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Resonant Scattering and Spontaneous Emission in Dielectrics: Microscopic Derivation of Local-Field Effects

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Resonant classical light scattering by impurity atoms inside dielectric cubic lattices is investigated in the point-dipole limit. Modifications to resonance frequencies and linewidths are shown to be different for substitutional and interstitial impurities. Spontaneous emission rates inside dielectrics exhibit the well-known empty-cavity and Lorentz local-field factors for substitutional and interstitial atoms, respectively. The results are generalized to disordered dielectrics, indicating that the substitutional case occurs prevalently for impurity atoms. [S0031-9007(98)06814-8]

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The properties of light in modulated dielectric structures have become a subject of intense interest. For instance, the fabrication of photonic band gap materials is aimed at achieving a gap in the optical band structure [1]. At the band gap the radiative density of states (RDOS) is zero and, therefore, spontaneous emission of excited atoms embedded in these materials is expected to be inhibited at those frequencies, since then coupling to propagating modes is absent. At other frequencies it has been shown that spontaneous emission can be either reduced or enhanced [1,2]. Note that the atoms in question are *different* from the ones constituting the dielectric. Quite generally, luminescence rates Γ can be modified by manipulating the optical mode structures. This has also been demonstrated experimentally and theoretically for several other systems, e.g., for excited atoms that are placed in cavities [3] consisting of metallic or dielectric mirrors or in systems [4,5] with dielectric interfaces. The placement of excited atoms inside a macroscopically homogeneous dielectric with dielectric constant ϵ also leads to a modification of Γ , which is then expected to follow the $\sqrt{\epsilon}$ dependence of the RDOS [6].

The latter problem has recently been reexamined in connection with possible local-field corrections that should enter expressions for Γ . The microscopic, or local, electric field an atom feels is usually different from the one following from classical and quantum-mechanical descriptions where the dielectric is considered on a coarse-grained or macroscopic scale. Determining the local field is an old problem in physics, for which Lorentz has given a mean-field-like argument in terms of a “virtual” cavity surrounding a dipole of the dielectric [7]. The resulting ratio of local and macroscopic fields is given by the Lorentz local-field factor

$$\mathcal{L}_{\text{Lor}} = (\epsilon + 2)/3, \quad (1)$$

which appears in expressions for several physical quantities, e.g., the Lorentz-Lorenz relation (LLR) or Clausius-Mossotti equation for the dielectric constant [7,8].

In the Onsager-Böttcher (OB) local-field approach [7] a cavity with volume V , in which a dipole with

polarizability α is placed, is considered inside a dielectric described by the macroscopic Maxwell’s equations. The local-field correction pertaining to the field in the cavity is

$$L_{\text{OB}}(\alpha/V) = \frac{3\epsilon}{2\epsilon + 1 - 2(\alpha/V)(\epsilon - 1)/3}. \quad (2)$$

In contrast to the Lorentz approach, two cases can now be distinguished: The dipole inside the cavity is either equivalent to or different from the ones forming the dielectric. In the former case, Eq. (2) reduces to the appropriate Lorentz factor \mathcal{L}_{Lor} when α/V satisfies the LLR. Hence, V^{-1} is equal to the density of dipoles inside the dielectric. In experiments [9] involving only *pure* systems, i.e., only *one* kind of atom or molecule is present, the observed local-field effects agree very well with the Lorentz description. If desired the influence of particle interactions not included in the LLR can be captured phenomenologically [7] using other values for V .

Considering the case of an impurity dipole inside the cavity one sometimes neglects the response of the dielectric on α . Equation (2) then simplifies to the, so-called, empty-cavity factor [10,11]

$$\mathcal{L}_{\text{emp}} = 3\epsilon/(2\epsilon + 1). \quad (3)$$

It has been proposed theoretically [10–17] that one of the factors, \mathcal{L}_{Lor} or \mathcal{L}_{emp} , should be included in a description of Γ of impurity atoms inside dielectrics. The majority of researchers in this field [11–15], in fact, endorse the Lorentz factor. On the other hand, experimental results [16] on atoms inside complexes indicate that the empty-cavity factor should be employed. These factors differ very much, especially for frequencies near a band gap or a material resonance where ϵ can become very large. It is not clear from the literature [10–17] which argument enables one to make a correct choice between the two factors. For pure systems, straightforward application of \mathcal{L}_{Lor} generally allows for an excellent description. Obviously, a microscopic theory is desired to clarify this issue for the case of impurity atoms.

To this end, we present in this Letter a microscopic analysis based on resonant classical light scattering by impurity atoms inside dielectric cubic lattices. The atoms forming the lattice, as well as the impurity atoms, are described by point dipoles, so that exact solutions can be obtained. In addition, we discuss how these results apply to the case of disordered dielectrics. By considering only elastic scattering the resonance linewidths are interpreted to equal the spontaneous emission rates of the impurities. The optical theorem, i.e., the equivalence of the amount of light taken out of the incident waves (extinction) and the amount of light redistributed over all angles (scattering), may be viewed as a microscopic analogon of Einstein's thermodynamic arguments relating extinction (and absorption), stimulated and spontaneous emission. Provided one distinguishes between interstitial and substitutional impurities, it will follow that the application of the factors \mathcal{L}_{Lor} and \mathcal{L}_{emp} , respectively, is appropriate.

We first discuss the case of the pure lattice in terms of a Green's function formalism [18]. The Green's function \mathcal{G} describing all dynamic properties of light in a periodic lattice of identical point dipoles can be expressed in terms of the free-space dyadic \mathcal{G}_0 and the t -matrix element t of a single dipole. The scattering to all orders in the potential is represented [18] by $t(\omega) = -(\omega/c_0)^2 \alpha(\omega)$, where α

is the complex dynamic polarizability of a single point dipole, and is assumed scalar for simplicity. The lattice t -matrix \mathcal{T} , defined by

$$\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 \cdot \mathcal{T} \cdot \mathcal{G}_0, \quad (4)$$

is easily obtained in momentum space from the expansion in \mathcal{G}_0 and t , and reads

$$\mathcal{T}(\omega; \mathbf{k}, \mathbf{k}') = \sum_{\mathbf{R}} T_{\mathbf{k}'}(\omega) e^{i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{R}}, \quad (5)$$

with

$$T_{\mathbf{k}}(\omega) = \left[t^{-1}(\omega) I - \sum_{\mathbf{R} \neq \mathbf{0}} e^{i\mathbf{k} \cdot \mathbf{R}} \mathcal{G}_0(\omega^+; \mathbf{R}) \right]^{-1}, \quad (6)$$

where $\{\mathbf{R}\}$ are lattice vectors, I is the unit dyadic in polarization space, and $\omega^+ \equiv \omega + i0$. The solutions $\omega(\mathbf{k})$ follow from considering the poles of $T_{\mathbf{k}}$.

From now on we consider lattices having cubic symmetry and frequencies lying in the lowest branch of the band structure, i.e., $\omega a/c_0 \ll 1$ and a is the lattice constant. The dispersion relation and the dyadic $T_{\mathbf{k}}$ can be determined explicitly in this case. The discrete Fourier transform appearing in $T_{\mathbf{k}}$ may be simplified using [12,14,18,19]

$$\Omega \sum_{\mathbf{R} \neq \mathbf{0}} e^{i\mathbf{k} \cdot \mathbf{R}} \mathcal{G}_0(\omega^+; \mathbf{R}) \xrightarrow{\omega a/c_0 \ll 1, ka \ll 1} \frac{1}{(\omega^+/c_0)^2 I - k^2 I + \mathbf{k}\mathbf{k}} - \frac{I}{3(\omega/c_0)^2} + i \frac{\Omega \omega I}{6\pi c_0}, \quad (7)$$

where Ω is the volume of the unit cell. The first term on the right-hand side equals the continuum Fourier transform of \mathcal{G}_0 , while the other terms express the exclusion of the singularity in the dipole-dipole part and the finite transverse part of $\mathcal{G}_0(\omega^+; \mathbf{R} = \mathbf{0})$, respectively. The latter term exactly cancels the imaginary part of t^{-1} , since for arbitrary t satisfying the optical theorem for a single dipole, $-c_0 \text{Im } t/\omega = |t|^2/6\pi$, one has $\text{Im } t^{-1} = \omega/6\pi c_0$. From the poles of $T_{\mathbf{k}}$ it then follows that $[kc_0/\omega(\mathbf{k})]^2 \equiv \varepsilon$ satisfies the well-known LLR [12,14,18,19]

$$\varepsilon(\omega) = 1 + \frac{\tilde{\alpha}}{\Omega} \times \mathcal{L}_{\text{Lor}}, \quad (8)$$

where $\tilde{\alpha}^{-1} = -(\omega/c_0)^2 \tilde{t}^{-1} \equiv \alpha^{-1} + i(\omega/c_0)^3/6\pi$ is a real quantity. We assume that ε describing the lattice does not have a resonance for the frequencies studied here.

It is expedient to introduce a real "bare" polarizability α_b that may be used to define the strength $v_b = -(\omega/c_0)^2 \alpha_b$ of a point-dipole potential $v_b I \delta(\mathbf{r} - \mathbf{R})$ entering the Maxwell-Helmholtz equation [18]. In this case one finds that the corresponding t -matrix element reads

$$t(\omega) I = [v_b^{-1} I - \mathcal{G}_0(\omega^+; \mathbf{0})]^{-1}, \quad (9)$$

where $\mathcal{G}_0(\omega^+; \mathbf{r})$ needs to be regularized since it diverges at $r = 0$. The latter can be done [18]. Here, it is not necessary to explicitly compute v_b . The resonances of

the impurities turn out not to depend on v_b . Equation (9) enables one to derive

$$\begin{aligned} \mathcal{G}(\omega^+; \mathbf{R}, \mathbf{R}') &= -\frac{1}{v_b} I \delta_{\mathbf{R}, \mathbf{R}'} \\ &+ \frac{\Omega}{v_b^2} \int_{\text{B.Z.}} \frac{d\mathbf{k}}{(2\pi)^3} e^{i\mathbf{k} \cdot (\mathbf{R} - \mathbf{R}')} T_{\mathbf{k}}(\omega), \end{aligned} \quad (10)$$

for positions on the lattice. The advantage of this expression lies in the fact that for $\mathbf{R} = \mathbf{R}'$ the integral in (10) is finite due to the finite support of the Brillouin zone. Contrary to the case of free space no singularities are present in the Green's function \mathcal{G} . Employing Eqs. (7) and (8) to calculate $T_{\mathbf{k}}$ one finally obtains the microscopic result

$$\mathcal{G}(\omega^+; \mathbf{R}, \mathbf{R}') = (\tilde{\alpha}/\alpha_b)^2 \mathcal{L}_{\text{Lor}}^2 \mathcal{G}_{\text{diel}}(\omega^+; \mathbf{R} - \mathbf{R}'), \quad (11)$$

where $\mathbf{R} \neq \mathbf{R}'$ and $\mathcal{G}_{\text{diel}}(\omega^+; \mathbf{r}) = \mathcal{G}_0(\sqrt{\varepsilon} \omega^+; \mathbf{r})$ describes [12,14,19] an ordered dielectric on a macroscopic scale. Generally, neither the factor $(\tilde{\alpha}/\alpha_b)^2$ in (11) nor the expression (10) for \mathcal{G} are given in the literature [12,14,19]. The electric field solutions can be obtained from \mathcal{G} . Note that the field solution \mathbf{E} at the position \mathbf{R} of a dipole includes depolarization effects and, therefore, is unequal to the conventionally used field \mathbf{E}^{exc} which excites the dipole [20]. Both fields are related through expressions for the dipole moment $\mathbf{d} = \tilde{\alpha} \mathbf{E}^{\text{exc}}$

and $\mathbf{d} = \alpha_b \mathbf{E}$, so $\tilde{\alpha}/\alpha_b$ is an internal-field factor [18]. Equation (11), except for the factors $\tilde{\alpha}/\alpha_b$ appropriate for lattice positions, also applies to other positions with cubic symmetry in the unit cell, or in an averaged sense for all space when *excluding* the depolarization fields at $\{\mathbf{R}\}$. These results show that the microscopic and macroscopic field solutions differ by a factor \mathcal{L}_{Lor} in accordance with textbook derivations.

We now treat the case of a lattice including either a substitutional or interstitial impurity atom. In an effective two-level or harmonic-oscillator approach the polarizability α_I of a single impurity in free space is [18]

$$\alpha_I(\omega) = \alpha_I(0) \times \frac{\omega_0^2}{\omega_0^2 - \omega^2 - i(\Gamma_0 \omega^3 / \omega_0^2)}, \quad (12)$$

with resonance frequency ω_0 and width Γ_0 . Invoking the optical theorem gives the relation $\Gamma_0 = \alpha_I(0) \omega_0^4 / 6\pi c_0^3$, which is consistent with Fermi's golden rule result for Γ_0 by taking the static polarizability $\alpha_I(0) = 2|\boldsymbol{\mu}|^2 / \varepsilon_0 \hbar \omega_0$ in terms of the dipole matrix element $\boldsymbol{\mu}$ of two atomic levels. One also has an (assumed given) point-potential strength $v_{b,I}$. The Green's function

$$\mathcal{G}_m = \mathcal{G} + \mathcal{G} \cdot \mathcal{T}_m \cdot \mathcal{G} \quad (13)$$

describes the system of lattice and impurity, in which, respectively, for a substitutional impurity at \mathbf{R}_s ($m = s$)

$$\mathcal{T}_s = [(v_{b,I} - v_b)^{-1} I - \mathcal{G}(\omega^+; \mathbf{R}_s, \mathbf{R}_s)]^{-1}, \quad (14)$$

and for an interstitial impurity at \mathbf{r}_i ($m = i$)

$$\mathcal{T}_i = [v_{b,I}^{-1} I - \mathcal{G}(\omega^+; \mathbf{r}_i, \mathbf{r}_i)]^{-1}. \quad (15)$$

Note the dependence on the difference of the relevant potential strengths in (14). The removal of a lattice dipole was not considered explicitly in the literature [11–15]. The imaginary parts of \mathcal{G} in (14) and (15) are connected to the RDOS and are proportional to $\sqrt{\varepsilon} \mathcal{L}_{\text{Lor}}^2$. On the other hand, the potential terms, as well as the real parts of the doubly on-site \mathcal{G} , differ from one another. Using Eqs. (4), (9), and (10) the t -matrices can be written as

$$\mathcal{T}_s = v_b^2 \left[(1/\tilde{t} - 1/\tilde{t}_I)^{-1} I - \Omega \int_{\text{B.Z.}} \frac{d\mathbf{k}}{(2\pi)^3} T_{\mathbf{k}} \right]^{-1}, \quad (16)$$

$$\mathcal{T}_i = \{t_i^{-1} I - [\mathcal{G}_0 \cdot \mathcal{T} \cdot \mathcal{G}_0](\omega^+; \mathbf{r}_i, \mathbf{r}_i)\}^{-1}, \quad (17)$$

from which the secular equations determining the impurity resonances readily follow. The resulting resonances turn out to be unequal. For static dipole-dipole interactions, these equations were derived in Ref. [19] in the description using exciting fields.

For the substitutional case the polarizability $\alpha_s(\omega) I \equiv -(c_0/\omega)^2 (\tilde{\alpha} \mathcal{L}_{\text{Lor}} / \alpha_b)^2 \mathcal{T}_s$ concurs with usual descriptions. The factor $(\tilde{\alpha} \mathcal{L}_{\text{Lor}} / \alpha_b)^2$ originates from $\mathcal{G} \cdot \mathcal{G}$ [see Eq. (13)]. This polarizability simplifies to

$$\alpha_s(0) = L_{\text{OB}}[\alpha_I(0)/\Omega] \mathcal{L}_{\text{Lor}} \times [\alpha_I(0) - \alpha(0)], \quad (18)$$

for $\omega \rightarrow 0$. Two important remarks concerning α_s are in order. In the first place, Eq. (18) can be written as

$$\alpha_s = L_{\text{OB}}(\alpha_I/\Omega) \mathcal{L}_{\text{emp}} \alpha_I - (\varepsilon - 1) \mathcal{L}_{\text{emp}} \Omega, \quad (19)$$

where the terms, respectively, describe the polarizabilities of a dielectric-modified impurity and of a cavity inside a macroscopic dielectric. Second, within the OB approach the combined polarizability of dipole α_I and cavity with volume Ω , observed outside the cavity, corresponds exactly with α_s demonstrating that the case of a substitutional can be investigated using a standard continuum description.

The desired properties at resonance are as follows. The polarizability of the “dressed” impurity near resonance has a Lorentzian structure analogous to Eq. (12): $\alpha_{s,I}(\omega) = \alpha_{s,I}(0) \omega_0^2(\varepsilon) / [\omega_0^2(\varepsilon) - \omega^2 - i\Gamma_0(\varepsilon) \omega^3 / \omega_0^2(\varepsilon)]$ with parameters given by

$$\omega_0^2(\varepsilon) / \omega_0^2 = [1 - 2\alpha_I(0)(\varepsilon - 1) \mathcal{L}_{\text{emp}} / 9\varepsilon \Omega], \quad (20)$$

$$\begin{aligned} \alpha_{s,I}(0) &= L_{\text{OB}}[\alpha_I(0)/\Omega] \mathcal{L}_{\text{emp}} \times \alpha_I(0), \\ &= \mathcal{L}_{\text{emp}}^2 \omega_0^2 / \omega_0^2(\varepsilon) \times \alpha_I(0), \end{aligned} \quad (21)$$

$$\Gamma_0(\varepsilon) / \Gamma_0 = \sqrt{\varepsilon} \mathcal{L}_{\text{emp}}^2 \times [\omega_0^2(\varepsilon) / \omega_0^2], \quad (22)$$

where $\omega_0(1) = \omega_0$ and $\Gamma_0(1) = \Gamma_0$. The resonance frequency is redshifted and the linewidth behaves in accordance with the empty-cavity description.

For an interstitial at a point with cubic symmetry one obtains an $\alpha_i(\omega)$ of the form (12) with parameters

$$\omega_0^2(\varepsilon) / \omega_0^2 = [1 - 2\alpha_I(0)(\varepsilon - 1) \mathcal{L}_{\text{Lor}} / 9\varepsilon \Omega], \quad (23)$$

$$\alpha_i(0) = \mathcal{L}_{\text{Lor}}^2 \omega_0^2 / \omega_0^2(\varepsilon) \times \alpha_I(0), \quad (24)$$

$$\Gamma_0(\varepsilon) / \Gamma_0 = \sqrt{\varepsilon} \mathcal{L}_{\text{Lor}}^2 \times [\omega_0^2(\varepsilon) / \omega_0^2]. \quad (25)$$

In the calculation sums such as $\sum_{\mathbf{R}} e^{i\mathbf{k} \cdot (\mathbf{R} - \mathbf{r}_i)} \mathcal{G}_0(\omega^+; \mathbf{r}_i - \mathbf{R})$ appearing in the term $\mathcal{G}_0 \cdot \mathcal{T} \cdot \mathcal{G}_0$ of \mathcal{T}_i have been replaced using expression (7). The width of the resonance now includes Lorentz local-field factors instead of the empty-cavity ones. The polarizability α_i clearly does not coincide with the results for a substitutional impurity. For other interstitial positions in the unit cell the widths will follow Eq. (25) apart from possible small corrections to (23). Obviously, these results apply only when atomic sizes compared to the lattice parameter a are such that interstitials may occur.

We now discuss the relevance of our results (20)–(25) to impurities in disordered dielectrics. The parameters a and Ω^{-1} are a measure of the mean interparticle distance and the density of particles forming the dielectric, respectively. The LLR (8), with α instead of $\tilde{\alpha}$, can be derived for disordered media using a microscopic multiple-scattering theory [8] in which the excluded-volume part of many-particle correlations is taken into account to all orders in the density. The remainder of the correlations as well as scattering more than once from the same particle, called recurrent scattering, is then neglected. For lattice systems, however, the latter are taken into account in Eqs. (6), (8), (14), and (15), since these expressions are

exact. Inside lattices interstitials have a different local environment compared to particles residing at lattice points $\{\mathbf{R}\}$. In particular, they have a *lower* coordination number. A substitutional impurity takes the place of a lattice particle, which then has to be removed. For disordered media it now becomes clear that in order to distinguish between “interstitial” and “substitutional” positions one has to consider correlations characterizing the local environment. Interstitial behavior occurs when impurities do not influence the many-particle correlations of the other particles *and* have lower coordination numbers. These impurities then interact with the usual Lorentz local field present inside a dielectric medium. On the other hand, an impurity inside a disordered system may be defined as a substitutional when it has comparable correlations and coordination numbers as the particles constituting the dielectric. A *larger* coordination number occurs when the size of the impurity is larger than the volume Ω . In the cases of comparable and larger coordination numbers the macroscopic OB approach is applicable. Subsequently, the empty-cavity behavior (21) and (22) will apply in agreement with recent experimental results [16]. At very low densities, the distinction between the two types of impurities will not be relevant, since any distinct features in correlations will disappear and $\mathcal{L}_{\text{Lor}} \approx \mathcal{L}_{\text{emp}}$ to first order in Ω^{-1} . The presence of interstitials is most easily realized in solid dielectrics. In the case of fluids, they will generally not occur.

It is interesting to remark on the effective dielectric constant of mixtures. To the same level of approximation used to obtain the LLR for pure fluids one can derive the LLR or Maxwell-Garnett expression [8] for a disordered collection of dipoles A and I with polarizabilities α and α_I , respectively. To lowest order in the density ρ_I of dipoles I one gets $\epsilon_{\text{eff}} = \epsilon + \rho_I \mathcal{L}_{\text{Lor}}^2 \alpha_I$, where ϵ is given by (8), suggesting that using \mathcal{L}_{Lor} seems appropriate for both types of impurities. However, for densities of dipoles A such that dipoles I are basically immersed in a host with dielectric constant ϵ another Maxwell-Garnett expression is appropriate [21]. Assuming for convenience that the dipoles I are dielectric spheres with volume V_{sp} one then obtains the following to lowest order in ρ_I : $\epsilon_{\text{eff}} = \epsilon + \rho_I \alpha'_I$, where α'_I is the effective polarizability of the dielectric sphere embedded in the host. Analogous to Eq. (19), $\alpha'_I = L_{\text{OB}}(\alpha_I/V_{\text{sp}})\mathcal{L}_{\text{emp}}\alpha_I - (\epsilon - 1)\mathcal{L}_{\text{emp}}V_{\text{sp}}$, where α_I is the free-space polarizability of the sphere. The latter Maxwell-Garnett description coincides with the OB approach and implies that for large volume fractions of dipoles A the impurities I have a substitutional character.

In summary, we have calculated the modifications to resonance properties of impurities induced by a dielectric host. The local-field corrections given in the literature are shown to be adequate, while conditions under which they occur have now been identified. The substitutional case

occurs prevalently. For the case of interstitials textbook local-field derivations are applicable.

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