Light-matter interaction

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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

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

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

$$P = I\cdot A.$$

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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 after ra-

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. Indeed, the condition of the dipole approximation is

$$\frac{\omega}{c}a \ll 1, \tag{28}$$

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

$$|\psi\rangle = c_a |a\rangle + c_b |b\rangle, \tag{29}$$

we get

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

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

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

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

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

Incoherent nonlinear processes

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and from the

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

we get

$$\frac{\mathrm{d}}{\mathrm{d}t}\gamma_k^{(0)} = 0, \tag{34}$$

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

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

and so on.

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

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

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

The time evolution caused by dipole coupling

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

therefore is

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

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 $\mathrm{e}^{\mathrm{i}(\omega+\omega_{kn})t}$; this is a good approximation when

$$|\omega + \omega_{kn}| \gg |\omega + \omega_{kn}|, \tag{40}$$

or in other words when the pumping $\boldsymbol{E} = \boldsymbol{E}_0\cos\omega t$ 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

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

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

Now after RWA, we find

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

where

$$\omega = \omega_k - \omega_n - \omega. \tag{43}$$

Therefore we find the probability to s

$$P_{n\to k}(t) = \tag{44}$$

The final result is the famous Fermi golden rule

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

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.

If we reflect on Fermi golden rule for a while, we find something not that physical: the spectrum of the final states may be discrete, and in this case the δ function looks suspicious: for continuous modes the final state spectrum is continuous and by saying we sum over final states we are actually integrating over the final states, so no singular function enters the final result of scattering probability; but for discrete systems this is not true. This is actually a motivation for quantization of the electromagnetic field for this provides us with a continuous energy spectrum of the composite system of matter and light.

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

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

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 $\omega = \omega_{\text{eg}}$, 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.

8 Elaboration on perturbation schemes

Suppose we have an *anhamornic* oscillator, and the EOM is

$$
m\ddot{x} + m\omega_0^2 x = -\beta x^2,
\tag{47}
$$

or in other words

$$
\ddot{x} + \omega_0^2 x = -\underbrace{\frac{\beta}{m}}_{=:\alpha} x^2.
\tag{48}
$$

Of course, when $\alpha = 0$, we go back to the harmonic case. This equation falls into the scheme

$$
Lx = \lambda N x,
\tag{49}
$$

where L is a linear operator and N is the nonlinear, "interacting" part. The perturbation series

$$
x(t) = x_0(t) + \lambda x_1(t) + \cdots
\tag{50}
$$

then gives

$$
\ddot{x}_0 + \omega_0^2 x_0 = 0,
\tag{51}
$$

$$
\ddot{x}_1 + \omega_0^2 x_1 = \alpha x_0^2,
\tag{52}
$$

$$
\ddot{x}_2 + \omega_0^2 x_2 = \alpha x_0 x_1,
\tag{53}
$$

and so on. $x_1(t)$ takes the $\cos(2\omega t)$ form, or from Feynman diagram it represents SHG from two excitons with frequency ω into one exciton with frequency 2ω.

In the case of a unitary x^3 term in the Hamiltonian, the perturbation theory seems to work fine; but now if we treat *damping* in the damped oscillator as a perturbation item, we get a pathological solution which *diverges* and doesn't converge, although the accuracy in the first several periods is good. This shows the limit of perturbation theory.

The solution of

$$
\ddot{a} + \gamma a + \omega_0^2 a = 0
\tag{54}
$$

Study dissipation and noise here; todo: spontaneous radiation; what's the difference between spontaneous radiation and the radiation shown here?

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is

$$a(t) = a_0 e^{-i\omega_\pm t}, \tag{55}$$

where

$$\omega_\pm = \pm\sqrt{\omega_0^2 - \left(\frac{\gamma}{2}\right)^2} - i\frac{\gamma}{2}, \tag{56}$$

where we observe an envelope

$$\langle a \rangle (t) = a_0 e^{-\gamma t/2} \tag{57}$$

under which we see a fast oscillation with its frequency modified by damping. When damping is strong compared with ω_0, it's hard to separate the two, but when damping is not that strong, separation between damping and oscillation – or more generally, separation between two time scales – is possible, and the oscillation shouldn't be too different from $e^{-i\omega_0 t}$.

With this in mind, assuming that $\gamma/\omega_0 \ll 1$, and rewriting the equation as

$$\ddot{a} + \varepsilon\gamma a + \omega_0^2 a = 0, \tag{58}$$

where ε is a unitless constant for bookkeeping purpose, we write down the ansatz

$$a(t) = \bar{a}(\underbrace{\varepsilon t}_{=:\tau})e^{-i\omega_0 t}, \tag{59}$$

which contains a fast variable $e^{-i\omega_0 t}$ and a slow variable $\bar{a}$; the argument of $\bar{a}$ is intentionally set to εt to remind us that $\bar{a}$ is very dull to how time passes by. In the more general case we use $f(t)$ in place of $e^{-i\omega_0 t}$. Putting it into (58), and noticing that

$$\dot{a} = -i\omega_0\bar{a}e^{-i\omega_0 t} + \varepsilon\frac{d\bar{a}}{d\tau}e^{-i\omega_0 t}, \tag{60}$$

and

$$\ddot{a} \approx (-i\omega_0)^2\bar{a}e^{-i\omega_0 t} + 2\varepsilon(-i\omega_0)e^{-i\omega_0 t}\frac{d\bar{a}}{d\tau}, \tag{61}$$

we have

$$2\varepsilon(-i\omega_0)e^{-i\omega_0 t} \cdot \dot{a}\frac{d\bar{a}}{d\tau} + \varepsilon(-i\omega_0)e^{-i\omega_0 t}\bar{a} = 0 \;\Rightarrow\; \frac{d\bar{a}}{d\tau} + \frac{\gamma}{2}\bar{a} = 0. \tag{62}$$

This is exactly what we expect. In this example we explicitly write $a(t)$ as the multiplication of $e^{-i\omega_0 t}$ and $\bar{a}(\tau)$; if we instead introduce $f(t)$, we will find the equation it follows is just the harmonic oscillator equation. Finally, we replace $\varepsilon\gamma$ with γ to go back to the original problem, and an approximate solution has been found.

The most formal procedure requires us to keep track of every ε without any ahead-of-time approximations like (61) or inserting the $\gamma = 0$ solution as $f(t)$ into the ansatz for a. In simple calculations however people tend to only keep track of the $\mathcal{O}(\varepsilon)$ term, and the above procedure is perfectly legit. The idea is the fast part of the solution is usually not perturbed at all. This approximation – tracking only $\mathcal{O}(\varepsilon)$ – is known as **slowly varying envelope approximation**.

Relation with ordinary PT; RG?

9 Rotating wave approximation revisited

Consider the driven harmonic oscillator

$$\ddot{a} + \omega_0^2 a = \varepsilon A \cos(\omega t), \tag{63}$$

where we have assumed that the driving term is weak enough; of course since there is no damping, the amplitude will increase infinitely, but since the driving force is small enough it doesn't explode fast.

The ansatz is

$$a(t) = \bar{a}(\varepsilon t)e^{-i\omega_0 t}. \tag{64}$$

Substituting this into the EOM, we get

$$(-\omega_0^2\bar{a} - 2\varepsilon i\omega_0\partial_\tau\bar{a} + \varepsilon^2\partial_\tau^2\bar{a})e^{-i\omega_0 t} + \omega_0^2\bar{a}e^{-i\omega_0 t} = \varepsilon A \cos(\omega t). \tag{65}$$

The $\mathcal{O}(\varepsilon)$ term is

$$-2\mathrm{i}\varepsilon\omega_0\partial_\tau\bar{a} = \varepsilon A\cos(\omega t) \;\Rightarrow\; \frac{\mathrm{d}\bar{a}}{\mathrm{d}t} = \frac{\mathrm{i}A}{2\omega_0}\cos(\omega t)\mathrm{e}^{\mathrm{i}\omega_0 t}. \tag{66}$$

Unfortunately $\bar{a}$ still has fast oscillation, but then we can average over time and get rid of the fast oscillation, and the eventual result is

$$\frac{\mathrm{d}\langle\bar{a}\rangle}{\mathrm{d}t} = \frac{\mathrm{i}A}{4\omega_0}. \tag{67}$$

To make this approximation make sense, we require

$$\frac{A}{4\omega_0} \ll \omega_0 \tag{68}$$

so that the increasing of the envelope is not of the same order of magnitude of ω_0.

10   Spontaneous emission

We can really think of the spontaneous rate as produced by an effective photon flux: the spontaneous emission rate can be rewritten as

$$R_{\mathrm{sp}} = \sigma \cdot \frac{c}{\hbar\omega} \cdot \frac{\hbar\omega^3}{\pi c^3}. \tag{69}$$

The spontaneous emission rate can also be obtained by replacing the thermal photon occupation in stimulated emission with 1.

<div style="float:right; border:1px solid; background:orange;">Relation between cross section and expectation of p</div>

11   Coupling with a quantized EM field

Consider an atom in a cavity. We assume that the size of the cavity is very large, almost infinite; this means once a photon is emitted it almost never goes back, and thus

<div style="float:right; border:1px solid; background:orange;">Rate equation</div>

- we don't need to consider anything like Poincaré recurrence, and can model the system as a dissipative one, and

- the out state spectrum is continuous and we can use Fermi golden rule.

The second point can be seen as a logical consequence of the first one, since Fermi golden rule is somehow "dissipative" in nature.

12   Interlude: quantization of LC circuit

As a demonstration of canonical quantization, let's consider the quantum version of an LC circuit. The magnetic energy is

$$U_{\mathrm{m}} = \frac{1}{2}LI^2 = \frac{1}{2}L\dot{Q}^2, \tag{70}$$

and the electric energy is

$$U_{\mathrm{e}} = \frac{1}{2}\frac{Q^2}{C}, \tag{71}$$

and if we consider the former as the kinetic energy and the latter as the potential energy, the Lagrangian is

$$L = \frac{1}{2}L\dot{Q}^2 - \frac{Q^2}{2C}, \tag{72}$$

and we can easily find that this Lagrangian gives the correct EOM. Going to the Hamiltonian formalism, after a Legendre transform we get

$$H = \frac{\Phi^2}{2L} + \frac{1}{2}\frac{Q^2}{C}, \tag{73}$$

where Φ is the canonical momentum of Q, i.e.

$$\Phi := \frac{\partial L}{\partial \dot{Q}} = L\dot{Q}. \tag{74}$$

Of course Φ has its own physical meaning: the magnetic flux. Applying canonical commutation relation

$$[Q, \Phi] = \mathrm{i}\hbar \tag{75}$$

we can already quantize the system. It looks like a quantum harmonic oscillator, also with different "mass" and "spring constant". So the final form of the Hamiltonian, in occupation number representation, is

> Deriving this from QED

$$H = \hbar\omega \left(a^\dagger a + \frac{1}{2} \right), \tag{76}$$

where

$$\omega = \frac{1}{\sqrt{LC}} \tag{77}$$

is the LC frequency.

It should be noted that to implement the quantum circuit, ordinary metals probably won't work, since they contain too many other degrees of freedom that couple strongly with the electromagnetic degrees of freedom and destroy the coherence of the latter even when the temperature is low. Superconducting wires are usually used in place of ordinary wires.

We are describing only one harmonic oscillator here; when there are many harmonic oscillators aligned in a certain way in space, it's possible to define a *field operator* $\hat{\phi}(\boldsymbol{r}, t)$, where $\boldsymbol{r}$ is equivalent to the label of harmonic oscillators, and the time evolution is treated in the same way as in ordinary canonical quantization.

Let's consider the electric and magnetic field in the LC circuit. It can be verified that

$$\boldsymbol{E}(\boldsymbol{r}) = \underbrace{\bar{\boldsymbol{E}}(\boldsymbol{r})\sqrt{\frac{\hbar}{2\omega_0}}}_{\boldsymbol{E}^0(\boldsymbol{r})}(a_{\boldsymbol{q}}^\dagger + a_{\boldsymbol{q}}), \tag{78}$$

and

$$\boldsymbol{B}(\boldsymbol{r}) = \underbrace{\bar{\boldsymbol{B}}(\boldsymbol{r})\mathrm{i}\sqrt{\frac{\hbar\omega_0}{2}}}_{\boldsymbol{B}^0(\boldsymbol{r})}(a_{\boldsymbol{q}}^\dagger - a_{\boldsymbol{q}}). \tag{79}$$

As a simple demonstration let's assume that there is no non-trivial polarization in the circuit: thus we can treat $\boldsymbol{E}$ and $\boldsymbol{B}$ as scalars in *this* case. Normalization of $\boldsymbol{E}^0$ and $\boldsymbol{B}^0$ can be decided by evaluating the total energy: we expect to get

$$\langle U_\mathrm{e} \rangle + \langle U_\mathrm{m} \rangle = \frac{1}{2}\hbar\omega_0 \tag{80}$$

when we are at the ground state. We can easily verify that the consequence of this equation is

$$\epsilon_0 \int \mathrm{d}^3\boldsymbol{r} \, |\boldsymbol{E}^0(\boldsymbol{r})| = \frac{1}{2}\hbar\omega_0. \tag{81}$$

When the shape of our system is not very nontrivial and we can essentially treat it as a box with volume V in which $|\boldsymbol{E}^0(\boldsymbol{r})|$ is completely uniform, this tells us

$$E^0(\boldsymbol{r}) = \sqrt{\frac{\hbar\omega_0}{2\epsilon_0 V}}. \tag{82}$$

So the electric field operator for only one mode in a large box is

$$\boldsymbol{E}(\boldsymbol{r}) = \hat{\boldsymbol{z}}\sqrt{\frac{\hbar\omega_0}{2\epsilon_0 V}}(a^\dagger + a). \tag{83}$$

13 Cavity light field modes and two-level atom

In principle (83) should be summed over to get the full electric field operator. But in real experimental settings the only active degrees of freedom in a system are two energy levels of an atom and one EM mode, and the Hamiltonian becomes

$$H = E_{\mathrm{g}} |\mathrm{g}\rangle\langle\mathrm{g}| + E_{\mathrm{e}} |\mathrm{e}\rangle\langle\mathrm{e}| - \boldsymbol{\mu} \cdot \boldsymbol{E} + \hbar\omega_0(a^\dagger a + 1/2), \tag{84}$$

where $\boldsymbol{E}$ is given by (83). The wave function is already huge: the Hilbert space of the cavity part is countably infinite. We can analyze possible transitions by calculating

$$\langle \text{final}| -\mu E_0 \cos\theta(a + a^\dagger)|\text{initial}\rangle . \tag{85}$$

The transition between $|\mathrm{g}, n = 1\rangle$ and $|\mathrm{e}, n = 0\rangle$, for example, gives us Rabi oscillation. It's impossible to get spontaneous decay in this model, because the number of optical modes is not large enough; we can find spontaneous decay when there are lots of $a_{\boldsymbol{q}}$.

14 Quantization of field

Now we consider the complete electric field operator. We assume it's the sum of something like (83), where a may be any annihilation operator corresponding to an oscillation mode of the system treated as a quantum oscillator, and $\hat{\boldsymbol{z}}$ needs to be replaced by the true polarization vector $\hat{\boldsymbol{f}}$, which may also contain phase factors like $\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}$, with the same normalization

$$\int \mathrm{d}^3\boldsymbol{r}\, |\hat{\boldsymbol{f}}(\boldsymbol{r})|^2 = V. \tag{86}$$

Note that the equivalence of this quantization and the "real" canonical quantization by imposing commutation relations on $\boldsymbol{A}$ is not guaranteed a priori, but is indeed true. The interaction between light and a two-level atom is then modeled by

$$H = H_{\mathrm{atom}} - \boldsymbol{\mu} \cdot \boldsymbol{E} + \hbar \sum_k \omega_k \left(a_k^\dagger a_k + \frac{1}{2} \right). \tag{87}$$

The basis of the Hilbert space is

$$\{ |\mathrm{atom}, \underbrace{n_1, n_2, \ldots, n_k, \ldots}_{\mathrm{light},\{n_i\}}\rangle \}. \tag{88}$$

The dipole interaction term still changes one photon occupation number by 1 at once; but now an excited state can evolve into many one-photon final states, and we will see we will get exponential decay of the atomic occupation.

15 Spontaneous emission in vacuum

Now we finally can have a quantitative description of spontaneous emission. The complete Hamiltonian is

$$H = H_{\text{two level atom}} + \sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} \left(a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \frac{1}{2} \right) - \sum_{\boldsymbol{k}} \boldsymbol{\mu} \cdot \boldsymbol{E}_0 \mathrm{i}(a_{\boldsymbol{k}}^\dagger - a_{\boldsymbol{k}}). \tag{89}$$

Consider only the single-photon process, we confine (88) to the subspace where there is at most one photon in the system. The total transition rate, i.e. the probability for an excited atom to emit one photon and goes back to the ground state, is the sum of the transition rates of all radiation channels. So we have

$$\begin{aligned}
P_{\mathrm{e}\to\mathrm{g}} &= \sum_{\boldsymbol{k}} \frac{2\pi}{\hbar^2} |W_{\mathrm{e}\to\mathrm{g},\boldsymbol{k}}|^2 \delta(\omega_{\boldsymbol{k}} - \omega_{\mathrm{eg}}) \\
&= \frac{2\pi}{\hbar^2} |W|^2 D(\omega_{\mathrm{eg}}),
\end{aligned} \tag{90}$$

When the light field is in a coherent state, we also have Rabi oscillation. Explore the situation under which we have Rabi frequency; especially if it has anything to do with MBPT.

Commutation relation between $\boldsymbol{E}$ and $\boldsymbol{B}$

where we have assumed that the wave vector of the output photon $\boldsymbol{k}$ has little influence on the transition amplitude $W_{\mathrm{e}\to\mathrm{g},\boldsymbol{k}}$ (this assumption of course fails in highly anistropic systems, like photonic crystals), and $D(\omega)$ is the density of states of EM modes. When $\boldsymbol{k}$ does have a strong influence on W, we need to rewrite W into

$$W_{\mathrm{e}\to\mathrm{g},\boldsymbol{k}} = W(\omega_{\boldsymbol{k}}, \hat{\boldsymbol{k}}), \tag{91}$$

and the transition probability becomes

$$P_{\mathrm{e}\to\mathrm{g}} = \frac{2\pi}{\hbar^2} \int \frac{\mathrm{d}\Omega}{4\pi} \left| W(\omega_{\mathrm{eg}}, \hat{\boldsymbol{k}}) \right|^2 D(\omega_{\mathrm{eg}}), \tag{92}$$

where $\mathrm{d}\Omega$ is the solid angle element of $\hat{\boldsymbol{k}}$.

In free space, we have

$$|W|^2 = |\mu_{\mathrm{eg}}|^2 |E_0|^2 \cos^2\theta = \frac{\hbar\omega}{2\epsilon_0 V} |\mu_{\mathrm{eg}}|^2 \cos^2\theta. \tag{93}$$

The DOS of light is

$$D(\omega) = \frac{V}{2\pi^2} \frac{\omega^2}{c^3}. \tag{94}$$

We can verify that

$$\int \frac{\mathrm{d}\Omega}{4\pi} \cos^2\theta = \frac{1}{3}, \tag{95}$$

and we still have double degeneracy of polarization. The final transition rate is

$$\begin{aligned}
\Gamma_{\mathrm{e}\to\mathrm{g}} &= 2 \cdot \frac{2\pi}{\hbar^2} \cdot \frac{\hbar\omega}{2\epsilon_0 V} |\mu_{\mathrm{eg}}|^2 \cdot \frac{1}{3} \cdot \frac{V}{2\pi^2} \frac{\omega^2}{c^3} \\
&= \frac{\omega^3 |\mu_{\mathrm{eg}}|^2}{3\pi\epsilon_0 \hbar c^3}.
\end{aligned} \tag{96}$$

It's possible that we also have a continuum of *initial* states: in this case we need to sum over initial states as well, resulting in a joint density of states.

The decay rate also gives an uncertainty of the energy levels.

16 Emission into a cavity

There is no dissipation in the EM field in vacuum, but there may be dissipation in a cavity. A cavity always have coupling with a continuum of degrees of freedom, which produces what is known as dissipation if we keep our eyes on the cavity itself. If we go to the example of the LC circuit, this time connected to a semi-infinite transmission line, which plays the role of resistance, we will observe the same phenomenon. In a leaky cavity we no longer have perfectly well defined frequencies, and when the atom is coupled to the cavity, we need to replace the old DOS by an effective DOS with finite lindwidths; note that for each mode in the cavity (leaky or not), we always have [Derivation]

$$\int \mathrm{d}\omega \, \mathrm{DOS}(\omega) = 1. \tag{97}$$

Now suppose we have an atom in a leaky cavity. It has to spontaneous emission targets: the free space (whose DOS is (94)) and the cavity modes. Assuming that only the *one* cavity mode nearby is relevant, we have

$$D_{\mathrm{leaky\ cavity}}(\omega) = \frac{1}{2\pi} \frac{\Delta\omega}{(\omega - \omega_0)^2 + (\Delta\omega/2)^2}, \tag{98}$$

where ω_0 is the frequency of that mode if there were no dissipation, and $\Delta\omega$ is the line width. Defining

$$\Delta\omega =: \frac{\omega_0}{Q}, \tag{99}$$

we find

$$\eta_{\mathrm{Purcell}} := \frac{\Gamma_{\mathrm{cavity}}}{\Gamma_{\mathrm{free\ space}}} \propto \frac{Q}{V} \cdot \text{something about } \omega. \tag{100}$$

So unexpectedly, if we increase Q and decrease V, we can make the atom selectively emits a photon whose behavior we know. This is known as **Purcell enhancement**; a Purcell device can be used as a single photon source. Note that Q shouldn't be too strong if we want to use it as a single photon source, or otherwise what we get is just Rabi oscillation.