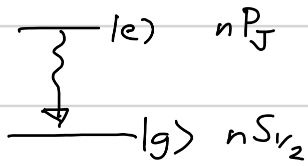
Physics 566 – Quantum Optics I
Lecture 16 – Spotaneous Emission

An atom in an excited state is not a stationary state because the atom couples to the quantum electromagnetic vacuum. We have seen that, when the atom is in a high-Q electromagnetic cavity, the atom *coherently* exchanges energy with the field, periodically emitting the photon into the cavity while the atom deexcites to a lower energy state, and reabsorbing the photon with a return to the excited state. These coherent "Vacuum Rabi oscillations" are indicative of "closed quantum system" in which there are only a few degrees of freedom and no perturbing effects of a quantum "enviroment." For the Jaynes–Cummings model, we need only consider a two-level atom and one mode of the quantum electromagnetic field.

For an atom in an excited state in free space we know that it undergoes a very different kind of evolution, it spontaneously and *irreversibly* decays to a lower energy state

Eg. First excited state → ground in an alkali $\qquad \Gamma = \frac{1}{\tau_{life}}$



An atom in free space is an open quantum system. That is, the atom is coupled to the continuum of modes associated with the electromagnetic field. This coupling allows the atom to radiate into a continuum of modes, so the photon radiates to infinity and never returns leading to irreviable decay of the atom. We will study this in much greater detail in later lectures and we study the general problem of open quantum systems. We begin our first foray here with a study of spontaneous emission.

Wigner – Weisskopf Theory

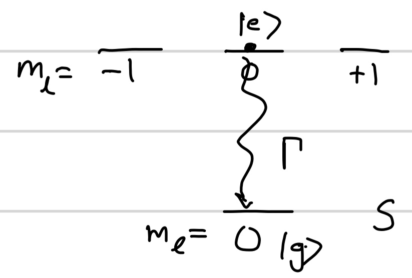
Our goal is to solve for the evolution of the Atom + Field system with the initial condition $|\Psi^{(0)}_{AF}\rangle = |e\rangle \otimes |0\rangle$ (atom in excited state, field in vacuum state). We are intrested in the evolution towards $|g\rangle \otimes |\{1_{k,\lambda}\}\rangle$. As there is some resonance frequency $\omega_{eg} = \frac{E_e - E_g}{\hbar}$, we have use our approximate atom–field interaction Hamiltonian in the RWA, i.e., Only the modes with $\omega_{\vec{k},\lambda} \approx \omega_{eg}$ will dominate the dynamics of interest and $\lambda \ll a_0$ dipole approximation. In the Schrödinger picture the Hamiltonian is

$$\Rightarrow \hat{H} = \underbrace{\hbar\omega_{eg}\hat{\sigma}_z}_{\hat{H}_A} + \underbrace{\sum_{\vec{k},\mu}\hbar\omega_k \hat{a}^\dagger_{\vec{k}\mu}\hat{a}_{\vec{k}\mu}}_{\hat{H}_F} + \underbrace{\sum_{\vec{k},\mu}\left(\hbar g_{\vec{k},\mu}\hat{a}_{\vec{k}\mu}\hat{\sigma}_+ + \hbar g^*_{\vec{k}\mu}\hat{a}^\dagger_{\vec{k}\mu}\hat{\sigma}_-\right)}_{\hat{H}_{AF}}$$

where $\hbar g_{\vec{k},\mu} = -\langle e|\hat{\vec{d}}|g\rangle \cdot \vec{\epsilon}_{\vec{k},\mu}\sqrt{\frac{2\pi\hbar\omega_k}{V_Q}}\, e^{i\vec{k}\cdot\vec{R}_A}$ ← atomic position

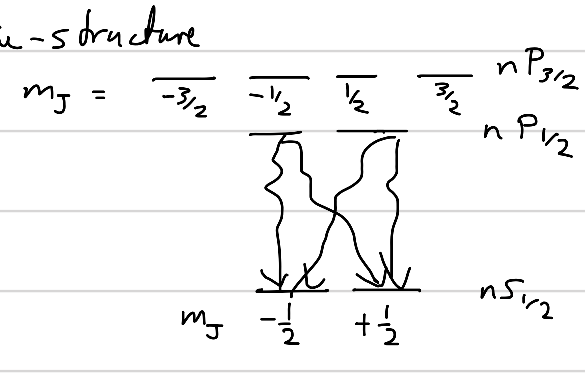
Quantization volume

Note: There is a subtle question of degeneracy of atomic sublevels and the polarization of the modes the couples $|g\rangle$ & $|e\rangle$ according to the dipole selection rules. Consider first the case of an atom with nondegenerate ground state ($l=0$, S-state), and excited ($l=1$, P state)



We can start the atom in any one of the excited m_l orbitals, and there is only one decay channel. Because the vacuum is isotropic, it cannot know in which direction the atom angular momentum is pointing. Thus the decay rate, Γ, must be independent of the excited m_l. For convenience, we will take $|e\rangle = |nP, m_l = 0\rangle$, $|g\rangle = |nS, m_l = 0\rangle$, so that $\langle e|\hat{\vec{d}}|g\rangle = d_{eg}\vec{e}_z$ ← Quantization axis.

If we include fine-structure



Decay channels for $nP_{1/2} \to nP_{3/2}$

The total decay rate from a level $|e, J m_J\rangle$ $\Gamma_{eJm_J} = \sum_{m_J'} \Gamma_{eJm_J \to gJ'm_J'}$. As before, Γ_{eJm_J} is independent of m_J (isotropic vacuum). If we are interested in a spontaneous transition from one specific sublevel to another we specify all the relevant quantum numbers.

E.g. $|e\rangle = |nP, J, m_J\rangle$, $|g\rangle = |nS, J', m_J'\rangle$

To solve for the time evolution, it is convenient to work in the interaction picture:

Joint A/F state evolution $\Rightarrow \frac{\hbar}{-i}\frac{\partial}{\partial t}|\Psi^{(I)}_{AF}\rangle = \hat{H}^{(I)}_{AF}(t)\,|\Psi_{AF}(t)\rangle$

$$\hat{H}^{(I)}_{AF}(t) = \hat{U}^{\dagger}_{Free}(t)\,\hat{H}_{AF}\,\hat{U}_{Free}(t) = \sum_{\vec{k},\mu} \hbar\left(g_{\vec{k}\mu}\,\hat{a}_{\vec{k},\mu}\,\sigma_{+}\,e^{-i(\omega_k-\omega_{eg})t} + g^{*}_{\vec{k}\mu}\,\hat{a}^{\dagger}_{\vec{k}\mu}\,\hat{\sigma}_{-}\,e^{+i(\omega_k-\omega_{eg})t}\right)$$

We solve this by decomposition in a basis: $\{|g\rangle\otimes|n_{\vec{k},\lambda}\rangle,\ |e\rangle\otimes|n_{\vec{k}\lambda}\rangle\}$

At $t=0: |\Psi_{AF}(0)\rangle = |e\rangle\otimes|0\rangle$

<u>Note</u>: We know from our study of the Jaynes-Cummings model, the total excitation in the atom + field system is conserved. Since we start in the excited state with field in vacuum, we have 1 excitation $\Rightarrow$ Dynamics are restricted to the subspace spanned by

$$\{\underset{\text{atom excited}}{|e\rangle}\otimes\underset{\text{vacuum}}{|0\rangle},\ \underset{\text{ground atom}}{|g\rangle}\otimes\underset{\text{one photon in mode }\vec{k},\mu}{|1_{\vec{k}\mu}\rangle}\}$$

We take as our Ansatz: $|\Psi^{(I)}_{AF}(t)\rangle = c_{e,0}(t)\,|e\rangle\otimes|0\rangle + \sum_{\vec{k},\mu} c_{g\,1_{\vec{k}\mu}}(t)\,|g\rangle\otimes|1_{\vec{k}\mu}\rangle$

The equations of motion for the probability amplitudes:

Infinite set of coupled ODEs
$$\begin{cases} \dot{c}_{e,0} = -\frac{i}{\hbar}\left(\langle e|\otimes\langle 0|\right)\hat{H}^{(I)}_{AF}(t)\,|\Psi^{(I)}_{AF}(t)\rangle = -i\sum_{\vec{k},\mu} g_{\vec{k}\mu}\,e^{-i(\omega_k-\omega_{eg})t}\,c_{g\,1_{\vec{k}\lambda}} \\ \dot{c}_{g,1_{\vec{k}\mu}} = -\frac{i}{\hbar}\left(\langle g|\otimes\langle 1_{\vec{k}\mu}|\right)\hat{H}^{(I)}_{AF}\,|\Psi^{(I)}_{AF}(t)\rangle = -i\,g^{*}_{\vec{k}\mu}\,e^{+i(\omega_k-\omega_{eg})t}\,c_{e,0} \end{cases}$$

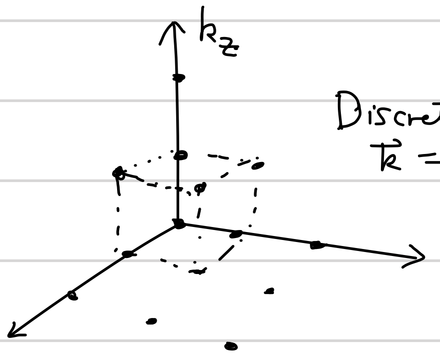
Formally integrating: $c_{g\,1_{\vec{k}\mu}}(t) = -i\,g^{*}_{\vec{k}\mu}\int_0^t dt'\,e^{i(\omega_k-\omega_{eg})t'}\,c_{e,0}(t')$

$\Rightarrow$ Formal integro-differential equation solution:

$$\boxed{\dot{c}_{e,0} = -\sum_{\vec{k},\mu}|g_{\vec{k}\mu}|^2\int_0^t dt'\,e^{-i(\omega_k-\omega_{eg})(t-t')}\,c_{e,0}(t')}$$

- <u>Perturbation theory</u>: $c_{e,0}(0)=1 \Rightarrow c_{e,0}(t)\simeq 1$ in integral $\Rightarrow$ Fermi's Golden Rule
- <u>Wigner-Weisskopf approximation</u>: $c_{e,0}(t)$ slowly varying $\Leftrightarrow$ Born-Markoff Approximation

We have an infinite, but countable set of coupled ODEs because we have defined the modes in terms a finite quantization volume. Our goal here is to describe the evolution in free space $\Rightarrow$ taking the quantization boundaries to infinity. In that situation, the sum over a discrete set of modes becomes an integral over a continuum of modes. We can count the number of modes using the concept of the "density of states": $\mathcal{D}(\vec{k})\, d^3k$ = # of modes in a volume d^3k of $\vec{k}$-space



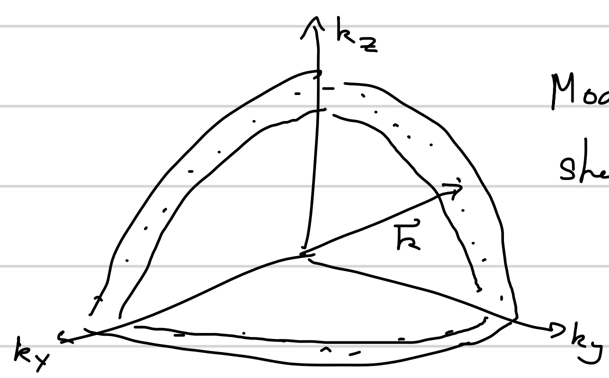
Discrete modes
$\vec{k} = \left(n_x \frac{2\pi}{L_x},\ n_y \frac{2\pi}{L_y},\ n_z \frac{2\pi}{L_z}\right)$

$\Rightarrow$ Density of states
$$\mathcal{D}(\vec{k}) = \frac{1 \text{ mode of given } \vec{k}}{\left(\frac{2\pi}{L_x}\right)\left(\frac{2\pi}{L_y}\right)\left(\frac{2\pi}{L_z}\right)}$$

$$\Rightarrow \mathcal{D}(\vec{k}) = \frac{V}{(2\pi)^3}$$

$$\sum_{\vec{k}} = \int d^3\vec{k}\, \mathcal{D}(\vec{k}) = \int \underset{\text{solid angle in } \vec{k}\text{-space}}{k^2 d\Omega_{\vec{k}}}\, dk\, \mathcal{D}(\vec{k}) = \int d\Omega_k\, d\omega_k \underbrace{\frac{\omega_k^2}{c^3}\mathcal{D}(\vec{k})}_{\mathcal{D}(\omega_k)}$$

$$\Rightarrow \mathcal{D}(\omega_k) = \frac{V}{(2\pi)^3}\frac{\omega_k^2}{c^3} = \text{Density of modes in frequency}$$

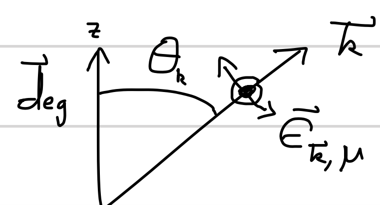


Mode density in a spherical shell in k-space: ω_k^2 dependence from 3D space

Thus, in the continuum limit,

$$\dot{c}_{e,0} = -\int_0^\infty d\omega_k\, \mathcal{D}(\omega_k)\left(\int d\Omega_k \sum_\mu |g_\mu(\vec{k})|^2\right)\int_0^t dt'\, e^{-i(\omega_k-\omega_{eg})(t-t')}\, c_{e0}(t')$$

Aside: $\int d\Omega_k \sum_\mu |g_\mu(\vec{k})|^2 = \left(\frac{1}{\hbar}\sqrt{\frac{2\pi\hbar\omega_k}{V}}\right)^2 \int d\Omega_k \sum_\mu |\vec{d}_{eg}\cdot\vec{\epsilon}_{\vec{k}\mu}|^2$



$$\Rightarrow \sum_\mu |\vec{d}_{eg}\cdot\vec{\epsilon}_{\vec{k}\mu}|^2 = \sin^2\theta_k\, |\vec{d}_{eg}|^2 \quad \text{(dipole emission pattern)}$$

$$\Rightarrow \int d\Omega_k \sum_\mu |\vec{d}_{eg}\cdot\vec{\epsilon}_{\vec{k},\mu}|^2 = \int \underbrace{2\pi\, d(\cos\theta_k)}_{d\Omega_k} \sin^2\theta_k |\vec{d}_{eg}|^2 = \frac{8\pi}{3}|\vec{d}_{eg}|^2$$

$$\Rightarrow \int d\Omega_k \sum_\mu |g_\mu(\vec{k})|^2 = \frac{16\pi^2\omega_k}{3\hbar V}|\vec{d}_{eg}|^2 \equiv \overline{g^2(\omega_k)}$$

Wigner - Weisskopf (Born-Markoff) Approximation

$$\dot{c}_{e,0} = -\int_0^\infty d\omega_k\, \overline{g^2(\omega_k)}\,\mathcal{D}(\omega_k) \int_0^t dt'\, e^{-i(\omega_k-\omega_{eg})(t-t')}\, c_{e,0}(t')$$

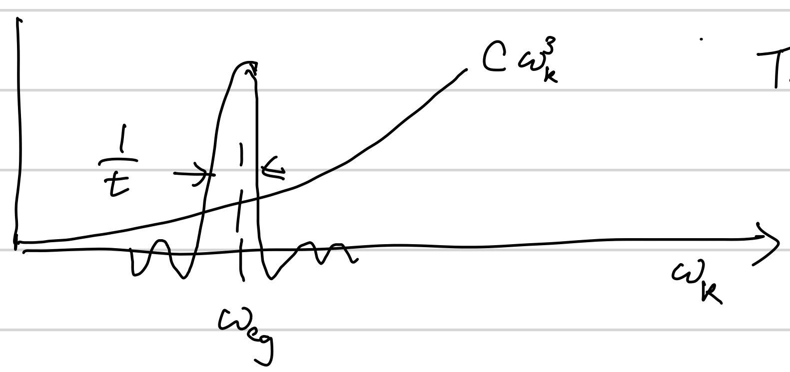
So far our calculation has been exact, up to the dipole + rotation wave approximation. We now make more radical approximations, originally due to Wigner & Weisskopf. This is based a two approximations: (1) The atom-light coupling is weak (i.e. a perturbation), so $c_{e,0}(t)$ changes much more slowly than ω_{eg} (in the interaction picture). (2) the "spectrum" $\overline{g^2(\omega_k)}\,\mathcal{D}(\omega_k)$ is very broad, and doesn't change much over frequencies that contribute to the integral.

Because the broad, the factor $e^{-i(\omega_k-\omega_{eg})(t-t')}$ will oscillate very fast for all times $t'<t$. Thus because $c_e(t')$ varies slowly compared to these oscillations, the value of the integral will be dominated by the end-point, where $t=t'$. Thus we can set $c_e(t') \Rightarrow c_e(t)$ and take it out of the integral. We recognize this as the Markov approximation, in that the evolution of $c_{e0}(t)$ depends only on the value of c_{e0} @ time t, and not the whole history. This also relied on weak coupling, and so in the theory of open quantum systems this is often called the "Born-Markov approximation," where the Born approximation is taken from perturbation theory of scattering.

$$\Rightarrow \dot{c}_{e,0} \approx -\left[\int_0^\infty d\omega_k\, \overline{g^2(\omega_k)}\,\mathcal{D}(\omega_k) \underbrace{\int_0^t dt'\, e^{-i(\omega_k-\omega_{eg})(t-t')}}_{\frac{1-e^{-i(\omega_k-\omega_{eg})t}}{-i(\omega_k-\omega_{eg})} = e^{-i\frac{(\omega_k-\omega_{eg})t}{2}}\,\mathrm{sinc}\left(\left[\frac{\omega_k-\omega_{eg}}{2}\right]t\right)}\right] c_{e,0}(t)$$

Here $\mathrm{sinc}\,x = \frac{\sin x}{x}$

We can sketch the integrand schematically



The width $\frac{1}{t} \ll$ the bandwidth of $\bar{g}^2(\omega_k)\,\mathcal{D}(\omega_k) \Rightarrow$ sinc function looks like a "delta function"

Thus, the next approximation is to take $t \to \infty$ in the limit of the integral. This is again justified by the assumption that spectrum is broad-band, and so the integral will vanish for $t >> t'$. Thus $\int_0^t dt' \, e^{-i(\omega_k - \omega_{eg})(t-t')} C_{e,0}(t') \Rightarrow \int_0^\infty d\tau \, e^{-i(\omega_k-\omega_{eg})\tau} C_{eg}(t)$

$$\dot{C}_{e,0} \approx -\left[\int_0^\infty d\omega_k \, \overline{g^2(\omega_k)} \mathcal{D}(\omega_k) \underbrace{\int_0^\infty d\tau \, e^{-i(\omega_k-\omega_{eg})\tau}}_{\zeta(\omega_k - \omega_{eg})}\right] C_{e,0}(t)$$

Said another way,

$$\dot{C}_{e,0} = -\int_0^t dt' \underbrace{\left[\int_0^\infty d\omega_k \, \overline{g^2(\omega_k)} \, \mathcal{D}(\omega_k) \, e^{-i(\omega_k-\omega_{eg})(t-t')}\right]}_{K(t-t')} C_{e,0}(t')$$

The function $K(t-t')$ is sometimes known as the "memory kernel" since it determines how the change in $C_{e,0}(t)$ depends on the whole history from $0 \to t$. It represents the Fourier transform of the ω_k-dependent coupling constant, weighted by the density of states. Because this is very broadband $K(t-t') \sim \delta(t-t') \Rightarrow$ Correlation time of the vacuum with the atom is very small. This is the Markov approximation – The atom-vacuum correlations lose "memory".

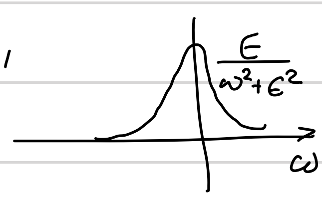
With the Wigner-Weisskopf approximation in hand, we can evaluate the expression

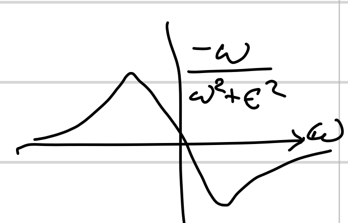
$$\dot{C}_{e,0} \approx -\left[\int_0^\infty d\omega \, \overline{g^2(\omega_k)} \, \mathcal{D}(\omega_k) \, \zeta(\omega_k - \omega_{eg})\right] C_{e0}(t)$$

Where $\zeta(\omega) \equiv \int_0^\infty e^{-i\omega\tau} d\tau$ = Fourier transform of a step-function

This is not a convergent function: Regularize: $\zeta(\omega) = \lim_{\epsilon \to 0^+} \int_0^\infty d\tau \, e^{-i\omega\tau - \epsilon\tau} = \lim_{\epsilon \to 0^+} \frac{1}{i\omega + \epsilon}$

$$\Rightarrow \zeta(\omega) = \lim_{\epsilon \to 0}\left[\frac{\epsilon}{\omega^2+\epsilon^2} - i\frac{\omega}{\omega^2+\epsilon^2}\right] \text{ "Complex Lorentzian"}$$





$$= \pi\delta(\omega) - i\mathcal{P}\left(\frac{1}{\omega}\right) \quad \text{Cauchy's Principle Part}$$

$$\mathcal{P}\left[\int_{-a}^{b} \frac{f(x)}{x} dx\right] = \lim_{\epsilon \to 0^+}\left[\int_{-a}^{-\epsilon} \frac{f(x)}{x} + \int_{\epsilon}^{b} \frac{f(x)}{x}\right] dx$$

$\Rightarrow$ In the Wigner–Weisskopf (Markoff approximation)

$$\dot{c}_{e,0} = -\left[\int d\omega_k \, \overline{g^2(\omega_k)} \, \mathcal{D}(\omega_k) \left(\pi \delta(\omega_k - \omega_{eg}) - i \mathbb{P} \frac{1}{\omega_k - \omega_{eg}} \right) \right] c_{e0}$$

$$\boxed{\begin{aligned} &\Rightarrow \dot{c}_{e0} = \left(i\delta - \frac{\Gamma}{2}\right) c_{e0} \Rightarrow c_{e0}(t) = e^{-\frac{\Gamma}{2}t} e^{-i\delta t} c_{e0}(0) \\ &|c_{e,0}(t)|^2 = e^{-\Gamma t} |c_{e,0}(0)|^2 : \text{ Exponential Decay! Spontaneous emission!} \\ &\Gamma = 2\pi \, \overline{g^2(\omega_{eg})} \, \mathcal{D}(\omega_{eg}) = \frac{4}{3} |d_{eg}|^2 \frac{\omega^3}{\hbar c^3} \quad \text{(Fermi's Golden Rule)} \\ &\text{Einstein-A coefficient.} \\ &\delta = \mathbb{P} \int_0^\infty d\omega_k \, \mathcal{D}(\omega_k) \frac{\overline{|g^2(\omega_k)|}}{\omega_{eg} - \omega_k} : \quad \text{Contribution to the Lamb-Shift from Near resonant modes in the RWA} \end{aligned}}$$

Note: $\dot{c}_{e0} = \left(i\delta - \frac{\Gamma}{2}\right) c_{e0}$ is irreversible.

How did we start with time-reversible Schrödinger evolution and end up with time-irreversible behavior? The "slide of hand" occurred when we made the Markoff approximation. By throwing away the atoms memory of its dynamical history, we throw away reversible behavior. We will return to study this in much more detail later when we study open quantum systems in more detail.

Given the solution $c_{e,0}(t) = c_{e,0} e^{-\Gamma t - i\delta t}$ (in the interaction picture), we can find $c_{g,1_k}(t)$.

- Firstly, we absorb δ in ω_{eg} to get a renormalized energy, $\omega_{eg} \rightarrow \frac{E_e - E_g + \delta}{\hbar} = \tilde{\omega}_{eg}$
- Next, we look at the solution when the atom is initially in the excited state, $c_{e,0}(0) = 1$

$$\dot{c}_{g,1_{k\mu}} = -i g^*_{k\mu} e^{i(\omega_k - \omega_{eg})t} c_{e0}(t) \Rightarrow c_{g,1_{k\mu}} = -i g^*_{k\mu} \int_0^t e^{i(\omega_k - \omega_{eg})t'} c_{e0}(t') = \frac{-i}{\hbar} \int_0^t e^{i(\omega_k - \tilde{\omega}_{eg} + i\frac{\Gamma}{2})t'}$$

$$\Rightarrow c_{g,1_{k,\lambda}}(t) \cong -i g^*_{k,\mu} \frac{e^{i(\omega_k - \tilde{\omega}_{eg})t} e^{-\frac{\Gamma}{2}t} - 1}{i(\omega_k - \tilde{\omega}_{eg}) - \frac{\Gamma}{2}}$$

Thus we have the (irreversible) evolution of the pure state of the system: atom + field

$$|\Psi_{AF}(t)\rangle = e^{-\frac{\Gamma}{2}t}|e\rangle\otimes|0\rangle + \sum_{\vec{k},\mu}{}' g^*_{\vec{k},\mu}\frac{-e^{i(\omega_k-\omega_{eg})t}e^{-\frac{\Gamma}{2}t}+1}{+(\omega_k-\omega_{eg})+i\frac{\Gamma}{2}}|g\rangle\otimes|1_{\vec{k}\mu}\rangle \quad \text{(unnormalized)}$$

In the long-time limit $t \gg \frac{1}{\Gamma}$

$$|\Psi_{AF}\rangle \to |g\rangle\otimes|\phi_{photon}(t)\rangle, \quad \text{where } |\phi_{photon}(t)\rangle = \sum_{\vec{k},\mu}{}' g^*_{\vec{k},\mu}\frac{1}{(\omega_k-\omega_{eg})+i\frac{\Gamma}{2}}|1_{\vec{k},\mu}\rangle$$

$|\phi_{photon}(t)\rangle$ is a 1-photon wave packet that is spontaneously emitted by the atom:

The momentum distribution $\tilde{\phi}_{photon}(\vec{k},t) = \dfrac{g^*_{\vec{k},\mu}}{(c|\vec{k}|-\omega_{eg})+i\frac{\Gamma}{2}} = \sqrt{\dfrac{2\pi\hbar c|\vec{k}|}{V}}\,\vec{\epsilon}^{\,*}_{\vec{k},\mu}\cdot\vec{d}_{eg}\,\dfrac{e^{-i\vec{k}\cdot\vec{R}}}{(\omega_k-\omega_{eg})+i\frac{\Gamma}{2}}$

We can look at the positive frequency component of the electric field associated with this wave packet that acts as a kind of "wavefunction" of the photon: $\hat{a}_{\vec{k}\mu} \to \tilde{\phi}_{\vec{k},\mu}$

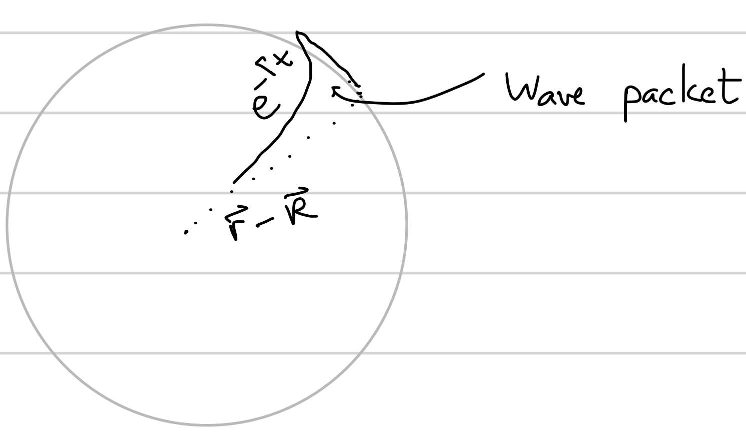
$$\vec{\mathcal{E}}_{photon}(\vec{r},t) = \sum_{\vec{k},\mu}{}'\sqrt{\frac{2\pi\hbar\omega_k}{V}}\,\vec{\epsilon}_{\vec{k},\mu}\,\tilde{\phi}_{\vec{k},\mu}\,e^{i(\vec{k}\cdot\vec{r}-\omega_k t)} = \sum_{\vec{k},\mu}{}'\frac{2\pi\hbar\omega_k}{V}\,\vec{\epsilon}_{\vec{k}\mu}\,\vec{\epsilon}^{\,*}_{\vec{k},\mu}\cdot\vec{d}_{eg}\,\frac{e^{i\vec{k}\cdot(\vec{r}-\vec{R})-\omega_k t}}{(\omega_k-\omega_{eg})+i\frac{\Gamma}{2}}$$

Going to the continuum limit an making the Wigner – Weisskopf approximation:

$$|\vec{\mathcal{E}}_{photon}(\vec{r},t)| = \frac{A\,e^{(-i\omega_{eg}-\frac{\Gamma}{2})(t-\frac{|\vec{r}-\vec{R}|}{c})}}{|\vec{r}-\vec{R}|}\;\underbrace{\mathbb{1}\left(t-\frac{|\vec{r}-\vec{R}|}{c}\right)}_{\text{step function}}$$

$$A = -\sin\theta\left(\frac{\omega_{eg}}{c}\right)^2|\vec{d}_{eg}|$$

Spherical wave (with dipole radiation pattern) with finite wave train:



Physical picture of spontaneous emission

From the point of view of Fermi's Golden rule, spontaneous emissions appears to be caused purely by vacuum fluctuations. Indeed, often one speaks of spontaneous emission as emission "stimulated by a vacuum photon". This picture is evolved from a rate equation picture, but does it capture all of the physics? When studying the classical Lorentz oscillator theory, we found that dipole oscillation decays due to "radiation reaction." What role does the self-field of the radiating atom play in quantum mechanics? The address this, it is convenient to work in the Heisenberg picture. Recall the Hamiltonian

$$\hat{H} = \frac{\hbar\omega_{eg}}{2}\hat{\sigma}_z + \sum_{\vec{k},\mu}\hbar\omega_k \hat{a}^\dagger_{\vec{k}\mu}\hat{a}_{\vec{k}\mu} + \sum \hbar\left(g_{\vec{k}\mu}\hat{\sigma}_+\hat{a}_{\vec{k}\mu} + g^*_{\vec{k}\mu}\hat{a}^\dagger_{\vec{k}\mu}\hat{\sigma}_-\right)$$

Note: The Heisenberg picture preserved equal-time commutators. Thus, for example

$$[\hat{\sigma}_+(t), \hat{a}_{\vec{k},\mu}(t)] = [\hat{\sigma}_+(0), a_{\vec{k}\mu}(0)] = 0,$$

so, as long as operators are taken at the same time, atomic and field operator commute. When at different times, we must be more careful, as $\hat{\sigma}_+(t')$ may depend on $\hat{a}^\dagger_{\vec{k}\mu}(t)$, for example, if which case $[\hat{\sigma}_+(t'), \hat{a}_{\vec{k},\mu}(t)] \neq 0$. The Hamiltonian written above is expressed in "normal order." That is all the $\hat{a}$'s to the right and $\hat{a}^\dagger$'s to the left. Because all the operators in $\hat{H}$ are at equal time, we can order at will.

Now consider the Heisenberg equations of motion:

$$\frac{d}{dt}\hat{a}_{\vec{k},\mu} = \frac{-i}{\hbar}[\hat{a}_{k,\mu}, \hat{H}] = -i\omega_k\hat{a}_{\vec{k},\mu} - ig^*_{\vec{k},\mu}\hat{\sigma}_-$$

$$\frac{d}{dt}\hat{\sigma}_+ = \frac{-i}{\hbar}[\hat{\sigma}_+, \hat{H}] = i\omega_{eg}\hat{\sigma}_+ - i\sum_{\vec{k}\mu} g^*_{\vec{k}\mu}\hat{a}^\dagger_{\vec{k}\mu}\hat{\sigma}_z$$

$$\frac{d}{dt}\hat{\sigma}_z = \frac{-i}{\hbar}[\hat{\sigma}_z, \hat{H}] = -2i\sum_{\vec{k},\mu}\left(g_{\vec{k}\mu}\hat{\sigma}_+\hat{a}_{\vec{k}\mu} - g_{\vec{k}\mu}\hat{a}^\dagger_{\vec{k}\mu}\hat{\sigma}_-\right)$$

We can formally integrate these equations. Consider first, $\hat{a}_{\vec{k},\lambda}(t)$

$$\hat{a}_{\vec{k},\lambda}(t) = \underbrace{\hat{a}_{\vec{k},\lambda}(0)\,e^{-i\omega_k t}}_{\text{"Vacuum field"}} \underbrace{- i g^*_{\vec{k},\mu}\int_0^t dt'\,e^{-i\omega_k(t-t')}\,\hat{\sigma}_-(t')}_{\text{"Source field"}}$$

We have interpreted the first term as the "vacuum field", i.e. the contribution to $\hat{a}(t)$ arising from vacuum fluctuations, that are present even in the absence of the atom. In contrast, the second term is the "source field", i.e. the contribution to $\hat{a}(t)$ arising from the atom. In fact, resumming over the modes, the total field

$$\hat{\vec{E}}^{(+)}(\vec{r},t) = \vec{E}^{(+)}_{vac}(\vec{r},t) + \vec{E}^{(+)}_{source}(\vec{r},t)$$

where $\vec{E}^{(+)}_{vac}(\vec{r},t) = \sum_{\vec{k},\mu} \sqrt{\frac{2\pi\hbar\omega_k}{V}}\, \vec{\epsilon}_{\vec{k},\mu}\, e^{i(\vec{k}\cdot\vec{r}-\omega_k t)}\, \hat{a}_{\vec{k},\mu}$: Vacuum field

$$\vec{E}^{(+)}_{source}(\vec{r},t) = -\frac{\omega_{eg}^2}{c^2}(\vec{d}_{eg}\times\vec{e}_r)\times\vec{e}_r\, \frac{\hat{\sigma}_-\left(t-\frac{|\vec{r}-\vec{R}|}{c}\right)}{|\vec{r}-\vec{R}|}$$: Dipole radiated field

We seek to explore the relative contributions of $\hat{a}_{vac}(t)$ and $\hat{a}_{source}(t)$ to spontaneous emission and level shifts. To lowest order in perturbation theory:

$$\hat{a}(t) = \underbrace{e^{-i\omega_k t}\hat{a}(0)}_{vacuum} + \underbrace{\delta\hat{a}(t)}_{source}, \quad \hat{\sigma}_+(t) = e^{i\omega_{eg}t}\hat{\sigma}_+(0) + \delta\hat{\sigma}_+(t), \quad \hat{\sigma}_z(t) = \hat{\sigma}_z(0) + \delta\hat{\sigma}_z(t)$$

Thus, to lowest order: $\frac{d}{dt}\hat{\sigma}_+ = i\omega_{eg}\hat{\sigma}_+ - i\sum_{\vec{k},\mu} g^*_{\vec{k},\mu}\left[\hat{a}^\dagger_{\vec{k}\mu}(0)e^{-i\omega_k t}\,\delta\hat{\sigma}_z(t) + \delta\hat{a}^\dagger_z(t)\,\hat{\sigma}_z(0)\right]$

Now take the expected value for the field in the vacuum, and the atom in arbitrary state $|\psi\rangle$.

Note: Because we have chosen normal order $\langle 0|\hat{a}^\dagger(0) = 0$ $\Rightarrow$ the vacuum field has no contribution to the evolution of $\langle\hat{\sigma}_+\rangle$ in normal order.

$$\Rightarrow \frac{d}{dt}\langle\hat{\sigma}_+\rangle = i\omega_{eg}\langle\hat{\sigma}_+\rangle - i\sum_{\vec{k},\mu}\langle\delta\hat{a}^\dagger_z(t)\hat{\sigma}_z(0)\rangle$$

Now, $\delta\hat{a}(t) = -i g^*_{\vec{k}\mu}\int_0^t dt' e^{-i\omega_k(t-t')}\hat{\sigma}_-(t') \approx -i g^*_{\vec{k}\mu} e^{-i\omega_k t}\int_0^t dt' e^{i(\omega_k-\omega_{eg})t'}\hat{\sigma}_-(0)$

$$\Rightarrow \frac{d}{dt}\langle\hat{\sigma}_+\rangle = i\omega_{eg}\langle\hat{\sigma}_+\rangle + \sum_{\vec{k}\mu}|g_{\vec{k}\mu}|^2 e^{-i\omega_k t}\int_0^t dt'\, e^{i(\omega_k-\omega_{eg})t'}\underbrace{\langle\hat{\sigma}_+(0)\hat{\sigma}_z(0)\rangle}_{=-\langle\hat{\sigma}_+(0)\rangle = -e^{-i\omega_{eg}t}\langle\hat{\sigma}_+(t)\rangle}$$

$$\Rightarrow \frac{d}{dt}\langle\hat{\sigma}_+\rangle = i\omega_{eg}\langle\hat{\sigma}_+\rangle - \underbrace{\int_0^{t'} dt\left(\sum_{\vec{k}\mu}|g_{\vec{k},\mu}|^2 e^{i\omega_k(t-t')}\right)e^{i\omega_{eg}(t-t')}}_{=\left(\frac{\Gamma}{2}+i\delta\right)\text{ in Born-Markoff approximation}}\langle\hat{\sigma}_+\rangle$$

$$\Rightarrow \boxed{\frac{d}{dt}\langle\hat{\sigma}_+\rangle = \left(i(\omega_{eg}+\delta) - \frac{\Gamma}{2}\right)\langle\hat{\sigma}_+\rangle \Rightarrow \text{Damped, oscillating coherence ?}}$$

Similarly, $\frac{d}{dt}\langle\hat{\sigma}_z\rangle = -2i\sum_{\vec{k},\mu}\left[g_{\vec{k}\mu}\langle\hat{\sigma}_+(0)\,\delta\hat{a}_{\vec{k}\mu}(t)\rangle e^{i\omega_{eg}t} + h.c.\right)$

$$= \underbrace{-2\int_0^t dt' \sum_{\vec{k}\mu}|g_{\vec{k}\mu}|^2 e^{-i\omega_k(t-t')}\; e^{i\omega_{eg}(t-t')}}_{\frac{\Gamma}{2}+i\delta}\;\underbrace{\langle\hat{\sigma}_+(0)\hat{\sigma}_-(0)\rangle}_{|e\rangle\langle e| \,=\, \frac{\mathbb{1}+\hat{\sigma}_z(0)}{2}} + h.c.,$$

$$\Rightarrow \boxed{\frac{d}{dt}\langle\hat{\sigma}_z\rangle = -\Gamma\left(\langle\hat{\sigma}_z\rangle+1\right), \quad \text{Damped Population}}$$

We see here that, using normal order, we can attribute the spontaneous decay and level shift completely to radiation reaction, with the vacuum field playing no role. In this picture, if we start in $|e\rangle$, the role of the vacuum fluctuations is to cause a random "tipping of the Bloch vector", giving the state ρ_{eg} coherence, which then oscillates and radiates, leading to decay. We should be cautious about this interpretation, for had we chosen anti-normal order, vacuum fluctuations completely determine the level shift and contribute to Γ. Moreover, it has been shown by Dalibard, Dupont-Roc, and Cohen-Tannoudji (J. Physique 43 1617 (1982)), that only in symmetric order can one associate physical meaning to the relative contribution of vacuum field & radiation reaction to level shifts & decay.

Nonetheless, a lesson to be drawn from the Heisenberg analysis is that the standard picture from Fermi's Golden Rule, typically used in laser physics, of vacuum photons "stimulating emission" misses some important physics. Spontaneous emission can largely be understood as "radiative decay" as we studied in the classical Lorentz model. Moreover, as we will see, when we drive the atom so that it scatters photons (resonance fluorescence), the relative role of radiation reaction and vacuum fluctuations becomes more apparent, and the classical picture of a radiating (decaying) antenna should be kept in mind.