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The quantized field in a dielectric and application to the radiative decay of an embedded atom

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Abstract

Weisskopf–Wigner theory applied to the macroscopically quantized field shows that the spontaneous emission rate of an excited atom is unaffected by a dielectric, rather than enhanced by a factor of the refractive index, as local-field effects are eliminated in the continuum approximation. Dielectric effects on an atom must be calculated microscopically.
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Macroscopic quantization of the field in a linear dielectric [1–6], introduced by Ginzburg in 1940 [1], reduces the complexity of the microscopic quantum electrodynamic theory by combining the matter degrees of freedom with the field. The macroscopic treatment of the spontaneous emission rate of an embedded atom consists of applying the usual quantum electrodynamic methods with the quantized macroscopic fields. Based on Fermi's golden rule, the effect of a continuous dielectric environment on an excited atom is to enhance the spontaneous emission rate by a factor of the refractive index n compared to the vacuum environment [4–8]. Denoting the spontaneous emission rate in the vacuum as Γ_v, we write this 'index effect' [9] as

$$\Gamma = n\Gamma_v. \tag{1}$$

However, Fermi's golden rule has no provision for the differences between field-mode operators and Ginzburg polariton operators. Here, we show by Weisskopf–Wigner theory that the macroscopic result for the rate of radiative decay, and spontaneous emission, of an impurity atom in a dielectric is

$$\Gamma = \Gamma_v \tag{2}$$

because the interaction that is responsible for the modification of the spontaneous emission rate is not present in the macroscopic theory. The spontaneous emission rate of an embedded atom is, in fact, affected by the dielectric and we conclude that a proper description of spontaneous emission in a dielectric requires the full microscopic theory [9,10]. The 'index effect' is not physical and does not validate or constrain the results of a more fundamental microscopic [9,10] theory of spontaneous emission in a dielectric.

The model system of a two-level atom in a linear dielectric is treated microscopically as enumerated localized oscillators and a localized atom interacting with the quantized vacuum field. The Hamiltonian

$$\begin{aligned} H = {} & \sum_{l\lambda} \hbar\omega_l a_l^\dagger a_l + \sum_n \hbar\omega_b b_n^\dagger b_n \\ & - i\hbar \sum_{nl\lambda} \left(h_l a_l b_n^\dagger e^{i\mathbf{k}_l\cdot\mathbf{r}_n} - h_l^* a_l^\dagger b_n e^{-i\mathbf{k}_l\cdot\mathbf{r}_n} \right) \\ & + \frac{\hbar\omega_a}{2}\sigma_3 - i\hbar \sum_{l\lambda} \left(g_l a_l \sigma_+ - g_l^* a_l^\dagger \sigma_- \right) \end{aligned} \tag{3}$$

consists of, in order, the free-field Hamiltonian H_{fld}, the free-oscillator Hamiltonian H_{osc}, the oscillator–field interaction Hamiltonian H_{ofi}, the free-atom Hamiltonian H_{atom}, and the

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atom–field interaction Hamiltonian $H_{\rm int}$. Here, $a_l^\dagger$ and a_l are the creation and destruction operators for field modes, ω_l is the frequency of the field in the mode l, $b_n^\dagger$ and b_n are the creation and destruction operators for the oscillator at the fixed position $\mathbf{r}_n$, ω_b is the resonance frequency associated with the oscillators, $h_l = (2\pi\omega_l/\hbar V)^{1/2}\hat{\boldsymbol{\mu}}_b \cdot \hat{\mathbf{e}}_{\mathbf{k}_l\lambda}$ is the electric dipole interaction in terms of the electric dipole moment $\boldsymbol{\mu}_b$, σ_3 is the inversion operator, $\sigma_\pm$ are the raising and lowering operators, ω_a is the resonance frequency of the atom, $g_l = (2\pi\omega_l/\hbar V)^{1/2}\boldsymbol{\mu}_a \cdot \hat{\mathbf{e}}_{\mathbf{k}_l\lambda}$ is the coupling between the field and the atom at $\mathbf{r} = \mathbf{0}$, and μ_a is the dipole moment for the atom. In order to condense the notation, the operator subscript λ for sums over the polarizations is combined with the mode index l and zero-point energies have been suppressed. The usual dipole and rotating-wave approximations have been employed. The rotating-wave approximation has a reasonable region of validity [11] for dielectrics and has been invoked [11], albeit implicitly, in quantum electrodynamic treatments of dielectrics by Hopfield [12] and by Knoester and Mukamel [8], among others.

The microscopic [9,10] quantum electrodynamic treatment of spontaneous emission in a dielectric using the Hamiltonian (3) is complex. Because the dielectric responds linearly to the field, considerable simplification is attained in a macroscopic treatment that combines the material response with the field. This can be accomplished either [11] by Ginzburg's [1] macroscopic analog of the Dirac quantization of the field in the vacuum or by a Fano–Hopfield [12,13] diagonalization of the Hamiltonian. One obtains the effective macroscopic Hamiltonian

$$H = \sum_{l\lambda} \hbar\omega_l \bar{\xi}_l^\dagger \bar{\xi}_l \tag{4}$$

in terms of macroscopic matter–field polariton operators, $\bar{\xi}_l^\dagger$ and $\bar{\xi}_l$ with the overbar denoting spatially averaged operators. The effective macroscopic electric field operator is

$$\bar{\mathbf{E}} = \frac{i}{n}\sum_{l\lambda}\sqrt{\frac{2\pi\hbar\omega_l}{V}}\left(\bar{\xi}_l e^{i\mathbf{k}\cdot\mathbf{r}} - \mathrm{H.c.}\right)\hat{\mathbf{e}}_{\mathbf{k}_l\lambda}. \tag{5}$$

The spontaneous emission rate of an impurity atom in a dielectric can then be obtained from the macroscopic analog of quantum electrodynamics by applying Fermi's golden rule

$$\Gamma = \frac{2\pi}{\hbar}\left|\langle f|H_{\rm int}|i\rangle\right|^2 D \tag{6}$$

to the effective interaction Hamiltonian $H_{\rm int} = -\boldsymbol{\mu}_a \cdot \bar{\mathbf{E}}$, where i labels the initial state and f denotes all available final states. As it is typically calculated, the dielectric renormalization of the vacuum spontaneous emission rate of an atom is found to be n [4–8] due to the dielectric renormalization of the electric field operator (5) by n^{-1}, which is squared, and the $D = n^3$ density-of-states factor.

Comparison of the effective Hamiltonian (4) and the field + dielectric part of the microscopic Hamiltonian (3) shows that the Ginzburg polariton operators $\bar{\xi}_l$ are not necessarily equivalent to the field-mode operators a_l. Then properties of these operators must be established as it is not apparent that the macroscopic Ginzburg polariton operators can be treated like the microscopic field-mode operators. Hopfield [12] formally diagonalized the Hamiltonian of a microscopic model of a dielectric, deriving the effective Hamiltonian

$$H = \sum_{l\lambda} \hbar E_{vl}\bar{\alpha}_l^\dagger\bar{\alpha}_l \tag{7}$$

in terms of the eigenvalues E_{vl} and polariton operators $\bar{\alpha}_l$ and $\bar{\alpha}_l^\dagger$. In typical usage [8,12], the Hopfield polariton operators are used as field-mode operators by fixing two of the eigenvalues as a consequence of substituting photon energies $\hbar\omega$ for a pair of polariton eigenenergies in the polariton dispersion relation, thereby reducing the rank of the eigenvalue equation and adiabatically eliminating the oscillator equations of motion [11]. This interpretation is enforced by the non-zero commutation relation

$$\left[\bar{\alpha}_l, \bar{\alpha}_{l'}^\dagger\right] = \delta_{ll'}\delta_{\lambda\lambda'}. \tag{8}$$

The transformation between operators, $\bar{\xi}_l = \sqrt{E_{vl}}\bar{\alpha}_l$, is obtained by comparison of Eqs. (7) and (4). Then the effective Hamiltonian

$$H = \sum_{l\lambda}\hbar\omega_l\bar{\xi}_l^\dagger\bar{\xi}_l + \frac{\hbar\omega_a}{2}\sigma_3 - \frac{i}{n}\hbar\sum_{l\lambda}\left(g_l\bar{\xi}_l\sigma_+ - g_l^*\bar{\xi}_l^\dagger\sigma_-\right) \tag{9}$$

becomes

$$H = \sum_{l\lambda}\hbar n\omega_l\bar{\alpha}_l^\dagger\bar{\alpha}_l + \frac{\hbar\omega_a}{2}\sigma_3 - \frac{i}{\sqrt{n}}\hbar\sum_{l\lambda}\left(g_l\bar{\alpha}_l\sigma_+ - g_l^*\bar{\alpha}_l^\dagger\sigma_-\right) \tag{10}$$

as $E_{vl} \to n\omega_l$ in the limit of small $\delta n = n - 1$.

Fermi's golden rule is a prescription for applying Weisskopf–Wigner theory to the spontaneous emission of an atom. Taking a more rigorous route, we calculate the radiative decay rate of an atom by applying Weisskopf–Wigner theory to the effective Hamiltonian. Writing the Heisenberg equations of motion that correspond to the Hamiltonian (10) and formally eliminating the Hopfield polariton operators, one obtains

$$\frac{d\sigma_3}{dt} = -2\sum_{l\lambda}\frac{g_l}{n}\tilde{\sigma}_+\int_{-\infty}^{t}dt'\, e^{-i(n\omega_l-\omega_a)(t-t')}g_l^*\tilde{\sigma}_-(t') + \mathrm{H.c.}, \tag{11}$$

where $\tilde{\sigma}_\pm = \sigma_\pm e^{\mp i\omega_a t}$. Evaluating the preceding equation in the Markov approximation and mode-continuum limit [14,15], we find that the decay rate of the impurity atom is suppressed by a factor of n due to the presence of the dielectric as the well-known n^3 factor for the enhanced density-of-states in a dielectric medium is offset by the well-known renormalization of the mode frequencies. The same result is obtained from the Ginzburg effective Hamiltonian (9) using the commutation relation for Ginzburg polaritons

$$\left[\bar{\xi}_l, \bar{\xi}_{l'}^\dagger\right] = n\delta_{ll'}\delta_{\lambda\lambda'} \tag{12}$$

derived from (8). The suppression of the spontaneous emission rate by n,

$$\Gamma = \frac{1}{n}\Gamma_v = \frac{4\omega_a^3\mu_a^2}{3n\hbar c^3}, \tag{13}$$