

Light-matter interaction

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TODO:

- There are rare cases where the energy velocity (i.e. the velocity that connects \mathbf{s} and u) is not the same as group velocity; for example in metal wave guide?
- From the current response of an external field to ϵ_r . (That's to say, from TD-aGW to dielectric function.)
- Specifically, noise exists when there is damping. Could this be modeled by some sort of non-equilibrium field theory? (I think I asked this question before ...)

1 Coarse-grained description of medium

A linearly polarized medium can be described by

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P} = \epsilon_0 \mathbf{E} + \epsilon_0 \chi_e \mathbf{E} = \epsilon_0 \underbrace{(1 + \chi_e)}_{=:\epsilon_r} \mathbf{E}. \quad (1)$$

Similarly we can deal with magnetization, although here the notation, due to historical reasons, needs to be altered slightly. Now we have

$$\mathbf{B} = \mu_0 (\mathbf{H} + \mathbf{M}) = \mu_0 \underbrace{(1 + \chi_m)}_{\mu_r} \mathbf{H}, \quad (2)$$

as if \mathbf{B} has the same status of \mathbf{D} , although it's \mathbf{H} that is the auxiliary field.

From the fourth Maxwell equation we directly find that time oscillation of polarization stimulates a magnetic field, which is correct since $\partial_t \mathbf{P}$ can be understood as a current: charges have to be rearranged so that the orientation of a dipole changes, which then give us a current.

The Poynting's theorem now reads

$$\nabla \cdot \mathbf{S} + \mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} + \mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} + \mathbf{J}_{\text{free}} \cdot \mathbf{E} = 0. \quad (3)$$

This is the conservation equation of energy only when the material is linear, where we can redefine the electromagnetic energy as

$$u = \frac{1}{2} (\mathbf{E} \cdot \mathbf{D} + \mathbf{B} \cdot \mathbf{H}), \quad (4)$$

which includes the energy stored in the medium. What we *always* have is

$$\nabla \cdot \mathbf{S} + \frac{\partial}{\partial t} \left(\frac{\epsilon_0}{2} \mathbf{E}^2 + \frac{\mu_0}{2} \mathbf{H}^2 \right) + \left(\mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t} + \mu_0 \mathbf{H} \cdot \frac{\partial \mathbf{M}}{\partial t} + \mathbf{J}_{\text{free}} \cdot \mathbf{E} \right) = 0, \quad (5)$$

which is the conservation equation for the “pure” electromagnetic energy, e.g. the energy stored in the electromagnetic field. Of course, the internal degrees of freedom of the medium may contain microscopic electromagnetic field modes, so by “the energy stored in the electromagnetic field” we mean “the energy stored in the electromagnetic modes we care”.

2 A more generalized description of linear response

The $\mathbf{D} = \epsilon_0 \epsilon_r \mathbf{E}$ case is almost trivial: they respond instantaneously and they don't absorb energy. In this section we still assume that the response is linear, but don't assume that the response is spontaneous. Thus

$$\mathbf{P}(t) = \int_{-\infty}^{\infty} \alpha(t - \tau) \mathbf{E}(\tau) d\tau. \quad (6)$$

Thus the polarization field is the convolution of the response function and the electric field. In principle things can be even more complicated: α can have momentum dependence in the Fourier space, and the consequence is that the response is non-local: this is important in, say, plasmon, where long-range Coulomb interaction requires a non-local description of the response.

The wave equation now is

$$\nabla \times \nabla \times \mathbf{E} = -\mu_0 \epsilon_0 \frac{\partial^2}{\partial t^2} \epsilon_r \otimes \mathbf{E}, \quad (7)$$

where \otimes means convolution. This equation is more clearly illustrated by its form in the frequency space:

$$\nabla \times \nabla \times \mathbf{E} = \mu_0 \epsilon_0 \omega^2 \epsilon_r(\omega) \mathbf{E}(\omega), \quad (8)$$

where

$$\epsilon_r(\omega) = \int_{-\infty}^{\infty} \epsilon_r(t) e^{i\omega t} dt, \quad (9)$$

and from the fact that $\epsilon_r(t)$ is real,

$$\epsilon_r(-\omega) = \epsilon_r(\omega)^*. \quad (10)$$

For clarity we may want to use $\tilde{\epsilon}_r$ to refer to $\epsilon_r(\omega)$. The damping behavior of $\epsilon_r(t)$ in the time domain is represented by the imaginary part of $\epsilon_r(\omega)$ in the frequency domain.

(8) tells us an important point: if the input is time harmonic, so is the output. Indeed we can measure $\epsilon_r(\omega)$ in this way.

The plane wave mode with a frequency-dependent $\epsilon_r(\omega)$ has the following dispersive relation:

$$\left(\mathbf{k}^2 - \frac{\omega^2}{c^2} \epsilon_r(\omega) \right) \mathbf{E} = 0 \Rightarrow \mathbf{k} = \pm \frac{\omega \tilde{n}(\omega)}{c} \hat{\mathbf{k}}, \quad (11)$$

where

$$\tilde{n}(\omega) = \sqrt{\epsilon_r(\omega)}. \quad (12)$$

From the dispersive relation we can define phase velocity and group velocity.

It should be noted that n – and hence \mathbf{k} – is allowed to have an imaginary part, which tells us absorption in the material. Defining

$$\tilde{n}(\omega) = n(\omega) + i\kappa(\omega), \quad (13)$$

we find

$$\mathbf{E}(\mathbf{r}, t) = e^{-\frac{\omega}{c} \kappa(\omega) \hat{\mathbf{k}} \cdot \mathbf{r}} e^{i \frac{\omega}{c} n(\omega) \hat{\mathbf{k}} \cdot \mathbf{r} - i\omega t} E \hat{\mathbf{e}}, \quad (14)$$

and therefore the decaying coefficient is

$$\alpha(\omega) = \frac{\omega}{c} \kappa(\omega), \quad (15)$$

from which we find

$$\langle S(z) \rangle = \frac{1}{2} |\text{Re } \mathbf{E}^* \times \mathbf{H}| \propto e^{-2\alpha z}. \quad (16)$$

We can also evaluate the impact of imaginary part of ϵ_r – equivalently, of χ_e – from the perspective of the energy of the electromagnetic field. Consider a region with no energy flow into or out of it. We have

$$\begin{aligned} \nabla \cdot \mathbf{S} + \frac{\partial u}{\partial t} &= -\mathbf{E} \cdot \mathbf{J} \\ \Rightarrow \frac{d}{dt} \int \langle u \rangle d^d \mathbf{r} &= - \int d^d \mathbf{r} \frac{1}{2} \text{Re } \mathbf{E}^* \cdot \mathbf{J} = - \frac{1}{2} \int d^d \mathbf{r} \omega \epsilon_0 \text{Im } \chi(\omega) |\mathbf{E}|^2. \end{aligned} \quad (17)$$

Thus, when χ_2 is positive, the field loses energy, and when χ_2 is negative, the field gets energy.

3 Microscopic model of dispersive media: the harmonic oscillator model

The harmonic oscillator is often used as a simplistic model of the atom. The physical picture seems wrong at the first glance, but if we regard the displacement in the harmonic oscillator as the dipole of the atom, then things begin to make sense; indeed, the “classical” model of harmonic oscillator can be shockingly accurate in certain limits.

The EOM is

$$m\ddot{\mathbf{r}} = -m\Omega^2\mathbf{r} - m\gamma\dot{\mathbf{r}} + q\mathbf{E}, \quad (18)$$

or in other words

$$\ddot{\mathbf{p}} + \gamma\dot{\mathbf{p}} + \Omega^2\mathbf{p} = \frac{q^2}{m}\mathbf{E}(t), \quad (19)$$

where $\mathbf{p} = q\mathbf{r}$ is the dipole. The polarizability can be routinely found as

$$\mathbf{p}(\omega) = \underbrace{\frac{q^2/m}{\Omega^2 - \omega^2 - i\omega\gamma}}_{\alpha(\omega)} \mathbf{E}(\omega). \quad (20)$$

This leads to expected limit cases: when $\omega \ll \Omega$, \mathbf{E} is parallel to \mathbf{p} , while when $\omega \gg \Omega$, \mathbf{E} is in the opposite direction to \mathbf{p} .

Now consider an assembly of atoms, and we find the total polarization field is now

$$\mathbf{P} = \left(\frac{N}{V}\right) \cdot \mathbf{p}, \quad (21)$$

and therefore the behavior of the media, in terms of quantities in the theory of electromagnetism, is now

$$(\ddot{\mathbf{P}} + \gamma\dot{\mathbf{P}} + \Omega^2\mathbf{P}) = \frac{N}{V} \frac{q^2}{m} \mathbf{E}. \quad (22)$$

Recall that from Maxwell’s equations we also have

$$\nabla \times \nabla \times \mathbf{E} = -\mu_0\epsilon_0 \frac{\partial^2}{\partial t^2} \mathbf{E} - \mu_0 \frac{\partial^2}{\partial t^2} \mathbf{P}. \quad (23)$$

The coupled EOMs, in the strong coupling regime, are a simple model of **polariton**.

Real materials have more than one internal modes, and the relation between the polarizability and the electric field becomes

4 Scattering cross sections

For a single oscillator, the absorption power is

$$\langle P_{\text{abs}} \rangle = \langle q\mathbf{E} \cdot \dot{\mathbf{d}} \rangle = \gamma |\mathbf{E}|^2 = \sigma_{\text{abs}} \underbrace{u_{\text{em}} v_g}_{\text{incident intensity}}. \quad (24)$$

From this equation we naturally find a constant measuring how strong the absorption is with area dimension, which is righteously named as the absorption cross section. From this linear relation between absorption and incident intensity we immediately get Beer’s law

$$I(z) = I(0)e^{-\frac{N}{V}\sigma_{\text{abs}}z}, \quad (25)$$

where N is the number of absorption centers. The equation can be derived straightforwardly by using the definition to find

$$\Delta P = -I(z)N\sigma_{\text{abs}}$$

and noticing that

$$P = I \cdot A.$$

Whether a more realistic atom model reduces to the above EOM requires further investigation.

How can this be used for light trapping???

Is it possible to use solely ϵ_r to capture the behaviors of a polariton? Note that with ϵ_r we still only have one mode but here we actually have two modes, EM field and polarization mode. Also, how could, say, phonon, be modeled as \mathbf{P} .

the state of the material is changed after re-

5 Local field correction

The good old “a hole in a material” argument.

6 Kramers-Kronig relations

It’s possible that we have dispersion but no loss (as in, say, a wave guide, caused by the so-called geometric dispersion); in this case K-K relation may fail altogether, due to some quirky properties of the response function.

7 Semiclassical field-atom coupling

In this section we consider the coupling between a semiclassical field and a degree of freedom that is expected to represent an atom, be it a two-level system or a quantum oscillator or something else. The coupling Hamiltonian reads

$$H_1 = -\boldsymbol{\mu} \cdot \mathbf{E} = -q\mathbf{r} \cdot \mathbf{E}, \quad (26)$$

which means we ignore the space dependence of \mathbf{E} and just apply a (possibly time-dependent) uniform electric field to the atom. For example, the EOM of a harmonic oscillator is now

$$\dot{p} = -m\omega_0^2 x + qE, \quad \dot{x} = \frac{p}{m}, \quad (27)$$

which means the toy model of driven harmonic oscillator is in fact quantitatively correct. Indeed, the condition of the dipole approximation is

$$\frac{\omega}{c}a \ll 1, \quad (28)$$

which can be justified either by Taylor expansion or by the physical intuition that the atom should not see the variance of the electric field. The dipole approximation is usually correct, but caveats are needed for Rydberg atoms (a is too large for ordinary wave length) and cases with artificially enhanced strong electric variance as in, say, a very thin tip near a molecule (wave length too small for ordinary a).

One thing that can be immediately noticed is that the dipole moment expectation is always constant when the system is at a stationary state, since the $e^{-i\omega t}$ factors of the bra and the ket cancel each other. This means if we treat the light field as a classical field, we don’t have spontaneous emission, since radiation requires a time evolving dipole moment. If

$$|\psi\rangle = c_a |a\rangle + c_b |b\rangle, \quad (29)$$

we get

$$\langle \boldsymbol{\mu}(t) \rangle_{\text{oscillating part}} = c_a^* c_b e^{-i(\omega_b - \omega_a)t} \langle a | \boldsymbol{\mu} | b \rangle + \text{c.c.}, \quad (30)$$

and radiative coupling is possible when the matrix element is non-zero. When a realistic atomic model is used, this means that the parity of a and b should be different (and thus it’s impossible to have transition between 1s and 2s), and similarly we have selection rules for m and l . Note that we can still have multiple dipole-allowed transitions with the help of some intermediate states to connect two states that, say, have the same parity; or we can make use of other transition channels when the gradient of the electric field is truly large.

The time-dependent perturbation usually can’t be solved exactly. Below we review time-dependent perturbation theory. The Schrodinger equation is

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = (H_0 + \lambda H_1(t)) |\psi(t)\rangle, \quad (31)$$

and we do the decomposition (by including the $e^{-i\omega_n t}$ factor we implicitly come into the interaction picture)

$$|\psi(t)\rangle = \sum_n \underbrace{\left(\gamma_n^{(0)} + \lambda \gamma_n^{(1)} + \lambda^2 \gamma_n^{(2)} + \dots \right)}_{\gamma_n} |n\rangle e^{-i\omega_n t}, \quad (32)$$

Incoherent
nonlinear
processes

and from the

$$\frac{d\gamma_k}{dt} = \frac{1}{i\hbar} \lambda \sum_n \langle k | H_1 | n \rangle \gamma_n(t) e^{i(\omega_k - \omega_n)t} \quad (33)$$

we get

$$\frac{d}{dt} \gamma_k^{(0)} = 0, \quad (34)$$

$$\frac{d}{dt} \gamma_k^{(1)} = \frac{1}{i\hbar} \sum_n H_{1,kn} \gamma_n^{(0)} e^{i(\omega_k - \omega_n)t}, \quad (35)$$

$$\frac{d}{dt} \gamma_k^{(2)} = \frac{1}{i\hbar} \sum_n H_{1,kn} \gamma_n^{(1)} e^{i(\omega_k - \omega_n)t}, \quad (36)$$

and so on.

We can organize the perturbed coefficients in the scattering matrix formalism:

$$\gamma_k(t) = \gamma_k^{(0)} + \sum_n \underbrace{(S_{kn}^{(0)}(t) + S_{kn}^{(1)}(t) + \dots)}_{S_{kn}} \gamma_n^{(0)}, \quad (37)$$

and the scattering matrix $S_{kn}(t)$ can be obtained from the aforementioned series of equations.

The time evolution caused by dipole coupling

$$H = \underbrace{-\boldsymbol{\mu} \cdot \mathbf{E}_0}_{=: W_{kn}} \cos \omega t \quad (38)$$

therefore is

$$\begin{aligned} \frac{d\gamma_k^{(1)}}{dt} &= \frac{1}{i\hbar} \sum_n \gamma_n^{(0)} W_{kn} e^{i\omega_{kn}t} \cos \omega t \\ &= \frac{1}{2i\hbar} \sum_n W_{kn} \gamma_n^{(0)} (e^{i(\omega_{kn} + \omega)t} + e^{i(\omega_{kn} - \omega)t}), \end{aligned} \quad (39)$$

and the equation can then be solved directly since $\gamma_n^{(0)}$ is a constant. An important approximation is **rotating wave approximation (RWA)**, in which we ignore the term $e^{i(\omega + \omega_{kn})t}$; this is a good approximation when

$$|\omega + \omega_{kn}| \gg |\omega + \omega_{kn}|, \quad (40)$$

or in other words when the pumping $\mathbf{E} = \mathbf{E}_0 \cos \omega t$ is nearly resonant. Basically, in RWA we are extracting the *envelope* of $\gamma_k^{(1)}$, since ignoring the fast oscillating term in a function $f(t)$ is equivalent to replacing $f(t)$ by

$$\tilde{f}(t) = \frac{1}{T} \int_{t-T/2}^{t+T/2} f(t') dt', \quad (41)$$

where T is much larger than the period of the fast oscillation.

Now after RWA, we find

$$\begin{aligned} \gamma_k^{(1)}(T) &= \frac{1}{2i\hbar} \sum_n W_{kn} \gamma_n^{(0)} \frac{1}{i(\omega_{kn} - \omega)} e^{i(\omega_{kn} - \omega)t} \Big|_0^T \\ &= T \cdot \frac{1}{i\hbar} \sum_n W_{kn} \gamma_n^{(0)} e^{i\Delta\omega T/2} \text{sinc}(\Delta\omega T/2), \end{aligned} \quad (42)$$

where

$$\omega = \omega_k - \omega_n - \omega. \quad (43)$$

Therefore we find the probability to s

$$P_{n \rightarrow k}(t) = \quad (44)$$

The final result is the famous Fermi golden rule

$$P_{n \rightarrow k}(t) = t \cdot \frac{2\pi}{\hbar} |W_{kn}|^2 (\gamma_n^{(0)})^2 \delta_T(\omega_k - \omega_n - \omega). \quad (45)$$

We see expectedly that the transition probability reaches its maximum when the pumping is resonant. The fact that we have a sinc function profile of the transition probability comes from the finite length of the driving field: from the mathematical uncertainty relation, when the pumping pulse has a finite length, we don't have a completely well-defined ω , and the sinc shape comes from the Fourier transform of the finite length sine wave.

Now we consider radiation from the atom. This means we should consider the $\omega < 0$ term which contributes to the amplitude of states with lower energies. We do a reverse RWA and ignore the $e^{i(\omega_{kn}-\omega)t}$ term.

The dipole can also be evaluated from the perturbed wave function. We have

$$\begin{aligned}\langle\psi|\boldsymbol{\mu}|\psi\rangle &= (\langle\psi^{(0)}| + \lambda\langle\psi^{(1)}| + \lambda^2\langle\psi^{(2)}| + \dots)\boldsymbol{\mu}(|\psi^{(0)}\rangle + \lambda|\psi^{(1)}\rangle + \lambda^2|\psi^{(2)}\rangle + \dots) \\ &= \underbrace{\langle\psi^{(0)}|\boldsymbol{\mu}|\psi^{(0)}\rangle}_{\langle\boldsymbol{\mu}\rangle^{(0)}} + \underbrace{\langle\psi^{(0)}|\boldsymbol{\mu}|\psi^{(1)}\rangle + \text{c.c.} + \dots}_{\langle\boldsymbol{\mu}\rangle^{(1)}},\end{aligned}\tag{46}$$

and we can then find the response of the atom dipole to the electric field. When we are doing this actually we already have to slightly deviate from the standard semiclassical coupling theory, or otherwise when $\omega = \omega_{eg}$, the response is infinite. The physical answer is spontaneous radiation.

The above procedure can also be seen as a procedure to integrate out the atomic degrees of freedom and obtain an effective theory of the light field.

Study dissipation and noise here; todo: spontaneous radiation