Appendix B

Spontaneous Emission in the Weak Coupling Regime

In this Appendix we derive the normalized spontaneous decay rate of an atomic system using quantum electrodynamics. The analysis is based in part on Ref. [1]. In what follows, we concentrate exclusively on the weak coupling regime. Section B.1 presents the derivation of the decay constant in free space using QED and the Weisskopf-Wigner approximation [2, 3]. Section B.2 is devoted to calculate the spontaneous emission decay constant in a linear and inhomogeneous medium using the Heisenberg picture [1] which renders a clear connection between classical theory and QED.

B.1 Weisskopf-Wigner theory

According to QED, the spontaneous emission of an atom in free space is due to vacuum fluctuations. We consider a two level atom interacting with an infinite number of field modes. Each mode is characterized by its wavevector $\mathbf{k}$. This atom-field system is described by the Jaynes-Cummings Hamiltonian [4]

$$\hat{H} \;=\; \hbar\omega_o|e\rangle\langle e| \;+\; \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}}\, \hat{a}^{\dagger}_{\mathbf{k}}\hat{a}_{\mathbf{k}} \;-\; \sum_{\mathbf{k}} \hbar g_{\mathbf{k}} \left[\hat{a}_{\mathbf{k}}|e\rangle\langle g| \;+\; \hat{a}^{\dagger}_{\mathbf{k}}|g\rangle\langle e|\right]. \tag{B.1}$$

Here $|e\rangle$ ($|g\rangle$) is the excited (ground) state of the atom, $\hat{a}_{\mathbf{k}}$ and $\hat{a}^{\dagger}_{\mathbf{k}}$ are the annihilation and creation operators for the mode $\mathbf{k}$,* and $g_{\mathbf{k}}$ is the atom-field coupling strength defined as

$$g_{\mathbf{k}} = \sqrt{\frac{\omega_{\mathbf{k}}}{2\varepsilon_o\hbar V}}\; \hat{\boldsymbol{\epsilon}}_{\mathbf{k}} \cdot \langle g|\hat{\boldsymbol{\mu}}|e\rangle, \tag{B.2}$$

*We use the compressed notation for which $\mathbf{k}$ designates simultaneously the $\mathbf{k}$-vector and the polarization state. Each $\mathbf{k}$-vector possesses two linearly independent polarization states.

where V is the volume, $\hat{\boldsymbol{\epsilon}}$ is the unit vector in the direction of the electric field mode $\mathbf{E}_{\mathbf{k}}$, and $\hat{\boldsymbol{\mu}}$ is the dipole moment operator.

We assume that at $t = 0$ the atom is in the excited state and no photons are present. The initial state is therefore $|e, 0\rangle$, e and 0 designating the excited atomic state and the initial photon number, respectively. At any later time t the wavefunction $|\psi(t)\rangle$ of the system can be expanded as

$$|\psi(t)\rangle \;=\; C_0^e(t)\,\mathrm{e}^{-i\omega_o t}|e,0\rangle \;+\; \sum_{\mathbf{k}} C_{1\mathbf{k}}^g(t)\,\mathrm{e}^{-i\omega_k t}|g,1_{\mathbf{k}}\rangle, \tag{B.3}$$

where the C's are time-dependent expansion coefficients. In the state $|g, 1_{\mathbf{k}}\rangle$ the atom is in the ground state and one photon of mode $\mathbf{k}$ is released. By inserting Eq. (B.3) into the Schrödinger equation, we obtain:

$$\frac{dC_0^e}{dt} = -\sum_{\mathbf{k}} |g_{\mathbf{k}}|^2 \int_o^t C_0^e(t_1)\;\mathrm{e}^{-i(\omega_k-\omega_o)(t-t_1)}\;dt_1. \tag{B.4}$$

In the large volume limit, that is, $V \to \infty$, the sum in Eq. (B.4) can be substituted as

$$\sum_{\mathbf{k}} \longrightarrow 2\,\frac{V}{(2\pi)^3}\int_0^{2\pi} d\phi \int_0^{\pi} d\theta \sin\theta \int_0^{\infty} dk\, k^2\;, \tag{B.5}$$

where the factor of 2 arises from summing over the two polarization states associated with each $\mathbf{k}$-vector. Assuming that the dipole is oriented along the z axis, i.e. $\boldsymbol{\mu} = \langle g|\hat{\boldsymbol{\mu}}|e\rangle = \mu\,\hat{\mathbf{n}}_z$, the field-atom coupling strength becomes

$$|g_{\mathbf{k}}|^2 = \frac{\omega_{\mathbf{k}}}{2\varepsilon_o\hbar V}\,\mu^2\cos^2\theta. \tag{B.6}$$

After solving the angular integrals, Eq. (B.4) reduces to

$$\frac{dC_0^e}{dt} \;=\; -\frac{\mu^2}{6\pi^2\varepsilon_o\hbar c^3}\int_0^{\infty}\omega_k^3\int_0^t C_0^e(t_1)\,\mathrm{e}^{-i(\omega_k-\omega_o)(t-t_1)}\;dt_1\;d\omega_k\,. \tag{B.7}$$

So far, the derivation has been exact. We now introduce the Weisskopf-Wigner approximation to solve Eq. (B.7). This approximation involves the following two assumptions: (1) the spectrum of the field modes is very broad, and (2) the coefficient C_0^e changes slowly in time. Therefore, for times $t_1 \ll t$ the integrand oscillates very rapidly and there is no significant contribution to the value of the integral. The most dominant contribution originates from times $t_1 \approx t$. We therefore evaluate $C_0^e(t_1)$ at the actual time t and move it out of the integrand. In this limit, the atomic decay becomes a memoryless process (Markov process). To evaluate the remaining integral we extend the upper integration limit to infinity since there is no significant contribution for $t_1 \gg t$. Eq. (B.7) reduces now to

$$\frac{dC_0^e}{dt} \;=\; -\frac{\mu^2}{6\pi^2\varepsilon_o\hbar c^3}C_0^e(t)\int_0^{\infty}\omega_k^3\int_0^{\infty}\mathrm{e}^{-i(\omega_k-\omega_o)(t-t_1)}\;dt_1\;d\omega_k\,. \tag{B.8}$$

The integration can now be carried out analytically and we obtain

$$\frac{dC_0^e}{dt} \;=\; -\Big(\frac{\gamma_o}{2} + i\Delta\omega\Big)\, C_0^e(t)\,. \tag{B.9}$$

Here, γ_o is the free space decay constant

$$\gamma_o = \frac{\omega_o^3 \mu^2}{3\pi\varepsilon_o \hbar c^3} = \frac{\pi\omega_o \mu^2}{3\varepsilon_o \hbar}\rho(\omega_o), \tag{B.10}$$

with $\rho(\omega_o)$ being the electromagnetic density of modes. The second term in Eq. (B.9) is the Lamb shift and reads as

$$\Delta\omega = \frac{1}{4\pi\varepsilon_o}\frac{\mu^2}{3\pi\hbar c^3}\;\mathrm{P}\left\{\int \frac{\omega_{\mathrm{k}}^3}{\omega_{\mathrm{k}} - \omega_{\mathrm{o}}}\,\mathrm{d}\omega_{\mathrm{k}}\right\} \tag{B.11}$$

where P denotes the principal value of the integral. Since the integral diverges it is necessary to introduce a cut-off frequency w_f according to $\hbar\omega_f = 2m_e c^2$ (energy for 'pair' creation). With this correction, the Lamb shift $\Delta\omega$ turns out to be in the range of a few GHz which is very small compared to the optical transition frequency.

B.2 Inhomogeneous environments

We apply QED to derive the spontaneous decay rate of an atomic system in an inhomogeneous medium which is characterized by the lossless dielectric constant $\varepsilon(\mathbf{r})$.

Let us consider the vector potential operator $\hat{\mathbf{A}}(\mathbf{r},t)$ which satisfies the generalized Coulomb gauge $\nabla\cdot[\varepsilon(\mathbf{r})\,\hat{\mathbf{A}}] = 0$. The transverse vector potential can be expanded in a complete set of orthogonal modes $\mathbf{a_k}$ as [5]

$$\hat{\mathbf{A}}(\mathbf{r},t) \;=\; \hat{\mathbf{A}}^+(\mathbf{r},t) + \hat{\mathbf{A}}^-(\mathbf{r},t), \tag{B.12}$$

$$\hat{\mathbf{A}}^-(\mathbf{r},t) \;=\; \sum_{\mathbf{k}} \sqrt{\hbar/(2\varepsilon_o\omega_{\mathbf{k}}V)}\;\hat{a}_{\mathbf{k}}(t)\,\mathbf{a_k}(\mathbf{r})\,, \tag{B.13}$$

$$\hat{\mathbf{A}}^+(\mathbf{r},t) \;=\; \sum_{\mathbf{k}} \sqrt{\hbar/(2\varepsilon_o\omega_{\mathbf{k}}V)}\;\hat{a}_{\mathbf{k}}^\dagger(t)\,\mathbf{a_k^*}(\mathbf{r}). \tag{B.14}$$

Here, $\hat{\mathbf{A}}^-$ and $\hat{\mathbf{A}}^+$ contain only negative and positive frequency components, respectively. The normal modes satisfy the Helmholtz equation

$$\nabla\times\nabla\times\mathbf{a_k}(\mathbf{r}) \;+\; \varepsilon_o\varepsilon(\mathbf{r})\,\frac{\omega_{\mathbf{k}}^2}{c^2}\,\mathbf{a_k}(\mathbf{r}) = \mathbf{0}\,, \tag{B.15}$$

and they form an orthonormal and complete set, namely

$$\int \varepsilon(\mathbf{r})\;\mathbf{a_{k'}}(\mathbf{r})\cdot\mathbf{a_k^*}(\mathbf{r})\,d^3\mathbf{r} \;=\; \delta_{\mathbf{k}\mathbf{k'}}\,, \tag{B.16}$$

$$\int \mathbf{a}_{\mathbf{k}}^{*}(\mathbf{r}')\,\mathbf{a}_{\mathbf{k}}(\mathbf{r})\,d^3\mathbf{k} \;=\; \overleftrightarrow{\delta}_{\perp}(\mathbf{r}'-\mathbf{r})\;. \tag{B.17}$$

We now express the interaction term in the Hamiltonian [c.f. Eq. (B.1)] in terms of the electron momentum operator $\hat{\mathbf{p}}$ and the vector potential operator $\hat{\mathbf{A}}$ and obtain

$$\hat{H}_{int} \;=\; -\hat{\mathbf{p}}\cdot\hat{\mathbf{A}} \;=\; \sum_{\mathbf{k}} \hbar\Big[\kappa_{\mathbf{k}}^{*}\,\hat{a}_{\mathbf{k}}^{\dagger}|g\rangle\langle e| \;+\; \kappa_{\mathbf{k}}\,\hat{a}_{\mathbf{k}}|e\rangle\langle g|\Big]\,, \tag{B.18}$$

where $\kappa_{\mathbf{k}}$ denotes the coupling constant defined as

$$\kappa_{\mathbf{k}} = -\frac{e}{\hbar m}\sqrt{\hbar/(2\varepsilon_o\omega_{\mathbf{k}}V)}\;\mathbf{p}_{12}\cdot\mathbf{a}_{\mathbf{k}}(\mathbf{r}_o), \tag{B.19}$$

and $\mathbf{p}_{12}$ is the matrix element $\langle g|\hat{\mathbf{p}}|e\rangle$.

In QED, spontaneous decay is generated by vacuum fluctuations of the field. These fluctuations give rise to a source current density whose operator is denoted as $\hat{\mathbf{J}}$. The frequency correlation of $\hat{\mathbf{J}}$ can be calculated as

$$\langle\hat{\mathbf{J}}_{\omega'}^{+}(\mathbf{r}')\,\hat{\mathbf{J}}_{\omega}^{-}(\mathbf{r})\rangle \;=\; \frac{e^2}{m^2}\mathbf{p}_{12}\,\mathbf{p}_{12}\;\delta(\omega-\omega')\,\delta(\omega-\omega_o)\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(\mathbf{r}-\mathbf{r}_o)\;\langle\hat{N}_e\rangle\;, \tag{B.20}$$

where $\mathbf{r}_o$ is the center of mass of the atom, ω_o the center frequency of the distribution, and $\hat{N}_e = |e\rangle\langle e|$ the number operator of the excited state. The number operator satisfies the equation

$$\frac{d\hat{N}_e}{dt} \;=\; \frac{i}{\hbar}\int\Big[\hat{\mathbf{J}}^{+}(\mathbf{r},t)\cdot\hat{\mathbf{A}}^{-}(\mathbf{r},t) \;-\; \hat{\mathbf{A}}^{+}(\mathbf{r},t)\cdot\hat{\mathbf{J}}^{-}(\mathbf{r},t)\Big]\;d^3\mathbf{r}\;, \tag{B.21}$$

which can be derived by using Heisenberg's equation of motion for the different operators.

Let us denote the Fourier transforms of $\hat{\mathbf{A}}(\mathbf{r},t)$ and $\hat{\mathbf{J}}(\mathbf{r},t)$ by $\hat{\mathbf{A}}_{\omega}(\mathbf{r})$ and $\hat{\mathbf{J}}_{\omega}(\mathbf{r})$, respectively. Then, as a consequence of Heisenberg's equations of motion and the restriction to the *weak-coupling* regime, we can derive the following *quantum* wave equation [1]

$$\nabla\times\nabla\times\hat{\mathbf{A}}_{\omega}^{-}(\mathbf{r}) \;-\; \varepsilon(\mathbf{r})\frac{\omega^2}{c^2}\hat{\mathbf{A}}_{\omega}^{-}(\mathbf{r}) = \frac{1}{\varepsilon_o c^2}\hat{\mathbf{J}}_{\omega}^{-}(\mathbf{r})\;. \tag{B.22}$$

Using the definition of the dyadic Green's function from Eq. (**??**) (c.f. Section **??**) the solution for $\hat{\mathbf{A}}_{\omega}^{-}$ can be represented as

$$\hat{\mathbf{A}}_{\omega}^{-}(\mathbf{r}) \;=\; \frac{1}{\varepsilon_o c^2}\int\overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega)\,\hat{\mathbf{J}}_{\omega}^{-}(\mathbf{r}')\,d^3\mathbf{r}', \tag{B.23}$$

where we included ω in the argument of $\overleftrightarrow{\mathbf{G}}$. By applying the inverse Fourier transform we can derive the corresponding solution $\hat{\mathbf{A}}^{-}(\mathbf{r},t)$ in time domain. Finally, combining this solution with Eq. (B.20) and Eq. (B.21) we obtain the simple equation

$$\frac{d\langle\hat{N}_e\rangle}{dt} \;=\; -\gamma\,\langle\hat{N}_e\rangle, \tag{B.24}$$

with γ being the spontaneous decay rate

$$\gamma = -\frac{2e^2}{\varepsilon_o \hbar c^2 m^2}\ \mathbf{p}_{12} \cdot \mathrm{Im}\left\{\overleftrightarrow{\mathbf{G}}(\mathbf{r}_\mathrm{o}, \mathbf{r}_\mathrm{o}; \omega_\mathrm{o})\right\} \cdot \mathbf{p}_{12}. \tag{B.25}$$

In the (generalized) Coulomb gauge the momentum matrix elements $\mathbf{p}_{12}$ are related to the dipole matrix elements $\boldsymbol{\mu}$ as

$$\mathbf{p}_{12} \ = \ (im\omega_o/e)\ \boldsymbol{\mu}\ , \tag{B.26}$$

which allows us to write Eq. (B.25) in terms of $\hat{\boldsymbol{\mu}}$. Furthermore, in an inhomogeneous medium, the Green's function can be split into a primary (free space) part $\overleftrightarrow{\mathbf{G}}_o$ and a scattering part $\overleftrightarrow{\mathbf{G}}_s$. Using the fact that the contribution of $\overleftrightarrow{\mathbf{G}}_o$ leads to the free space decay rate γ_o [c.f. Eq. (B.10)] we can write the ratio γ/γ_o as

$$\frac{\gamma}{\gamma_o} \ = \ 1 \ + \ \frac{6\pi c}{\omega_o \mu^2}\ \boldsymbol{\mu} \cdot \mathrm{Im}\left\{\overleftrightarrow{\mathbf{G}}_\mathrm{s}(\mathbf{r}_\mathrm{o}, \mathbf{r}_\mathrm{o}; \omega_\mathrm{o})\right\} \cdot \boldsymbol{\mu}\ , \tag{B.27}$$

consistent with the classical derivation [Eq. (**??**)] in Section **??**.

References

[1] Yong Xu, Reginald K. Lee, and Amnon Yariv, *Phys. Rev. A* **61**, 33807 (2000).

[2] V. Weisskopf and E. Wigner, *Z. Phys.* **63**, 54 (1930).

[3] Y. Yamamoto, and A. Imamoglu, *Mesoscopic Quantum Optics*, New York: John Wiley & Sons (1999).

[4] E. T. Jaynes and F. W. Cummings, *Proc. IEEE* **51**, 89 (1963).

[5] R. J. Glauber and M. Lewenstein, *Phys. Rev. A* **43**, 467 (1991).