad -1 = \lim_{\omega,q\to 0}\chi_\mathrm{m}^\mathrm{London}(q,\omega)\,, \tag{5.144}$$

which is far off the London result and cannot reproduce the Meißner effect as discussed in Sct. 4.2. An intruding question is now how the "real" direct susceptibilities look like. Since relation (2.112) between magnetic susceptibility and current response is valid for both, direct and proper functions, we can without loss of generality work with the Dysonian relation (2.92) in form of Eq. (2.95), i. e.

$$\chi_\mathrm{T}^{-1}(\boldsymbol{q},\omega) = \widetilde{\chi}_\mathrm{T}^{-1}(\boldsymbol{q},\omega) - \mathbb{D}_0(\boldsymbol{q},\omega)\,, \tag{5.145}$$

which reverts in the static limit and after dividing both sides by μ_0/q^2 to the astonishingly simple relation

$$\chi_\mathrm{m}^{-1}(q) = \widetilde{\chi}_\mathrm{m}^{-1}(q) - 1 \quad \Leftrightarrow \quad \chi_\mathrm{m}(q) = \frac{\widetilde{\chi}_\mathrm{m}(q)}{1-\widetilde{\chi}_\mathrm{m}(q)}\,. \tag{5.146}$$

Following the previous discussion, we now re-interpret Eqs. (5.140) and (5.141) as approximations to the respective *proper* responses,

$$\widetilde{\chi}_\mathrm{m}^\mathrm{ns}(q) = \chi_\mathrm{m}^\mathrm{Landau} + \mathcal{O}(q^2)\,, \tag{5.147}$$

$$\widetilde{\chi}_\mathrm{m}^\mathrm{spin}(q) = \chi_\mathrm{m}^\mathrm{Pauli} + \mathcal{O}(q^2)\,. \tag{5.148}$$

After some more algebra, the following expressions for the *direct* magnetic susceptibilities can

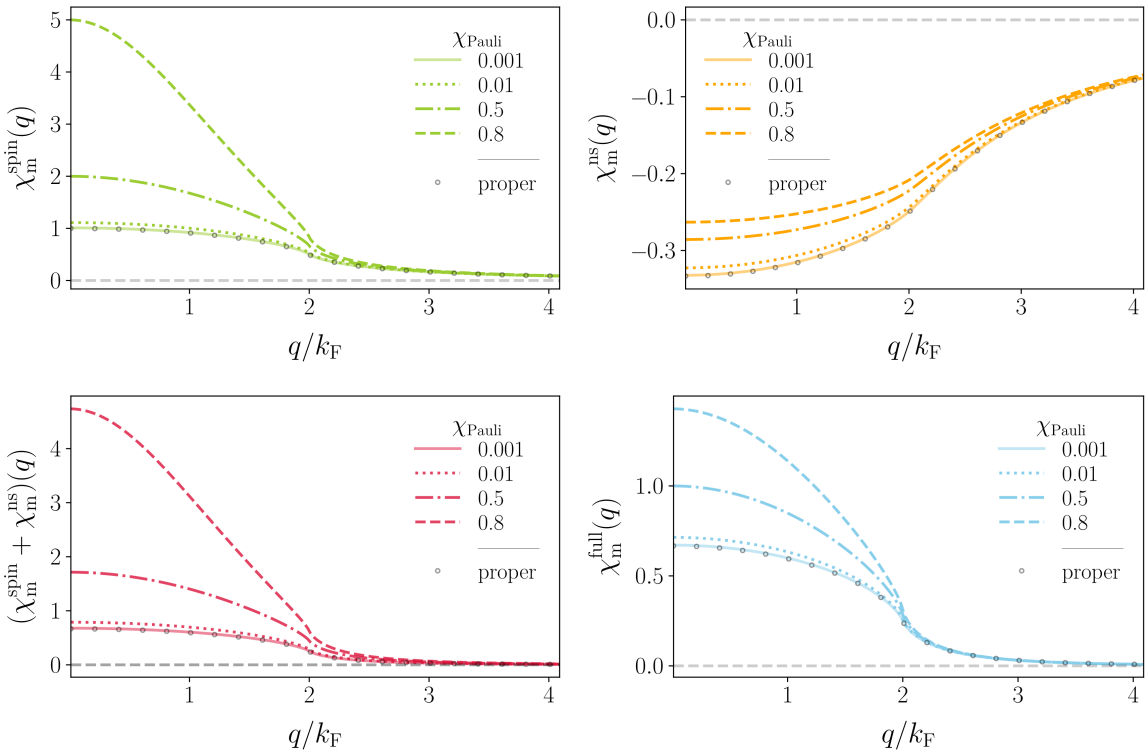


Fig. 5.3.: Diamagnetic and paramagnetic contributions to the *direct* magnetic susceptibility for different reference Pauli susceptibilities. In contrast to their proper counterparts (indicated by grey circles in each plot), the direct susceptibilities depend on the value of $\chi_{\mathrm{m}}^{\mathrm{Pauli}}(g(E_{\mathrm{F}}))$, even after normalizing to this very reference value. This has to be attributed to the non-linearity of the Dysonian relation (5.145). For small Pauli susceptibilities (and thus relatively small density of states at the Fermi level as typical for most materials), the direct versions do not differ significantly from their proper counterparts. However, as apparent from the bottom two plots, separated direct susceptibilities cannot be naïvely added to a total one, again because of the said non-linearity. Instead, the Dysonian equation has to be solved for the *full proper* magnetic susceptibility.

be derived:[4]

$$\chi_{\mathrm{m}}^{\mathrm{ns}}(q) = \left(\frac{1}{1-\chi_{\mathrm{m}}^{\mathrm{Landau}}}\right)\chi_{\mathrm{m}}^{\mathrm{Landau}} + \mathcal{O}(q^2)\,, \tag{5.149}$$

$$\chi_{\mathrm{m}}^{\mathrm{spin}}(q) = \left(\frac{1}{1-\chi_{\mathrm{m}}^{\mathrm{Pauli}}}\right)\chi_{\mathrm{m}}^{\mathrm{Pauli}} + \mathcal{O}(q^2)\,. \tag{5.150}$$

Regarding the fact that measured and theoretically calculated Pauli susceptibilities are quite small dimensionless numbers of order 10^{-6} to 10^{-5} for many metals (cf. e. g. [45, Tab. 31.5]), the small wavevector limits of spinless and spinorial direct responses apparently do not differ considerably from their proper counterparts. Only in case of a large density of states at the Fermi level as typical for some 3d transition metals like iron or nickel (see e. g. [47, Abb. 12.19]), the difference between proper and direct magnetic susceptibilities becomes significant as plotted in Fig. 5.3. In the theory of itinerant magnetism, this effect is known as Stoner enhancement[5] (see e. g. [71, §4 - §5] and [72]), which formally agrees with Eq. (5.145) when the so-called molecular field coefficient γ is set to one (cf. [71, Eq. (8.23)]). This is again not surprising since the "effective molecular field" inherent to this model (see e. g. [47,

[4] We skip these rather monotonous calculations since they do not bear any new difficulties and do not add anything relevant to the discussion.

Eq. (12.6.7)]),

$$\boldsymbol{B}_{\text{eff}} = \boldsymbol{B}_{\text{ext}} + \gamma\,\mu_0\,\boldsymbol{M}\,, \tag{5.151}$$

reverts in this case to the fundamental definition of the *total* magnetic field (2.8).

Although we treated spinorial and spinless parts separately in Eqs. (5.149) and (5.150) in order to compare the results with the standard literature, the two *direct* contributions cannot be naïvely combined into a corresponding total susceptibility as done for the proper versions via Eq. (5.143). The reason for this is that the Dysonian equation is simply not linear and thus

$$\chi_{\text{m}}^{\text{full}} = \frac{\widetilde{\chi}_{\text{m}}^{\text{ns}} + \widetilde{\chi}_{\text{m}}^{\text{spin}}}{1 - \left(\widetilde{\chi}_{\text{m}}^{\text{ns}} + \widetilde{\chi}_{\text{m}}^{\text{spin}}\right)} \quad \neq \quad \frac{\widetilde{\chi}_{\text{m}}^{\text{ns}}}{1 - \widetilde{\chi}_{\text{m}}^{\text{ns}}} + \frac{\widetilde{\chi}_{\text{m}}^{\text{spin}}}{1 - \widetilde{\chi}_{\text{m}}^{\text{spin}}} = \chi_{\text{m}}^{\text{ns}} + \chi_{\text{m}}^{\text{spin}}\,. \tag{5.152}$$

Instead, Eq. (5.145) has to be solved using the *total* proper magnetic susceptibility (5.143) right from the start. This procedure is in accord with the quote from the beginning of this section and confirms once again our stance that electromagnetic materials properties have in general to be derived from the *full* current response, which is based on Kubo formalism for the *total* current operator. However, Fig. 5.3 shows that as long as the magnitudes of the proper susceptibilities (i. e. Pauli and Landau terms) are very much less than one, Eq. (5.143) is still a good approximation even for the direct responses.

Last but not least it should not remain unmentioned that in practice, when the model of the free electron gas is applied to real metals, theoretically calculated susceptibilities may vary up to several magnitudes. This is usually fixed by introducing the (in general anisotropic) effective mass *tensor* [70, Eq. (3.178)],

$$\left(\frac{1}{m^*}\right)_{ij} = \frac{1}{\hbar^2}\,\frac{\partial^2}{\partial q_i\,\partial q_j}\,\epsilon_n(\boldsymbol{q})\,, \tag{5.153}$$

which can then be utilized to account for the fact that band structures of real materials are much more complex than the simple parabola of the free electron model. For the pure Pauli and Landau terms, the adapted total magnetic susceptibility is then given by (cf. [68, Eq. (7.64)])

$$\widetilde{\chi}_{\text{m}} = \chi_{\text{Pauli}}\left(1 - \frac{1}{3}\left(\frac{m}{m^*}\right)^2\right)\,, \tag{5.154}$$

where the scalar and constant parameter m^* is "some kind of an *average* performed over the 'Fermi layer'" [70, p. 120].

We further remark that all presented calculations are only valid for $T = 0\,\text{K}$. Hence, temperature-related effects as predicted by the Curie law are not covered for obvious reasons.

[5] Named after the British theoretical physicist Edmund Clifton Stoner (1899 - 1968).

Conclusion

> In constructing a new theory, we shall be careful to insist that they should be precise theories, giving a description from which definite conclusions can be drawn. We do not want to proceed in a fashion that would allow us to change the details of the theory at every place that we find it in conflict with experiment, or with our initial postulates. Any vague theory that is not completely absurd can be patched up by more vague talk at every point that brings up inconsistencies—and if we begin to believe in the talk rather than in the evidence we will be in a sorry state. [73, p. 22]
>
> *R. P. Feynman*

In this thesis, the general form of the current-current response tensor for the free, homogeneous and non-relativistic electron gas has been derived and studied, with particular attention to magnetic and optical materials properties. One of the key objectives was to prove our Central Claim, stating that all linear electromagnetic response functions can and should be, in principle, derived from the *full* frequency- and wavevector-dependent current-current reponse tensor. In this context, also the distinction between proper and direct response functions is emphasized and illustrated on the example of the magnetic susceptibility as representative of direct responses, and the (inverse) relative permittivity as a proper tensor.

After a profound review of electrodynamics in media and the closely related linear response theory in Part I, the most general non-relativistic expression for the quantum mechanical current operator has been derived in Sct. 3.1, based on the Pauli equation. With the help of a generalized version of the Kubo formula, the full current-current response tensor was then inferred. Compared to the results which can be found in standard literature on this topic, our version of the current response contains two additional terms besides the well-known diamagnetic and orbital contribution. While the spin-orbit cross-correlations vanish in case of the free electron gas because of its spin-unpolarized number density, the contribution of the spinorial part must in general not be neglected.

Traditionally, spin-based magnetism is accessed via a genuine spin-susceptibility. In Sct. 3.4, this object is compared with the spinorial current response from our approach. We demonstrate, that the latter two are indeed different response functions which must not be confused with each other. Further, we argue that by Ampère's law every magnetization (i. e. every induced magnetic field) without exception is generated by a microscopic current density as a matter of principle. This includes in particular a possibly spin-based magnetism as well. Additionally, this hypothesis is also backed by the fact, that the spinorial contribution is perfectly capable of reproducing well-known standard results like the Pauli paramagnetism,

even for the more general $T > 0$ case.

In Chpt. 4, this assertion is further investigated using the phenomenological London model for superconductors. After the latter has been reinterpreted in context of linear response theory as the simplest possible material model for which the fundamental covariant wave equation in media does not revert to its free counterpart, a selection of universal response relations are applied to the corresponding London conductivity. This way, we recovered some important standard results like the electromagnetic dispersion relations for the cold plasma and the Meißner-Ochsenfeld effect for type-I superconductors. Further, this example was perfectly suitable to apply our Gauge Claim, according to which the temporal gauge seems to be the preferred one for applications in the field of ab initio materials science. More specifically, only by using this gauge, the London model in terms of the current response, and the formulation in terms of conductivity tensor, can be converted into each other. Moreover, by adding a self-consistent spinorial correction to the latter, we could anticipate two essential findings of this thesis:

(i) Optical properties are hardly affected by the spinorial current response. This is exemplified using the transverse dispersion relation, which simply recovers the initial London result.

(ii) Magnetic properties are subject to more considerable changes, although phenomenological limiting cases like the aforementioned Meißner-Ochsenfeld effect are conserved. In fact, the magnetic susceptibility is even improved in a way that renders it constant in the static limit, while the original (direct) London susceptibility still depends on the wavevector.

This corroborates again our Central Claim by proving that a response theoretical treatment based on the full instead of the spinless conductivity tensor does not introduce effects contradictory to the standard results, but rather improves the established description.

Continuing with the main chapter 5 of this thesis, we start by reformulating the longitudinal and transverse parts of the full current response in terms of the Lindhard function, the (constant) number density and 12 scalar and material-dependent auxiliary functions. While processing the latter further we showed that only three of the initial 12 functions integrate to non-zero values. Moreover, we even solve them analytically in the zero-temperature limit. This is where the Lindhard Integral Theorem comes into play. In Sct. 5.2, we prove that the characteristic integrals corresponding to the three remaining scalar auxiliary functions can all be expressed in terms of the characteristic Lindhard integral and the latter's dependencies (i. e. wavevector and frequency). This is a fact which has (to the best of our knowledge) not been stated so far in the standard literature.

Supported by an extensive appendix on the Laurent series of the complex natural logarithm, we were able to reproduce the standard results of the electron gas for the spinless conductivity in the optical limit, as well as for the magnetic susceptibility in the static limit. En passant, we spot significant errors in the standard literature for these very response functions. For the magnetic susceptibility we further showed explicitly, that by incorporating the spinorial current response on the same level as the diamagnetic and orbital contributions, Pauli para-

and Landau diamagnetism emerge in a very natural way. For optical applications, we could also verify the anticipated London conclusion by numerically calculating the transverse dispersion relation for the spinless (i. e. diamagnetic plus orbital), and the full current response, all of which produced no significant corrections to the London result. This can be attributed to the fact, that a supposed spin correction enters the optical conductivity with not less than the forth power of the wavevector. Such terms are effectively discarded in the optical limit, where $|\boldsymbol{k}| \to 0$. In fact, referring to the dispersion relation of the orbital-corrected model, this seems to apply already for terms proportional to q^2.

Regarding the outlook of this thesis, the Lindhard theory constitutes again a good starting point to extend the study of the homogeneous electron gas to temperature-dependent effects. In this case, the Fermi-Dirac distributions in the auxiliary integrals do not revert to Heaviside step functions anymore and thus have to be treated numerically from the outset. In fact, a numerically stable code version for this purpose has already been written by the author of this thesis. Particularly interesting in this respect is a recent publication by Ancarani and Jouin [74], who propose a numerically more efficient technique to evaluate the Lindhard dielectric function. Drawing on their "mathematical trick", we can also improve our own code basis in this respect.

Besides this, we have already put our efforts into the evaluation and post-processing of wavevector-dependent response functions based on ab initio calculated proper conductivity tensors (see our publication [1]). Empirically, this object seems to be wavevector-independent for a wide class of materials. However, although a specific response function may be independent of the wavelength, this property can—according to the universal response relations and as a result of our Central Claim—not be upheld for all response functions simultaneously. Hence, we are in principle able to compute the wavevector-dependent ordinary and extraordinary refractive index from an optical conductivity tensor given only as a function of the frequency.

—Appendices—

Appendix A - Notation

Special objects

Operators	Operators $\hat{O}$ are marked with a hat
Spatial vectors	Spatial vectors are printed bold and indexed by latin numbers ranging from 1 to 3, e. g. $\boldsymbol{x} = (x_i) = (x_1, x_2, x_3)^\mathsf{T}$ is a space vector
Lorentz vectors	Lorentz vectors are printed in normal weight and indexed by greek letters ranging from 0 to 3, e. g. $x = (x^\mu) = (ct, \boldsymbol{x})^\mathsf{T}$ is a space-time coordinate
Spatial tensors	Tensors in space are overset with a double arrow and indexed by two latin numbers, each ranging from 1 to 3, e. g. $(\chi_{ij}) = \overset{\leftrightarrow}{\chi} \in \mathbb{M}^{3\times3}$
Lorentz tensors	Tensors in space-time are printed in normal weight and indexed by two greek letters, each ranging from 0 to 3, e. g. $(\chi^\mu{}_\nu) = \chi \in \mathbb{M}^{4\times4}$
Fourier transform	Fourier transforms of fields and kernels are denoted by the same symbol as the initial functions. Both can be distinguished by their arguments.

Constants

$\mathrm{i}, \quad \mathrm{i}^2 = -1$	Imaginary unit
$\mathrm{e} \approx 2.718\,282$	Base of the natural logarithm or Euler's number
$\pi \approx 3.141\,593$	Ratio of a circle's circumference to its diameter
$\varepsilon_0 \approx 8.854\,188 \times 10^{-12}\,\mathrm{C\,V^{-1}\,m^{-1}}$	Vacuum permittivity or electric constant
$\mu_0 \approx 1.256\,637 \times 10^{-6}\,\mathrm{N\,A^{-2}}$	Vacuum permeability or magnetic constant
$\mu_\mathrm{B} \approx 9.274\,010 \times 10^{-24}\,\mathrm{J\,T^{-1}}$	Bohr magneton or magnetic moment of the electron
$e \approx 1.602\,177 \times 10^{-19}\,\mathrm{C}$	Elementary charge, electrons have charge $(-e)$
$\hbar \approx 1.054\,572 \times 10^{-34}\,\mathrm{J\,s}$	Reduced Planck constant
$m \approx 9.109\,384 \times 10^{-31}\,\mathrm{kg}$	Electron rest mass
$c = 299\,792\,458\,\mathrm{m\,s^{-1}}$	Speed of light in vacuum

$\delta_{ij}, \delta^{\mu}_{\nu}$	Delta Kronecker in three and four dimensions
$\epsilon_{ijk}, \epsilon_{\alpha\beta\mu\nu}$	Levi-Civita Tensor in three and four dimensions

Field quantities and integral kernels[1]

$[\varphi] = \mathrm{V}$	Scalar potential
$[\boldsymbol{A}] = \frac{\mathrm{V\,s}}{\mathrm{m}}$	Vector potential
$[\boldsymbol{E}] = \frac{\mathrm{V}}{\mathrm{m}} = \frac{\mathrm{N}}{\mathrm{C}}$	Electric field (Electric fields strength)
$[\boldsymbol{P}] = [\boldsymbol{D}] = \frac{\mathrm{C}}{\mathrm{m}^2}$	Polarization density / Electric displacement field
$[\boldsymbol{B}] = \frac{\mathrm{V\,s}}{\mathrm{m}^2} = \frac{\mathrm{N}}{\mathrm{A\,m}} = \mathrm{T}$	Magnetic field (Magnetic flux density)
$[\boldsymbol{M}] = [\boldsymbol{H}] = \frac{\mathrm{A}}{\mathrm{m}}$	Magnetization / Magnetic field strength
$[\rho] = \frac{\mathrm{C}}{\mathrm{m}^3}$	Spatial charge density
$[\boldsymbol{j}] = \frac{\mathrm{A}}{\mathrm{m}^2}$	Current density
$[\sigma] = \frac{\mathrm{A}}{\mathrm{V\,m}} = \frac{\mathrm{S}}{\mathrm{m}}$	Conductivity
$[\chi] = \frac{\mathrm{A}}{\mathrm{V\,m\,s}} = \frac{\mathrm{S}}{\mathrm{m\,s}}$	Fundamental / Current response
$[\chi] = \frac{\mathrm{A\,s}}{\mathrm{V\,m}^3}, \quad [\chi_{nn}] = \frac{1}{\mathrm{J\,m}^3}$	Number / Charge density response
$[\chi_{\mu_3\mu_3}] = \frac{\mathrm{A\,m}}{\mathrm{V\,s}}, \quad [\chi] = \frac{1}{\mathrm{J\,m}^3}$	Spin response
$[\varepsilon] = \frac{\mathrm{F}}{\mathrm{m}} = \frac{\mathrm{A\,s}}{\mathrm{V\,m}}$	Permittivity
$[\mu] = \frac{\mathrm{H}}{\mathrm{m}} = \frac{\mathrm{N}}{\mathrm{A}^2} = \frac{\mathrm{V\,s}}{\mathrm{A\,m}}$	Permeability
$[\chi_\mathrm{e}] = [\chi_\mathrm{m}] = 1$	Electric and magnetic susceptibility
$[\varepsilon_\mathrm{r}] = [\mu_\mathrm{r}] = 1$	Relative permittivity and permeability
$[g(E)] = \frac{1}{\mathrm{J\,m}^3}$	Density of states

[1] Note that the deprecated historical terms do not necessarily reflect the true physical character of some fields. See Sct. 2.1 for details.

SI derived units

$\mathrm{C} = \mathrm{A\,s}$	Electric charge
$\mathrm{N} = \frac{\mathrm{J}}{\mathrm{m}} = \frac{\mathrm{kg\,m}}{\mathrm{s}^2}$	Force
$\mathrm{J} = \mathrm{N\,m} = \mathrm{W\,s} = \frac{\mathrm{kg\,m}^2}{\mathrm{s}^2}$	Energy
$\mathrm{W} = \frac{\mathrm{J}}{\mathrm{s}} = \mathrm{V\,A} = \frac{\mathrm{kg\,m}^2}{\mathrm{s}^3}$	Power
$\mathrm{V} = \frac{\mathrm{W}}{\mathrm{A}} = \frac{\mathrm{J}}{\mathrm{C}} = \frac{\mathrm{kg\,m}^2}{\mathrm{s}^3\,\mathrm{A}}$	Electric potential difference (voltage)
$\mathrm{F} = \frac{\mathrm{C}}{\mathrm{V}} = \frac{\mathrm{s}^4\,\mathrm{A}^2}{\mathrm{kg\,m}^2}$	Capacitance
$\Omega = \frac{1}{\mathrm{S}} = \frac{\mathrm{V}}{\mathrm{A}} = \frac{\mathrm{kg\,m}^2}{\mathrm{s}^3\,\mathrm{A}^2}$	Resistance / Impedance
$\mathrm{Wb} = \mathrm{V\,s} = \frac{\mathrm{kg\,m}^2}{\mathrm{s}^2\,\mathrm{A}}$	Magnetic flux
$\mathrm{T} = \frac{\mathrm{Wb}}{\mathrm{m}^2} = \frac{\mathrm{kg}}{\mathrm{s}^2\,\mathrm{A}}$	Magnetic flux density
$\mathrm{H} = \frac{\mathrm{Wb}}{\mathrm{A}} = \frac{\mathrm{kg\,m}^2}{\mathrm{s}^2\,\mathrm{A}^2}$	Inductance

Appendix B - Formulary

B.1. Basic analysis and vector calculus

Einstein notation

$$\boldsymbol{a} \cdot \boldsymbol{b} = \sum_{i=1}^{N=3} a_i b_i \overset{\text{EN}}{=} a_i b_i \tag{B.1}$$

Partial derivatives

$$\frac{\partial f(x,y)}{\partial x} \equiv \partial_x f \equiv f_x \quad \longrightarrow \quad y = \text{const} \tag{B.2}$$

$$\frac{\partial f(x,y)}{\partial y} \equiv \partial_y f \equiv f_y \quad \longrightarrow \quad x = \text{const} \tag{B.3}$$

Schwarz integrability condition

Let the second partial derivatives f_{xy}, f_{yx} be continuous, then for multiple derivatives:

$$\frac{\partial}{\partial x}\left(\frac{\partial f(x,y)}{\partial y}\right) \equiv \frac{\partial}{\partial y}\left(\frac{\partial f(x,y)}{\partial x}\right) \tag{B.4}$$

Fubini's theorem

Let $f : [a,b] \times [c,d] \to \mathbb{R}$ be continuous, then for multiple integrations:

$$\int_c^d \mathrm{d}y \left(\int_a^b \mathrm{d}x\, f(x,y)\right) \equiv \int_a^b \mathrm{d}x \left(\int_c^d \mathrm{d}y\, f(x,y)\right) \tag{B.5}$$

Total derivatives

$$\frac{\mathrm{d}}{\mathrm{d}t} f\big(\{g_1(t), \ldots, g_n(t)\}, t\big) = \frac{\partial f}{\partial t} + \sum_{k=1}^{n} \frac{\partial f}{\partial g_k} \frac{\mathrm{d}g_k(t)}{\mathrm{d}t} \tag{B.6}$$

Gradient of scalar fields

Definition

$$\operatorname{grad} \phi(\boldsymbol{x}) := \nabla \phi(x_1, x_2, x_3) = \begin{pmatrix} \partial_1 \phi \\ \partial_2 \phi \\ \partial_3 \phi \end{pmatrix} \tag{B.7}$$

Linearity

$$\nabla(\alpha f + \beta g) = \alpha\,(\nabla f) + \beta\,(\nabla g) \tag{B.8}$$

Curl of vector fields

Definition

$$\operatorname{curl} \boldsymbol{A}(\boldsymbol{x}) := \nabla \times \boldsymbol{A}(x_1, x_2, x_3) = \begin{pmatrix} \partial_1 \\ \partial_2 \\ \partial_3 \end{pmatrix} \times \begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix} = \begin{pmatrix} \partial_2 A_3 - \partial_3 A_2 \\ \partial_3 A_1 - \partial_1 A_3 \\ \partial_1 A_2 - \partial_2 A_1 \end{pmatrix} \tag{B.9}$$

Linearity

$$\nabla \times (\alpha \boldsymbol{A} + \beta \boldsymbol{B}) = \alpha \, (\nabla \times \boldsymbol{A}) + \beta \, (\nabla \times \boldsymbol{B}) \tag{B.10}$$

Product rule

$$\nabla \times (\phi \boldsymbol{A}) = \phi \, (\nabla \times \boldsymbol{A}) + (\nabla \phi) \times \boldsymbol{A} \tag{B.11}$$

Curl of a pure curl $\longrightarrow$ see "Vector Laplacian"
Curl of a pure gradient field

$$\operatorname{curl} \operatorname{grad} \phi = \nabla \times (\nabla \phi) = \boldsymbol{0} \tag{B.12}$$

Implication (for well-behaving functions on a simply connected set)

$$\nabla \times \boldsymbol{A} = \boldsymbol{0} \quad \Longleftrightarrow \quad \exists \, \phi : \boldsymbol{A} = -\nabla \phi \tag{B.13}$$

Divergence of vector fields

Definition

$$\operatorname{div} \boldsymbol{A}(\boldsymbol{x}) := \nabla \cdot \boldsymbol{A}(x_1, x_2, x_3) = \begin{pmatrix} \partial_1 \\ \partial_2 \\ \partial_3 \end{pmatrix} \cdot \begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix} = \partial_1 A_1 + \partial_2 A_2 + \partial_3 A_3 \tag{B.14}$$

Linearity

$$\nabla \cdot (\alpha \boldsymbol{A} + \beta \boldsymbol{B}) = \alpha \, (\nabla \cdot \boldsymbol{A}) + \beta \, (\nabla \cdot \boldsymbol{B}) \tag{B.15}$$

Product rule

$$\nabla \cdot (\phi \boldsymbol{A}) = \phi \, (\nabla \cdot \boldsymbol{A}) + \boldsymbol{A} \cdot (\nabla \phi) \tag{B.16}$$

Divergence of a pure gradient field $\longrightarrow$ (scalar) Laplace operator Δ

$$\operatorname{div} \operatorname{grad} \phi = \nabla \cdot (\nabla \phi) = \frac{\partial}{\partial x_i} \left(\frac{\partial \phi}{\partial x_i} \right) = \left(\frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2} + \frac{\partial^2}{\partial x_3^2} \right) \phi =: \Delta \phi \tag{B.17}$$

Divergence of a pure curl field

$$\operatorname{div} \operatorname{curl} \boldsymbol{A} = \nabla \cdot (\nabla \times \boldsymbol{A}) = \boldsymbol{0} \tag{B.18}$$

Implication (for well-behaving functions on a simply connected set)

$$\nabla \cdot \boldsymbol{B} = \boldsymbol{0} \quad \Longleftrightarrow \quad \exists \, \boldsymbol{A} : \boldsymbol{B} = \nabla \times \boldsymbol{A} \tag{B.19}$$

Laplace operator

Laplace-Beltrami operator for general curvilinear coordinates ($g_{\mu\nu}$ = metric)

$$\Delta_{\mathrm{LB}} = \frac{1}{\sqrt{|g|}}\,\partial_\mu \left(g^{\mu\nu}\sqrt{|g|}\,\partial_\nu \right) , \quad g = \det(g_{\mu\nu}) \tag{B.20}$$

Cartesian coordinates

$$\Delta \doteq \frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2} + \frac{\partial^2}{\partial x_3^2} \tag{B.21}$$

Spherical coordinates

$$\Delta \doteq \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial}{\partial r} \right) + \frac{1}{r^2 \sin\theta}\frac{\partial}{\partial \theta}\left(\sin\theta \frac{\partial}{\partial \theta} \right) + \frac{1}{r^2 \sin^2\theta}\frac{\partial^2}{\partial \phi^2} \tag{B.22}$$

Vector Laplacian (Graßmann identity for nabla)

$$\Delta\, \boldsymbol{A} = \nabla(\nabla \cdot \boldsymbol{A}) - \nabla \times (\nabla \times \boldsymbol{A}) \overset{\text{cart.}}{=} (\Delta\, A_1, \Delta\, A_2, \Delta\, A_3)^{\mathsf{T}} \tag{B.23}$$

B.2. Special relativity theory

Metric (general)

Bilinearity

$$g : V \times V \to \mathbb{C}\,, \quad (x, y) \mapsto g(x, y) \equiv x \cdot y \tag{B.24}$$

Symmetry

$$g_{\mu\nu} = g_{\nu\mu} \tag{B.25}$$

Inverse

$$g_{\mu\nu} g^{\nu\alpha} = \delta^{\alpha}_{\mu} \tag{B.26}$$

Minkowskian metric tensor

East side signature

$$\eta = \mathrm{diag}(-1, 1, 1, 1) = \begin{pmatrix} -1 & \mathbf{0}^{\mathsf{T}} \\ \mathbf{0} & \overset{\leftrightarrow}{\mathbb{1}}_{3\times 3} \end{pmatrix}\,, \quad \eta_{\mu\nu} = \eta^{\mu\nu} \tag{B.27}$$

Tensor contraction

Einstein notation

$$C^{\alpha}{}_{\beta} = \sum_{\nu=0}^{3} A^{\alpha\nu} B_{\nu\beta} \overset{\text{EN}}{=} A^{\alpha\nu} B_{\nu\beta} \tag{B.28}$$

Symmetric vs. antisymmetric

$$\text{if} \quad A_{\alpha\beta} = -A_{\beta\alpha}\,, \;\; B_{\alpha\beta} = B_{\beta\alpha} \quad \to \quad A_{\alpha\beta} B^{\alpha\beta} = 0 \tag{B.29}$$

Raising and lowering indices

First order tensor

$$x^{\mu} = g^{\mu\nu} x_{\nu}\,, \quad x_{\mu} = g_{\mu\nu} x^{\nu} \tag{B.30}$$

Second order tensor

$$F^{\mu\nu} = g^{\mu\alpha} g^{\nu\beta} F_{\alpha\beta}\,, \quad F_{\mu\nu} = g_{\mu\alpha} g_{\nu\beta} F^{\alpha\beta} \tag{B.31}$$

Mixed-variance tensor

$$T^{\alpha_1 \ldots \alpha_n}{}_{\beta_1 \ldots \beta_m} = g^{\alpha_1\mu_1} \cdots g^{\alpha_n\mu_n}\, g_{\beta_1\nu_1} \cdots g_{\beta_m\nu_m}\, T_{\mu_1 \ldots \mu_n}{}^{\nu_1 \ldots \nu_m} \tag{B.32}$$

Derivatives

Abbreviation

$$\partial^{\mu} \coloneqq \frac{\partial}{\partial x_{\mu}}\,, \quad \partial_{\mu} \coloneqq \frac{\partial}{\partial x^{\mu}} \tag{B.33}$$

D'Alembert operator

$$\Box \coloneqq -\partial_{\mu} \partial^{\mu} \tag{B.34}$$

Compact Ricci index notation

Symmetric parts (index parantheses notation)

$$T_{(\alpha\beta)\gamma\cdots\xi} \coloneqq \frac{1}{2!}\left(T_{\alpha\beta\gamma\cdots\xi} + T_{\beta\alpha\gamma\cdots\xi}\right) \tag{B.35}$$

$$T_{(\alpha\beta\gamma)\delta\cdots\xi} \coloneqq \frac{1}{3!}\left(T_{\alpha\beta\gamma\delta\cdots\xi} + T_{\gamma\alpha\beta\delta\cdots\xi} + T_{\beta\gamma\alpha\delta\cdots\xi} + T_{\alpha\gamma\beta\delta\cdots\xi} + T_{\beta\alpha\gamma\delta\cdots\xi} + T_{\gamma\beta\alpha\delta\cdots\xi}\right) \tag{B.36}$$

Antisymmetric parts (index bracket notation)

$$T_{[\alpha\beta]\gamma\cdots\xi} \coloneqq \frac{1}{2!}\left(T_{\alpha\beta\gamma\cdots\xi} - T_{\alpha\beta\gamma\cdots\xi}\right) \tag{B.37}$$

$$T_{[\alpha\beta\gamma]\delta\cdots\xi} \coloneqq \frac{1}{3!}\left(T_{\alpha\beta\gamma\delta\cdots\xi} + T_{\gamma\alpha\beta\delta\cdots\xi} + T_{\beta\gamma\alpha\delta\cdots\xi} - T_{\alpha\gamma\beta\delta\cdots\xi} - T_{\beta\alpha\gamma\delta\cdots\xi} - T_{\gamma\beta\alpha\delta\cdots\xi}\right) \tag{B.38}$$

Sum (only for two indices)

$$T_{\alpha\beta\gamma\cdots\xi} = T_{(\alpha\beta)\gamma\cdots\xi} + T_{[\alpha\beta]\gamma\cdots\xi} \tag{B.39}$$

Partial derivatives

$$T^{\alpha\beta}{}_{,\mu} \coloneqq \partial_\mu T^{\alpha\beta} = \frac{\partial}{\partial x^\mu} T^{\alpha\beta} \tag{B.40}$$

$$T^{\alpha\beta,\mu} \coloneqq \partial^\mu T^{\alpha\beta} = \frac{\partial}{\partial x_\mu} T^{\alpha\beta} \tag{B.41}$$

Covariant derivatives

$$T^{\alpha\beta}{}_{;\mu} \coloneqq \nabla_\mu T^{\alpha\beta} \tag{B.42}$$

$$T^{\alpha\beta;\mu} \coloneqq \nabla^\mu T^{\alpha\beta} \tag{B.43}$$

Multiple derivatives

$$T^{\alpha\beta}{}_{,\mu\nu} \coloneqq \partial_\nu \partial_\mu T^{\alpha\beta} \tag{B.44}$$

Combined notation

$$3!\, T_{[\alpha\beta,\gamma]} = \partial_\gamma T_{\alpha\beta} + \partial_\beta T_{\gamma\alpha} + \partial_\alpha T_{\beta\gamma} - \partial_\gamma T_{\beta\alpha} - \partial_\beta T_{\alpha\gamma} - \partial_\alpha T_{\gamma\beta} \tag{B.45}$$

Important four-vectors

Name	Contravariant	Covariant	
Four-position	$x^\mu = (ct, \boldsymbol{x})^\mathsf{T}$,	$x_\mu = (-ct, \boldsymbol{x})^\mathsf{T}$	(B.46)
Four-wavevector	$k^\mu = (\omega/c, \boldsymbol{k})^\mathsf{T}$,	$k_\mu = (-\omega/c, \boldsymbol{k})^\mathsf{T}$	(B.47)
Four-momentum	$p^\mu = (E/c, \boldsymbol{p})^\mathsf{T}$,	$p_\mu = (-E/c, \boldsymbol{p})^\mathsf{T}$	(B.48)
Four-potential	$A^\mu = (\varphi/c, \boldsymbol{A})^\mathsf{T}$,	$A_\mu = (-\varphi/c, \boldsymbol{A})^\mathsf{T}$	(B.49)
Four-current	$j^\mu = (c\rho, \boldsymbol{j})^\mathsf{T}$,	$j_\mu = (-c\rho, \boldsymbol{j})^\mathsf{T}$	(B.50)
Four-gradient (sign!)	$\partial^\mu = (-1/c\,\partial_t, \nabla)^\mathsf{T}$,	$\partial_\mu = (1/c\,\partial_t, \nabla)^\mathsf{T}$	(B.51)

Lorentz transformation

Linear map

$$\Lambda : \mathbb{R}^4 \to \mathbb{R}^4 \tag{B.52}$$

$$x \mapsto \Lambda x \tag{B.53}$$

Inverse

$$\Lambda^{\mu'}{}_{\mu} \Lambda_{\mu'}{}^{\nu} = \delta^{\nu}_{\mu} \tag{B.54}$$

Passive transformation

$$x = x^{\mu'} \boldsymbol{e}_{\mu'} = x^{\mu} \boldsymbol{e}_{\mu} \tag{B.55}$$

Contragredient matrix

$$\begin{pmatrix} x^{0'} \\ x^{1'} \\ x^{2'} \\ x^{3'} \end{pmatrix} = \Lambda \begin{pmatrix} x^{0} \\ x^{1} \\ x^{2} \\ x^{3} \end{pmatrix}, \quad \begin{pmatrix} \boldsymbol{e}^{0'} \\ \boldsymbol{e}^{1'} \\ \boldsymbol{e}^{2'} \\ \boldsymbol{e}^{3'} \end{pmatrix} = \left(\Lambda^{\mathsf{T}}\right)^{-1} \begin{pmatrix} \boldsymbol{e}^{0} \\ \boldsymbol{e}^{1} \\ \boldsymbol{e}^{2} \\ \boldsymbol{e}^{3} \end{pmatrix} \tag{B.56}$$

Coordinates

$$x^{\mu'} = \Lambda^{\mu'}{}_{\mu} x^{\mu} \tag{B.57}$$

Basis vectors

$$\boldsymbol{e}_{\mu'} = \Lambda_{\mu'}{}^{\mu} \boldsymbol{e}_{\mu} \tag{B.58}$$

Derivatives

$$\partial_{\mu'} = \Lambda_{\mu'}{}^{\mu} \partial_{\mu} \tag{B.59}$$

Scalar fields

$$\varphi(x) = \varphi'(x') \tag{B.60}$$

Vector fields

$$A^{\mu'}(x') = \Lambda^{\mu'}{}_{\mu} A^{\mu}(x) \tag{B.61}$$

General tensor fields

$$T^{\mu'_1 \cdots \mu'_n}{}_{\nu'_1 \ldots \nu'_m} = \Lambda^{\mu'_1}{}_{\mu_1} \cdots \Lambda^{\mu'_n}{}_{\mu_n} \Lambda_{\nu'_1}{}^{\nu_1} \cdots \Lambda_{\nu'_m}{}^{\nu_m} T^{\mu_1 \ldots \mu_n}{}_{\nu_1 \ldots \nu_m} \tag{B.62}$$

In contrast to general relativity theory, the special theory of relativity considers only linear transformations from and to inertial systems which leave the Minkowskian scalar product invariant. This requirement is just met by the Lorentz transformations Λ with

$$\eta(x', y') = \eta(\Lambda x, \Lambda y) = x^{\mathsf{T}} (\Lambda^{\mathsf{T}} \eta' \Lambda) y \stackrel{!}{=} x^{\mathsf{T}} \eta\, y\,, \tag{B.63}$$

and more specifically with the Minkowski metric explicitly defined in Eq. (B.27),

$$\eta(x, y) = -x^0 y^0 + \boldsymbol{x} \cdot \boldsymbol{y}\,. \tag{B.64}$$

All such transformations build the Lorentz group $\mathscr{L}$ whose elements can be categorized into four so-called connected components, namely proper and improper orthochronous and non-orthochronous ones denoted by $\mathscr{L}_+^\uparrow$, $\mathscr{L}_+^\downarrow$, $\mathscr{L}_-^\downarrow$, $\mathscr{L}_-^\uparrow$, where $(\pm)$ specifies the determinant and $(\uparrow\downarrow)$ tells if the 00-component is greater equal 1 (uparrow) or less equal -1 (downarrow). Orthochronous transformations $\mathrm{O}^+(1,3)$ ($\Lambda^0{}_0 = +1$) preserve the direction of time while proper transformations ($\det \mathscr{L} = +1$) preserve spatial orientation instead of inverting it. Therefore, the proper orthochronous set $\mathscr{L}_+^\uparrow$ or $\mathrm{SO}^+(1,3)$, which naturally includes the one-element $\mathbb{1}_{4x4}$, is a proper subgroup of $\mathscr{L}$. Every element of this subgroup can be generated from a combination of two more basic transformations, namely spatial rotation,

$$\Lambda(R) = \begin{pmatrix} 1 & \mathbf{0}^\mathsf{T} \\ \mathbf{0} & \overleftrightarrow{R} \end{pmatrix} , \tag{B.65}$$

with inverse $\Lambda^{-1}(R) = \Lambda(R^{-1})$, and Lorentz-boost,

$$\Lambda(\boldsymbol{v}) = \begin{pmatrix} \gamma & -\gamma\,\boldsymbol{v}^\mathsf{T}/c \\ -\gamma\,\boldsymbol{v}/c & \overleftrightarrow{\Lambda}(\boldsymbol{v}) \end{pmatrix} , \tag{B.66}$$

with the spatial part

$$\overleftrightarrow{\Lambda}(\boldsymbol{v}) = \overleftrightarrow{\mathbb{1}} + (\gamma - 1)\,\frac{\boldsymbol{v}\boldsymbol{v}^\mathsf{T}}{|\boldsymbol{v}|^2} , \tag{B.67}$$

and the inverse $\Lambda^{-1}(\boldsymbol{v}) = \Lambda(-\boldsymbol{v})$. Here, the boost parameter $\boldsymbol{v}$ is the relative velocity between primed and unprimed inertial frame, and γ denotes the famous Lorentz factor,

$$\gamma = \left(\sqrt{1 - \frac{|\boldsymbol{v}|^2}{c^2}} \right)^{-1} . \tag{B.68}$$

In Eq. (B.65), R is the orthogonal rotation matrix $R^\mathsf{T} = R^{-1}$ with determinant $\det(R) = 1$ (i. e. without roto-reflection). Such proper rotations build the special othogonal group SO(3) and embedded in four-dimensional space they form a subgroup of $\mathscr{L}$ themselves. For Lorentz boosts, the general transformation rule (B.57) becomes

$$t' = \gamma \left(t - \frac{\boldsymbol{v} \cdot \boldsymbol{x}}{c^2} \right) , \tag{B.69}$$

$$\boldsymbol{x}' = \boldsymbol{x} + \boldsymbol{v} \left(-\gamma t + (\gamma - 1)\,\frac{\boldsymbol{v} \cdot \boldsymbol{x}}{|\boldsymbol{v}|^2} \right) . \tag{B.70}$$

Further, it can be shown that two successive Lorentz boosts *in same direction* $\boldsymbol{n}$ with velocities v_1 and v_2 combine into another one in this very direction,

$$\Lambda(v_1 \boldsymbol{n})\,\Lambda(v_2 \boldsymbol{n}) = \Lambda(v\,\boldsymbol{n}) , \tag{B.71}$$

where the new velocity v is obtained by the relativistic velocity addition

$$v = \frac{v_1 + v_2}{1 + v_1 v_2/c^2}\,, \tag{B.72}$$

which follows directly from Eq. (B.66) and does not allow any velocity to exceed the speed of light c.

Now being able to build the proper orthochronous subgroup with Lorentz boosts and spatial rotations, the other three connected components may easily be derived with help of two further fundamental transformations, namely the spatial reflection (a. k. a. parity transformation),

$$P = \mathrm{diag}(1,-1,-1,-1) = \begin{pmatrix} 1 & \mathbf{0}^\mathsf{T} \\ \mathbf{0} & \overset{\leftrightarrow}{-\mathbb{1}} \end{pmatrix}\,, \tag{B.73}$$

and time-reversal (which is not to be confused with the Minkowski metric),

$$T = \mathrm{diag}(-1,1,1,1) = \begin{pmatrix} -1 & \mathbf{0}^\mathsf{T} \\ \mathbf{0} & \overset{\leftrightarrow}{\mathbb{1}} \end{pmatrix}\,, \tag{B.74}$$

such that

$$\mathscr{L}_+^\downarrow = TP\mathscr{L}_+^\uparrow\,, \tag{B.75}$$
$$\mathscr{L}_-^\uparrow = P\mathscr{L}_+^\uparrow\,, \tag{B.76}$$
$$\mathscr{L}_-^\downarrow = T\mathscr{L}_+^\uparrow\,. \tag{B.77}$$

As a consequence, it suffices to characterize the proper orthochronous subgroup in special relativity theory.

Covariant derivative

In order to become *general* covariant, partial derivatives in tensorial equations have to be replaced by covariant derivatives (i. e. the torsion-free Levi-Civita connection to the tangent bundle preserving a given Riemannian metric) according to

$$A^\mu{}_{,\nu} = A^\mu{}_{;\nu} - \Gamma^\mu{}_{\nu\lambda} A^\lambda\,, \tag{B.78}$$
$$A_{\mu,\nu} = A_{\mu;\nu} + \Gamma^\lambda{}_{\nu\mu} A_\lambda\,, \tag{B.79}$$

which applies to all rank-1 tensors like j^μ as well. Here, Γ denotes the so-called Christoffel symbols of the second kind (a. k. a. connection coefficients) defined through the metric $g_{\mu\nu}$ by

$$\Gamma^\mu{}_{\nu\lambda} = \frac{1}{2}\, g^{\mu\alpha}(g_{\alpha\lambda,\nu} + g_{\alpha\nu,\lambda} - g_{\nu\lambda,\alpha})\,, \tag{B.80}$$

which in turn follows from the vanishing covariant derivative of the metric tensor itself. For torsion-free metrics as assumed in general relativity (i. e. $\Gamma^\mu{}_{\nu\lambda} = \Gamma^\mu{}_{\lambda\nu}$), Eq. (C.1) for instance

can equivalently expressed using covariant derivatives,

$$F_{\mu\nu} = (A_{\nu;\mu} + \Gamma^{\lambda}{}_{\mu\nu}) - (A_{\mu;\nu} + \Gamma^{\lambda}{}_{\nu\mu}A_{\mu}) = A_{\nu;\mu} - A_{\mu;\nu} = 2!\, A_{[\nu;\mu]}\,, \tag{B.81}$$

which now constitutes a manifestly *general* covariant equation as opposed to being only (manifestly) Lorentz-covariant. A very important difference to simple partial derivatives is that Schwartz' condition does not apply anymore for covariant derivatives, i. e.

$$A^{a;bc} \neq A^{a;cb}\,. \tag{B.82}$$

Instead, their commutator is given by the famous Riemann curvature tensor,

$$[\nabla_\mu, \nabla_\nu]A^\alpha = R^{\alpha}{}_{\beta\mu\nu}\,A^\beta \tag{B.83}$$

which is in turn can be build from the Christoffel symbols using

$$R^{\alpha}{}_{\beta\mu\nu} = \partial_\mu \Gamma^{\alpha}{}_{\nu\beta} - \partial_\nu \Gamma^{\alpha}{}_{\mu\beta} + \Gamma^{\alpha}{}_{\mu\lambda}\Gamma^{\lambda}{}_{\nu\beta} - \Gamma^{\alpha}{}_{\nu\lambda}\Gamma^{\lambda}{}_{\mu\beta}\,, \tag{B.84}$$

such that it is ultimately based on the metric itself.

In view of their similar name it is most important to distinguish between the covariant derivative from general relativity and the (gauge) covariant derivative introduced in App. C.3. While the former is used to replace partial derivatives when transitioning from flat to "curved" space-time (actually local $\mathbb{R}^4$ on manifolds), where instead of Lorentz transformations the much wider class of general linear transformations GL(4) is allowed, the latter comes into play only in Yang-Mills-type gauge theories like (quantum) electrodynamics with symmetry group U(1), weak interaction with SU(2) or quantum chromodynamics with SU(3). Nevertheless, from a higher mathematical point of view both types can be unified into the generalized theory of principal bundles. For Yang-Mills theories, the potential A^μ then takes the rôle of the Christoffel symbols and the field strength tensor $F^{\mu\nu}$ can be regarded as curvature tensor.

B.3. Fourier transformation

Topology

We postulate that all field quantities should be elements of the Schwartz space,

$$\mathcal{S}(\mathbb{R}^n) \coloneqq \left\{ f \in C^\infty(\mathbb{R}^n) \,|\, \forall \alpha, \beta \in \mathbb{N}_0^n : x^\alpha D^\beta f(x) \in L(\mathbb{R}) \; \forall x \in \mathbb{R}^n \right\} , \tag{B.85}$$

i. e. the vector space of C^∞-functions $f(x)$ on $\mathbb{R}^n$, where $x^\alpha D^\beta f(x)$ is bounded for all non-negative multi-indices α and β. These fields are then called rapidly decreasing, which is a property required for the majority of theorems and rules given in the next sections. In particular, only for such fields the Helmholtz theorem can be applied and an invertible Fourier transform is defined as the automorphism $\mathcal{F} : \mathcal{S}(\mathbb{R}^n) \to \mathcal{S}(\mathbb{R}^n)$. [75]

The topological dual space of $\mathcal{S}$ is the space of tempered distributions $\mathcal{S}'$ (also called space of slowly increasing distributions) and slightly larger than the space of smooth functions with compact support $C_0^\infty(\mathbb{R}^n)$. Each element of this vector space is a continuous linear map $\mathcal{S}(\mathbb{R}^n) \to \mathbb{C}$. The domain of the Fourier transform can be enlarged to include such generalized functions as well [76]. This is important in so far as some of the response functions discussed in the main text are not covered by $L^1(\mathbb{R}^n)$ or $L^2(\mathbb{R}^n)$ but by $\mathcal{S}'(\mathbb{R}^n)$.

Conventions

For classical fields taking a single space-time vector as argument we define the Fourier transformation and its inverse by

$$\rho(\boldsymbol{k}, \omega) = c \int \frac{\mathrm{d}t}{(2\pi)^{1/2}} \int \frac{\mathrm{d}^3\boldsymbol{x}}{(2\pi)^{3/2}} \, \rho(\boldsymbol{x}, t) \, \mathrm{e}^{\mathrm{i}\omega t - \mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}} , \tag{B.86}$$

$$\rho(\boldsymbol{x}, t) = \frac{1}{c} \int \frac{\mathrm{d}\omega}{(2\pi)^{1/2}} \int \frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^{3/2}} \, \rho(\boldsymbol{k}, t) \, \mathrm{e}^{-\mathrm{i}\omega t + \mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x}} , \tag{B.87}$$

or in terms of the corresponding four-vectors,

$$\rho(k) = \int \frac{\mathrm{d}^4 x}{(2\pi)^2} \, \rho(x) \mathrm{e}^{-\mathrm{i}kx} , \tag{B.88}$$

$$\rho(x) = \int \frac{\mathrm{d}^4 k}{(2\pi)^2} \, \rho(k) \mathrm{e}^{\mathrm{i}kx} , \tag{B.89}$$

with the relativistic volume elements $\mathrm{d}^4 x = \mathrm{d}x^0 \mathrm{d}^3\boldsymbol{x}$ and $\mathrm{d}^4 k = \mathrm{d}k^0 \mathrm{d}^3\boldsymbol{k}$, and where the short notation

$$kx \equiv k_\mu x^\mu = -\omega t + \boldsymbol{k} \cdot \boldsymbol{x} \tag{B.90}$$

has been used. If we require "covariance under Fourier transformation" for response relations,

$$\rho_{\mathrm{ind}}(x) = \int \mathrm{d}^4 x' \; \chi(x, x') \, \varphi_{\mathrm{ext}}(x') , \tag{B.91}$$

$$\rho_{\mathrm{ind}}(k) = \int \mathrm{d}^4 k' \; \chi(k, k') \, \varphi_{\mathrm{ext}}(k') , \tag{B.92}$$

the integral kernels necessarily have to obey the following transformation rules,

$$\chi(k,k') = \int \frac{\mathrm{d}^4x}{(2\pi)^2} \int \frac{\mathrm{d}^4x'}{(2\pi)^2}\, \mathrm{e}^{-\mathrm{i}kx}\, \chi(x,x')\, \mathrm{e}^{\mathrm{i}k'x'}\,, \tag{B.93}$$

$$\chi(x,x') = \int \frac{\mathrm{d}^4k}{(2\pi)^2} \int \frac{\mathrm{d}^4k'}{(2\pi)^2}\, \mathrm{e}^{\mathrm{i}kx}\, \chi(k,k')\, \mathrm{e}^{-\mathrm{i}k'x'}\,, \tag{B.94}$$

which are in accordance with the functional chain rule

$$\chi(k,k') = \frac{\delta\rho(k)}{\delta\varphi(k')} = \int \mathrm{d}^4x \int \mathrm{d}^4x'\, \frac{\delta\rho(k)}{\delta\rho(x)}\, \frac{\delta\rho(x)}{\delta\varphi(x')}\, \frac{\delta\varphi(x')}{\delta\varphi(k')} \tag{B.95}$$

$$= \int \mathrm{d}^4x \int \mathrm{d}^4x'\, \frac{\mathrm{e}^{-\mathrm{i}kx}}{(2\pi)^2}\, \chi(x,x')\, \frac{\mathrm{e}^{\mathrm{i}k'x'}}{(2\pi)^2}\,, \tag{B.96}$$

and moreover agree with the conventions commonly applied in numerical implementations like `genspchi0.f90` in "The Elk Code" [31].

Integral representation of the Dirac distribution

Combining the Fourier transformation with its inverse (Eqs. (B.88) and (B.89)) yields

$$\rho(x) = \int \frac{\mathrm{d}^4k}{(2\pi)^2} \left(\int \frac{\mathrm{d}^4x}{(2\pi)^2}\, \rho(x')\, \mathrm{e}^{\mathrm{i}k(x-x')} \right), \tag{B.97}$$

which is equivalent to

$$\rho(x) = \int \mathrm{d}^4x'\, \rho(x') \left(\int \frac{\mathrm{d}^4k}{(2\pi)^4}\, \mathrm{e}^{\mathrm{i}k(x-x')} \right), \tag{B.98}$$

and thus

$$\delta^4(x-x') = \int \frac{\mathrm{d}^4k}{(2\pi)^4}\, \mathrm{e}^{\mathrm{i}k(x-x')}\,, \tag{B.99}$$

with

$$\delta^4(x-x') = \delta(ct-ct')\, \delta^3(\boldsymbol{x}-\boldsymbol{x}')\,. \tag{B.100}$$

Performing this calculation the other way around similarly yields

$$\delta^4(k-k') = \int \frac{\mathrm{d}^4x}{(2\pi)^4}\, \mathrm{e}^{\mathrm{i}x(k-k')}\,, \tag{B.101}$$

with

$$\delta^4(k-k') = c\,\delta(\omega-\omega')\, \delta^3(\boldsymbol{k}-\boldsymbol{k}')\,. \tag{B.102}$$

Homogeneous integral kernels

When homogeneity in space and time is assumed, integral kernels (i. e. functions of two space-time variables) only depend on the difference of their arguments,

$$\chi(x,x') = \chi(x-x') = \chi(\boldsymbol{x}-\boldsymbol{x}', t-t')\,, \tag{B.103}$$

and consequently their Fourier transformation needs to be of the form

$$\chi(k,k') = c\,\delta(\omega-\omega')\,\delta^3(\boldsymbol{k}-\boldsymbol{k}')\,\chi(\boldsymbol{k},\omega)\,. \tag{B.104}$$

With $\boldsymbol{r} = \boldsymbol{x}-\boldsymbol{x}'$ and $\tau = t-t'$, this implies the following explicit transformation rules for homogeneous response functions:

$$\chi(\boldsymbol{k},\omega) = c\int \mathrm{d}\tau \int \mathrm{d}^3\boldsymbol{r}\;\chi(\boldsymbol{r},\tau)\,\mathrm{e}^{\mathrm{i}\omega\tau-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\,, \tag{B.105}$$

$$\chi(\boldsymbol{r},\tau) = \frac{1}{c}\int \frac{\mathrm{d}\omega}{2\pi}\int \frac{\mathrm{d}^3\boldsymbol{q}}{(2\pi)^3}\;\chi(\boldsymbol{k},\omega)\,\mathrm{e}^{-\mathrm{i}\omega\tau+\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\,. \tag{B.106}$$

A comparison with Eqs. (B.86) and (B.87) shows that $\chi(\boldsymbol{k},\omega)$ does not Fourier transform in the same way as the field quantity $\rho(\boldsymbol{k},\omega)$. Simultaneously, the prefactor choice in Eq. (B.104) guarantees that response relations in the Fourier domain are of the conventional form,

$$\rho_{\mathrm{ind}}(\boldsymbol{k},\omega) = \chi(\boldsymbol{k},\omega)\,\varphi_{\mathrm{ext}}(\boldsymbol{k},\omega)\,, \tag{B.107}$$

which transforms into a convolution in real space,

$$\rho_{\mathrm{ind}}(\boldsymbol{x},t) = c\int \mathrm{d}t' \int \mathrm{d}^3\boldsymbol{x}'\;\chi(\boldsymbol{x}-\boldsymbol{x}',t-t')\,\varphi_{\mathrm{ext}}(\boldsymbol{x}',t')\,. \tag{B.108}$$

This shows in particular, that the Fourier covariance from Eqs. (B.93) and (B.94) is lost when working with the reduced transformations.

Regularization of response functions

Being Green functions, response integral kernels have in general distributional character. In order to Fourier transform such objects, they usually have to be regularized first. A central identity from complex analysis in this respect is the Sokhotski-Plemelj formula (a. k. a. Dirac identity) (see e. g. [77, §2.5.7]),

$$\lim_{\eta\to 0^+}\frac{1}{\omega\pm\mathrm{i}\eta} = \mp\mathrm{i}\pi\,\delta(\omega)+\mathcal{P}\frac{1}{\omega}\,, \tag{B.109}$$

where $\mathcal{P}$ denotes the Cauchy principal value. By infinitesimally shifting the pole into the complex plane, improper integrals like

$$\lim_{\eta\to 0^+}\int \mathrm{d}\omega'\;\frac{f(\omega')}{\omega'-\omega\pm\mathrm{i}\eta} = \mp\mathrm{i}\pi\,f(\omega)+\mathcal{P}\int \mathrm{d}\omega'\;\frac{f(\omega')}{\omega'-\omega}\,, \tag{B.110}$$

which otherwise would not exist in the Riemann or Lebesgue sense, can be "made convergent". In physics, this is also known as "regularization". The downside of this procedure however is, that the order of integral evaluation and taking the limit is not arbitrary anymore.

In this context, the Heaviside step function plays an important rôle as well as the most trivial retarded response kernel. According to Eq. (B.105) (restricted to the time domain and

leaving out the constant c consistently), it should Fourier transform as

$$\Theta(\omega) = \int_{-\infty}^{\infty} \mathrm{d}\tau\, \mathrm{e}^{\mathrm{i}\omega\tau}\, \Theta(\tau) = \int_{0}^{\infty} \mathrm{d}\tau\, \mathrm{e}^{\mathrm{i}\omega\tau} = \frac{\mathrm{e}^{\mathrm{i}\omega\tau}}{\mathrm{i}\omega}\bigg|_{0}^{\infty}. \tag{B.111}$$

Apparently, this integral does not converge because the exponential function with purely imaginary argument oscillates for $\tau \to \infty$. By performing the following replacement,

$$\Theta(\omega) \mapsto \lim_{\eta\to 0^+} \int_{0}^{\infty} \mathrm{d}\tau\, \mathrm{e}^{\mathrm{i}(\omega+\mathrm{i}\eta)\tau} = \lim_{\eta\to 0^+} \frac{\mathrm{e}^{\mathrm{i}(\omega+\mathrm{i}\eta)\tau}}{\mathrm{i}(\omega+\mathrm{i}\eta)}\bigg|_{0}^{\infty} = \lim_{\eta\to 0^+} \frac{\mathrm{i}}{\omega+\mathrm{i}\eta}\,, \tag{B.112}$$

this behavior can be "fixed". Fourier back-transforming this expression then yields the integral representation of the Heaviside step function (cf. [75, p. 1229]),

$$\Theta(\tau) = -\frac{1}{2\pi\mathrm{i}} \lim_{\eta\to 0^+} \int \mathrm{d}\omega\, \frac{\mathrm{e}^{-\mathrm{i}\omega\tau}}{\omega+\mathrm{i}\eta}\,, \tag{B.113}$$

which can alternatively be derived directly from Cauchy's integral formula. In the main text, this identity is used in form of

$$\mp\frac{\mathrm{i}}{\hbar}\,\Theta(\pm\tau)\,\mathrm{e}^{-\mathrm{i}\omega_0\tau} = \lim_{\eta\to 0^+} \int \frac{\mathrm{d}\omega}{2\pi}\, \frac{\mathrm{e}^{-\mathrm{i}\omega\tau}}{\omega-\omega_0\pm\mathrm{i}\eta}\,, \tag{B.114}$$

in order to derive the Fourier transform of the spectral representation of a general response function within the Kubo formalism.

Now, any retarded response function with $\chi(\tau) \equiv 0$ for $\tau < 0$ evidently satisfies the following identity,

$$\chi(\tau) = \Theta(\tau)\,\chi(\tau)\,. \tag{B.115}$$

By utilizing the integral representation of the Dirac delta distribution in the time domain,

$$\delta(\tau-\tau') = \int \frac{\mathrm{d}\omega}{2\pi}\, \mathrm{e}^{-\mathrm{i}\omega'(\tau-\tau')}\,, \tag{B.116}$$

and performing some rearrangements of terms, we further find

$$\chi(\omega) = \int \mathrm{d}\tau \int \mathrm{d}\tau'\, \delta(\tau-\tau')\,\Theta(\tau)\,\chi(\tau')\,\mathrm{e}^{\mathrm{i}\omega\tau} \tag{B.117}$$

$$= \int \frac{\mathrm{d}\omega'}{2\pi}\, \Theta(\omega-\omega')\,\chi(\omega') \tag{B.118}$$

$$= -\frac{1}{2\pi\mathrm{i}} \lim_{\eta\to 0^+} \int \mathrm{d}\omega'\, \frac{\chi(\omega')}{\omega-\omega'+\mathrm{i}\eta}\,. \tag{B.119}$$

After inserting the Sokhotski-Plemelj identity (B.110), the latter equation then produces the famous Kramers-Kronig relation,

$$\chi(\omega) = \frac{1}{\mathrm{i}\pi}\,\mathcal{P}\int \mathrm{d}\omega'\, \frac{\chi(\omega')}{\omega-\omega'}\,, \tag{B.120}$$

which translates to the following two Hilbert transformations for the real (χ_R) and imaginary

part (χ_I) of the response function,

$$\chi_\mathrm{R}(\omega) = -\mathcal{P}\int \frac{\mathrm{d}\omega'}{\pi}\,\frac{\chi_\mathrm{I}(\omega')}{\omega-\omega'}\,, \tag{B.121}$$

$$\chi_\mathrm{I}(\omega) = +\mathcal{P}\int \frac{\mathrm{d}\omega'}{\pi}\,\frac{\chi_\mathrm{R}(\omega')}{\omega-\omega'}\,. \tag{B.122}$$

This form of the Kramers-Kronig relations in particular show that real and imaginary parts of response functions are not independent from each other.

Lastly, we remark that explicitly making a Green function retarded ($\omega \mapsto \omega + \mathrm{i}\eta$) or advanced ($\omega \mapsto \omega - \mathrm{i}\eta$) are not the only (physically reasonable) options for regularization. In fact, since Green functions provide solutions to inhomogeneous linear differential equations, they are never uniquely defined. Instead, another form can always be constructed by adding an arbitrary homogeneous solution. Other regularization choices for example lead to the time-ordered Green function, which is an important object in quantum field theory.

Important properties and theorems

Let $g(k) = \mathcal{F}[f(x)](k)$ be the Fourier transform and $f(x) = \mathcal{F}^{-1}[g(k)](x)$ its well-defined inverse. Further, let c_1, c_2, x_0, k_0 and α be real constants. Then the following properties apply:

Linearity

$$\mathcal{F}[c_1 f_1 + c_2 f_2] = c_1\mathcal{F}[f_1] + c_2\mathcal{F}[f_2] \tag{B.123}$$

Translation

$$\mathcal{F}[f(x-x_0)] = \mathrm{e}^{-\mathrm{i}kx_0} g(k) \tag{B.124}$$

Modulation

$$\mathcal{F}[e^{\mathrm{i}k_0 x} f(x)] = g(k-k_0) \tag{B.125}$$

Scaling

$$\mathcal{F}[f(\alpha x)] = \frac{1}{|\alpha|}\, g\left(\frac{k}{\alpha}\right) \tag{B.126}$$

Conjugation

$$\mathcal{F}[f^*(x)] = g^*(-k) \tag{B.127}$$

Reality and imaginary condition

$$f \text{ is purely real} \quad \Rightarrow \quad g(-k) = g^*(k) \tag{B.128}$$

$$f \text{ is purely imaginary} \quad \Rightarrow \quad g(-k) = -g^*(k) \tag{B.129}$$

Derivative

$$\mathcal{F}[f'(x)] = \mathrm{i}k\, g(k) \tag{B.130}$$

Integration

$$g(0) = \frac{1}{\sqrt{2\pi}}\int \mathrm{d}x\; f(x) \tag{B.131}$$

Parseval identity

$$\int \mathrm{d}x\; f_1^*(x)\, f_2(x) = \int \mathrm{d}k\; g_1^*(k)\, g_2(k) \tag{B.132}$$

Convolution theorem

$$\mathcal{F}[f_1 * f_2] = \mathcal{F}[f_1]\,\mathcal{F}[f_2]\,, \quad \mathcal{F}[f_1 f_2] = \mathcal{F}[f_1] * \mathcal{F}[f_2] \tag{B.133}$$

with

$$(f * g)(y) = \frac{1}{\sqrt{2\pi}} \int \mathrm{d}x\; f(x) g(y - x) \tag{B.134}$$

Note, that the latter definition only applies to the convolution of two fields. For response relations, Eq. (B.108) has to be used.

Differential operator mapping rules

Minkowskian derivative

$$\partial_\mu \mapsto \mathrm{i} k_\mu \tag{B.135}$$

Cartesian and temporal derivatives

$$\partial_t \mapsto -\mathrm{i}\omega\,, \tag{B.136}$$

$$\partial_i \mapsto \mathrm{i} k_i \tag{B.137}$$

Laplace and d'Alembert operator

$$\Delta \mapsto (\mathrm{i}\boldsymbol{k}) \cdot (\mathrm{i}\boldsymbol{k}) = -|\boldsymbol{k}|^2\,, \tag{B.138}$$

$$\Box \mapsto -\omega^2/c^2 + |\boldsymbol{k}|^2 \tag{B.139}$$

Nabla applications

$$\nabla \circ \mapsto \mathrm{i}\boldsymbol{k} \circ \tag{B.140}$$

where $\circ$ can be either of $\cdot\boldsymbol{A}$, $\times\boldsymbol{A}$ or φ.

B.4. Functional derivatives

Definition

The equation

$$\int \mathrm{d}x \, \frac{\delta F[y]}{\delta y(x)} \, h(x) = \lim_{\epsilon \to 0} \frac{F[y + \epsilon h] - F[y]}{\epsilon} \tag{B.141}$$

has to apply for all (arbitrary) test functions $h(x)$.

Properties and rules

Linearity

$$\frac{\delta}{\delta y(x)} \left(\alpha F[y] + \beta G[y] \right) = \alpha \, \frac{\delta F[y]}{\delta y(x)} + \beta \, \frac{\delta G[y]}{\delta y(x)} \tag{B.142}$$

Product rule

$$\frac{\delta}{\delta y(x)} \left(F[y] G[y] \right) = F[y] \, \frac{\delta G[y]}{\delta y(x)} + \frac{\delta F[y]}{\delta y(x)} \, G[y] \tag{B.143}$$

Chain rule

$$\frac{\delta F[y]}{\delta y(x)} = \int \mathrm{d}x' \, \frac{\delta F[G]}{\delta G(x')} \, \frac{\delta G[y](x')}{\delta y(x)} \tag{B.144}$$

Local functionals

$$F[y] = \int \mathrm{d}x \, g(y(x)) \quad \Rightarrow \quad \frac{\delta F[y]}{\delta y(x)} = g'(y(x)) \, . \tag{B.145}$$

Linear functionals

$$F[y] = \int \mathrm{d}x \, g(x) y(x) = \left\langle y, \frac{\delta F[y]}{\delta y} \right\rangle \quad \Rightarrow \quad \frac{\delta F[y]}{\delta y(x)} = g(x) \tag{B.146}$$

Distributions

$$\frac{\delta f(x)}{\delta f(x')} = \delta(x - x') \tag{B.147}$$

Fourier space

$$\frac{\delta F[y]}{\delta y(x)} \; \overset{\mathcal{F}}{\longmapsto} \; \frac{\partial F(y)}{\partial y} \tag{B.148}$$

B.5. Projectors and Helmholtz theorem

Definition

Let V be a vector space. The endomorphism $P : V \to V$ is then called a projection if it is idempotent, i. e. $P^2 = P$. Consequently, the only possible eigenvalues are 0 and 1 with corresponding eigenspaces $\ker P$ and $\operatorname{im} P$. The vector space V can then be expressed as the direct sum

$$V = \ker P \oplus \operatorname{im} P \,. \tag{B.149}$$

If P is a projection then

$$\bar{P} = \mathbb{1} - P \tag{B.150}$$

is one as well. Both are further related by

$$\ker P = \operatorname{im} \bar{P} \,, \tag{B.151}$$

$$\operatorname{im} P = \ker \bar{P} \,, \tag{B.152}$$

Suppose the subspaces V_1 and V_2 denote the range and kernel of P respectively, then V_1 is the complement of V_2. In particular, P acts on V_1 as identity operator,

$$\forall v_1 \in V_1 : P v_1 = v_1 \,, \tag{B.153}$$

just as $\bar{P}$ does on the subspace V_2,

$$\forall v_2 \in V_2 : \bar{P} v_2 = v_2 \,. \tag{B.154}$$

By contrast, applying any of these two projectors to a vector $v \in V$ will project v onto the respective image subspace,

$$P v = \begin{pmatrix} v_1 \\ 0 \end{pmatrix} \,, \quad \bar{P} v = \begin{pmatrix} 0 \\ v_2 \end{pmatrix} \tag{B.155}$$

such that every vector $v \in V$ is uniquely determined by the sum

$$v = v_1 \oplus v_2 = \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} \tag{B.156}$$

and the vector space V can be written as

$$V = V_1 \oplus V_2 = \left\{ \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} , v_1 \in V_1, v_2 \in V_2 \right\} \,. \tag{B.157}$$

In case V is complete and has a scalar product (i. e. if V is a Hilbert space), then $P = P^\dagger$ is called orthogonal projection if it additionally satisfies

$$\forall v, w \in V : \langle Pv, w \rangle = \langle Pv, Pw \rangle = \langle v, Pw \rangle \,. \tag{B.158}$$

Further, the inner product of V can be decomposes into

$$\left\langle \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} \middle| \begin{pmatrix} w_1 \\ w_2 \end{pmatrix} \right\rangle_{V_1 \oplus V_2} = \langle v_1 \,|\, w_1 \rangle_{V_1} + \langle v_2 \,|\, w_2 \rangle_{V_2} \,. \tag{B.159}$$

and $V_2 = V_1^{\perp}$ is then called *orthogonal* complement of V_1.

Longitudinal and transverse projectors in Fourier space

We define the longitudinal projection operator in index and matrix notation by

$$\overset{\leftrightarrow}{P}_{\mathrm{L}}(\boldsymbol{k}) = \frac{\boldsymbol{k}\boldsymbol{k}^{\mathsf{T}}}{|\boldsymbol{k}|^2}\,, \quad (P_{\mathrm{L}})_{ij}(\boldsymbol{k}) = \frac{k_i k_j}{|\boldsymbol{k}|^2}\,, \tag{B.160}$$

together with the transverse projection operator

$$\overset{\leftrightarrow}{P}_{\mathrm{T}}(\boldsymbol{k}) = \overset{\leftrightarrow}{\mathbb{1}} - \overset{\leftrightarrow}{P}_{\mathrm{L}}(\boldsymbol{k})\,, \quad (P_{\mathrm{T}})_{ij}(\boldsymbol{k}) = \delta_{ij} - \frac{k_i k_j}{|\boldsymbol{k}|^2}\,. \tag{B.161}$$

By rewriting the latter using the Graßmann identity, the explicit application of $\overset{\leftrightarrow}{P}_{\mathrm{T}}$ to a vector field $\boldsymbol{E} = \boldsymbol{E}(\boldsymbol{k})$ in Fourier space is given by

$$\overset{\leftrightarrow}{P}_{\mathrm{T}}(\boldsymbol{k})\,\boldsymbol{E} = \frac{|\boldsymbol{k}|^2 \boldsymbol{E} - \boldsymbol{k}(\boldsymbol{k}\cdot\boldsymbol{E})}{|\boldsymbol{k}|^2} = \frac{\boldsymbol{k}\times(\boldsymbol{k}\times\boldsymbol{E})}{|\boldsymbol{k}|^2}\,. \tag{B.162}$$

In addition, we define the transverse rotation operator

$$(R_{\mathrm{T}})_{ij}(\boldsymbol{k}) = \epsilon_{ilj}\,\frac{k_l}{|\boldsymbol{k}|}\,, \tag{B.163}$$

and its action on a vector,

$$\overset{\leftrightarrow}{R}_{\mathrm{T}}(\boldsymbol{k})\,\boldsymbol{E} = \frac{\boldsymbol{k}\times\boldsymbol{E}}{|\boldsymbol{k}|} = \frac{1}{|\boldsymbol{k}|}\begin{pmatrix} 0 & -k_3 & k_2 \\ k_3 & 0 & -k_1 \\ -k_2 & k_1 & 0 \end{pmatrix}\begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix}\,. \tag{B.164}$$

In case of $\boldsymbol{k}$ being a unit vector, e.g. $\boldsymbol{n} = \boldsymbol{k}/|\boldsymbol{k}|$, these three operators simplify to

$$\overset{\leftrightarrow}{P}_{\mathrm{L}}\,\boldsymbol{E} = \boldsymbol{n}(\boldsymbol{n}\cdot\boldsymbol{E})\,, \quad \overset{\leftrightarrow}{P}_{\mathrm{T}}\,\boldsymbol{E} = -\boldsymbol{n}\times(\boldsymbol{n}\times\boldsymbol{E})\,, \quad \overset{\leftrightarrow}{R}_{\mathrm{T}}\,\boldsymbol{E} = \boldsymbol{n}\times\boldsymbol{E}\,, \tag{B.165}$$

from which is obvious that the three resulting vectors are orthogonal. Moreover, $\{\overset{\leftrightarrow}{P}_{\mathrm{L}}, \overset{\leftrightarrow}{P}_{\mathrm{T}}, \overset{\leftrightarrow}{R}_{\mathrm{T}}\}$ form a commutative algebra which is represented by the following multiplication table (see [6, Tab. 1]):

$\cdot$	$\overset{\leftrightarrow}{P}_{\mathrm{L}}$	$\overset{\leftrightarrow}{P}_{\mathrm{T}}$	$\overset{\leftrightarrow}{R}_{\mathrm{T}}$
$\overset{\leftrightarrow}{P}_{\mathrm{L}}$	$\overset{\leftrightarrow}{P}_{\mathrm{L}}$	0	0
$\overset{\leftrightarrow}{P}_{\mathrm{T}}$	0	$\overset{\leftrightarrow}{P}_{\mathrm{T}}$	$\overset{\leftrightarrow}{R}_{\mathrm{T}}$
$\overset{\leftrightarrow}{R}_{\mathrm{T}}$	0	$\overset{\leftrightarrow}{R}_{\mathrm{T}}$	$-\overset{\leftrightarrow}{P}_{\mathrm{T}}$

(B.166)

Helmholtz's theorem

The fundamental theorem of vector calculus (also known as Helmholtz's Theorem) states, that each (well behaving) vectorial field is uniquely determined by its sources and curls, or more precisely, can be written as a sum of a pure curl and a gradient field,

$$\boldsymbol{E}(\boldsymbol{x},t) = -\nabla\varphi(\boldsymbol{x},t) + \nabla \times \boldsymbol{W}(\boldsymbol{x},t)\,, \tag{B.167}$$

where the field's sources and curl enter in the definition of the components [24, eq. 3.40]

$$\varphi(\boldsymbol{x},t) = \frac{1}{4\pi}\int \mathrm{d}^3\boldsymbol{x}'\ \frac{(\nabla'\cdot\boldsymbol{E})(\boldsymbol{x}',t)}{|\boldsymbol{x}-\boldsymbol{x}'|}\,, \tag{B.168}$$

and

$$\boldsymbol{W}(\boldsymbol{x},t) = \frac{1}{4\pi}\int \mathrm{d}^3\boldsymbol{x}'\ \frac{(\nabla'\times\boldsymbol{E})(\boldsymbol{x}',t)}{|\boldsymbol{x}-\boldsymbol{x}'|}\,. \tag{B.169}$$

Since pure curl fields are divergence-free (solenoidal) and gradient fields are curl-free (irrotational), the Helmholtz theorem can also be interpreted as a decomposition into transverse and longitudinal components,

$$\boldsymbol{E}(\boldsymbol{x},t) = \boldsymbol{E}_{\mathrm{L}}(\boldsymbol{x},t) + \boldsymbol{E}_{\mathrm{T}}(\boldsymbol{x},t)\,. \tag{B.170}$$

By making use of the longitudinal and transverse projection operators (B.160) and (B.161), Eq. (B.170) can be equivalently formulated in Fourier space as

$$\boldsymbol{E}(\boldsymbol{k},\omega) = \overleftrightarrow{P}_{\mathrm{L}}(\boldsymbol{k})\,\boldsymbol{E}(\boldsymbol{k},\omega) + \overleftrightarrow{P}_{\mathrm{T}}(\boldsymbol{k})\,\boldsymbol{E}(\boldsymbol{k},\omega) \equiv \boldsymbol{E}_{\mathrm{L}}(\boldsymbol{k},\omega) + \boldsymbol{E}_{\mathrm{T}}(\boldsymbol{k},\omega)\,, \tag{B.171}$$

where the two latter factors are explicitly given by

$$\boldsymbol{E}_{\mathrm{L}}(\boldsymbol{k},\omega) = \frac{\boldsymbol{k}(\boldsymbol{k}\cdot\boldsymbol{E}(\boldsymbol{k},\omega))}{|\boldsymbol{k}|^2} \tag{B.172}$$

and

$$\boldsymbol{E}_{\mathrm{T}}(\boldsymbol{k},\omega) = -\frac{\boldsymbol{k}\times(\boldsymbol{k}\times\boldsymbol{E}(\boldsymbol{k},\omega))}{|\boldsymbol{k}|^2}\,. \tag{B.173}$$

Both parts are orthogonal in Fourier space w. r. t. the euclidean scalar product

$$\boldsymbol{E}_{\mathrm{L}}^*(\boldsymbol{k})\cdot\boldsymbol{E}_{\mathrm{T}}(\boldsymbol{k})\,, \tag{B.174}$$

and in real space w. r. t. the inner product

$$\int \mathrm{d}^3\boldsymbol{x}\ \ \boldsymbol{E}_{\mathrm{L}}(\boldsymbol{x},t)\cdot\boldsymbol{E}_{\mathrm{T}}(\boldsymbol{x},t) = 0\,, \tag{B.175}$$

where we assumed that $\boldsymbol{E}(\boldsymbol{x},t)$ is a real vector field.

Inversion

In general, projectors cannot be inverted because their determinant is always zero. However, objects containing contributions from two complementary projectors can. In particular, this is not restricted to the prominent example of response tensors in the isotropic limit (see Sct. 2.5) as we will prove now for the projector pair $\{\overleftrightarrow{P}_{\mathrm{L}}, \overleftrightarrow{P}_{\mathrm{T}}\}$.

Let M be a general tensor-valued object of the form

$$\overleftrightarrow{M} = a\overleftrightarrow{P}_{\mathrm{L}} + b\overleftrightarrow{P}_{\mathrm{T}}\,, \tag{B.176}$$

with functions $a, b \neq 0$. Then its invese is given by

$$\overleftrightarrow{M}^{-1} = \frac{1}{a}\overleftrightarrow{P}_{\mathrm{L}} + \frac{1}{b}\overleftrightarrow{P}_{\mathrm{T}}\,. \tag{B.177}$$

This can be verified easily by multiplying both tensors with each other,

$$\overleftrightarrow{M}\overleftrightarrow{M}^{-1} = \frac{a}{a}(\overleftrightarrow{P}_{\mathrm{L}})^2 + \frac{a}{b}\overleftrightarrow{P}_{\mathrm{L}}\overleftrightarrow{P}_{\mathrm{T}} + \frac{b}{a}\overleftrightarrow{P}_{\mathrm{T}}\overleftrightarrow{P}_{\mathrm{L}} + \frac{b}{b}(\overleftrightarrow{P}_{\mathrm{T}})^2\,. \tag{B.178}$$

With help of the multiplication table (B.166) this can be simplified for all $a, b \neq$ to

$$\overleftrightarrow{M}\overleftrightarrow{M}^{-1} = \overleftrightarrow{M}^{-1}\overleftrightarrow{M} = \overleftrightarrow{P}_{\mathrm{L}} + \overleftrightarrow{P}_{\mathrm{T}} = \overleftrightarrow{\mathbb{1}}\,, \tag{B.179}$$

which completes the proof.

The subtle catch here is that the inverse is only defined as long as both functions a and b are non-zero as is already obvious from Eq. (B.177). For response tensors and other tensorial objects this means that even if they are invertible in their general isotropic form, this feature can be lost by taking a specific limit or evaluating the function for a specific set of arguments where one of the prefactors vanish. For example the electric solution generator (1.54) in the static limit is identical to $\overleftrightarrow{P}_{\mathrm{L}}$ and thus not of the required form (B.176) anymore.

B.6. Complex analysis

All following calculations are based on the rationales of complex analysis a. k. a. theory of functions of one complex variable. Every justification used below can be found in standard literature on this topic, preferably Ref. [78].

Complex logarithm

The natural logarithm of a non-zero complex number can naïvely be calculated as follows:

$$\ln(z) = \ln(r\mathrm{e}^{\mathrm{i}\varphi}) = \ln(r) + \mathrm{i}\arg(\mathrm{e}^{\mathrm{i}\varphi}) \stackrel{?}{=} \ln(r) + \mathrm{i}\varphi\,. \tag{B.180}$$

However, because this equation is fulfilled for every other angle $\varphi' = \varphi + n(2\pi)$ with $n \in \mathbb{Z}$ as well, the complex logarithm can only be defined as a multivalued function. That is, every complex number z has infinitely many complex logarithms as visualized in Fig. B.1b. Since this is not particularly useful in practical calculations, one has to fix a so-called branch of the function, which works as follows: Let G be a connected open subset of $\mathbb{C}$ not containing the origin. Then a branch of $\arg(z)$ in G is a *continuous* function $\alpha(z)$ for which $\alpha(z) = \arg(z)$ for each $z \in G$. The easiest way to remove the discontinuity of $\arg(z)$ is to exclude the negative real axis from $\mathbb{C}$ such that $G = \mathbb{C} \setminus \{\, x \in \mathbb{R} \colon x \leq 0 \,\}$. The associated branch of $\arg(z)$ and $\ln(z)$ are then called *principal branches* of the respective functions. In particular, we then have

$$\mathrm{Ln}(z) = \mathrm{Ln}\left(|z|\,\mathrm{e}^{\mathrm{i}\arg(z)}\right) \stackrel{\mathrm{def}}{=} \mathrm{Ln}(|z|) + i\,\mathrm{Arg}(z) \tag{B.181}$$

where $\mathrm{Arg}(z)$ denotes the *principal argument* of z, i. e. the angle which lies in the interval $(-\pi, \pi]$.[1]

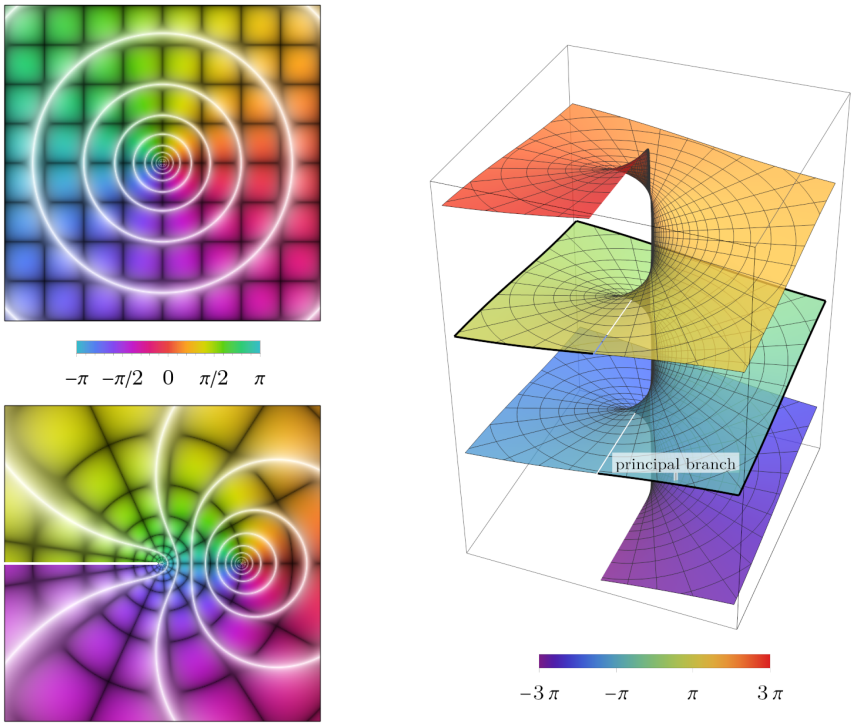
Having chosen a particular branch of $\ln(z)$, it can be shown that $\mathrm{Ln}(z)$ is holomorphic (i. e. satisfies the Cauchy-Riemann equations) and its derivative is given by

$$\mathrm{Ln}'(z) = \frac{1}{z}\,. \tag{B.182}$$

Being holomorphic in particular means that a function can be locally, say at point z_0, represented by power series with center z_0. The region of convergence has then to be determined appropriately. The radius of convergence R is then defined as the supremum of $|z - z_0|$ over all z for which the series converges. In case $R > 0$, the series converges absolutely and locally uniformly in the disk defined by $|z - z_0| < R$. Likewise, if $R < \infty$, the series diverges everywhere outside this disk.

In general, specific branches $l(f)$ of logarithms of holomorphic functions $\ln(f)$ can be constructed by a special integration. This holds in particular for $f(z) = z$, i. e. for the natural logarithm itself. If G is a *simply* connected open subset of $\mathbb{C}$ not containing the

[1] This is in fact the common definition implemented in modern programming languages like Mathematica, python/numpy, C, etc.. It must not be confused with a related definition where the argument runs from 0 to 2π, in which case the branch cut would be on the positive real axis.



(a) Phase portrait of $\mathrm{Ln}(z)$ **(b)** Riemann surface of $\ln(z)$

Fig. B.1.: Visualization of the complex logarithm. The upper part of (a) shows the phase portrait of the complex domain, i. e. $f(z) = z$, as reference. There, $\mathrm{Re}(f)$ and $\mathrm{Im}(f)$ are darkened periodically (black grid), whereas $\ln|f|$ is periodically brightened (concentric circles). Principal phases $\mathrm{Arg}(z)$ are coloured according to the legend. The bottom part of (a) is the image of $z \mapsto \mathrm{Ln}(z)$ using the same coloring scheme. By contrast, (b) shows the imaginary part of the multivalued complex logarithm $\ln(z)$, which is a non-compact Riemann surface provided by analytic continuation. The white strip along the negative real axis emphasizes the branch cut in both latter plots, which emerge from the indetermination of $\arg(z)$ along at these points.

origin, then a specific branch of $\ln(z)$ on G can be constructed by

$$l(z) = \ln(a) + \int_a^z \frac{\mathrm{d}z'}{z'}\,, \tag{B.183}$$

where a is some point in G and $\ln(a)$ can be any logarithm of the complex number a. The catch here is that, in contrast to a one-dimensional integral on the real axis where only the direction of integration has to be fixed, in order to determine a complex integral an entire curve in $\mathbb{C}$ or G needs to be specified in general. More explicitly, the Riemann integral of a complex-valued function with sufficiently good properties along a curve parametrized by $\gamma\colon[a,b] \to \mathbb{C}$ is defind by the line integral

$$\int_\gamma \mathrm{d}z\, f(z) = \int_a^b \mathrm{d}t\, f(\gamma(t))\gamma'(t)\,. \tag{B.184}$$

If the integrand is the derivative of a holomorphic function (which is then holomorphic itself),

[1] A nice introduction to different methods of creating such (enhanced) phase portraits has been written by E. Wegert [79]. The yearly released "Math Calendar" [80] composed from such domain colored plots is published by Wegert et al. on the author's alma mater as well.

the generalized complex version of the fundamental theorem of calculus,

$$\int_\gamma \mathrm{d}z\, f'(z) = f(\gamma(b)) - f(\gamma(a))\,, \tag{B.185}$$

is obtained. This follows from the property that for holomorphic functions, the value of the integral in Eq. (B.184) is invariant under continuous deformation of the path γ while holding the endpoint fixed. This is particulary relevant for *simply* connected sets, where *any* path can be deformed continuously into *any* other. Consequently, it is sufficient to fix the starting point a in Eq. (B.183). Adjacent branches can then be entered by analytic continuation.

Mercator series

At some point in the main text we need to refer to the following complex power series:

$$\mathrm{Ln}\left(\frac{z+1}{z-1}\right) = 2\left(\frac{1}{z} + \frac{1}{3z^3} + \frac{1}{5z^5} + \cdots\right) \quad \text{for } z \to \infty\,. \tag{B.186}$$

This is evidently a special case of a Laurent series,

$$f(z; z_0) = \underbrace{\sum_{n=0}^{\infty} a_n\,(z-z_0)^n}_{\text{analytic part}} + \underbrace{\sum_{n=1}^{\infty} b_n\,(z-z_0)^{(-n)}}_{\text{principal part}}\,, \tag{B.187}$$

centered around $z_0 = 0$ and with all $a_n \equiv 0$, i. e. consisting only of a principal part. Laurent series are a handy tool from complex analysis to find power series of functions which cannot be represented by Taylor series. This is especially useful when the point of expansion is an isolated singularity. Apparently, in case all b_n are zero, the regular Taylor series is retained and hence Laurent series are a generalization of the latter. The coefficients a_n and b_n are formally defined via Cauchy's differentiation formula[2] such that

$$a_n = \frac{1}{2\pi\mathrm{i}} \oint_\gamma \mathrm{d}s\, \frac{f(s)}{(s-z_0)^{n+1}}\,, \qquad b_n = \frac{1}{2\pi\mathrm{i}} \oint_\gamma \mathrm{d}s\, f(s)\,(s-z_0)^{n-1}\,, \tag{B.188}$$

where γ describes a Jordan curve in the complex plane enclosing the expansion point z_0. If γ lies in an annulus in which $f(z)$ is holomorphic, then the Laurent series is valid everywhere inside this annulus. In pactice however, the above formulae for a_n and b_n have only conceptional value. Instead, Laurent series are usually obtained by manipulating known Taylor series, which we will do in the following as well.

We start by reformulating the derivative of some branch of the complex logarithm as follows,

$$l'(z) = \frac{1}{z} = \frac{1}{a}\,\frac{1}{1-\left(-\frac{z-a}{a}\right)}\,, \quad \text{for } a \neq 0\,. \tag{B.189}$$

The first equality has already been disussed above, wheras the second one is just a trivial rearrangement of terms. The right-hand side can now be formulated as a geometric series in

[2] This follows from Cauchy's integral formula, which in turn follows from Cauchy's integral theorem.

the variable $w = -\frac{z-a}{a}$,

$$\frac{1}{a}\frac{1}{1-w} = \frac{1}{a}\sum_{n=0}^{\infty} w^k \quad \text{for } |w| < 1\,, \tag{B.190}$$

and hence

$$\frac{1}{z} = \sum_{n=0}^{\infty} \frac{(-1)^n\,(z-a)^n}{a^{n+1}} \quad \text{for } |z-a| < |a|\,. \tag{B.191}$$

Now applying the construction rule (B.183) for new branches of $\ln(z)$ on the left-hand side and regular term-wise integration for the complex polynomial on the right-hand side leads to

$$l(z) - \ln(a) = \int_a^z \frac{\mathrm{d}z'}{z'} = \sum_{n=0}^{\infty} \frac{(-1)^n\,(z-a)^{n+1}}{(n+1)\,a^{n+1}} \quad \text{for } |z-a| < |a|\,. \tag{B.192}$$

In order to obtain the principal logarithm, all we have to do now is specify that a has to be chosen from G as defined above for the principal branch. By re-indexing $n \mapsto n+1$ we then find

$$\mathrm{Ln}(z) = \mathrm{Ln}(a) + \sum_{n=1}^{\infty} \frac{(-1)^{n+1}\,(z-a)^n}{n\,a^n} \quad \text{for } |z-a| < |a|\,, \tag{B.193}$$

and after replacing $z \mapsto z+a$ with $a = 1$ and $\mathrm{Ln}(1) = 0$ we finally arrive at

$$\mathrm{Ln}(1+z) = \sum_{n=1}^{\infty} \frac{(-1)^{n+1}\,z^n}{n} \quad \text{for } |z| < 1\,, \tag{B.194}$$

which is known as the complex Mercator series[3], i. e. the Maclaurin series of $\mathrm{Ln}(1+z)$, or equivalently, the Taylor series of $\mathrm{Ln}(z)$ around $z_0 = 1$. This is the first of two series we need to show Eq. (B.186). Substituting again $z \mapsto -z$ yields the second one,

$$-\mathrm{Ln}(1-z) = \sum_{n=1}^{\infty} \frac{z^n}{n} \quad \text{for } |z| < 1\,, \tag{B.195}$$

where $(-1)^{2n+1} = -1$ has been used. The expression resulting from the sum of both series can now be read-off from their expansions,

$$\mathrm{Ln}\left(\frac{1+z}{1-z}\right) = \mathrm{Ln}(1+z) - \mathrm{Ln}(1-z) \tag{B.196}$$

$$= \left(z - \frac{z^2}{2} + \frac{z^3}{3} - \frac{z^4}{4} \pm \cdots\right) + \left(z + \frac{z^2}{2} + \frac{z^3}{3} + \frac{z^4}{4} + \cdots\right) \tag{B.197}$$

$$= 2\left(z + \frac{z^3}{3} + \frac{z^5}{5} + \cdots\right) \tag{B.198}$$

$$= 2\sum_{n=0}^{\infty} \frac{z^{2n+1}}{2n+1}\,, \tag{B.199}$$

[3] Named after the German mathematician Nicholas Mercator (a. k. a. Nikolaus Kauffmann) who discovered this series in 1668. Isaac Newton made the same discovery around the same time but independently from Mercator.

valid for $|z| < \min(1,1) = 1$. Of course, the same result can be obtained analytically by splitting each series for even and odd n, where the even ones exactly cancel each other. Finally, performing another change of variables, $z \mapsto 1/z$, we arrive at the searched-for Laurent series,

$$\operatorname{Ln}\left(\frac{z+1}{z-1}\right) = \operatorname{Ln}\left(\frac{1+1/z}{1-1/z}\right) = \sum_{n=0}^{\infty} \frac{2}{(2n+1)\,z^{2n+1}} \quad \text{for } |z| > 1\,, \tag{B.200}$$

which evaluates to (B.186) and is centered at $z_0 = \infty$.

Note, that the first equality in Eq. (B.196) is in fact not always true as can be seen in the two different approaches for the following case,

$$\operatorname{Ln}(-\mathrm{i}) = \ln(1) - \frac{\pi}{2}\,\mathrm{i} = -\frac{\pi}{2}\,\mathrm{i}\,, \tag{B.201}$$

$$\operatorname{Ln}(-\mathrm{i}) = \operatorname{Ln}(-1) + \operatorname{Ln}(\mathrm{i}) = (\ln(1) + \pi\mathrm{i}) + \left(\ln(1) + \frac{\pi}{2}\,\mathrm{i}\right) = \frac{3\pi}{2}\mathrm{i}\,, \tag{B.202}$$

which obviously differ by 2π. This is again due to the branch cut along the negative real axis. Because of these issues we initially excluded this interval from the set G, such that the principal logarithm is actually not defined there. Therefore, we have to make sure that in Eq. (B.196) and Eq. (B.200) not only the fractions in the arguments of the left-hand side, but also $1 \pm z$ and $1 \pm 1/z$ separately never evaluate to a negative real number. Fortunately, this is already guaranteed by the radius of convergence of the respective series. In particular, $z \pm 1$ for negative real z does not lead to any further problems or restrictions since we never used any series in the respective terms.

In the main text where this series is eventually applied, we additionally bypass this case by explicitly including the regularization factor $\mathrm{i}\eta$ with $\eta > 0$ in the relevant places, which makes z always a complex number with non-zero imaginary part. Arguments are then mostly of the form $z = \pm|a| + \mathrm{i}\eta|b|$, i. e. essentially real with only a small positive imaginary part. Furthermore, we assume that η is closer to zero than any other factor (even in limits). Consequently,

$$\lim_{\eta\to 0^+} \operatorname{Ln}(+|a| + \mathrm{i}\eta|b|) = \ln(|a|)\,, \tag{B.203}$$

$$\lim_{\eta\to 0^+} \operatorname{Ln}(-|a| + \mathrm{i}\eta|b|) = \ln(|a|) + \mathrm{i}\pi\,, \tag{B.204}$$

may be approximately used according to Eq. (B.181).

Exploiting the latter relation we can infer a third important series which is required to find the equivalent of Eq. (B.200) valid for $|z| < 1$. After replacing $z \mapsto z + a$ in Eq. (B.193), we now choose $a = -1 + \mathrm{i}\eta$ with $\eta > 0$ instead of $a = 1$. This yields

$$\operatorname{Ln}(z - 1 + \mathrm{i}\eta) = \operatorname{Ln}(-1 + \mathrm{i}\eta) + \sum_{n=1}^{\infty} \frac{(-1)^{n+1}\,z^n}{n\,(-1+\mathrm{i}\eta)^n} \quad \text{for } |z| < |-1 + \mathrm{i}\eta|\,, \tag{B.205}$$

[3] In fact, the exact region of convergence of all these series needs to be determined carefully. The complex mercator series for instance is not only valid for $|z| < 1$, but also converges on every nibbled disk with $|z| \leq 1$ and $z \neq 1$ as follows from Abel's theorem. However, this is not relevant for our purposes.

which reduces in the limit $\eta \to 0^+$ to

$$\mathrm{Ln}(z-1) = \lim_{\eta \to 0^+} \mathrm{Ln}(z-1+\mathrm{i}\eta) = \mathrm{i}\pi - \sum_{n=1}^{\infty} \frac{z^n}{n} \qquad \text{for } |z| < 1 \wedge \mathrm{Im}(z) \geq 0\,. \tag{B.206}$$

By combining the latter result again with Eq. (B.194) we find

$$\mathrm{Ln}\left(\frac{z+1}{z-1}\right) = \mathrm{Ln}(z+1) - \mathrm{Ln}(z-1) \tag{B.207}$$

$$= \left(z - \frac{z^2}{2} + \frac{z^3}{3} - \frac{z^4}{4} \pm \cdots\right) + \left(-\mathrm{i}\pi + z + \frac{z^2}{2} + \frac{z^3}{3} + \frac{z^4}{4} + \cdots\right) \tag{B.208}$$

$$= -\mathrm{i}\pi + 2\left(z + \frac{z^3}{3} + \frac{z^5}{5} + \cdots\right) \tag{B.209}$$

$$= -\mathrm{i}\pi + 2\sum_{n=0}^{\infty} \frac{z^{2n+1}}{2n+1}\,, \tag{B.210}$$

which is valid for $|z| < 1 \wedge \mathrm{Im}(z) \geq 0$.

Appendix C - Yang-Mills gauge theory

C.1. Field strength tensor

Given the Maxwell Eqs. (1.1) to (1.4), it is clear that the electromagnetic fields themselves could never transform as simply as a Lorentz vector field. This is most obvious in the electrostatic case, where a transition to a relatively moved inertial frame would instantly lead to moving charges and thereby to currents which in turn generate magnetic fields. As discussed in more detail in Sct. 1.4, electric and magnetic fields are thus far from independent from each other and have to be handled by other means.

For that reason, we first introduce the potentials $A^\mu = (\varphi/c, \boldsymbol{A})^\mathsf{T}$ as given in Eqs. (1.5) and (1.6) and define another quantity known as field strength tensor by

$$F^{\mu\nu} \stackrel{\text{def}}{=} A^{\nu,\mu} - A^{\mu,\nu} = 2!\, A^{[\nu,\mu]}\,. \tag{C.1}$$

Since $F^{\mu\nu}$ obviously is a four-tensor of second order, its two indices simply transform separately,

$$F^{\mu'\nu'}(x) = \Lambda^{\mu'}{}_{\mu}\Lambda^{\nu'}{}_{\nu}F^{\mu\nu}(x) \qquad \text{or} \qquad F' = \Lambda F \Lambda^\mathsf{T}\,, \tag{C.2}$$

similar to general n-rank tensors as shown in Eq. (B.62).

In (3+1)-formalism, the field strength tensor is given explicitly in terms of $\boldsymbol{E}$ and $\boldsymbol{B}$ by

$$(F^{\mu\nu}) = \begin{pmatrix} 0 & \boldsymbol{E}^\mathsf{T}/c \\ -\boldsymbol{E}/c & \overleftrightarrow{-B} \end{pmatrix}\,, \qquad (F_{\mu\nu}) = \begin{pmatrix} 0 & -\boldsymbol{E}^\mathsf{T}/c \\ \boldsymbol{E}/c & \overleftrightarrow{-B} \end{pmatrix}\,, \tag{C.3}$$

where the magnetic field matrix,

$$\overleftrightarrow{\boldsymbol{B}} = \epsilon_{ikj}B_k\,(\boldsymbol{e}_i \otimes \boldsymbol{e}_j) = \begin{pmatrix} 0 & -B_3 & B_2 \\ B_3 & 0 & -B_1 \\ -B_2 & B_1 & 0 \end{pmatrix}\,, \tag{C.4}$$

has been defined with a sign convention similar to the transverse rotation operator (Eq. (B.163)) such that $\overleftrightarrow{\boldsymbol{B}}\boldsymbol{x} = \boldsymbol{B} \times \boldsymbol{x}$ and $\boldsymbol{x}^\mathsf{T}\overleftrightarrow{\boldsymbol{B}} = (\boldsymbol{x} \times \boldsymbol{B})^\mathsf{T}$. Equivalently, the relations in Eq. (C.3) may be expressed as

$$F^{0i} = E^i/c\,, \tag{C.5}$$

$$F^{ij} = \epsilon_{ijk}\,B_k\,. \tag{C.6}$$

and similarly for the covariant version, $F_{\mu\nu} = \eta_{\mu\alpha}\eta_{\nu\beta}F^{\alpha\beta}$, where ϵ_{ijk} is the Levi-Civita symbol in three dimensions and it is understood that there is no difference between co-

and contravariant versions of spatial vectors and tensors, i. e. $E^i \equiv E_i$. Conversely, the electromagnetic field components can be extracted again using the relations

$$E_i = cF^{0i}\,, \tag{C.7}$$

$$B_i = \frac{1}{2}\,\epsilon_{ikl}F^{kl}\,. \tag{C.8}$$

which follow directly from Eqs. (C.5) and (C.6) with the identity

$$\epsilon_{ijk}\,\epsilon_{lmn} = \begin{vmatrix} \delta_{il} & \delta_{im} & \delta_{in} \\ \delta_{jl} & \delta_{jm} & \delta_{jn} \\ \delta_{kl} & \delta_{km} & \delta_{kn} \end{vmatrix} \quad \rightarrow \quad \epsilon_{ikl}\,\epsilon_{klm} = 2\,\delta_{im}\,. \tag{C.9}$$

Using these conversion rules, the simple transformation of the field strength tensor in Eq. (C.2) translates after some algebraic manipulations to the far more complicated transformation (cf. e. g. [12, §A.2])

$$\boldsymbol{E}' = \gamma(\boldsymbol{E} + \boldsymbol{v}\times\boldsymbol{B}) - (\gamma-1)\,\frac{\boldsymbol{v}(\boldsymbol{v}\cdot\boldsymbol{E})}{|\boldsymbol{v}|^2}\,, \tag{C.10}$$

$$\boldsymbol{B}' = \gamma\left(\boldsymbol{B} - \frac{\boldsymbol{v}\times\boldsymbol{E}}{c^2}\right) - (\gamma-1)\,\frac{\boldsymbol{v}(\boldsymbol{v}\cdot\boldsymbol{B})}{|\boldsymbol{v}|^2}\,. \tag{C.11}$$

Consquently it is not very reasonable to stick with Maxwell's equations in the Heaviside form but convert them into manifestly (Lorentz-)covariant equations. As shown in Sct. 1.1, this can be achieved by reformulating the wave equation for the potentials in terms of the field strength tensor which eventually leads to Eq. (1.13). The latter can also be written using compact Ricci notation and then reads

$$F^{\mu\nu}{}_{,\nu} = \mu_0 j^\mu\,, \tag{C.12}$$

where $j^\mu = (c\rho, \boldsymbol{j})$ is the four-current. As an equation consisting only of four-tensors, the latter equation is invariant under Lorentz transformation itself and thus manifestly covariant. Applying again the partial derivative to Eq. (C.12) implies charge conservation in (flat) space-time[1],

$$j^\mu{}_{,\mu} = 0\,, \tag{C.13}$$

where the mixed derivatives $F^{\mu\nu}{}_{,\nu\mu}$ vanished according to Eq. (B.29).

Since Eq. (C.12) is equivalent only to the inhomogeneous Maxwell equations (1.1) (1.2), some authors prefer to additionally introduce the Hodge dual of the field strength tensor,

$$\star F^{\mu\nu} = \frac{1}{2}\,\epsilon^{\mu\nu\alpha\beta}\,F_{\alpha\beta}\,, \tag{C.14}$$

where ϵ is the rank-4 Levi-Civita pseudo-scalar with sign convention $\epsilon^{0123} = +1$. In terms of

[1] Similarly, taking the mixed second *covariant* derivative of the *manifest covariant* definition of the field strength tensor implies charge conservation in *curved* space-time, $j^\mu{}_{;\mu} = 0$.

the components of $\boldsymbol{E}$ and $\boldsymbol{B}$, this tensor is given by

$$(\star F^{\mu\nu}) = \begin{pmatrix} 0 & -\boldsymbol{B}^{\mathsf{T}} \\ \boldsymbol{B} & -\overset{\leftrightarrow}{E}/c \end{pmatrix}, \quad (\star F_{\mu\nu}) = \begin{pmatrix} 0 & \boldsymbol{B}^{\mathsf{T}} \\ -\boldsymbol{B} & -\overset{\leftrightarrow}{E}/c \end{pmatrix}, \tag{C.15}$$

and the electric field matrix

$$\overset{\leftrightarrow}{E} = \epsilon_{ikj} E_k \, (\boldsymbol{e}_i \otimes \boldsymbol{e}_j) = \begin{pmatrix} 0 & -E_3 & E_2 \\ E_3 & 0 & -E_1 \\ -E_2 & E_1 & 0 \end{pmatrix}. \tag{C.16}$$

These expressions given, it is easy to verify that the homogeneous Maxwell equations (1.3) and (1.4) can be equivalently expressed as the partial derivatives of this dual,

$$\star F^{\mu\nu}{}_{,\nu} = 0 \,. \tag{C.17}$$

In fact, evaluating the latter equation for fixed μ yields again the geometric Bianchi identity (cf. Eq. (1.20) and Eq. (B.45)),

$$F_{[\alpha\beta,\gamma]} = 0 \,. \tag{C.18}$$

Further, Eq. (C.17) is automatically solved when inserting Eq. (C.1) as the defining equation for the field strength tensor. It is, however, important to keep in mind that $F^{\mu\nu}$ is (in contrast to $\{\boldsymbol{E}, \boldsymbol{B}\}$ and A^μ) no *fundamental* field, because its derivatives, Eqs. (C.12) and (C.17), do not represent new wave equations for the field strength tensor $F^{\mu\nu}$ but instead determine the dynamics of the four-potential A^μ.

For the sake of completeness, we also give the two genuine invariants of F,

$$F^{\mu\nu} F_{\mu\nu} = 2 \left(|\boldsymbol{B}|^2 - \frac{1}{c^2} |\boldsymbol{E}|^2 \right), \tag{C.19}$$

$$\star F^{\mu\nu} F_{\mu\nu} = -\frac{4}{c} \, \boldsymbol{B} \cdot \boldsymbol{E} \,, \tag{C.20}$$

whereby $(\star F^{\mu\nu})(\star F_{\mu\nu}) = -F^{\mu\nu} F_{\mu\nu}$ does not lead to any new Lorentz scalar [81, Eq. (18.69)]. Besides their obvious meaningful content, at least one of these invariants emerge again in context of the relativistic lagrangian formulation of electrodynamics, which starts from the fundamental Yang-Mills Lagrange density (cf. [49, §5.4])

$$\mathcal{L}(A_\alpha, \partial_\beta A_\alpha) = -\frac{1}{4\mu_0} F_{\mu\nu} F^{\mu\nu} + j^\mu A_\mu = -\frac{1}{4\mu_0} F_{\mu\nu} \left(\eta_{\mu\gamma} \, \eta^{\nu\delta} \, F_{\gamma\delta} \right) + j^\mu A_\mu \,. \tag{C.21}$$

Applying the Euler-Lagrange equations to this,

$$\partial_\beta \left(\frac{\partial \mathcal{L}(A_\alpha, \partial_\beta A_\alpha)}{\partial (\partial_\beta A_\alpha)} \right) - \frac{\partial \mathcal{L}(A_\alpha, \partial_\beta A_\beta)}{\partial A_\alpha} = 0 \tag{C.22}$$

with

$$\frac{\partial \mathcal{L}}{\partial (\partial_\beta A_\alpha)} = -\frac{1}{4\mu_0} \, \eta^{\mu\gamma} \, \eta^{\nu\delta} \left(F_{\gamma\delta} \left(\delta^\beta_\mu \, \delta^\alpha_\nu - \delta^\beta_\nu \, \delta^\alpha_\mu \right) + F_{\mu\nu} \left(\delta^\beta_\gamma \, \delta^\alpha_\delta - \delta^\beta_\nu \, \delta^\alpha_\gamma \right) \right) = \frac{1}{\mu_0} \, F^{\alpha\beta} \tag{C.23}$$

recovers the Maxwell equations in form of Eq. (C.12).

C.2. Minimal coupling principle

Minimal coupling in quantum field theory refers to the way how fields couple to other fields, in particular how electromagnetic fields couple to so-called matter fields.

An important object in this respect is the (gauge) covariant derivative defined in terms of the partial derivative ∂_μ and the external vector potential A_μ,

$$\partial_\mu \mapsto D_\mu = \partial_\mu + \frac{\mathrm{i}}{\hbar}\, eA_\mu \,. \tag{C.24}$$

Equivalently, this replacement reads in terms of spatial and temporal components

$$\nabla \mapsto \nabla + \frac{\mathrm{i}}{\hbar}\, e\boldsymbol{A} \,, \tag{C.25}$$

$$\partial_t \mapsto \partial_t - \frac{\mathrm{i}}{\hbar}\, e\varphi \,, \tag{C.26}$$

where φ is the scalar potential and $A_\mu = (-\varphi/c, \boldsymbol{A})^\mathsf{T}$. In contrast to the situation for partial derivatives, covariant derivatives do not commute,

$$[D_\mu, D_\nu] = \frac{\mathrm{i}}{\hbar}\, e\, F_{\mu\nu} \,. \tag{C.27}$$

The gauge transformation in classical electrodynamics, where the symmetry group is U(1), is defined by the unitary phase factor

$$U_f(x) = \exp -\frac{\mathrm{i}}{\hbar}\, ef(x) \,, \tag{C.28}$$

where f is a real-valued scalar function. Gauge transformations always have to be carried out for all involved fields. In this particular case, the joint transformation reads

$$A_\mu(x) \mapsto A'_\mu(x) = U_f(x)\, A_\mu(x) U_f^*(x) + \frac{\hbar}{\mathrm{i}e}\, U_f(x)\, \partial_\mu U_f^*(x) \,, \tag{C.29}$$

$$\psi(x) \mapsto \psi'(x) = U_f(x)\, \psi(x) \,, \tag{C.30}$$

Evaluated for the vector potential, this yields

$$A'^\mu(x) = A^\mu(x) + \partial^\mu f(x) \,. \tag{C.31}$$

The covariant derivative on the other hand transforms via

$$D_\mu \mapsto D'_\mu \equiv U_f(x)\, D_\mu U_f^*(x) = \partial_\mu + \frac{\mathrm{i}}{\hbar}\, A'_\mu \,, \tag{C.32}$$

such that

$$D'_\mu \psi'(x) = U_f(x)\, D_\mu \psi(x) \,. \tag{C.33}$$

In commutative gauge theory, the field strength tensor stays invariant under gauge transformation

$$F'_{\mu\nu}(x) \stackrel{\mathrm{def}}{=} U_f(x)\, F_{\mu\nu}(x) U_f^*(x) \equiv F_{\mu\nu}(x). \tag{C.34}$$

C.3. Gauge invariant quantities and equations

Gauge-invariant current density

We want to prove the gauge invariance of the current density as given by Eq. (3.8), i. e. with only orbital and diamagnetic parts. For convenience, we will work without the explicit spin summation. It may be added to the result afterwards and has no further relevance regarding the derivation.

Inserting the U(1) transformation rules from App. C.2 for the primed wave functions and potential on the right hand side of

$$\boldsymbol{j}'(\boldsymbol{x},t) = \frac{e}{m}\,\boldsymbol{A}'(\boldsymbol{x},t)\,\rho'(\boldsymbol{x},t) + \frac{(-e)\hbar}{2m\mathrm{i}}\left[(\psi')^*(\boldsymbol{x},t)\,(\nabla\psi')(\boldsymbol{x},t) - \psi'(\boldsymbol{x},t)\,(\nabla\psi')^*(\boldsymbol{x},t)\right], \tag{C.35}$$

yields

$$\begin{aligned}\boldsymbol{j}'(\boldsymbol{x},t) = \frac{e}{m}\,&[\boldsymbol{A}(\boldsymbol{x},t)\,\rho(\boldsymbol{x},t) + (\nabla f)(\boldsymbol{x},t)\,\rho(\boldsymbol{x},t)] \\ &+\frac{(-e)\hbar}{2m\mathrm{i}}\Big[\left(\mathrm{e}^{\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi^*(\boldsymbol{x},t)\right)\nabla\left(\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi(\boldsymbol{x},t)\right) \\ &\qquad-\left(\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi(\boldsymbol{x},t)\right)\nabla\left(\mathrm{e}^{\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi^*(\boldsymbol{x},t)\right)\Big],\end{aligned} \tag{C.36}$$

where the charge density transforms trivially,

$$\rho'(\boldsymbol{x},t) = (-e)\,(\psi')^*(\boldsymbol{x},t)\,\psi'(\boldsymbol{x},t) = (-e)\,\psi^*(\boldsymbol{x},t)\,\psi(\boldsymbol{x},t) = \rho(\boldsymbol{x},t)\,. \tag{C.37}$$

Applying Leibniz' rule to the gradients in the last two lines, the current density becomes

$$\begin{aligned}\boldsymbol{j}'(\boldsymbol{x},t) = \frac{e}{m}\,&\boldsymbol{A}(\boldsymbol{x},t)\,\rho(\boldsymbol{x},t) - \frac{(-e)\hbar}{2mi}\left[2\,\frac{\mathrm{i}}{\hbar}\,\rho(\boldsymbol{x},t)\,(\nabla f)(\boldsymbol{x},t))\right] \\ &+\frac{(-e)\hbar}{2m\mathrm{i}}\Bigg[\left(\mathrm{e}^{\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,(\psi^*\psi)(\boldsymbol{x},t)\left(-\frac{\mathrm{i}}{\hbar}\,e\,(\nabla f)(\boldsymbol{x},t)\right)\right) \\ &\qquad-\left(\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\mathrm{e}^{\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,(\psi\psi^*)(\boldsymbol{x},t)\left(\frac{\mathrm{i}}{\hbar}\,e\,(\nabla f)(\boldsymbol{x},t)\right)\right) \\ &\qquad+\left(\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\mathrm{e}^{\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi^*(\boldsymbol{x},t)\,(\nabla\psi)(\boldsymbol{x},t)\right) \\ &\qquad-\left(\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\mathrm{e}^{\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi(\boldsymbol{x},t)\,(\nabla\psi)^*(\boldsymbol{x},t)\right)\Bigg],\end{aligned} \tag{C.38}$$

which eventually reduces to

$$\boldsymbol{j}'(\boldsymbol{x},t) = \frac{(-e)\hbar}{2m\mathrm{i}}\left[\psi^*(\boldsymbol{x},t)\,(\nabla\psi)(\boldsymbol{x},t) - \psi(\boldsymbol{x},t)\,(\nabla\psi)^*(\boldsymbol{x},t)\right] = \boldsymbol{j}(\boldsymbol{x},t)\,. \tag{C.39}$$

Apparently, this is exactly the equal to Eq. (C.35) for the unprimed wave functions and potential and thus equal to the umprimed current density, which completes the proof.

Adding the spinorial part in Eq. (C.35) does not alter this proof qualitatively since the

exponential functions of the transformed wave functions cancel each other,

$$\begin{aligned}
\boldsymbol{j}'_{\text{spin}}(\boldsymbol{x},t) &= \nabla\times\left(\frac{(-e)\hbar}{2m}\sum_{s,s'=\uparrow,\downarrow}(\psi')^*_s(\boldsymbol{x},t)\,\boldsymbol{\sigma}_{ss'}\,\psi'_{s'}(\boldsymbol{x},t)\right)\\
&= \nabla\times\left(\frac{(-e)\hbar}{2m}\sum_{s,s'=\uparrow,\downarrow}\left(\mathrm{e}^{\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi^*_s(\boldsymbol{x},t)\right)\boldsymbol{\sigma}_{ss'}\left(\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\psi_{s'}(\boldsymbol{x},t)\right)\right)\\
&= \nabla\times\left(\frac{(-e)\hbar}{2m}\sum_{s,s'=\uparrow,\downarrow}\psi^*_s(\boldsymbol{x},t)\,\boldsymbol{\sigma}_{ss'}\,\psi_{s'}(\boldsymbol{x},t)\right)\\
&= \boldsymbol{j}_{\text{spin}}(\boldsymbol{x},t)\,,
\end{aligned}\tag{C.40}$$

such that the curl does not apply to any of these terms and the spinorial current density hence transforms trivially. Note, however, that this is not the case for separated orbital and diamagnetic parts. Only combined they stay invarinat under local gauge transformation.

Gauge-invariant Schrödinger equation

We want to prove the gauge invariance of the (free) Schrödinger equation,

$$\mathrm{i}\hbar\,\partial_t\psi(\boldsymbol{x},t) = \frac{|\hat{\boldsymbol{p}}|^2}{2m}\,\psi(\boldsymbol{x},t)\,.\tag{C.41}$$

Therefore, we first replace the partial derivatives with their covariant equivalents, which directly leads to the Schrödinger equation with minimal coupled potentials,

$$\mathrm{i}\hbar\left(\partial_t - \frac{\mathrm{i}}{\hbar}\,e\,\varphi(\boldsymbol{x},t)\right)\psi(\boldsymbol{x},t) = \frac{|\hat{\boldsymbol{p}}+e\boldsymbol{A}(\boldsymbol{x},t)|^2}{2m}\,\psi(\boldsymbol{x},t)\,.\tag{C.42}$$

Setting $\boldsymbol{A}\equiv\boldsymbol{0}$ and $\varphi\equiv 0$, the Eq. (C.41) is retained.

Since this proof is quite cumbersome, we will process the left and right hand side of the latter equation separately in order to prove its gauge invariance. Starting with the easy part, we insert the transformation rules from App. C.2 for the scalar potential and the wave function, apply Leibniz' rule where necessary and factor out the exponential function. This way, we straight-forwardly obtain the unprimed version of the left-hand side multiplied with an exponential factor:

$$\begin{aligned}
&\mathrm{i}\hbar\left(\partial_t - \frac{\mathrm{i}}{\hbar}\,e\,\varphi'(\boldsymbol{x},t)\right)\psi'(\boldsymbol{x},t)\\
&= \mathrm{i}\hbar\left(\partial_t - \frac{\mathrm{i}}{\hbar}\,e\left(\varphi(\boldsymbol{x},t) - (\partial_t f)(\boldsymbol{x},t)\right)\right)\left(\mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\psi(\boldsymbol{x},t)\right)\\
&= \mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\mathrm{i}\hbar\left((\partial_t\psi)(\boldsymbol{x},t) - \frac{\mathrm{i}}{\hbar}\,e\,(\partial_t f)(\boldsymbol{x},t)\,\psi(\boldsymbol{x},t) - \frac{\mathrm{i}}{\hbar}\,e\,\varphi(\boldsymbol{x},t)\psi(\boldsymbol{x},t) + \frac{\mathrm{i}}{\hbar}\,e\,(\partial_t f)(\boldsymbol{x},t)\,\psi(\boldsymbol{x},t)\right)\\
&= \mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,\mathrm{i}\hbar\left(\partial_t - \frac{\mathrm{i}}{\hbar}\,e\,\varphi(\boldsymbol{x},t)\right)\psi(\boldsymbol{x},t)
\end{aligned}\tag{C.43}$$

In other words, the partial time derivative of the wave function transforms just like the wave

function itself,

$$\mathrm{i}\hbar\,(\partial_t\psi)'(\boldsymbol{x},t) = \mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\,(\partial_t\psi)(\boldsymbol{x},t)\,, \tag{C.44}$$

which is especially true for vanishing potentials.

For the right-hand side, we first note that the numerator may be expanded such that the operator-valued terms can be identified more easily,

$$\begin{aligned}|\hat{\boldsymbol{p}} + e(\boldsymbol{A}+\nabla f)|^2 &= |\hat{\boldsymbol{p}}|^2 + \hat{\boldsymbol{p}}\cdot e(\boldsymbol{A}+\nabla f) + e(\boldsymbol{A}+\nabla f)\cdot\hat{\boldsymbol{p}} + e^2\left(|\boldsymbol{A}|^2 + 2\boldsymbol{A}\cdot\nabla f + (\nabla f)^2\right)\\ &= |\hat{\boldsymbol{p}} + e\boldsymbol{A}|^2 + 2e\,(\nabla f)\cdot\hat{\boldsymbol{p}} + \frac{\hbar}{\mathrm{i}}\,e\,(\Delta f) + 2e^2\boldsymbol{A}\cdot(\nabla f) + e^2\,|\nabla f|^2\,,\end{aligned} \tag{C.45}$$

where the identity

$$\hat{\boldsymbol{p}}\cdot(\nabla f) = (\nabla f)\cdot\hat{\boldsymbol{p}} + \frac{\hbar}{\mathrm{i}}\,\Delta f \tag{C.46}$$

has been used. Further, the first term in the second line has been substituted in place of its expansion

$$\begin{aligned}|\hat{\boldsymbol{p}} + e\boldsymbol{A}|^2 &= |\hat{\boldsymbol{p}}|^2 + \hat{\boldsymbol{p}}\cdot e\boldsymbol{A} + e\boldsymbol{A}\cdot\hat{\boldsymbol{p}} + |\boldsymbol{A}|^2\\ &= |\hat{\boldsymbol{p}}|^2 + 2e\boldsymbol{A}\cdot\hat{\boldsymbol{p}} + \frac{\hbar}{\mathrm{i}}\,e\nabla\cdot\boldsymbol{A} + |\boldsymbol{A}|^2\,,\end{aligned} \tag{C.47}$$

where the two operator-valued terms have been written again in front of the multiplicative ones. Looking at the two expansions in Eqs. (C.45) and (C.47), we see that only the action of $\hat{\boldsymbol{p}}$ and $|\hat{\boldsymbol{p}}|^2$ onto the transformed wave functions are left to be determined.

For the momentum operator we find

$$\begin{aligned}\hat{\boldsymbol{p}}\left(\mathrm{e}^{-\mathrm{i}ef/\hbar}\,\psi\right) &= \mathrm{e}^{-\mathrm{i}ef/\hbar}\,\hat{\boldsymbol{p}}\psi + \mathrm{e}^{-\mathrm{i}ef/\hbar}\,(-e)\,(\nabla f)\,\psi\\ &= \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(\hat{\boldsymbol{p}} - e\,(\nabla f)\right)\psi\,,\end{aligned} \tag{C.48}$$

and applying $\hat{\boldsymbol{p}}$ a second time to both sides of this result yields

$$\begin{aligned}|\hat{\boldsymbol{p}}|^2\left(\mathrm{e}^{-\mathrm{i}ef/\hbar}\,\psi\right) &= \hat{\boldsymbol{p}}\cdot\left(\mathrm{e}^{-\mathrm{i}ef/\hbar}\left(\hat{\boldsymbol{p}}\psi - e\,(\nabla f)\,\psi\right)\right)\\ &= \mathrm{e}^{-\mathrm{i}ef/\hbar}\,(-e)\,(\nabla f)\cdot\left(\hat{\boldsymbol{p}}\psi - e\,(\nabla f)\,\psi\right) + \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(|\hat{\boldsymbol{p}}|^2\psi - e\hat{\boldsymbol{p}}\cdot(\nabla f)\,\psi\right)\\ &= \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(-2e\,(\nabla f)\cdot\hat{\boldsymbol{p}} + e^2\,|\nabla f|^2 + |\hat{\boldsymbol{p}}|^2 - \frac{\hbar}{\mathrm{i}}\,e\,(\Delta f)\right)\psi\,,\end{aligned} \tag{C.49}$$

where in the last line an identity similar to Eq. (C.46),

$$\hat{\boldsymbol{p}}\cdot\boldsymbol{A} = \boldsymbol{A}\cdot\hat{\boldsymbol{p}} + \frac{\hbar}{\mathrm{i}}\,\nabla\cdot\boldsymbol{A}\,. \tag{C.50}$$

has been used.

The action of the untransformed operator from Eq. (C.47) can now be easily determined by simply inserting the latter results. In the resulting expression, the the exponential factor can again be factored out such that the remaining operators now act on the unprimed wave

function,

$$
\begin{aligned}
|\hat{\boldsymbol{p}} + e\boldsymbol{A}|^2 \left(\mathrm{e}^{-\mathrm{i}ef/\hbar}\psi\right) &= \left(|\hat{\boldsymbol{p}}|^2 + 2e\boldsymbol{A}\cdot\hat{\boldsymbol{p}} + \frac{\hbar}{\mathrm{i}}\, e\nabla\cdot\boldsymbol{A} + |\boldsymbol{A}|^2\right)\left(\mathrm{e}^{-\mathrm{i}ef/\hbar}\psi\right) \\
&= \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(-2e\,(\nabla f)\cdot\hat{\boldsymbol{p}} + e^2\,|\nabla f|^2 + |\hat{\boldsymbol{p}}|^2 - \frac{\hbar}{\mathrm{i}}\, e\,(\Delta f)\right)\psi \\
&\quad + \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(2e\boldsymbol{A}\cdot\hat{\boldsymbol{p}} - 2e^2\boldsymbol{A}\cdot(\nabla f)\right)\psi \\
&\quad + \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(\frac{\hbar}{\mathrm{i}}\, e\nabla\cdot\boldsymbol{A} + |\boldsymbol{A}|^2\right)\psi \\
&= \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(-2e\,(\nabla f)\cdot\hat{\boldsymbol{p}} + e^2\,|\nabla f|^2 - \frac{\hbar}{\mathrm{i}}\, e\,(\Delta f) - 2e^2\boldsymbol{A}\cdot(\nabla f)\right)\psi \\
&\quad + \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(|\hat{\boldsymbol{p}} + e\boldsymbol{A}|^2\right)\psi\,. \qquad \text{(C.51)}
\end{aligned}
$$

This result in turn, can now be compared to the expansion in Eq. (C.45). Apparently, all multiplicative factors will just cancel each other, whereas for the operator-valued ones one has to consider again their action on the transformed wave function. Eventually, this leads to the following expression

$$
\begin{aligned}
|\hat{\boldsymbol{p}} + e(\boldsymbol{A} + \nabla f)|^2 \left(\mathrm{e}^{-\mathrm{i}ef/\hbar}\psi\right) &= \mathrm{e}^{-\mathrm{i}ef/\hbar}\left(|\hat{\boldsymbol{p}} + e\boldsymbol{A}|^2 - 2e\,(\nabla f)\cdot\hat{\boldsymbol{p}} + 2e^2|\nabla f|^2\right)\psi \\
&\quad + 2e\,(\nabla f)\cdot\left(\mathrm{e}^{-\mathrm{i}ef/\hbar}\left(\hat{\boldsymbol{p}} - e\,(\nabla f)\right)\psi\right) \\
&= \mathrm{e}^{-\mathrm{i}ef/\hbar}|\hat{\boldsymbol{p}} + e\boldsymbol{A}|^2\,\psi \qquad \text{(C.52)}
\end{aligned}
$$

from where it is obvious that the covariant momentum operator (a. k. a. kinetic momentum as opposed to canonical momentum) applied to the wave function transforms again like the wave function itself,

$$
\left(\frac{|\hat{\boldsymbol{p}} + e\boldsymbol{A}(\boldsymbol{x},t)|^2}{2m}\,\psi(\boldsymbol{x},t)\right)' = \mathrm{e}^{-\mathrm{i}ef(\boldsymbol{x},t)/\hbar}\left(\frac{|\hat{\boldsymbol{p}} + e\boldsymbol{A}(\boldsymbol{x},t)|^2}{2m}\,\psi(\boldsymbol{x},t)\right)\,. \qquad \text{(C.53)}
$$

Combined with the transformation property of the left-hand side we see that the transformed Schrödinger is equal to the unprimed version after multiplying through with the exponential factor that occurs in the transformation rule of the wave function.

Gauve invariant continuity equation

We want to prove the gauge invariance of the continuity equation. First, we show that the continuity equation can only be fulfilled using the full current density as given by Eq. (3.1) in combination with the minimal coupling Hamiltonian (3.3). For that reason, consider the divergence term of the continuity equation, which in case of the full current density reads

$$
\nabla\cdot\boldsymbol{j}(\boldsymbol{x},t) = \nabla\cdot\left(\boldsymbol{j}_{\mathrm{orb}}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{dia}}(\boldsymbol{x},t) + \boldsymbol{j}_{\mathrm{spin}}(\boldsymbol{x},t)\right)\,. \qquad \text{(C.54)}
$$

For the first contribution we obtain the familiar expression,

$$\nabla \cdot \boldsymbol{j}_{\mathrm{orb}}(\boldsymbol{x},t) = \frac{(-e)\hbar}{2m\mathrm{i}} \left(\psi^*(\boldsymbol{x},t)\,(\Delta\,\psi)(\boldsymbol{x},t) - \psi(\boldsymbol{x},t)\,(\Delta\,\psi)^*(\boldsymbol{x},t) \right). \tag{C.55}$$

In the diamagnetic term, nabla applies according to Leibniz' rule once to the vector potential as divergence and once to the charge density as gradient,

$$\nabla \cdot \boldsymbol{j}_{\mathrm{dia}}(\boldsymbol{x},t) = \frac{e}{m} \left(\nabla \cdot \boldsymbol{A}(\boldsymbol{x},t) + \boldsymbol{A}(\boldsymbol{x},t) \cdot (\nabla\rho)(\boldsymbol{x},t) \right). \tag{C.56}$$

Since the spinorial third part is a pure curl field (see e. g. Eq. (C.40)), its divergence simply vanishes identically (cf. App. B),

$$\nabla \cdot \boldsymbol{j}_{\mathrm{spin}}(\boldsymbol{x},t) = 0\,. \tag{C.57}$$

Similar to the standard derivation, the charge density is first expressed in terms of the wave function on which the time derivative then acts according to Leibniz' rule. These derivatives are then replaced using the Schrödinger equation,

$$(-e)\,\partial_t\psi = \frac{(-e)}{\mathrm{i}\hbar}\,\hat{H}\psi = \left(-\frac{(-e)\hbar}{2m\mathrm{i}}\,\Delta + \frac{e^2}{2m}\left(\nabla\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\nabla\right) - \frac{e^3}{2m\mathrm{i}\hbar}\,|\boldsymbol{A}|^2 + \frac{e^2}{\mathrm{i}\hbar}\,\varphi \right)\psi\,. \tag{C.58}$$

Inserting this into the expansion of the charge density yields

$$\begin{aligned}
\partial_t\rho &= (-e)\,\psi^*\,(\partial_t\psi) + (-e)\,\psi\,(\partial_t\psi^*) \\
&= \psi^* \left(-\frac{(-e)\hbar}{2m\mathrm{i}}\,\Delta + \frac{e^2}{2m}\left(\nabla\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\nabla\right) + \frac{e^3}{2m\mathrm{i}\hbar}\,|\boldsymbol{A}|^2 + \frac{e^2}{\mathrm{i}\hbar}\,\varphi \right)\psi \\
&\quad + \psi \left(\frac{(-e)\hbar}{2m\mathrm{i}}\,\Delta + \frac{e^2}{2m}\left(\nabla\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\nabla\right) - \frac{e^3}{2m\mathrm{i}\hbar}\,|\boldsymbol{A}|^2 - \frac{e^2}{\mathrm{i}\hbar}\,\varphi \right)\psi^* \\
&= -\nabla\cdot\boldsymbol{j}_{\mathrm{orb}} + \frac{e^2}{2m}\Big(\psi^* \left(\nabla\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\nabla\right)\psi + \psi\left(\nabla\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\nabla\right)\psi^* \Big) \\
&= -\nabla\cdot\boldsymbol{j}_{\mathrm{orb}} + \frac{e^2}{2m}\Big(\psi^*\psi\,(\nabla\cdot\boldsymbol{A}) + 2\boldsymbol{A}\cdot(\psi^*\nabla\psi) + \psi\psi^*(\nabla\cdot\boldsymbol{A}) + 2\boldsymbol{A}\,(\psi\nabla\psi^*) \Big) \\
&= -\nabla\cdot\boldsymbol{j}_{\mathrm{orb}} - \frac{e}{m}\left(\rho(\nabla\cdot\boldsymbol{A}) + \boldsymbol{A}\cdot(\nabla\rho) \right) \\
&= -\nabla\cdot\left(\boldsymbol{j}_{\mathrm{orb}} + \boldsymbol{j}_{\mathrm{dia}} \right),
\end{aligned} \tag{C.59}$$

which just cancels the divergence term Eq. (C.54).

The invariance under local gauge transformation of the continuity equation can now easily be justified with the invariance of the current itself as shown in the beginning of this section, combined with the trivial transformation of the charge density, such that

$$\partial_t\rho'(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{j}'(\boldsymbol{x},t) = \partial_t\rho(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{j}(\boldsymbol{x},t) = 0\,. \tag{C.60}$$

At this point it is important to note, that the partial derivatives in the continuity equation do not have to be replaced by (gauge) covariant derivatives, at least not for U(1) transformations. Heuristically, a "real" divergence and time derivative is required in order to derive (global)

charge conservation via Gauß' divergence theorem. In fact, according to Noether's theorem (local) charge conservation is actually directly related to the invariance under (global) U(1) symmetry transformations [49, §10]. Analytically, this circumstance can be attributed to the fact that different objects like scalars, vectors and tensors transform differently. With respect to U(1) gauge transformation, the vector behaves like a scalar while it transforms like a vector within the Lorentz symmetry group. Within the noncommutative SU(2) and SU(3) gauge theories, the vector potential becomes a tensor itself therefore changes its transformation behavior (w. r. t. gauge transformations). There, the partial derivatives are then indeed replaced by their covariant equivalents, but the current is then not given anymore by Eq. (3.1). Instead, it has to be replaced by the Yang-Mills current (see e. g. [82, §11.2f]).

Appendix D - Periodic solids

D.1. Partitioning of reciprocal space

Real space

Crystalline solids can be modelled as an infinite lattice of spatial points with an orientation and structure that stays identical independently of the chosen reference lattice point (e. g. the origin), or in other words, is invariant under translation by integral multiples of a lattice vector. Such structures are commonly known as Bravais lattice. Mathematically, the position of a lattice point is given by

$$\boldsymbol{R_n} = n_1\boldsymbol{a}_1 + n_2\boldsymbol{a}_2 + n_3\boldsymbol{a}_3\,, \tag{D.1}$$

where $\boldsymbol{a}_i$ denotes the three so called primitive lattice vectors (as opposed to conventional lattice vectors). The ideal and infinitely large crystal lattice can then be regarded as the set of all such lattice points,

$$\Gamma = \left\{\boldsymbol{R_n}\middle|\, \boldsymbol{n} = (n_1, n_2, n_3) \in \mathbb{Z}^3\right\}\,. \tag{D.2}$$

Further, the condition $V_{\text{cell}} = \boldsymbol{a}_1 \cdot (\boldsymbol{a}_2 \times \boldsymbol{a}_3) \neq 0$ is required, i. e. the $\boldsymbol{a}_i$ must not be linearly dependent. The magnitudes of these three *primitive* lattice vectors and the three angles they enclose pairwise are called lattice constants[1]. In this context, primitive means that a single cell contains exactly one lattice point (that must not necessarily lie in its center). By contrast, the conventional cell is usually larger and contains more than one lattice point, but also illustrates symmetries more obvious. Since conventional cells do not have any further conceptional meaning for this thesis, we restrict to primitive cells in the following. More precisely, we only regard the Wigner-Seitz cell, which is uniquely defined by the condition that the enclosed lattice point must lie exactly in its center. In other words, the Wigner-Seitz cell is the region in real space, which is closer to a specific lattice point than to any other point of the Bravais lattice.

Reciprocal space

In perfect crystals, the electronic charge density $\rho \equiv \rho_{\text{e}}$ should comprise the same periodicity as the underlying lattice, i. e. it should be invariant under translations by lattice vectors

[1] For cubic lattices the lattice constants $|\boldsymbol{a}_i|$ are not to be confused with *the* lattice constant a, which is usually tabulated for materials and identical to the edge length of the *conventional* lattice. Instead, we have $\left|\boldsymbol{a}_i^{\text{fcc}}\right| = \sqrt{2}/2\,a$ and $\left|\boldsymbol{a}_i^{\text{bcc}}\right| = \sqrt{3}/2\,a$.

$\rho(\boldsymbol{x}+\boldsymbol{R_n}) = \rho(\boldsymbol{x})\,,\ \forall \boldsymbol{R_n} \in \Gamma$. Since every periodic function can be expanded in a Fourier series, we may postulate w. l. o. g.

$$\rho(\boldsymbol{x}) = \sum_{\boldsymbol{G}} \rho_{\boldsymbol{G}} \mathrm{e}^{\mathrm{i}\boldsymbol{G}\cdot\boldsymbol{x}}\,, \tag{D.3}$$

with an infinite number of (in general complex) expansion coefficients $\rho_{\boldsymbol{G}}$. The associated index-vectors $\boldsymbol{G}$ themselves have to fulfill the condition

$$\mathrm{e}^{\mathrm{i}\boldsymbol{G_m}\cdot\boldsymbol{R_n}} = 1\,, \tag{D.4}$$

in order to respect the lattice periodicity (indices n and m left out for better readability),

$$\rho(\boldsymbol{x}+\boldsymbol{R}) = \sum_{G} \rho_{\boldsymbol{G}} \mathrm{e}^{\mathrm{i}\boldsymbol{G}\cdot\boldsymbol{x}} \mathrm{e}^{\mathrm{i}\boldsymbol{G}\cdot\boldsymbol{R}} \overset{!}{=} \rho(\boldsymbol{x})\,. \tag{D.5}$$

Obviously, this can only be true if $\boldsymbol{G_m}\cdot\boldsymbol{R_n} = 2\pi l$, $l \in \mathbb{Z}$. Taking the definitions for real lattice points, Eq. (D.1), and postulating w. l. o. g. that every $\boldsymbol{G}$ may be constructed in a similar way, i. e.

$$\boldsymbol{G_m} = m_1\boldsymbol{b}_1 + m_2\boldsymbol{b}_2 + m_3\boldsymbol{b}_3\,, \tag{D.6}$$

then one possible choice for the primitive reciprocal lattice vectors $\boldsymbol{b}_i$ is (using Einstein notation!)

$$b_i = \pi\epsilon_{ijk}\,\frac{\boldsymbol{a}_j\times\boldsymbol{a}_k}{|\boldsymbol{a}_1\cdot(\boldsymbol{a}_2\times\boldsymbol{a}_3)|} = \pi\epsilon_{ijk}\,\frac{\boldsymbol{a}_j\times\boldsymbol{a}_k}{V_c}\,. \tag{D.7}$$

Both, real and reciprocal primitive lattice vectors then fulfill the Laue conditions

$$\boldsymbol{a}_i\cdot\boldsymbol{b}_j = 2\pi\delta_{ij}\,, \tag{D.8}$$

and consequently, $m_i \in \mathbb{Z}$. If the function's periodicity is just the lattice periodicity of the crystal, then the grid formed by $\boldsymbol{G}$-vectors is called reciprocal or *Fourier lattice*

$$\Gamma^{-1} = \left\{ \boldsymbol{G_m} \,\middle|\, \boldsymbol{m} = (m_1, m_2, m_3) \in \mathbb{Z}^3 \ \wedge\ \mathrm{e}^{\mathrm{i}\boldsymbol{G_m}\cdot\boldsymbol{R_n}} = 1\ \forall \boldsymbol{R_n} \in \Gamma \right\}. \tag{D.9}$$

Since there is no restriction for the number of $\boldsymbol{G}$-vectors, the reciprocal lattice is a (infinitely large) Bravais lattice as well.

Dual lattice and thermodynamic limit

The lattice periodicity discussed before is only one of two important periodicities used in theoretical solid state physics. While the former is an inherent characteristic of idealized crystalline materials, the Born-von Kármán periodic boundary conditions on the other hand reflect the fact that each real sample is finite. Because every periodicity in real space leads to a discretization in reciprocal space, we initially found the (infinitely large) Fourier lattice as a result of the real-space lattice periodicity. Likewise, the Born-von Kármán boundary conditions will discretize the Fourier domain even further as we will see shortly.

First, we regard the N-electron wave function Ψ^N. Its name already indicates that in

the corresponding Schrödinger equation, the number of particles N should remain finite[2] (in fact, even constant). On the other hand, we required lattice periodicity for the electronic charge density on the infinitely large Bravais lattice in Eq. (D.5). The solution to this alleged contradiction is stipulating periodic boundary conditions for the electronic wave function,

$$\Psi^N(\ldots, \boldsymbol{x} + N_i \boldsymbol{a}_i, \ldots) \stackrel{!}{=} \Psi^N(\ldots, \boldsymbol{x}, \ldots)\,, \quad i = 1,2,3\,, \tag{D.10}$$

where N_i is the number of unit cells in each direction of the crystal sample. The infinitely large real-space lattice from Eq. (D.2) is then replaced by a spatially limited one known as direct lattice,

$$\Gamma = \{\boldsymbol{R_n} | \, \boldsymbol{n} = (n_1, n_2, n_3) \ \wedge \ n_i = 0 \ldots N_i - 1 \ (i = 1,2,3)\}\ . \tag{D.11}$$

which becomes a Bravais lattice again in the so called thermodynamic limit, $N_i \to \infty$ for $i = 1,2,3$.[3]

Following the same procedure as given by Eqs. (D.4) to (D.6), we find the Born-von Kármán vectors

$$\boldsymbol{k_m} = \frac{m_1}{N_1}\,\boldsymbol{b}_1 + \frac{m_2}{N_2}\,\boldsymbol{b}_2 + \frac{m_3}{N_3}\,\boldsymbol{b}_3\,, \tag{D.12}$$

discretizing the reciprocal space on an even finer grid. Obviously, the Fourier lattice is a subset of the latter, namely for integral expansion coefficients $m_i = nN_i\,,\ n \in \mathbb{Z}$. In order to eliminate this redundancy, the dual lattice $\Gamma^* \subset \{\boldsymbol{k_m}\}$ is introduced as[4]

$$\Gamma^* = \left\{\boldsymbol{q_m} | \, \boldsymbol{m} = (m_1, m_2, m_3) \ \wedge \ m_i = 0 \ldots N_i - 1 \ \wedge \ \mathrm{e}^{\mathrm{i}N_i \boldsymbol{q_m}\cdot\boldsymbol{a_i}} = 1 \ (i = 1,2,3)\right\}\ , \tag{D.13}$$

with the same N_i used in the definition of the direct lattice. Consequently, there are exactly as many points in dual space as points that form the direct lattice, namely $N = N_1 N_2 N_3$. The limitation for m_i in Eq. (D.13) in fact picks only those vectors, that are unique up to a translation by a Fourier lattice vector $\boldsymbol{G}$. In the thermodynamic limit $N_i \to \infty$, where the direct lattice becomes an infinite Bravais lattice, the dual lattice instead becomes continuous and is known as the first Brillouin zone,

$$\mathcal{B} = \{\boldsymbol{q} \,|\, \boldsymbol{q} = v_1 \boldsymbol{b}_1 + v_2 \boldsymbol{b}_2 + v_3 \boldsymbol{b}_3,\ 0 \le v_i < 1 \ (i = 1,2,3)\}\ . \tag{D.14}$$

Thus, the thermodynamic limit in real space corresponds to a *continuum limit* in Fourier space. From the definition of dual and reciprocal lattice vectors it is obvious that an arbitrary point in Fourier space can now be constructed by taking any reciprocal vector and adding a dual vector,

$$\boldsymbol{k} = \boldsymbol{G} + \boldsymbol{q}\,. \tag{D.15}$$

[2] In fact, this is also quite important for the numerical treatment, because realizing infinitely large arrays in finite memory has proven to be quite hard.

[3] The distinction between direct and real-space Bravais lattice is subtle and usually not discussed in textbooks. Therefore, and because there is no common symbol for the former, we will mark both with Γ.

[4] Note, that in order to prevent confusion between arbitrary (allowed) wavevectors $\boldsymbol{k_m}$ and dual lattice points $\boldsymbol{q_m}$, we will use different symbols for both throughout this thesis. Unfortunately, common literature and text books are quite inconsequent regarding this differentiation.

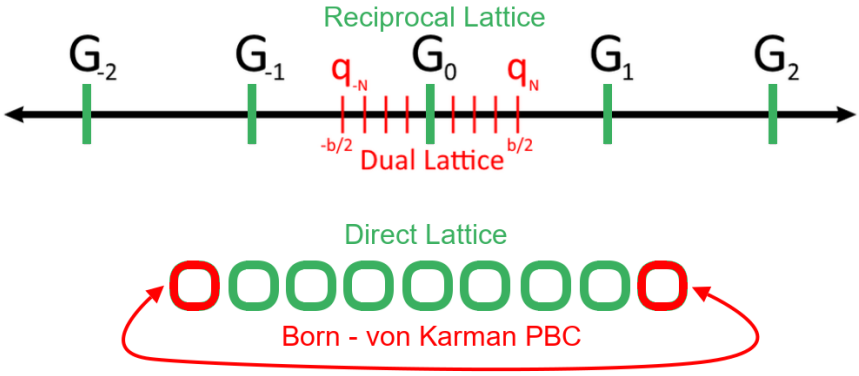


Fig. D.1.: Schematic view on the partition of a one-dimensional crystal in direct and reciprocal space. Red lines symbolize $\boldsymbol{q}$-points that are supported only in the first Brillouin zone and form the dual lattice. Their number resemble the number of joint unit cells in real space and they originate from the postulation of Born-von Kármán periodic boundary conditions. In the thermodynamic limit, the set becomes continuous. By contrast, $\boldsymbol{G}$-vectors, indicated by blue green ticks, form the reciprocal or Fourier lattice. In principle, there are infinitely many of them and they are virtually always discrete, but for computational reasons their number is capped at a specific $|\boldsymbol{G}_{\max}|$ in numerical applications. From these two basic types of vectors a general point in reciprocal space can be composed as $\boldsymbol{k} = \boldsymbol{q} + \boldsymbol{G}$.

By suitably adding reciprocal lattice vectors $\boldsymbol{G_m}$, the dual lattice can also be chosen to be more symmetric w.r.t. the origin. This is what is usually illustrated in textbooks and also shown in fig. D.1.

This partitioning of reciprocal space has a number of notable side-effects. For instance, the dispersion relation can now be visualized in the so called "reduced zone scheme", where $\omega(\boldsymbol{k}) = \omega(\boldsymbol{q} + \boldsymbol{G}) \to \omega_n(\boldsymbol{q})$ is plotted as n functions of $\boldsymbol{q}$ along a specific path in the first Brillouin zone. Then for a particular $\boldsymbol{q}$, bands corresponding to vectors $\boldsymbol{k}_n = \boldsymbol{q} + \boldsymbol{G}_n$ from the n-th Brillouin zone will be moved to first Brillouin zone, such that different $\boldsymbol{G}$ emerge as band index n. Further and more important for this thesis, response functions $\chi(\boldsymbol{k}, \boldsymbol{k}', \omega)$ simplify in a way that they do not depend on two general vectors in Fourier space anymore but merely require a vector from first Brillouin zone and two reciprocal lattice vectors. The latter two are usually set to zero such that the response function becomes $\chi_{\boldsymbol{G}=\boldsymbol{0},\boldsymbol{G}'=\boldsymbol{0}}(\boldsymbol{q})$ as discussed in App. D.2. For strictly homogeneous systems like the free electron gas where no periodic potential is present, there is also no Fourier lattice and consequently the entire Fourier space may be regarded as first Brillouin zone. In such a case, the simplified argumentation from App. B.3 applies to response functions.

A very thorough discussion of all different limites, their combination and their interpretation can be found in App. A and §2.2.1 of Ref. [8].

D.2. Homogeneous limit

Microscopic response function in real space are in general of the form Eq. (B.91), i. e. they constitute tensorial integral kernels as opposed to sheer scalar numbers as suggested by the commonly adduced "material relations", Eqs. (2.11) and (2.12). In particular, they are in general non-local and inhomogeneous in real space as well as in Fourier space, as becomes obvious from the transformation rules Eqs. (B.93) and (B.94): In order to find the induced current at a single point in space-time, the potential A^μ has to be known on the entire support of the integral kernel $\chi^\mu{}_\nu(\,\cdot\,, x')$. On a fundamental level, two essentially different physical limitations contribute to this mathematical circumstance. As a matter of fact, real samples do not occupy the entire space but possess a (in general highly non-trivial) geometry. Therefore, it is impossible for a sample to be spatially homogeneous already for that reason. However, theoretical materials science is usually concerned with the description of bulk properties instead of geometry-dependent effects[5] and therefore, all physical properties have to be calculated in the thermodynamic limit as a matter of principle. Conceptually, this corresponds then again to a material filling all of space homogeneously (see App. D.1 for details). On the other hand, real materials are naturally inhomogeneous on the microscopic level due to their atomic structure. This is reflected in the non-locality of their response functions. For ideal crystals, however, the situation simplifies quite drastically provided one accepts certain premises. Since we always assume homogeneity in time, the following discussion restricts to the spatial part and thus, time or frequency arguments are left out.

Within the bulk, crystalline solids stay invariant under translations by direct lattice vectors, i. e.

$$\chi(\boldsymbol{x}, \boldsymbol{x}') = \chi(\boldsymbol{x} + \boldsymbol{a}, \boldsymbol{x}' + \boldsymbol{a}) \tag{D.16}$$

in real space or (by Eq. (B.93) and Eq. (B.124))

$$\chi(\boldsymbol{k}, \boldsymbol{k}') = \mathrm{e}^{\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{a}}\chi(\boldsymbol{k}, \boldsymbol{k}')\,, \tag{D.17}$$

equivalently in the Fourier domain. From the last equation it follows immediately, that the two wavevectors can only differ by an arbitrary reciprocal lattice vector. As a consequence, the two initial wavevectors can be decomposed into a common vector $\boldsymbol{q}$ that lies within the first Brillouin zone and two reciprocal lattice vectors $\boldsymbol{G}$ and $\boldsymbol{G}'$ (see Eq. (D.15) and Fig. D.1),

$$\boldsymbol{k} = \boldsymbol{q} + \boldsymbol{G}\,, \tag{D.18}$$

$$\boldsymbol{k}' = \boldsymbol{q} + \boldsymbol{G}'\,, \tag{D.19}$$

such that the response functions now effectively depend on three vectors,

$$\chi(\boldsymbol{q} + \boldsymbol{G}, \boldsymbol{q}' + \boldsymbol{G}') = \chi_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{q})\,\delta^3(\boldsymbol{q} - \boldsymbol{q}')\,. \tag{D.20}$$

[5] There is a quite busy field of research on surface effects nonetheless, although not relevant for this thesis.

For a general response law like Eq. (2.37) or Eq. (B.91) this translates to

$$\rho_{\mathrm{ind}}(\boldsymbol{q}+\boldsymbol{G}) = \sum_{\boldsymbol{G}'} \chi_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{q})\,\varphi_{\mathrm{ext}}(\boldsymbol{q}+\boldsymbol{G}')\,. \tag{D.21}$$

Now assuming that the Fourier transform of the fields ρ and φ are supported in the first Brillouin Zone anyway, this response law significantly reduces to the simple product

$$\rho_{\mathrm{ind}}(\boldsymbol{k}) = \chi_{\boldsymbol{0}\boldsymbol{0}}(\boldsymbol{k})\,\varphi_{\mathrm{ext}}(\boldsymbol{k})\,, \tag{D.22}$$

which recovers the result from Eq. (B.107) provided one identifies $\chi \equiv \chi_{\boldsymbol{0}\boldsymbol{0}}$. As discussed in App. B.3, the response function then appears indeed strictly homogeneous in real space, i. e.

$$\chi(\boldsymbol{x},\boldsymbol{x}';t,t') = \chi(\boldsymbol{x}-\boldsymbol{x}',t-t') \quad \overset{\mathcal{F}}{\to} \quad \chi(\boldsymbol{k},\omega)\,. \tag{D.23}$$

Correspondingly, we call the transition $\boldsymbol{G},\boldsymbol{G}' \to \boldsymbol{0}$ for response functions *homogeneous limit*.

The justification for this limit on physical grounds coincidently reveals an interesting interpretation of the corresponding experimental situation as well. Since wavevectors are restricted to the first Brillouin zone, their moduli have to fulfill the approximate inequality $|\boldsymbol{k}| \lessapprox \pi/a$, where a is a typical primitive lattice constant. By $|\boldsymbol{k}| = 2\pi/\lambda$, the associated wavelengths have to be larger than at least twice this lattice constant, $2a \lessapprox \lambda$. This is of course a version of the famous Nyquist-Shannon sampling theorem, where the external field corresponds to the signal and the crystal plays the rôle of the sampler [1, p. 3]. Lattice constants of simple crystals usually lie in the range of a few angstrom, whereas wavelengths used in different spectroscopical applications are typically in the range of nanometers and thereby much larger in comparison. Therefore, it is highly intuitive that at such wavelengths the material – even if crystalline on the microscopic level – appears to be homogeneous on the macroscopic level. Only for experiments involving soft X-rays with wavelengths around 1 Å, or equivalently, energies in the range of keV, this is not the case anymore. Thus, it is not surprising that exactly this part of the electromagnetic spectrum is used in diffraction experiments to determine the structure of single-crystals (XRC) or crystallite powders (XRPD). In the phenomenological explanation of such experiments, this condition corresponds to Bragg's law,

$$n\lambda = 2d\sin\theta\,, \tag{D.24}$$

which cannot be fulfilled if $\lambda > 2d$ for typical interplanar distances d. [47, §2.3.1]

Consequently, we can simply assume that the external fields considered in this thesis do not lie in the X-ray range, which certainly applies for that kind of spectroscopical experiments relevant to linear response theory. Thus, we will stick to the notation $\varphi(\boldsymbol{k})$ instead of the more exact $\varphi(\boldsymbol{q})$ and implicitly restrict these vectors to the first Brillouin zone. Moreover, this simultaneously saves us from otherwise unspecified or complicated averaging procedures as required in the traditional approach (see Sct. 2.1). Instead, it is the response function that is subject to a "macroscopical transition", which is simply implemented by the evaluation at small wavevectors in the sense of the homogeneous limit, $\boldsymbol{G},\boldsymbol{G}' \to \boldsymbol{0}$. Hence, a general

response function behaves in our case exactly as described in App. B.3 and in particular Eq. (B.104).

Appendix E - Electromagnetic spectrum

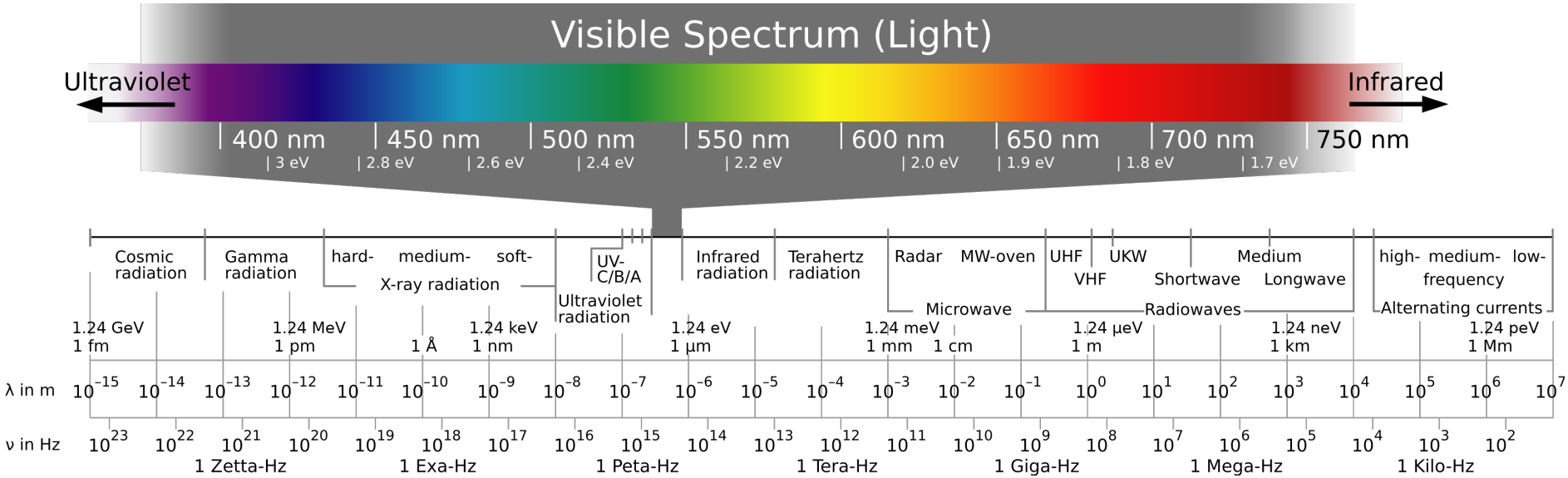


Fig. E.1.: Overview of the electromagnetic spectrum with indicated wavelength, frequency and energy ranges.

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Hiermit versichere ich, dass ich die vorliegende Arbeit ohne unzulässige Hilfe Dritter und ohne Benutzung anderer als der angegebenen Hilfsmittel angefertigt habe; die aus fremden Quellen direkt oder indirekt übernommenen Gedanken sind als solche kenntlich gemacht.

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Declaration

I hereby declare that I completed this work without any improper help from a third party and without using any aids other than those cited. All ideas derived directly or indirectly from other sources are identified as such.

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