

QUANTUM DESCRIPTION OF DECAY PHENOMENA BETWEEN MIRRORS

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We compare quantum-mechanically spontaneous emission and resonance transfer of excitation between mirrors. The comparison has the advantage of helping us to understand decay phenomena in microstructures better in terms of real and virtual photons.

INTRODUCTION

Decay phenomena are among the most basic problems in the quantum theory of radiation. For they not only are prototypes to show the perturbation method in solving quantum-transition problems, but also related to some fundamental questions in quantum electrodynamics (QED), such as the properties of the quantum vacuum¹ and causality².

The effects of electromagnetic boundary conditions on atomic decay properties have been extensively studied over the past decades, which are of interest both from the theoretical point of view and because of potential applications for techniques in the mesoscopic and nanoscopic regimes³⁻¹⁶. In this paper we discuss and compare quantum-mechanically spontaneous emission and resonance transfer of excitation in a microcavity. Such a comparison has the advantage, not only of helping us to understand the connection between these two processes better, but even more, of exhibiting very clearly different roles played by real and virtual photons in decay phenomena between mirrors. In the discussion, considerable emphasis is placed on the near field effects which become more and more important in optics of nanostructures, for example, techniques like scanning near field optical microscopy (SNOM)¹⁷. The outline is as follows: in Section II and III we discuss modified spontaneous emission and resonance transfer of excitation in a microcavity, respectively. From the discussion one will see that the radiative properties of an atom are drastically influenced by the surrounding atoms in a microcavity, because resonant dipole-dipole interaction (RDDI) between atoms with small separation provides an additional pathway for the radiating atom to decay. A summary of the comparison between the two processes

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is contained in Section IV. Appendix A gives some mathematical details regarding the evaluation of integrals in Sec. III, and Appendix B shows how the same result in Sec. III with the multipolar form of Hamiltonian can be obtained using the minimal coupling formalism.

MODIFIED SPONTANEOUS EMISSION BETWEEN MIRRORS

According to perturbation theory in quantum mechanics, spontaneous decay is described as a first-order transition problem, in which the initial state $|E_i\rangle$ is discrete (excited atom plus vacuum field) and the final state $|E_f\rangle$ continuous. The total transition probability can be obtained by summing over the range of continuum states, and the spontaneous emission rate is defined by

$$P_{sp} = \frac{2\pi}{\hbar} |\langle E_f | H_{int} | E_i \rangle|^2 \delta(E_f - E_i) \quad (1)$$

where H_{int} is the interaction Hamiltonian. This is the well-known *Fermi Golden Rule*.

If the multipolar Hamiltonian in dipole approximation is chosen¹⁸,

$$H_{int} = -\frac{1}{\epsilon_0} \boldsymbol{\mu} \cdot \mathbf{d}^\perp, \quad (2)$$

the Fermi rule gives the decay rate:

$$P_{sp} = \frac{2\pi}{\hbar} \sum_{k n \lambda} |\langle G; 1(\mathbf{k}, n, \lambda) | -\frac{1}{\epsilon_0} \boldsymbol{\mu} \cdot \mathbf{d}^\perp | E; 0 \rangle|^2 \delta(E_{eg} - \hbar\Omega_{kn}). \quad (3)$$

Here, $\boldsymbol{\mu}$ is the electric dipole and \mathbf{d}^\perp is the Maxwell displacement vector, which is the canonical conjugate to vector potential \mathbf{A} (to within constant factors) satisfying the boundary conditions in a cavity consisting of two parallel mirrors placed at $z = 0$ and $z = L$,⁶

$$\begin{aligned} \mathbf{d}^\perp(\boldsymbol{\rho}, z) = & i \sum_k \sum_{n=0}^{\infty} \sqrt{\frac{\hbar\Omega_{kn}\epsilon_0}{2AL}} (\delta_{n0} + \sqrt{2}(1 - \delta_{n0})) e^{i\mathbf{k} \cdot \boldsymbol{\rho}} \{ \hat{\mathbf{k}} \times \hat{\mathbf{z}} \sin \frac{n\pi z}{L} a_{kn1} \\ & + (\hat{k} \frac{i n \pi c}{L \Omega_{kn}} \sin \frac{n\pi z}{L} - \hat{z} \frac{kc}{\Omega_{kn}} \cos \frac{n\pi z}{L}) a_{kn2} + h.c., \} \end{aligned} \quad (4)$$

where $\boldsymbol{\rho}$ is in the xy plane, \mathbf{k} is a two-dimensional wave vector parallel to the mirrors, $a_{kn\lambda}$ are annihilation operators specified by a polarization index $\lambda = 1, 2$. In the initial state the atom is in the excited state $|E\rangle$ with no photon present, and in the final state it is in the ground state $|G\rangle$ with one occupied photon in the field. In the delta function E_{eg} is the transition energy and Ω_{kn} are the existing photon frequencies in the cavity :

$$\Omega_{kn}^2 = c^2 k^2 + \left(\frac{n\pi c}{L}\right)^2 \quad (5)$$

Without loss of generality, one can assume that the atom is located at position $\mathbf{R} = (0, 0, z)$ with dipole moment $\mu = (\mu_x, 0, \mu_z)$, then the decay rate is calculated as

$$P_{sp} = \frac{c}{4\epsilon_0 L^3} \sum_{n=0}^{\infty} (\delta_{n0} + 2(1 - \delta_{n0}))^2 \int_{n\pi/L}^{\infty} q dq \{ \mu_x^{ge}(z)^2 \sin^2 \left(\frac{n\pi z}{L} \right) \frac{q^2 + (n\pi/L)^2}{q} + 2\mu_z^{ge}(z)^2 \cos^2 \left(\frac{n\pi z}{L} \right) \frac{q^2 - (n\pi/L)^2}{q} \} \delta(\hbar c q - E_{eg}) \quad (6)$$

where $q^2 = k^2 + (n\pi/L)^2$.

Because of the existence of the delta function in Eq. (6), only those terms satisfying

$$\frac{E_{eg}}{\hbar c} \geq \frac{n\pi}{L} \quad (7)$$

or $n = n^*$ ($= 2L/\lambda^*$, where λ^* is the transition wavelength) contribute to the decay rate. Thus

$$P_{sp} = \frac{\pi^2}{4\hbar\epsilon_0 L^3} \sum_{n=0}^N (\delta_{n0} + \sqrt{2}(1 - \delta_{n0}))^2 \{ \mu_x^{ge}(z)^2 \sin^2 \left(\frac{n\pi z}{L} \right) (n^2 + n^{*2}) + 2\mu_z^{ge}(z)^2 \cos^2 \left(\frac{n\pi z}{L} \right) (n^{*2} - n^2) \} \quad (8)$$

where N is the maximal integral part no larger than n^* . Since there is the following identity for series

$$\sum_{n=1}^N \sin^2 nx = \frac{N}{2} - \frac{\cos(N+1)x \sin Nx}{2 \sin x}, \quad (9)$$

(an identical relation holds also for cosine function), differentiating them with respect to x twice leads Eq. (8) to the same result found by G. Barton⁶ and M. R. Philpott⁷, in which an alternative Hamiltonian in the minimal coupling formalism $\frac{e}{m} \mathbf{p} \cdot \mathbf{A}$ was used. Two Hamiltonians are related to each other by a canonical transformation¹⁸.

Therefore, there is a striking feature of spontaneous decay being completely inhibited for the cavity length $L < \lambda^*/2$ when the dipole is parallel to the mirrors ($\mu_z = 0$). In quantum theory this results from the fact that the delta function $\delta(E_{eg} - \eta\Omega_{kn})$ in Eq. (3) requires conservation of energy and momentum in the process, i.e., *spontaneous decay is due to the emission of real photons of resonant frequencies into the space irreversibly propagating away.*

Indeed, the complete inhibition of spontaneous emission is only possible with perfect mirrors. In the classical electrodynamical description of a dipole between mirrors,¹¹⁻¹³ the correction of the radiative properties is attributed to the radiation reaction (the self-field and the reflected field by the mirrors) on the dipole. The quadrature component of the reaction field, which remains finite, causes a modification

of the decay rate, whereas the component in phase with the dipole oscillation, which diverges at the dipole location, shifts the oscillation frequency (after renormalization). Generally, the effects of mirrors is modeled by a complex reflection coefficient of a real amplitude with a phase shift. For imperfect mirrors, the extra phase shift can admix the divergent (near field) component to the decay rate of the dipole; as a result, and the decay rate diverges for a parallel dipole between mirrors with sufficiently small separation.

The near field effects have recently become more and more important for understanding physical phenomena in the nanoscopic regime, due to much of recent progress in SNOM. At sufficiently short distances the near field plays a significant role in the interaction of atoms with their environment, such as the tip-sample interaction in SNOM¹⁷ and dipole-phase conjugate mirror coupling.¹⁹ However, as far as the above first-order perturbation theory in quantum mechanics is concerned, spontaneous emission of Eq. (3) cannot account for the effects of the near field in decay processes, which need second-order perturbation theory. As a matter of fact, an additional phase shift in the reflection coefficient phenomenologically describes energy losses to the mirrors. Thus, in the following section we shall discuss the excitation (energy) transfer of an atom-pair between mirrors, which will be helpful for us to understand not only the role of *virtual* photons in decay processes, but also the relationship between spontaneous emission and resonance transfer of excitation.

MODIFIED EXCITATION TRANSFER IN A CAVITY

In the problem of resonance transfer of excitation, the interaction Hamiltonian for the system is best given in the multipolar form. In electric-dipole approximation, for a pair of atoms, it can be written as¹⁸

$$H_{int} = -\frac{1}{\epsilon_0}\boldsymbol{\mu}(A) \cdot \mathbf{d}^\perp(\mathbf{R}_A) - \frac{1}{\epsilon_0}\boldsymbol{\mu}(B) \cdot \mathbf{d}^\perp(\mathbf{R}_B) \quad (10)$$

Let A and B be two-level identical atoms one of which is in an excited state. With second-order perturbation theory, the Fermi golden rule also gives the rate of excitation transfer $P_{tr} = \frac{2\pi}{\hbar}|M|^2\delta(E_i - E_f)$ with the matrix element for the transition

$$M = \sum_I \frac{\langle E_A G_B; 0 | H_{int} | I \rangle \langle I | H_{int} | G_A E_B; 0 \rangle}{E_i - E_I - i\epsilon} \quad (11)$$

in the limit $\epsilon \rightarrow 0$, where the intermediate states are of two types: $|G_A G_B; 1(\mathbf{k}, n, \lambda)\rangle$ and $|E_A E_B; 1(\mathbf{k}, n, \lambda)\rangle$, E_i , E_I , and E_f are energies of the initial, intermediate, and final states. Eq. (11) can be divided into two parts²⁰

$$M = V + i\Gamma, \quad (12)$$

with

$$V = \sum_I \frac{\langle G_A E_B; 0 | H_{int} | I \rangle \langle I | H_{int} | E_A G_B; 0 \rangle}{E_i - E_I} \quad (13)$$

which is a principal value integral representing the potential energy between the two atoms, and

$$\Gamma = \sum_I \langle G_A E_B; 0 | H_{int} | I \rangle \langle I | H_{int} | E_A G_B; 0 \rangle \pi \delta(E_i - E_I), \quad (14)$$

which corresponds to the case when a real photon is transferred from one atom to the other, in terms of the spontaneous emission and induced absorption of the real photon.

Consider that two atoms are located at $\mathbf{R}(A) = (0, 0, z_A)$ and $\mathbf{R}(B) = (x, 0, z_B)$ in the xz plane between the mirrors. By taking into consideration two possible polarization indexes of the photon of the intermediate states, we have

$$\begin{aligned} V = & \sum_{\mathbf{k}n} \frac{\hbar \Omega_{kn}}{2\epsilon_0 AL} (\delta_{n0} + \sqrt{2}(1 - \delta_{n0}))^2 \left[\sin \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \right. \\ & \times \sum_{ij} \mu_i^{ge}(A) \mu_j^{ge}(B) (\hat{\mathbf{k}} \times \hat{\mathbf{z}})_i (\hat{\mathbf{k}} \times \hat{\mathbf{z}})_j \left(\frac{e^{ik \cos \theta}}{E_{eg} - \hbar \Omega_{kn}} - \frac{e^{-ik \cos \theta}}{E_{eg} + \hbar \Omega_{kn}} \right) \\ & + \left(\frac{n\pi c}{L \Omega_{kn}} \right)^2 \sin \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \sum_{ij} \mu_i^{ge}(A) \mu_j^{ge}(B) \hat{k}_i \hat{k}_j \left(\frac{e^{ik \cos \theta}}{E_{eg} - \hbar \Omega_{kn}} - \frac{e^{-ik \cos \theta}}{E_{eg} + \hbar \Omega_{kn}} \right) \\ & + \left(\frac{kc}{\Omega_{kn}} \right)^2 \cos \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \mu_z^{ge}(A) \mu_z^{ge}(B) \left(\frac{e^{ik \cos \theta}}{E_{eg} - \hbar \Omega_{kn}} - \frac{e^{-ik \cos \theta}}{E_{eg} + \hbar \Omega_{kn}} \right) \\ & + \frac{i n \pi k c^2}{L \Omega_{kn}^2} \sin \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \sum_i \mu_i^{ge}(A) \mu_z^{ge}(B) \hat{k}_i \left(\frac{e^{ik \cos \theta}}{E_{eg} - \hbar \Omega_{kn}} + \frac{e^{-ik \cos \theta}}{E_{eg} + \hbar \Omega_{kn}} \right) \\ & - \frac{i n \pi k c^2}{L \Omega_{kn}^2} \cos \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \\ & \left. \times \sum_i \mu_z^{ge}(A) \mu_i^{ge}(B) \hat{k}_i \left(\frac{e^{ik \cos \theta}}{E_{eg} - \hbar \Omega_{kn}} + \frac{e^{-ik \cos \theta}}{E_{eg} + \hbar \Omega_{kn}} \right) \right] \quad (15) \end{aligned}$$

where $\mu_i^{ge}(A) = \langle G_A | \mu_i(A) | E_A \rangle$ and $\mu_j^{eg}(B) = \langle E_B | \mu_j(B) | G_B \rangle$, and $\mathbf{k} = k(\cos \theta, \sin \theta, 0)$.

After conversion of the \mathbf{k} -sum into an integral, for example, the first term in the large square bracket in the right-hand side of Eq. (15) (denoted by V_1) becomes

$$\begin{aligned} V_1 = & \frac{\hbar^2}{4\pi\epsilon_0 L} \sum_n (\delta_{n0} + \sqrt{2}(1 - \delta_{n0}))^2 \sin \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \\ & \times \int_0^\infty k dk \frac{c^2 k^2 + (n\pi c/L)^2}{E_{eg}^2 - (\hbar ck)^2 - (\hbar n\pi c/L)^2} [\mu_x^{ge}(A) \mu_x^{ge}(B) (J_0(kx) + J_2(kx))] \end{aligned}$$

$$+ \mu_y^{ge}(A)\mu_y^{ge}(B)(J_0(kx) + J_2(kx)) \quad (16)$$

where $J_n(z)$ are the Bessel functions of the first kind.²¹ The different terms of the integral can be evaluated by the residue theorem as shown in Appendix A.

Because *the potential energy V arises from the exchange of virtual photons of all frequencies* — the principal value of the integral over frequency, it can be shown as consisting of two parts of contribution both from evanescent and propagating modes in the cavity. The latter can be merged into $i\Gamma$ of Eq. (14) (evaluated in a similar way), and the matrix element M finally becomes

$$M = M_{n>n^*} + iM_{n<n^*} \quad (17)$$

with the evanescent-mode part

$$\begin{aligned} M_{n>n^*} &= \frac{\pi}{2\epsilon_0 L^3} \sum_{n>n^*} \sin \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \\ &\times \left[-(\mu_x^{ge}(A)\mu_x^{ge}(B) + \mu_y^{ge}(A)\mu_y^{ge}(B))(n^{*2} + n^2)K_0\left(\frac{\pi x}{L}\sqrt{n^2 - n^{*2}}\right) \right. \\ &+ \left. (\mu_x^{ge}(A)\mu_x^{ge}(B) - \mu_y^{ge}(A)\mu_y^{ge}(B))(n^{*2} - n^2)K_2\left(\frac{\pi x}{L}\sqrt{n^2 - n^{*2}}\right) \right] \\ &+ \frac{\pi}{\epsilon_0 L^3} \sum_{n>n^*} \cos \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \\ &\times \mu_z^{ge}(A)\mu_z^{ge}(B)(n^2 - n^{*2})K_0\left(\frac{\pi x}{L}\sqrt{n^2 - n^{*2}}\right) \\ &+ \frac{\pi}{2\epsilon_0 L^3} \sum_{n>n^*} \sin \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \\ &\times \mu_x^{ge}(A)\mu_z^{ge}(B)n\sqrt{(n^2 - n^{*2})}K_1\left(\frac{\pi x}{L}\sqrt{n^2 - n^{*2}}\right) \\ &- \frac{\pi}{2\epsilon_0 L^3} \sum_{n>n^*} \cos \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \\ &\times \mu_z^{ge}(A)\mu_x^{ge}(B)n\sqrt{(n^2 - n^{*2})}K_1\left(\frac{\pi x}{L}\sqrt{n^2 - n^{*2}}\right), \quad (18) \end{aligned}$$

where $K_n(z)$ are the modified Bessel functions, and the propagating-mode part

$$\begin{aligned} M_{n<n^*} &= \frac{\pi^2}{4\epsilon_0 L^3} \sum_{n<n^*} \sin \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \\ &\times \left[(\mu_x^{ge}(A)\mu_x^{ge}(B) + \mu_y^{ge}(A)\mu_y^{ge}(B))(n^{*2} + n^2)H_0^{(2)}\left(\frac{\pi x}{L}\sqrt{n^{*2} - n^2}\right) \right. \\ &+ \left. (\mu_x^{ge}(A)\mu_x^{ge}(B) - \mu_y^{ge}(A)\mu_y^{ge}(B))(n^{*2} - n^2)H_2^{(2)}\left(\frac{\pi x}{L}\sqrt{n^{*2} - n^2}\right) \right] \end{aligned}$$

$$\begin{aligned}
& + \frac{\pi^2}{4\epsilon_0 L^3} \sum_{n < n^*} (\delta_{n0} + \sqrt{2}(1 - \delta_{n0}))^2 \cos \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \\
& \times \mu_z^{ge}(A) \mu_z^{ge}(B) (n^2 - n^{*2}) H_0^{(2)}\left(\frac{\pi x}{L} \sqrt{n^{*2} - n^2}\right) \\
& - \frac{\pi^2}{2\epsilon_0 L^3} \sum_{n < n^*} \sin \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \\
& \times \mu_x^{ge}(A) \mu_z^{ge}(B) n \sqrt{(n^{*2} - n^2)} H_1^{(2)}\left(\frac{\pi x}{L} \sqrt{n^{*2} - n^2}\right) \\
& + \frac{\pi^2}{2\epsilon_0 L^3} \sum_{n < n^*} \cos \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \\
& \times \mu_z^{ge}(A) \mu_x^{ge}(B) n \sqrt{(n^{*2} - n^2)} H_1^{(2)}\left(\frac{\pi x}{L} \sqrt{n^{*2} - n^2}\right), \tag{19}
\end{aligned}$$

where $H_n^{(1)}(z)$, $H_n^{(2)}(z)$ are the Hankel functions.

Thus, the most dramatic difference between spontaneous emission and excitation transfer between mirrors is the contribution from RDDI to the latter. The excitation transfer can be assisted by evanescent waves even in cavities in which spontaneous emission by the atoms is inhibited because the atomic transition frequency is much less than the cavity cutoff frequency. ²²

REMARKS

The decay properties of an atom are usually dependent on its environment. Classically, the effects of electromagnetic boundary conditions on radiative properties of a dipole between mirrors are phenomenologically modeled by a complex reflection coefficient of the mirrors. For perfect conducting mirrors, it is shown that the decay of a dipole parallel to the mirrors can be completely suppressed when the transition frequency is much less than the cutoff frequency. However, for real mirrors, the divergence of the decay rate at a small gap happens because the phase shift couples the divergent near field of the dipole into the decay rate of the dipole radiation.

The microscopic description of decay phenomena, provided by QED, makes a distinction between spontaneous decay and resonance transfer of excitation. Spontaneous decay is due to a coupling between a single dipole and the electromagnetic field, the rate of which is calculated by first order perturbation theory. Since the decay rate is proportional to a delta function part in Eq. (3), the process is described in terms of emission of real photons of resonant frequencies. On the other hand, the calculation of the rate of excitation transfer requires second order perturbation theory. The process not only is a result of real photons transferred from one atom to the other, but also involves resonant dipole-dipole interaction by exchange of virtual photons of all frequencies.

Thus, the quantum description depicts a clear picture to us how decay phenomena take place in terms of real and virtual photons. Resonance transfer of excitation due to the RDDI part by exchange of virtual photons plays a significant role in decay phenomena in physical systems with microscopic scale, like microcavities and SNOM devices, which not only explains the divergence of radiation between imperfect mirrors with small gap, because realistic mirrors are constituted of atoms, but also is the underlying mechanism of using a pointed detector in SNOM devices to convert the non-radiative field distribution near the sample surface into radiative fields detectable in the far-field region (the tip-sample interaction).

Appendix A

The definite integral in Eq. (16) involves four terms taking the following forms:

$$\int_0^{\infty} \frac{k^3 J_0(kx)}{k^2 - r^2} dk \quad (\text{A.1})$$

$$\int_0^{\infty} \frac{k^3 J_2(kx)}{k^2 - r^2} dk \quad (\text{A.2})$$

$$\int_0^{\infty} \frac{k J_0(kx)}{k^2 - r^2} dk \quad (\text{A.3})$$

$$\int_0^{\infty} \frac{k J_2(kx)}{k^2 - r^2} dk \quad (\text{A.4})$$

The contours are closed in Fig. 1, for example, for the term (A.1) with an infinite quadrant in the upper and lower half-plane, respectively. When $r^2 > 0$, there is a pole ($z = r$) on the real axis (Fig. 1(a)). Then

$$\int_0^{\infty} \frac{k^3 H_0^{(1)}(kx)}{k^2 - r^2} e^{-\alpha k} dk + \int_0^{\infty} \frac{h^3 H_0^{(1)}(e^{i\pi/2} hx)}{h^2 + r^2} e^{-i\alpha h} dh - i\pi \frac{r^2}{2} H_0^{(1)}(rx) e^{-\alpha r} = 0 \quad (\text{A.5})$$

and

$$\int_0^{\infty} \frac{k^3 H_0^{(2)}(kx)}{k^2 - r^2} e^{-\alpha k} dk + \int_0^{\infty} \frac{h^3 H_0^{(2)}(e^{-i\pi/2} hx)}{h^2 + r^2} e^{i\alpha h} dh + i\pi \frac{r^2}{2} H_0^{(2)}(rx) e^{-\alpha r} = 0 \quad (\text{A.6})$$

Adding them, we have

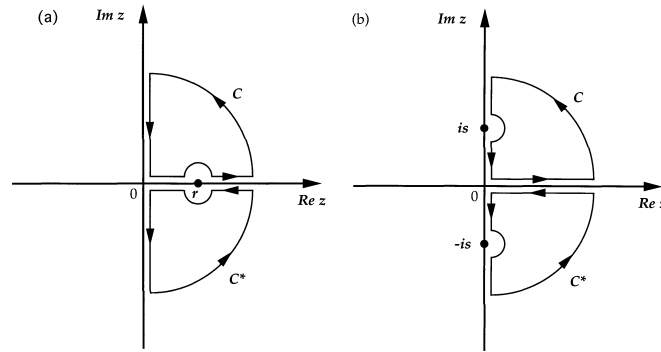


Fig. 1 Contours for the integral (A.1). (a) For $r^2 > 0$; (b) For $r^2 < 0$.

$$\int_0^\infty \frac{2k^3 J_0(kx)}{k^2 - r^2} e^{-\alpha k} dk + \int_0^\infty \frac{h^3 (H_0^{(1)}(e^{i\pi/2} hx) e^{-i\alpha h} + H_0^{(2)}(e^{-i\pi/2} hx) e^{i\alpha h})}{h^2 + r^2} dh + \pi r^2 Y_0(rx) e^{-\alpha r} = 0 \tag{A.7}$$

where identities

$$H_0^{(1)}(rx) + H_0^{(2)}(rx) = 2J_0(rx) \tag{A.8}$$

$$H_0^{(1)}(rx) - H_0^{(2)}(rx) = 2iY_0(rx) \tag{A.9}$$

are used. $Y_n(z)$ are the Bessel functions of the second kind. Taking $\alpha \rightarrow 0$ in Eq. (A.7) leads to

$$\int_0^\infty \frac{k^3 J_0(kx)}{k^2 - r^2} dk = -\frac{\pi r^2}{2} \mathcal{Y}(s) \tag{A.10}$$

If $r^2 < 0$, there are two poles ($z = \pm is$) on the imaginary axis as shown in Fig. 1(b), and we have

$$\int_0^\infty \frac{k^3 H_0^{(1)}(kx)}{k^2 + s^2} e^{-\alpha k} dk + \int_0^\infty \frac{h^3 H_0^{(1)}(e^{i\pi/2} hx)}{h^2 - s^2} e^{-i\alpha h} dh + i\pi \frac{s^2}{2} H_0^{(1)}(e^{i\pi/2} sx) e^{-i\alpha s} = 0 \tag{A.11}$$

and

$$\int_0^\infty \frac{k^3 H_0^{(2)}(kx)}{k^2 + s^2} e^{-\alpha k} dk + \int_0^\infty \frac{h^3 H_0^{(2)}(e^{-i\pi/2} hx)}{h^2 - s^2} e^{i\alpha h} dh - i\pi \frac{s^2}{2} H_0^{(2)}(e^{-i\pi/2} sx) e^{i\alpha s} = 0 \tag{A.12}$$

where $s^2 = -r^2$. Adding Eqs. (A.11) and (A.12) and using

$$H_0^{(1)}(e^{i\pi/2}sx) - H_0^{(2)}(e^{-i\pi/2}sx) = -\frac{4i}{\pi}K_0(sx), \quad (\text{A.13})$$

we obtain

$$\int_0^\infty \frac{k^3 J_0(kx)}{k^2 + s^2} dk = -s^2 K_0(sx) \quad (\text{A.14})$$

If $r^2 = 0$, both Eqs. (A.10) and (A.14) become zero.

Other terms (A.2)-(A.4) can be evaluated in a similar way. For example, there is a pole at $z = 0$ for the term (A.4). Choosing contours C and C^* that detour around the pole, we have

$$\int_0^\infty \frac{k J_2(kx)}{k^2 - r^2} dk = -\frac{\pi}{2} Y_2(sx) - \frac{2}{x^2 r^2} \quad (\text{A.15})$$

for $r^2 > 0$, and

$$\int_0^\infty \frac{k J_2(kx)}{k^2 - r^2} dk = -K_0(sx) + \frac{2}{x^2 r^2} \quad (\text{A.16})$$

for $r^2 = -s^2 < 0$, where the second terms in the right-hand sides are the residue at $z = 0$. If $r^2 = 0$, both Eqs. (A.15) and (A.16) take the value $1/2$.

Appendix B

We now show how the same expression for the rate of excitation transfer can be obtained using the minimal coupling Hamiltonian

$$H_{int} = \frac{e}{m} \sum_{\alpha} \mathbf{p}_{\alpha A} \cdot \mathbf{A}(\mathbf{R}_A) - \frac{e}{m} \sum_{\alpha} \mathbf{p}_{\alpha B} \cdot \mathbf{A}(\mathbf{R}_B) + V_{inter}, \quad (\text{B.1})$$

where the summation of momentum \mathbf{p}_{α} is over the electrons of atoms A and B, and V_{inter} is the Coulomb coupling term. In Eq. (B.1) we neglect terms quadratic in e since they do not contribute to inter-atomic coupling in this order.

To calculate V_{inter} , we first give the Green's function in the space inside two infinite parallel mirrors a distance L apart:

$$G(xyz|XYZ) = \frac{1}{\pi L} \sum_{n=1}^{\infty} \sin \frac{n\pi z}{L} \sin \frac{n\pi Z}{L} K_0\left(\frac{n\pi}{L} \sqrt{(x-X)^2 + (y-Y)^2}\right) \quad (\text{B.2})$$

Therefore, the scalar potential related with the charge density $\rho(XYZ)$ is

$$\phi(xyz) = \frac{1}{\epsilon_0} \iiint G(xyz|XYZ) \rho(XYZ) dXdYdZ \quad (\text{B.3})$$

Consider a dipole, A, to be at $(0, 0, z_A)$, then the corresponding potential becomes

$$\begin{aligned} \phi(xyz) &= \frac{1}{\epsilon_0 L^2} \sum_{n=1}^{\infty} n \sin \frac{n\pi z}{L} [\mu_z(A) \cos \frac{n\pi z_A}{L} \sin \frac{n\pi Z}{L} K_0(\frac{n\pi}{L} \sqrt{x^2 + y^2}) \\ &- \frac{x\mu_x(A) + y\mu_y(A)}{\sqrt{x^2 + y^2}} \sin \frac{n\pi z_A}{L} K'_0(\frac{n\pi}{L} \sqrt{x^2 + y^2})] \end{aligned} \quad (\text{B.4})$$

If another dipole, B, is added, the electrostatic energy of the system formed by the two dipoles is given by

$$V_{inter} = -\mu(A) \cdot \mathbf{E}_A(B), \quad (\text{B.5})$$

where $\mathbf{E}_A(B)$ is the electric field by dipole A at the position $(x, 0, z_B)$ of dipole B. Using $\mathbf{E}(xyz) = -\text{grad}\phi$, Eq. (B.5) becomes

$$\begin{aligned} V_{inter} &= -\frac{\pi}{2\epsilon_0 L^3} \sum_{n=1}^{\infty} \sin \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \\ &\times \left[-(\mu_x(A)\mu_x(B) + \mu_y(A)\mu_y(B))n^2 K_0(\frac{n\pi x}{L}) \right. \\ &+ \left. (\mu_x(A)\mu_x(B) - \mu_y(A)\mu_y(B))n^2 K_2(\frac{n\pi x}{L}) \right] \\ &+ \frac{\pi}{\epsilon_0 L^3} \sum_{n=1}^{\infty} \cos \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \mu_z(A)\mu_z(B)n^2 K_0(\frac{n\pi x}{L}) \\ &+ \frac{\pi}{\epsilon_0 L^3} \sum_{n=1}^{\infty} \sin \frac{n\pi z_A}{L} \cos \frac{n\pi z_B}{L} \mu_x(A)\mu_z(B)n^2 K_1(\frac{n\pi x}{L}) \\ &- \frac{\pi}{\epsilon_0 L^3} \sum_{n=1}^{\infty} \cos \frac{n\pi z_A}{L} \sin \frac{n\pi z_B}{L} \mu_z(A)\mu_x(B)n^2 K_1(\frac{n\pi x}{L}) \end{aligned} \quad (\text{B.6})$$

Then the V_{inter} -related matrix elements can be calculated using Eq. (B.6). For example, after substituting $\mu_i(A)$ with $\mu_x^{ge}(A)$ ($= \langle G_A | \mu_i(A) | E_A \rangle$) and $\mu_i(B)$ with $\mu_j^{eg}(B)$ ($= \langle E_B | \mu_j(B) | G_B \rangle$) in the right-hand side of Eq. (B.6), one gets the expression for $\langle G_A E_B | V_{inter} | E_A G_B \rangle$.

Next we calculate the term $\underline{p} \underline{\mathcal{A}}$ due to atom-electromagnetic field interaction. This requires second-order perturbation theory in the same manner to the multipolar formalism. The momentum transition matrix elements p_i^{ge} and p_i^{eg} can be connected to transition dipole moments in the following way¹⁸

$$p_i^{ge} = -imc \frac{2\pi}{e\lambda} \mu_i^{ge} \quad (\text{B.7})$$

and

$$p_i^{eg} = imc \frac{2\pi}{e\lambda} \mu_i^{eg} \quad (\text{B.8})$$

With the above relations in calculating the matrix element for transition, additional poles arise at $k = \pm i F(n\pi, L)$, when evaluating the principal value required as before. This comes from the fact that Ω_{kn} appears in the denominator of $\underline{\mathcal{A}}$.⁶ Those poles give a wholly non-retarded contribution which exactly cancels the electrostatic contribution related with V_{inter} , leading to the result Eq. (3.8) found before.

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