Light-matter interaction

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September 13, 2023

TODO:

- There are rare cases where the energy velocity (i.e. the velocity that connects $\boldsymbol{s}$ 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

$$\boldsymbol{D} = \epsilon_0 \boldsymbol{E} + \boldsymbol{P} = \epsilon_0 \boldsymbol{E} + \epsilon_0 \chi_\mathrm{e} \boldsymbol{E} = \epsilon_0 \underbrace{(1 + \chi_\mathrm{e})}_{=:\epsilon_\mathrm{r}} \boldsymbol{E}. \tag{1}$$

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

$$\boldsymbol{B} = \mu_0 (\boldsymbol{H} + \boldsymbol{M}) = \mu_0 \underbrace{(1 + \chi_\mathrm{m})}_{\mu_\mathrm{r}} \boldsymbol{H}, \tag{2}$$

as if $\boldsymbol{B}$ has the same status of $\boldsymbol{D}$, although it's $\boldsymbol{H}$ 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 $\partial_t \boldsymbol{P}$ 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

$$\boldsymbol{\nabla} \cdot \boldsymbol{S} + \boldsymbol{H} \cdot \frac{\partial \boldsymbol{B}}{\partial t} + \boldsymbol{E} \cdot \frac{\partial \boldsymbol{D}}{\partial t} + \boldsymbol{J}_\mathrm{free} \cdot \boldsymbol{E} = 0. \tag{3}$$

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

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

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

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

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 $\boldsymbol{D} = \epsilon_0\epsilon_\mathrm{r}\boldsymbol{E}$ 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

$$\boldsymbol{P}(t) = \int_{-\infty}^{\infty} \alpha(t-\tau)\boldsymbol{E}(\tau)\,\mathrm{d}\tau\,. \tag{6}$$

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

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E} = -\mu_0\epsilon_0\frac{\partial^2}{\partial t^2}\epsilon_\mathrm{r}\otimes\boldsymbol{E}, \tag{7}$$

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

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E} = \mu_0\epsilon_0\omega^2\epsilon_\mathrm{r}(\omega)\boldsymbol{E}(\omega), \tag{8}$$

where

$$\epsilon_\mathrm{r}(\omega) = \int_{-\infty}^{\infty}\epsilon_\mathrm{r}(t)\mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{d}t\,, \tag{9}$$

and from the fact that $\epsilon_\mathrm{r}(t)$ is real,

$$\epsilon_\mathrm{r}(-\omega) = \epsilon_\mathrm{r}(\omega)^*. \tag{10}$$

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

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

The plane wave mode with a frequency-dependent $\epsilon_\mathrm{r}(\omega)$ has the following dispersive relation:

$$\left(\boldsymbol{k}^2 - \frac{\omega^2}{c^2}\epsilon_\mathrm{r}(\omega)\right)\boldsymbol{E} = 0 \Rightarrow \boldsymbol{k} = \pm\frac{\omega\tilde{n}(\omega)}{c}\hat{\boldsymbol{k}}, \tag{11}$$

where

$$\tilde{n}(\omega) = \sqrt{\epsilon_\mathrm{r}(\omega)}. \tag{12}$$

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

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

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

we find

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

and therefore the decaying coefficient is

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

from which we find

$$\langle S(z)\rangle = \frac{1}{2}|\mathrm{Re}\,\boldsymbol{E}^*\times\boldsymbol{H}| \propto \mathrm{e}^{-2\alpha z}. \tag{16}$$

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

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

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

$$m\ddot{\boldsymbol{r}} = -m\Omega^2\boldsymbol{r} - m\gamma\dot{\boldsymbol{r}} + q\boldsymbol{E}, \tag{18}$$

or in other words

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

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

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

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

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

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

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

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

Recall that from Maxwell's equations we also have

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E} = -\mu_0\epsilon_0\frac{\partial^2}{\partial t^2}\boldsymbol{E} - \mu_0\frac{\partial^2}{\partial t^2}\boldsymbol{P}. \tag{23}$$

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

$$\langle P_{\mathrm{abs}}\rangle = \left\langle q\boldsymbol{E}\cdot\dot{\boldsymbol{d}}\right\rangle = \gamma|\boldsymbol{E}|^2 = \sigma_{\mathrm{abs}}\underbrace{u_{\mathrm{em}}v_{\mathrm{g}}}_{\text{incident intensity}}. \tag{24}$$

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 immediate get Beer's law

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

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

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

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 $\boldsymbol{P}$.

the state of the material is changed

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

$$H_1 = -\boldsymbol{\mu} \cdot \boldsymbol{E} = -q\boldsymbol{r} \cdot \boldsymbol{E}, \tag{26}$$

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

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

which means the toy model of driven harmonic oscillator is in fact quantitatively correct.

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 $\mathrm{e}^{-\mathrm{i}\omega t}$ 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

$$|\psi\rangle = c_a |a\rangle + c_b |b\rangle, \tag{28}$$

we get

$$\langle \boldsymbol{\mu}(t) \rangle_{\text{oscillating part}} = c_a^* c_b \mathrm{e}^{-\mathrm{i}(\omega_b - \omega_a)t} \langle a|\boldsymbol{\mu}|b\rangle + \text{c.c.}, \tag{29}$$

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.

Incoherent nonlinear processes

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

$$\mathrm{i}\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = (H_0 + \lambda H_1(t)) |\psi(t)\rangle, \tag{30}$$

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

$$|\psi(t)\rangle = \sum_n \underbrace{\left(\gamma_n^{(0)} + \lambda\gamma_n^{(1)} + \lambda^2\gamma_n^{(2)} + \cdots\right)}_{\gamma_n} |n\rangle \mathrm{e}^{-\mathrm{i}\omega_n t}, \tag{31}$$

and from the

$$\frac{\mathrm{d}\gamma_k}{\mathrm{d}t} = \frac{1}{\mathrm{i}\hbar}\lambda \sum_n \langle k|H_1|n\rangle \gamma_n(t) \mathrm{e}^{\mathrm{i}(\omega_k - \omega_n)t} \tag{32}$$

we get

$$\frac{\mathrm{d}}{\mathrm{d}t}\gamma_k^{(0)} = 0, \tag{33}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\gamma_k^{(1)} = \frac{1}{\mathrm{i}\hbar} \sum_n H_{1,kn}\gamma_n^{(0)} \mathrm{e}^{\mathrm{i}(\omega_k - \omega_n)t}, \tag{34}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\gamma_k^{(2)} = \frac{1}{\mathrm{i}\hbar}\sum_n H_{1,kn}\gamma_n^{(1)}\mathrm{e}^{\mathrm{i}(\omega_k-\omega_n)t}, \tag{35}$$

and so on.

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

$$\gamma_k(t) = \gamma_k^{(0)} + \sum_n \underbrace{(S_{kn}^{(0)}(t) + S_{kn}^{(1)}(t) + \cdots)}_{S_{kn}}\gamma_n^{(0)}, \tag{36}$$

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

Relation with off-shell time independent PT; time ordering?