Local-field corrections to the decay rate of excited molecules in absorbing cavities: The Onsager model

M. S. Tomaš
Rudjer Bošković Institute, P.O. Box 180, 10002 Zagreb, Croatia
(Received 31 August 2000; published 18 April 2001)

The decay rate and the classical radiation power of an excited molecule (atom) in the center of a dispersive and absorbing dielectric sphere, taken as a simple model of a cavity, are calculated adopting the Onsager model for the local field. The local-field correction factor to the external (radiation and absorption) power loss of the molecule is found to be $|3\varepsilon(\omega)/[3\varepsilon(\omega)+1]|^2$, with $\varepsilon(\omega)$ being the dielectric function of the sphere. However, local-field corrections to the total decay rate (power loss) of the molecule are found to be much more complex including those to the decay rate in the infinite cavity medium, as derived recently by Scheel *et al.* [Phys. Rev. A **60**, 4094 (1999)], and similiar corrections to the cavity-induced decay rate. The results obtained can be cast into general forms that do not rely on the specific cavity shape and molecule position considered. This suggests the general results for the local-field corrections to the decay rate and to the external power loss of a molecule in an absorbing cavity that are valid for molecule positions away from the cavity walls.

DOI: 10.1103/PhysRevA.63.053811 PACS number(s): 42.50.Lc, 42.60.Da, 33.50.−j

I. INTRODUCTION

Within macroscopic electrodynamics, the decay rate Γ of an excited molecule (atom) at a position $\mathbf{r}_0$ in an absorbing cavity is given by

$$\Gamma = \frac{2\omega^2}{\hbar c^2}\mathbf{p}_{fi}^{*}\cdot \mathrm{Im}\,\vec{\mathbf{G}}(\mathbf{r}_0,\mathbf{r}_0;\omega)\cdot\mathbf{p}_{fi}, \tag{1}$$

with $\vec{\mathbf{G}}(\mathbf{r},\mathbf{r}_0;\omega)$ being the (classical) dyadic Green function for the system, $\mathbf{p}_{fi}$ the relevant dipole matrix element, and ω the transition frequency. This result is most simply obtained using the classical theory of molecular (radiative) decay [1] in conjuction with the correspondence principle and extending it straightforwardly to absorbing systems [2]. Within QED, however, it is derived by employing the quantized form of the macroscopic field in absorbing systems obtained rather recently [3–5] and using the Fermi golden rule [6,7] or by solving Heisenberg's equations of motion for the molecule and the field in the Markov approximation [8,9].

Splitting the Green function into the translationally invariant part $\vec{\mathbf{G}}^0(\mathbf{r}-\mathbf{r}_0;\omega)$ and the scattering part $\vec{\mathbf{G}}^{\mathrm{sc}}(\mathbf{r},\mathbf{r}_0;\omega)$, the decay rate can be generally expressed as

$$\Gamma = \Gamma^0 + \Gamma^{\mathrm{sc}}, \tag{2}$$

where Γ^0 is the decay rate as would be in the infinite cavity medium and Γ^{sc} is the corresponding cavity-induced decay rate. Upon an appropriate regularization of $\vec{\mathbf{G}}^0(\mathbf{r}-\mathbf{r}_0;\omega)$, the rate Γ^0 is given by [2,7]

$$\Gamma^0 = \Gamma_{\mathrm{free}}\left[\frac{3}{2}\frac{\varepsilon''(\omega)}{|\varepsilon(\omega)|^2}\left(\frac{c}{\omega R_m}\right)^3 + \eta(\omega)\right], \tag{3}$$

where

$$\varepsilon(\omega) = [\eta(\omega) + i\kappa(\omega)]^2 \tag{4}$$

is the dielectric function of the cavity medium, R_m an effective molecule-medium distance, and

$$\Gamma_{\mathrm{free}} = \frac{4\omega^3|\mathbf{p}_{fi}|^2}{3\hbar c^3} \tag{5}$$

is the free-space spontaneous emission (SE) rate. The first contribution to Γ^0 in Eq. (3) is identified as the nonradiative decay rate Γ^0_{nr} due to the near-field mediated transfer of the molecular energy to the surrounding medium, whereas the second one is the familiar decay rate Γ^0_{rad} due to the radiation losses of the molecule in the medium, i.e., the SE rate Γ^0_{SE} [6,10,11].

For an optically dense cavity medium, the above result has to be improved by accounting for the difference between the macroscopic field used in its derivation and the actual (local) field with which the molecule interacts. Restricting ourselves to low-density cavity media in our previous consideration of the molecular decay [2] as well as of the spontaneous emission spectrum [12] in an absorbing planar cavity, we ignored this difference implicitly assuming, however, that the decay rate corrected for the effect of the local field in an isotropic cavity was of the form

$$\tilde{\Gamma}_{\mathrm{loc}} = \mathcal{L}[\Gamma^0 + \Gamma^{\mathrm{sc}}], \tag{6}$$

where $\mathcal{L}$ is an appropriate generalization of the local-field correction factor in the lossless case. Thus, as suggested by Barnett *et al.* for the decay rate $\tilde{\Gamma}^0_{\mathrm{loc}}$ in the infinite cavity [6,7], one would have

$$\mathcal{L}_{\mathrm{Lor}}(\varepsilon) = \left|\frac{\varepsilon(\omega)+2}{3}\right|^2 \tag{7}$$

in the Lorentz (virtual-cavity) model [13–17] and

$$\mathcal{L}_{\mathrm{Ons}}(\varepsilon)=\left|\frac{3\varepsilon(\omega)}{2\varepsilon(\omega)+1}\right|^2 \tag{8}$$

in the Onsager (real-cavity) model [18–21] for the local field. In either case, the effect of the local field would cancel when dealing with the normalized rate $\hat{\Gamma}=\tilde{\Gamma}_{\mathrm{loc}}/\tilde{\Gamma}^0_{\mathrm{loc}}=\Gamma/\Gamma^0$.

The conjecture expressed by Eqs. (6)–(8) has recently been shown to be incorrect for both models for the local field, which we indicate in Eq. (6) by putting the tilde sign on Γ. First, Scheel et al. [22] demonstrated that, in the Lorentz model, a proper inclusion of the (quantum) noise polarization in the local field led to a more complex ω-dependence of the decay rate Γ^0_{loc} in an absorbing, medium than that given by the simple product $\mathcal{L}_{\mathrm{Lor}}\Gamma^0$ (see Eqs. (49) and (50) of Ref. [22]). Their result is in full agreement with Γ^0_{loc} in absorbing dielectrics derived by Fleischhauer using a microscopic approach [23]. In a subsequent work Scheel et al. [8] showed that the decay rate Γ^0_{loc} in the Onsager model for the local field was also much more complex in the absorption case than that given by the product $\mathcal{L}_{\mathrm{Ons}}\Gamma^0$ [see Eq. (27) below].

Knowing the decay rate Γ^0_{loc} in an infinite cavity (medium), it is natural to seek the decay rate Γ_{loc} in a finite cavity, i.e., in view of Eq. (2), the cavity-induced rate $\Gamma^{\mathrm{sc}}_{\mathrm{loc}}$. Formally, the calculation of $\Gamma^{\mathrm{sc}}_{\mathrm{loc}}$ is most straightforwardly performed if one adopts the Onsager model for the local field. In this model, one assumes the molecule in the center of a spherical hole of radius small compared with the transition wavelength often referred to as the Onsager cavity. In view of Eq. (1), the problem then reduces to the calculation of the Green function for the system, with the Onsager cavity at the source position. Owing to the combined symmetry of such a composite system, however, this represents a difficult task, which for most relevant cavity geometries demands approximative or numerical methods [24]. Therefore, in order to take a (first) step towards determination of $\Gamma^{\mathrm{sc}}_{\mathrm{loc}}$, in this work we consider a special case where the molecule is located in the center of an absorbing sphere taken as a simple model of a dielectric cavity. High symmetry of this configuration enables one to perform a simple and exact calculation of $\Gamma^{\mathrm{sc}}_{\mathrm{loc}}$ in the Onsager model as well as of the power loss $W^{\mathrm{loc}}_{\mathrm{ext}}$ of the molecule outside the sphere. In turn, these results (may) provide a lead to the corresponding solutions for a more general configuration. We note that this special configuration has recently been considered by Cao *et al*. [21] in their calculation of the local-field corrections to the SE rate in a purely dispersive medium. We partially adopt their approach, generalizing it to an absorbing multilayered spherical system. It should also be noted that owing to the existence of high-Q resonances and, accordingly, great ability of enhancing optical processes [26], dielectric microspheres are very attractive objects for cavity QED studies. Thus, modification of the decay rate and the radiation intensity of an excited molecule (atom) in or near a (lossless) microsphere has been theoretically considered in both the weak [9,27–31] and the strong [9,31,32] molecule-field coupling limit, and experimental observations of modified fluorescence intensity have also been reported [33,34].

Owing to its intuitive clarity and easy visualization, we prefer a classical discourse in this work. Therefore, in Sec. II we recall the classical approach to the problem of the decaying molecule and rederive Eq. (3) raising, through a plausible argument, a question on the completeness of this result for absorbing media. In Sec. III we first obtain formal results for the decay rate and the power loss in the external layer of an excited molecule in the center of an absorbing multilayered spherical system. We then apply these general results to a medium with the Onsager cavity and provide an alternative derivation of Γ^0_{loc} to that given in Ref. [8]. Subsequently, by considering an absorbing dielectric sphere with and without the Onsager cavity, we determine $\Gamma^{\mathrm{sc}}_{\mathrm{loc}}$ and $W^{\mathrm{loc}}_{\mathrm{ext}}$ in terms of the corresponding quantities for the bare sphere. In Sec. IV we briefly illustrate the effect of the local field in this model on $\Gamma^{\mathrm{sc}}_{\mathrm{loc}}$ and the total rate Γ_{loc}. The main points of this work are summarized in Sec. V and the necessary mathematical background is given in Appendices A and B.

II. PRELIMINARIES

In the classical approach, an excited molecule is represented by a point dipole $\mathbf{p}\exp(-i\omega t)$ at the molecular position $\mathbf{r}_0$ oscillating with the frequency of the transition, ω. The molecular decay rate Γ is then related through $\Gamma=W/\hbar\omega$ to the power

$$W=\frac{\omega}{2}\mathrm{Im}[\mathbf{p}^*\cdot\mathbf{E}(\mathbf{r}_0,\mathbf{r}_0;\omega)] \tag{9}$$

lost by the dipole in supporting its own field. Equation (1) is then obtained, introducing the Green function of the system through

$$\mathbf{E}(\mathbf{r},\mathbf{r}_0;\omega)=\frac{\omega^2}{c^2}\vec{\mathbf{G}}(\mathbf{r},\mathbf{r}_0;\omega)\cdot\mathbf{p}, \tag{10}$$

noting that $\vec{\mathbf{G}}(\mathbf{r}_0,\mathbf{r}_0;\omega)$ is the diagonal dyadic and using the correspondence principle to let $\mathbf{p}\rightarrow 2\mathbf{p}_{fi}$ [35].

The dipole field (Green function) in a cavity can always be written as

$$\mathbf{E}(\mathbf{r},\mathbf{r}_0;\omega)=\mathbf{E}^0(\mathbf{R};\omega)+\mathbf{E}^{\mathrm{sc}}(\mathbf{r},\mathbf{r}_0;\omega), \tag{11}$$

where [2]

$$\mathbf{E}^0(\mathbf{R};\omega)=\frac{1}{\varepsilon}\left[\frac{3\hat{\mathbf{R}}\hat{\mathbf{R}}-\vec{\mathbf{I}}}{R^3}(1-ikR)-\frac{4\pi}{3}\vec{\mathbf{I}}\delta(\mathbf{R})+k^2\frac{\vec{\mathbf{I}}-\hat{\mathbf{R}}\hat{\mathbf{R}}}{R}\right]\cdot\mathbf{p}e^{ikR}, \tag{12}$$

with $\vec{\mathbf{I}}$ being the unit dyadic, $\mathbf{R}=\mathbf{r}-\mathbf{r}_0$, $\hat{\mathbf{R}}=\mathbf{R}/R$, and

$$k(\omega)\equiv\sqrt{\varepsilon(\omega)}\,\frac{\omega}{c}=k'(\omega)+ik''(\omega), \tag{13}$$

is the field of the dipole as would be in the infinite cavity (medium), and $\mathbf{E}^{\mathrm{sc}}(\mathbf{r},\mathbf{r}_0;\omega)$ is the component of the dipole

field scattered from the cavity walls. Owing to the singular longitudinal (near-field) component of $\mathbf{E}^0(\mathbf{R};\omega)$

$$\mathbf{E}^0_{\parallel}(\mathbf{R};\omega)=\frac{1}{\varepsilon}\left[\frac{3\hat{\mathbf{R}}\hat{\mathbf{R}}-\vec{\mathbf{I}}}{R^3}-\frac{4\pi}{3}\vec{\mathbf{I}}\delta(\mathbf{R})\right]\cdot\mathbf{p}, \tag{14}$$

it is the calculation of Γ^0 that represents a difficult step in determining the decay rate in absorbing systems.

To remove the singularity from $\mathbf{E}^0_{\parallel}(\mathbf{R};\omega)$ in the spirit of the macroscopic-field approach, one may average this component of the dipole field over an appropriately chosen spherical volume $V_m=(4\pi/3)R_m^3$ around the molecule [7] or regularize it by letting $\delta(\mathbf{R})\to 1/V_m$ [2,25]. In either case, one finds that

$$\mathbf{E}^0_{\parallel}(\mathbf{R};\omega)|_{R\to 0}=-\frac{1}{\varepsilon}\frac{4\pi}{3V_m}\mathbf{p}. \tag{15}$$

Since for the dipole transverse field $\mathbf{E}^0_{\perp}(\mathbf{R};\omega)=\mathbf{E}^0(\mathbf{R};\omega)-\mathbf{E}^0_{\parallel}(\mathbf{R};\omega)$ we find

$$\mathbf{E}^0_{\perp}(\mathbf{R};\omega)|_{R\to 0}=ik\frac{\omega^2}{c^2}\frac{2}{3}\mathbf{p}, \tag{16}$$

this leads to the total classical dipole power loss

$$W^0=\frac{\omega^4|\mathbf{p}|^2}{3c^3}\left[\frac{3}{2}\frac{\varepsilon''(\omega)}{|\varepsilon(\omega)|^2}\left(\frac{c}{\omega R_m}\right)^3+\eta(\omega)\right] \tag{17}$$

and, accordingly, to the decay rate Γ^0 given by Eq. (3).

That the above result oversimplifies the frequency dependence of the molecular power loss (decay rate) in absorbing media becomes clear if one tries to obtain W^0 using the Poynting theorem. In this way, by calculating the dipole energy flow W^0_f through a spherical surface around the dipole and the energy W^0_a absorbed per second in the enclosed volume (see Appendix A), we find

$$W^0=\frac{\omega^4|\mathbf{p}|^2}{3c^3}\left[\frac{\varepsilon''}{|\varepsilon|^2}|(1-ikR_c)e^{ikR_c}|^2\left(\frac{c}{\omega R_c}\right)^3+\eta e^{-2k''R_c}\right], \tag{18}$$

where R_c is (formally) the lower limit of the radial integration in W^0_a. Owing to their characteristic dependence on the dielectric function of the medium, we refer to two terms in this equation as the absorption ($\sim\varepsilon''$) and the radiation ($\sim\eta$) contribution to W^0, respectively. Since this notation may associate to W^0_a and W^0_f as the respective origins of these contributions, we stress that both W^0_a and W^0_f are needed to obtain each of them, as is clear from the derivation in Appendix A.

To obtain the dipole power loss from Eq. (18), the $R_c\to 0$ limit should eventually be taken. However, consider R_c as a small ($R_c\ll\lambda$) but finite cutoff for the moment. In this case, expanding W^0 in powers of $\omega R_c/c$, we find

$$\tilde{W}^0=\frac{\omega^4|\mathbf{p}|^2}{3c^3}\left\{\frac{\varepsilon''(\omega)}{|\varepsilon(\omega)|^2}\left\{\left(\frac{c}{\omega R_c}\right)^3+\varepsilon'(\omega)\frac{c}{\omega R_c}-\frac{2}{3}[\eta(\omega)\varepsilon''(\omega)+\kappa(\omega)\varepsilon'(\omega)]\right\}+\eta(\omega)+O\left(\frac{\omega R_c}{c}\right)\right\}. \tag{19}$$

In addition to a near-field term, which very much resembles the corresponding term in Eq. (17), now we have two new absorption terms. The most striking is the appearance of another R_c-free term, which therefore persists even in the $R_c\to 0$ limit. This implies that a corresponding term must appear in any calculation of W^0, which indicates that Eq. (17) is, in this respect, incomplete.

Taking R_c in Eq. (18) as a cutoff is equivalent to setting $\mathbf{E}^0(\mathbf{R};\omega)=0$ for $R<R_c$. Since this field does not obey Maxwell's equations, it is clearly incorrect to regard $\tilde{W}^0$ as the dipole power loss, which we have emphasized by using the tilde. On the other hand, extending the macroscopic field down to intermolecular distances ($R_c\to 0$) is not justified as the actual field acting on the molecule may largely differ from it. These ambiguities concerning W^0 are naturally resolved within an exact macroscopic-field approach in the following section, where we adopt the Onsager (real cavity) model for the local field and therefore assume the molecule in the center of an empty spherical cavity with the radius R_c small compared with the transition wavelength λ. Since in this case, the longitudinal component $\mathbf{E}^0_{\parallel}(\mathbf{R};\omega)$ of the dipole field does not contribute to the molecular power loss, no singularity appears in the theory.

III. LOCAL-FIELD CORRECTIONS

Consider an excited molecule (dipole) in the center of an N-layered spherical system as depicted in Fig. 1. In this case, the power loss W^{sc} of the molecule may be written as

$$W^{\mathrm{sc}}=\frac{\omega}{2}\mathrm{Im}[\mathbf{p}^*\cdot\mathbf{E}^{\mathrm{sc}}_1(\mathbf{r};\omega)|_{r\to 0}], \tag{20}$$

where $\mathbf{E}^{\mathrm{sc}}_1(\mathbf{r};\omega)=\mathbf{E}^{\mathrm{sc}}_1(\mathbf{r},\mathbf{r}_0;\omega)|_{r_0\to 0}$ is the scattered part of the dipole field in the central region. The calculation of the dipole field in this configuration is outlined in Appendix B, assuming, for simplicity, that $\mathbf{p}=p\hat{\mathbf{z}}$. Using Eq. (B6), we find from Eq. (20) that ($k_0=\omega/c$)

$$W^{\mathrm{sc}}=W_{\mathrm{free}}\mathrm{Re}\,\sqrt{\varepsilon_1}C_1^N,\quad W_{\mathrm{free}}=\frac{ck_0^4|\mathbf{p}|^2}{3}, \tag{21}$$

where C_1^N is the corresponding reflection coefficient. In the case of the empty central region ($W^0=W_{\mathrm{free}}$), we therefore have for the normalized total decay rate $\hat{\Gamma}=W/W_{\mathrm{free}}$ of the molecule

$$\hat{\Gamma}=1+\mathrm{Re}[C_1^N]_{\varepsilon_1=1}. \tag{22}$$

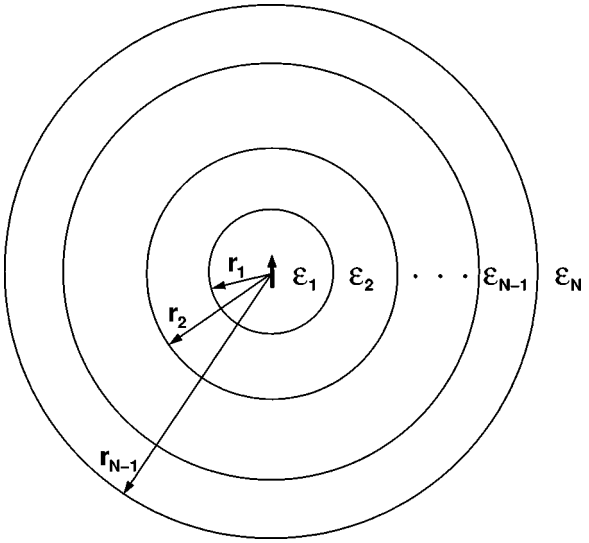

FIG. 1. System considered in this paper. All layers are assumed absorbing and are described by the complex dielectric function $\varepsilon_i(\omega)$.

Identifying the central sphere with the Onsager cavity, this general result provides a direct way for inclusion of the local-field corrections to the decay rate in spherical multilayered systems.

Of obvious interest is also the radiation power $W_N^{\rm rad}$ of the molecule or, generally, the total power loss W_N in the outer region of the multilayer. By comparing Eqs. (A1a) and (B5), we see that the dipole field $\mathbf{E}_N(\mathbf{r};\omega)$ in this region is the same as the field produced in the infinite medium (N) of the dipole ($C_N^N \equiv C_{N+}^N$)

$$\mathbf{p}_N = \frac{\varepsilon_1}{\varepsilon_N} C_N^N \mathbf{p}. \tag{23}$$

Accordingly, provided that we let $\mathbf{p}\to\mathbf{p}_N$, $\varepsilon\to\varepsilon_N$, and $R_c \to r_{N-1}$, we can adopt all results concerning the dipole power loss in an infinite medium derived in Appendix A. For example, with these replacements, the angular distribution of radiation $dW_N^{\rm rad}/d\Omega$ is obtained by keeping only the radiation field ($\sim 1/r^2$) contribution to Eq. (A2),

$$\frac{dW_N^{\rm rad}}{d\Omega} = \eta_N \frac{ck_0^4|\mathbf{p}|^2}{8\pi}\left|\frac{\varepsilon_1}{\varepsilon_N} C_N^N\right|^2 e^{-2k_N'' r}\sin^2\vartheta, \tag{24}$$

the radiation power $W_N^{\rm rad}$ is given by the last term in Eq. (A3) and the total power loss W_N by Eq. (18). Regarding the central sphere as the Onsager cavity, we see that the local-field correction factor to W_N (and $W_N^{\rm rad}$) is given by $|C_N^N/\varepsilon_N|^2$ for $\varepsilon_1 = 1$ and in the limit $k_0 r_1 = k_0 R_c \ll 1$.

A. Infinite medium

To calculate local-field corrections to the decay rate in an infinite medium with the dielectric function $\varepsilon(\omega)$, we consider a two-layered system consisting of this medium with the Onsager cavity cut around the origin. Then, as was done by Scheel *et al.* [8], $\hat{\Gamma}_{\rm loc}^0$ is straightforwardly obtained using Eq. (22) and expanding the reflection coefficient $C_1^2(1,\varepsilon;R_c)$ given by Eq. (B7) in powers of $\rho_1 = k_0 R_c \ll 1$. It is very instructive to rederive this result by applying the Poynting theorem to a spherical surface around the molecule enclosing the Onsager cavity and thus determine the power loss $W_{\rm loc}^0$ of the molecule. We note that we have performed such a calculation in Appendix A to obtain Eq. (18). Thus, $W_{\rm loc}^0$ is given by this equation provided that we let $\mathbf{p}\to\mathbf{p}_{\rm eff}$, where [cf. Eq. (23)]

$$\mathbf{p}_{\rm eff} = \frac{1}{\varepsilon} C_2^2(1,\varepsilon;R_c)\mathbf{p}$$

and regard R_c as the Onsager cavity radius.

Expanding the coefficient $C_2^2(1,\varepsilon;R_c)$ given by Eq. (B8) for small $\rho_1 = k_0 R_c$, we find

$$\mathbf{p}_{\rm eff} = \frac{3\varepsilon}{2\varepsilon+1}\left[1 - \frac{10\varepsilon^2 - 9\varepsilon - 1}{10(2\varepsilon+1)}(k_0R_c)^2 - i\frac{2}{3}\frac{\varepsilon^{3/2}(\varepsilon-1)}{2\varepsilon+1}(k_0R_c)^3 + O\{(k_0R_c)^4\}\right]\mathbf{p}. \tag{25}$$

Owing to the $(k_0R_c)^{-3}$ factor in the first term on the right-hand side (rhs) of Eq. (18), in this term we must use

$$\left|(1 - i\sqrt{\varepsilon}k_0R_c)e^{i\sqrt{\varepsilon}k_0R_c}\mathbf{p}_{\rm eff}\right|^2 = \left|\frac{3\varepsilon}{2\varepsilon+1}\right|^2\left[1 + \frac{1}{5}{\rm Re}\frac{14\varepsilon+1}{2\varepsilon+1}(k_0R_c)^2 - 2\,{\rm Im}\frac{\varepsilon^{3/2}}{2\varepsilon+1}(k_0R_c)^3 + O\{(k_0R_c)^4\}\right]|\mathbf{p}|^2. \tag{26}$$

Since the second term on the rhs of Eq. (18) is a well-behaved function of k_0R_c, in this term it is sufficient to let $\mathbf{p}_{\rm eff} = 3\varepsilon\mathbf{p}/(2\varepsilon+1)$. In this way, for the normalized decay rate $\hat{\Gamma}_{\rm loc}^0 = W_{\rm loc}^0/W_{\rm free}$ we obtain

$$\hat{\Gamma}_{\rm loc}^0 = \left|\frac{3\varepsilon(\omega)}{2\varepsilon(\omega)+1}\right|^2\left\{\frac{\varepsilon''(\omega)}{|\varepsilon(\omega)|^2}\left[\left(\frac{c}{\omega R_c}\right)^3 + \frac{28|\varepsilon(\omega)|^2 + 16\varepsilon'(\omega) + 1}{5|2\varepsilon(\omega)+1|^2}\left(\frac{c}{\omega R_c}\right) - 2\frac{2\kappa(\omega)|\varepsilon(\omega)|^2 + \kappa(\omega)\varepsilon'(\omega) + \eta(\omega)\varepsilon''(\omega)}{|2\varepsilon(\omega)+1|^2}\right] + \eta(\omega) + O\left(\frac{\omega R_c}{c}\right)\right\}. \tag{27}$$

The above result coincides with that of Scheel *et al.* [22]. This time, however, the origin of various contributions to $\hat{\Gamma}_{\rm loc}^0$ along with their separate local-field corrections can be clearly identified. As seen from comparison with Eq. (17), while the near-field and the radiation-field terms get multiplied by $\mathcal{L}_{\rm Ons}$ as expected [7], it is the appearance of the

additional absorption terms already signaled in Eq. (19) that represents essentially new corrections to $\hat{\Gamma}^0$ in absorbing media. Of these two terms, the most interesting is the (usually) negative R_c-free contribution. This term effectively adds to the radiation-field contribution (η) to the decay rate in absorbing media and tends to diminish the overall rate.

B. Cavity

Having determined $\Gamma^0_{\rm loc}$, the next step is to consider the decay rate $\Gamma_{\rm loc}$ in the general case when the molecule is embedded in an inhomogeneous system, i.e., in a cavity. In order to determine $\Gamma_{\rm loc}$, we consider the decaying molecule in the center of a dielectric sphere of radius R and the dielectric function $\varepsilon(\omega)$ immersed in an external medium with the dielectric function $\varepsilon_{\rm ext}(\omega)$. In this case, the relevant reflection coefficient to be inserted in Eq. (22) is $C_1^3(1,\varepsilon,\varepsilon_{\rm ext};R_c,R)$ [Eq. (B10)] corresponding to the sphere with the Onsager cavity. Expanding this coefficient in powers of $\rho_{11}=k_0R_c$, we find that

$$C_1^3(1,\varepsilon,\varepsilon_{\rm ext};R_c,R)=-i\frac{9\varepsilon}{2\varepsilon+1}(k_0R_c)^{-3} -i\frac{9\varepsilon(8\varepsilon+1)}{5(2\varepsilon+1)^2}(k_0R_c)^{-1} -\frac{9\varepsilon^{5/2}}{(2\varepsilon+1)^2}\frac{\beta_1-\beta_2}{\beta_1+\beta_2}-1+O(k_0R_c), \tag{28}$$

with β_j given by Eq. (B14). One may recognize that

$$\frac{2\beta_1}{\beta_1+\beta_2}=-C_1^2(\varepsilon,\varepsilon_{\rm ext};R), \tag{29}$$

where $C_1^2(\varepsilon,\varepsilon_{\rm ext};R)$ [Eq. (B7)] is the reflection coefficient of the system without the Onsager cavity. One may also see that

$$\mathrm{Re}\frac{9\varepsilon^{5/2}}{(2\varepsilon+1)^2}=\left|\frac{3\varepsilon}{2\varepsilon+1}\right|^2\eta-\frac{18\varepsilon''}{|2\varepsilon+1|^4}[(2|\varepsilon|^2+\varepsilon')\kappa+\varepsilon''\eta] \tag{30}$$

is equal to the R_c-free contribution to $\hat{\Gamma}^0_{\rm loc}$ [Eq. (27)] and that analogous results hold for the real parts of the first two terms in Eq. (28). Therefore, from Eq. (22) we find

$$\hat{\Gamma}_{\rm loc}=\hat{\Gamma}^0_{\rm loc}+\hat{\Gamma}^{\rm sc}_{\rm loc}, \tag{31}$$

where

$$\hat{\Gamma}^{\rm sc}_{\rm loc}=\mathrm{Re}\frac{9\varepsilon^{5/2}}{(2\varepsilon+1)^2}C_1^2(\varepsilon,\varepsilon_{\rm ext};R) \tag{32}$$

is the normalized cavity-induced decay rate with the local-field corrections.

The above result for $\hat{\Gamma}^{\rm sc}_{\rm loc}$ can be transformed into a form similiar to Eq. (27) for $\hat{\Gamma}^0_{\rm loc}$. Letting $\sqrt{\varepsilon}\to\sqrt{\varepsilon}C_1^2(\varepsilon,\varepsilon_{\rm ext};R)$ in Eq. (30), we see that $\hat{\Gamma}^{\rm sc}_{\rm loc}$ is obtained from the R_c-free contribution to $\hat{\Gamma}^0_{\rm loc}$ upon replacements $\eta\to\hat{\Gamma}^{\rm sc}$ and $\kappa\to 2\hat{\Delta}^{\rm sc}$, where

$$\hat{\Gamma}^{\rm sc}=\mathrm{Re}\,\sqrt{\varepsilon_1}C_1^2(\varepsilon,\varepsilon_{\rm ext};R) \tag{33}$$

is the normalized cavity-induced decay rate [Eq. (21)] and

$$\hat{\Delta}^{\rm sc}=\frac{1}{2}\mathrm{Im}\,\sqrt{\varepsilon_1}C_1^2(\varepsilon,\varepsilon_{\rm ext};R) \tag{34}$$

is the normalized classical cavity-induced level shift [36] of the molecule in the bare sphere. Accordingly, from Eq. (27) we finally have

$$\hat{\Gamma}^{\rm sc}_{\rm loc}=\left|\frac{3\varepsilon(\omega)}{2\varepsilon(\omega)+1}\right|^2\left\{\hat{\Gamma}^{\rm sc}-2\frac{\varepsilon''(\omega)}{|\varepsilon(\omega)|^2} \times\frac{2[2|\varepsilon(\omega)|^2+\varepsilon'(\omega)]\hat{\Delta}^{\rm sc}+\varepsilon''(\omega)\hat{\Gamma}^{\rm sc}}{|2\varepsilon(\omega)+1|^2}\right\}. \tag{35}$$

We note that this is not an unexpected result once we have learned the correct form of the decay rate $\hat{\Gamma}^0_{\rm loc}$ in the infinite medium. In its derivation it is implicitly assumed that the radius of the sphere, i.e., the molecule-mirror distance is much larger than the transition wavelength ($k_0R\gg 1$). Under these circumstances, the molecule-mirror interaction goes through the radiation-field component of the scattered field and therefore only this field component determines the cavity-induced rate. Thus, the rate $\mathcal{L}_{\rm Ons}\hat{\Gamma}^{\rm sc}$, as would be obtained by letting $\mathbf{p}\to 3\varepsilon\mathbf{p}/(2\varepsilon+1)$ in Eq. (21), is corrected for an absorption contribution in the same way as is the radiation-field contribution to the rate $\hat{\Gamma}^0_{\rm loc}$ in the infinite cavity.

C. External region

To find the local-field corrections to the molecular losses in the external region, we consider the field $\mathbf{E}^{\rm loc}_{\rm ext}(\mathbf{r};\omega)$ outside the sphere with the Onsager cavity. As already noted, this field is equal to the field in the infinite external medium [Eq. (A1a), with $k=k_{\rm ext}$] of the dipole [cf. Eq. (23)],

$$\mathbf{p}^{\rm loc}_{\rm ext}=\frac{1}{\varepsilon_{\rm ext}}C_3^3(1,\varepsilon,\varepsilon_{\rm ext};R_c,R)\mathbf{p}, \tag{36}$$

with the coefficient $C_3^3(1,\varepsilon,\varepsilon_{\rm ext};R_c,R)$ given by Eq. (B12). For small $\rho_{11}=k_0R_c$, we find that

$$C_3^3(1,\varepsilon,\varepsilon_{\rm ext};R_c,R)=-\frac{i\varepsilon_{\rm ext}}{\sqrt{\varepsilon}k_0R}\frac{3\varepsilon^2}{2\varepsilon+1}\frac{2}{\beta_1+\beta_2} +O[(k_0R_c)^2], \tag{37}$$

with β_j given by Eq. (B14). Now

$$\frac{i\varepsilon_{\rm ext}}{\sqrt{\varepsilon}k_0R}\frac{2}{\beta_1+\beta_2}=-C_2^2(\varepsilon,\varepsilon_{\rm ext};R), \tag{38}$$

where $C_2^2(\varepsilon,\varepsilon_{\rm ext};R)$ is the external-field coefficient of the system without the Onsager cavity [cf. Eq. (B8)]. To the leading term in k_0R_c, we therefore have

$$\mathbf{p}_{\rm ext}^{\rm loc}=\frac{3\varepsilon}{2\varepsilon+1}\frac{\varepsilon}{\varepsilon_{\rm ext}}C_2^2(\varepsilon,\varepsilon_{\rm ext};R)\mathbf{p}=\frac{3\varepsilon}{2\varepsilon+1}\mathbf{p}_{\rm ext}, \tag{39}$$

where $\mathbf{p}_{\rm ext}$ is the corresponding effective dipole moment for the bare sphere [cf. Eq. (23)]. Accordingly, for the external field we have

$$\mathbf{E}_{\rm ext}^{\rm loc}(\mathbf{r};\omega)=\frac{3\varepsilon(\omega)}{2\varepsilon(\omega)+1}\mathbf{E}_{\rm ext}(\mathbf{r};\omega), \tag{40}$$

with $\mathbf{E}_{\rm ext}(\mathbf{r};\omega)$ being the external field for the bare sphere. Consequently, this implies $\mathcal{L}_{\rm Ons}$ as the proper local-field correction factor to the external power loss

$$W_{\rm ext}^{\rm loc}=\left|\frac{3\varepsilon(\omega)}{2\varepsilon(\omega)+1}\right|^2W_{\rm ext}. \tag{41}$$

Note that here $W_{\rm ext}$ is generally given by Eq. (18), with $k=k_{\rm ext}$ and $R_c=R$.

D. Other systems

We end this section with a remark on a (plausible) generalization of the results obtained. Using Eqs. (10) and (B6), we see that Eqs. (28) and (29) lead to the following relation:

$$\vec{\mathbf{G}}_{\rm loc}^{\rm sc}(\mathbf{r}_0,\mathbf{r}_0;\omega)=ik_0\left[-i\frac{9\varepsilon}{2\varepsilon+1}(k_0R_c)^{-3}-i\frac{9\varepsilon(8\varepsilon+1)}{5(2\varepsilon+1)^2}(k_0R_c)^{-1}+\frac{9\varepsilon^{5/2}}{(2\varepsilon+1)^2}-1\right]\frac{2}{3}\vec{\mathbf{I}}+\left(\frac{3\varepsilon}{2\varepsilon+1}\right)^2\vec{\mathbf{G}}^{\rm sc}(\mathbf{r}_0,\mathbf{r}_0;\omega) \tag{42}$$

between the Green-function element

$$\vec{\mathbf{G}}_{\rm loc}^{\rm sc}(\mathbf{r}_0,\mathbf{r}_0;\omega)=ik_0C_1^3(1,\varepsilon,\varepsilon_{\rm ext};R_c,R)\frac{2}{3}\vec{\mathbf{I}} \tag{43}$$

for the sphere with the Onsager cavity, and the Green-function element

$$\vec{\mathbf{G}}^{\rm sc}(\mathbf{r}_0,\mathbf{r}_0;\omega)=ik_0\sqrt{\varepsilon}C_1^2(\varepsilon,\varepsilon_{\rm ext};R)\frac{2}{3}\vec{\mathbf{I}} \tag{44}$$

for the bare sphere. In Eq. (42) any reference to the specific system considered in its derivation is formally lost, which implies that this equation could be taken as a general relationship between the equal-point Green-function elements for a system with and without the Onsager cavity valid for $\mathbf{r}_0$ away from a system interface. Complementing $\vec{\mathbf{G}}_{\rm loc}^{\rm sc}(\mathbf{r}_0,\mathbf{r}_0;\omega)$ with the free-space Green-function element $\vec{\mathbf{G}}^{\rm free}(\mathbf{r}_0,\mathbf{r}_0;\omega)$ and noting that $\mathrm{Im}\,\vec{\mathbf{G}}^{\rm free}(\mathbf{r}_0,\mathbf{r}_0;\omega)=ik_0(2/3)\vec{\mathbf{I}}$, through Eq. (1) one immediately obtains the normalized molecular decay rate as given by Eqs. (27), (31), and (35), with $\hat{\Gamma}^{\rm sc}$ and $\hat{\Delta}^{\rm sc}$ in Eq. (35) now expressed in terms of the Green function for the bare system as

$$\begin{aligned}\hat{\Gamma}^{\rm sc}&=\frac{3c}{2\omega}\hat{\mathbf{p}}\cdot\mathrm{Im}\vec{\mathbf{G}}^{\rm sc}(\mathbf{r}_0,\mathbf{r}_0;\omega)\cdot\hat{\mathbf{p}},\\ \hat{\Delta}^{\rm sc}&=-\frac{3c}{4\omega}\hat{\mathbf{p}}\cdot\mathrm{Re}\,\vec{\mathbf{G}}^{\rm sc}(\mathbf{r}_0,\mathbf{r}_0;\omega)\cdot\hat{\mathbf{p}},\end{aligned} \tag{45}$$

where $\hat{\mathbf{p}}$ gives the direction of the transition. Consequently, at first sight, there is no reason why Eqs. (35) and (45) should not be taken as the general result for the molecular decay rate in an absorbing cavity valid for all positions of the molecule away from the cavity walls and for all transition-dipole orientations. Similarly, in view of Eq. (10), it appears that Eq. (40) indicates a rather general relationship between the Green-function elements for a system with and without the Onsager cavity,

$$\vec{\mathbf{G}}_{\rm loc}(\mathbf{r},\mathbf{r}_0;\omega)=\frac{3\varepsilon(\omega)}{2\varepsilon(\omega)+1}\vec{\mathbf{G}}^{\rm sc}(\mathbf{r},\mathbf{r}_0;\omega), \tag{46}$$

where $\mathbf{r}$ is in the external layer and $\mathbf{r}_0$ is in the cavity. If this relation holds, $\mathcal{L}_{\rm Ons}(\varepsilon)$ as the local-field correction factor for the molecular external power loss in the general case would then be its immediate consequence. Of course, as already stressed, these conjectures cannot be proved without the calculation of the exact Green function for the system including the Onsager cavity at the source position.

IV. DISCUSSION

To illustrate the effect of the local field on the decay rate in absorbing cavities, we exploit the above simple model and consider the decaying molecule in the center of an absorbing dielectric sphere surrounded by air ($\varepsilon_{\rm ext}=1$). The dielectric function of the sphere is modeled as

$$\varepsilon(\omega)=\varepsilon_b+\frac{\Omega^2}{\omega_0^2-\omega^2-i\omega\gamma}, \tag{47}$$

where ε_b is the background (high-frequency) dielectric constant, ω_0 and γ are, respectively, the center frequency and the width of the absorption resonance, and its strength is

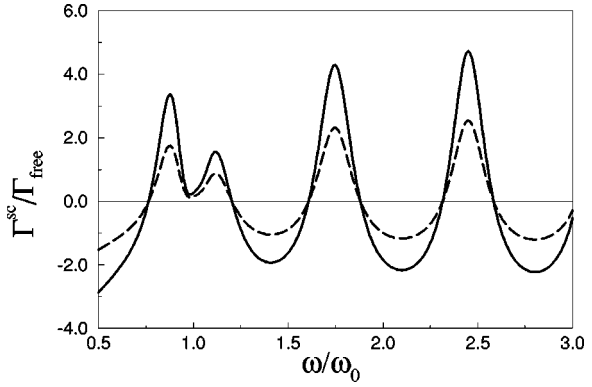

FIG. 2. Cavity-induced decay rate with (solid line) and without (dashed line) local-field corrections. The radius of the sphere is $R=\lambda_0/\pi$ and the material parameters are $\varepsilon_b=5$, $\Omega=0.5\omega_0$, and $\gamma=0.1\omega_0$.

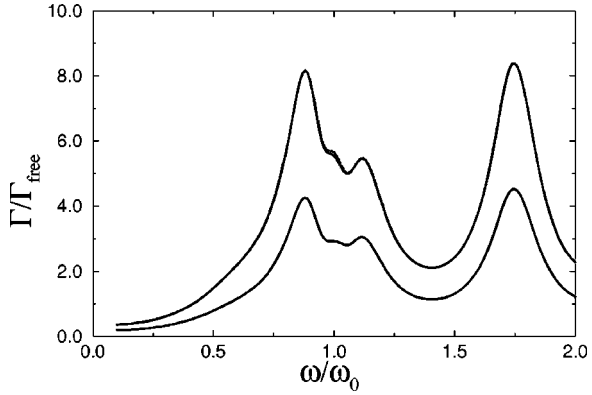

FIG. 3. Total decay rate with (upper solid line) and without (lower solid line) local-field corrections for the system described in Fig. 2. The dotted line represents the macroscopic decay rate with the usual local-field correction factor. The Onsager cavity radius is $R_c=0.1\lambda$ and $R_m=R_c$.

controlled by Ω^2. In this analysis, a relatively large background dielectric constant $\varepsilon_b=5$ is chosen to strengthen the cavity effect of the sphere.

The effects of the local field on the cavity-induced rate are illustrated in Fig. 2, where the rates $\hat{\Gamma}^{sc}_{loc}$ [Eq.(32)] and $\hat{\Gamma}^{sc}$ [Eq. (33)] are compared for the system with (solid line) and without (dashed line) the Onsager cavity, respectively. Away from the resonance, the system is lossless and, owing to the cavity effect of the sphere, the two (SE) rates exhibit familiar oscillations with ω, with the amplitudes of oscillations scaled by the usual local-field correction factor $\mathcal{L}_{Ons}(\varepsilon_b)\simeq 1.85$ [cf. Eq. (35)]. A somewhat different effect of the local field is observed in the region of the resonance $\omega\simeq\omega_0$. For the medium parameters chosen, the second (absorption) term on the rhs of Eq. (35) is small compared with the first one. Moreover, according to our calculations, $\hat{\Gamma}^{sc}_{loc}$ cannot be distinguished on this scale from $\mathcal{L}_{Ons}\hat{\Gamma}^{sc}$ over a wide range of the parameters Ω and γ. The cavity-induced rate in this frequency region is therefore (again) predominantly determined by the product of $\mathcal{L}_{Ons}$ and $\hat{\Gamma}^{sc}$. Each of these quantities exhibits a characteristic asymmetric dispersion around ω_0. In addition, since for this radius $(R\omega_0/c=2)$ the sphere nearly corresponds to the first-order enhancement cavity with respect to the SE rate at $\omega=\omega_0$ in the lossless $(\Omega=0)$ case, the medium is nearly in resonance with the sphere. As a consequence, instead of a peak near $\omega=\omega_0$, as would appear in the nonabsorbing $(\Omega=0)$ case, owing to the resonant absorption, the rates $\hat{\Gamma}^{sc}_{loc}$ and $\hat{\Gamma}^{sc}$ exhibit asymmetric double-peak structures, which very much resemble the ''cavity-polariton'' part of the SE spectrum in the bad-cavity case [12].

Figure 3 represents the total decay rates $\hat{\Gamma}_{loc}$ [Eq. (31) in conjuction with Eqs. (27) and (32)] and $\hat{\Gamma}$ [Eq. (2) in conjuction with Eqs. (3) and (33)] with and without the local-field corrections, respectively, along with the decay rate $\hat{\tilde{\Gamma}}_{loc}=\mathcal{L}_{Ons}\hat{\Gamma}$ [Eqs. (6) and (8)] with the local-field correction factor assumed in our previous work [2,12]. In plotting these curves, we have taken that the molecule-medium distance R_m figuring in $\hat{\Gamma}^0$ [Eq. (3)] is equal to the Onsager cavity radius, $R_m=R_c$. Away from the resonance, $\hat{\Gamma}_{loc}$ and $\hat{\Gamma}$ merely reproduce the corresponding curves of Fig. 2, with values enhanced by the off-resonance infinite-cavity decay rates $\hat{\Gamma}^0_{loc}=\mathcal{L}_{Ons}(\varepsilon_b)\sqrt{\varepsilon_b}\simeq 4.1$ and $\hat{\Gamma}^0=\sqrt{\varepsilon_b}\simeq 2.2$, respectively. As before, the situation around ω_0 is different. For this R_c and the medium parameters chosen, the absorption contributions to $\hat{\Gamma}^0_{loc}$ [Eq. (27)] and $\hat{\Gamma}^{sc}_{loc}$ [Eq. (35)] are (still) small compared with the corresponding radiation contributions. Accordingly,

$$\hat{\Gamma}_{loc}\approx\mathcal{L}_{Ons}(\eta+\hat{\Gamma}^{sc}) \tag{48}$$

holds. For $R_m\approx R_c$, the same conclusion applies to the decay rate $\hat{\Gamma}$ as well, so that

$$\hat{\Gamma}\approx\eta+\hat{\Gamma}^{sc}. \tag{49}$$

The different shapes of the curves in Fig. 3 when compared with those in Fig. 2 in this region are therefore caused by the (anomalous) dispersion of the superimposed $\mathcal{L}_{Ons}\eta$ and η, respectively. This also explains why $\hat{\Gamma}_{loc}$ (upper solid line) practically cannot be distinguished on this scale from the decay rate $\hat{\tilde{\Gamma}}_{loc}=\mathcal{L}_{Ons}\hat{\Gamma}$ (dotted line), although the corresponding absorption contributions to these rates are different [cf. Eqs. (27) and (3)]. We note that Eqs. (48) and (49) hold for a range of the Onsager cavity radii and/or medium parameters. In these cases, our previous conjecture concerning the local-field corrections to the decay rate, as expressed by Eq. (6), remains (approximately) valid.

With decreasing the Onsager cavity radius, the absorption contribution to the decay rate becomes gradually the dominant contribution with the leading near-field term equal to the nonradiative rate of the molecule. In Fig. 4 we represent the situation where R_c is still large enough, so that nonradi-

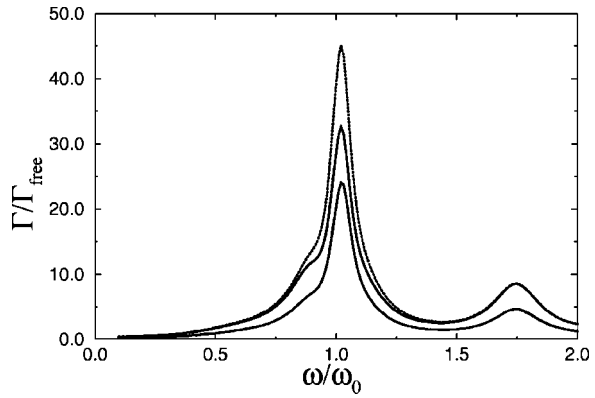

FIG. 4. Same as in Fig. 3. but for the Onsager cavity radius $R_c=0.03\lambda$.

ative and radiative contributions to $\hat{\Gamma}_{\rm loc}$ are of the same order of magnitude. In this regime, the decay rate may approximately be written as

$$\hat{\Gamma}_{\rm loc}\approx\mathcal{L}_{\rm Ons}\left[\frac{\varepsilon''}{|\varepsilon|^2}(k_0R_c)^{-3}+\eta+\hat{\Gamma}^{\rm sc}\right]. \tag{50}$$

The difference between $\hat{\Gamma}_{\rm loc}$ (upper solid line) and $\hat{\Gamma}$ (lower solid line) this time arises not only because of the overall extra factor $\mathcal{L}_{\rm Ons}$, but also because of the 3/2 times larger nonradiative contribution to $\hat{\Gamma}$ [cf. Eqs. (27) and (3)]. This 3/2 factor in the corresponding nonradiative rates also leads to a significantly larger decay rate $\hat{\tilde{\Gamma}}_{\rm loc}$ (dotted line) than the true rate $\hat{\Gamma}_{\rm loc}$ (upper solid line). We note, however, that the comparison between $\hat{\Gamma}_{\rm loc}$, $\hat{\Gamma}$, and $\hat{\tilde{\Gamma}}_{\rm loc}$ is given here only for illustrative purposes as their difference is a consequence of our arbitrary input $R_m=R_c$. Indeed, in addition to demanding, on physical grounds, the same order of magnitude for R_m and R_c, any other relation between these parameters may also be assumed.

Of course, owing to the strong increase of the R_c-dependent terms, for even smaller R_c the decay rate $\Gamma^0_{\rm loc}$ largely exceeds the cavity-induced rate $\Gamma^{\rm sc}_{\rm loc}$, so that $\Gamma_{\rm loc}\approx\Gamma^0_{\rm loc}$ holds. For a detailed discussion of the decay rate in this regime, we therefore refer the reader to Ref. [8].

V. SUMMARY

In this work we have calculated the decay rate and the classical external power loss of an excited molecule located in the center of an absorbing and dispersive dielectric sphere by adopting the Onsager (real cavity) model for the local field. We have found that the external fields of a dipole (classically representing the molecule) calculated with and without the Onsager cavity scale with $3\varepsilon(\omega)/[2\varepsilon(\omega)+1)]$. This immediately gives $|3\varepsilon(\omega)/[2\varepsilon(\omega)+1]|^2$ as the correct local-field correction factor for the (radiation and absorption) power loss of the molecule outside the sphere in the absorbing case. Whereas this result could have been guessed on the basis of a straightforward analytical continuation of the corresponding result in the lossless case, the local-field corrections to the total decay rate (power loss) of the molecule are found to be much more complex. The total decay rate is found to consist of the decay rate for the infinite sphere, as very recently obtained for an absorbing medium in Ref. [8], and of the cavity-induced rate for which we have obtained a similar expression. When expressed in terms of the Green function for the sphere, these results become formally system independent. This suggests the general results for the local-field corrections to the decay rate and to the external power loss of a molecule in an absorbing cavity and located away from the cavity walls.

APPENDIX A: DIPOLE POWER LOSS

To calculate W^0 using the Poynting theorem, it is convenient to rewrite the dipole electromagnetic field in spherical coordinates. With $\vec{\mathbf{I}}=\hat{\mathbf{r}}\hat{\mathbf{r}}+\hat{\boldsymbol{\theta}}\hat{\boldsymbol{\theta}}+\hat{\boldsymbol{\phi}}\hat{\boldsymbol{\phi}}$ in Eq. (12), letting the origin be at the dipole site and assuming, for simplicity, that $\mathbf{p}=p\hat{\mathbf{z}}$, we have ($k_0=\omega/c$)

$$\mathbf{E}^0(\mathbf{r};\omega)=ik k_0^2 p\left\{\frac{2h_1^{(1)}(kr)}{kr}\cos\vartheta\hat{\mathbf{r}}+\left[\frac{h_1^{(1)}(kr)}{kr}-h_0^{(1)}(kr)\right]\sin\vartheta\hat{\boldsymbol{\theta}}\right\}, \tag{A1a}$$

$$\mathbf{B}^0(\mathbf{r};\omega)=k^2k_0h_1^{(1)}(kr)\mathbf{p}\times\hat{\mathbf{r}}=k^2k_0ph_1^{(1)}(kr)\sin\vartheta\hat{\boldsymbol{\phi}}, \tag{A1b}$$

where

$$h_0^{(1)}(z)=-i\frac{e^{iz}}{z},\quad h_1^{(1)}(z)=-\frac{e^{iz}}{z}\left(1+\frac{i}{z}\right)$$

are the spherical Hankel functions of the first kind. The radial component of the Poynting vector

$$\mathbf{P}(\mathbf{r};\omega)=\frac{c}{8\pi}\mathrm{Re}\,\mathbf{E}^0(\mathbf{r};\omega)\times[\mathbf{B}^0(\mathbf{r};\omega)]^*$$

is then easily found to be

$$\hat{\mathbf{r}}\cdot\mathbf{P}(\mathbf{r};\omega)=\frac{c}{8\pi}\mathrm{Im}\frac{|k|^6|\mathbf{p}|^2}{\varepsilon\sqrt{\varepsilon^*}}\left[-\frac{h_1^{(1)}(kr)}{kr}+h_0^{(1)}(kr)\right]\times[h_1^{(1)}(kr)]^*\sin^2\vartheta. \tag{A2}$$

This determines the angular distribution of the energy flow $dW_f^0/d\Omega=r^2\hat{\mathbf{r}}\cdot\mathbf{P}(\mathbf{r};\omega)$ through a spherical surface of radius r around the dipole. Upon integration over the angles, we therefore have

$$W_f^0(r)=\frac{\omega|\mathbf{p}|^2}{3}\frac{\varepsilon''}{|\varepsilon|^2}\frac{|(1-ikr)e^{ikr}|^2}{r^3}+\eta\frac{\omega k_0^3|\mathbf{p}|^2}{3}e^{-2k''r}. \tag{A3}$$

The dipole energy absorbed per second within the volume V_r of the sphere is given by

$$W_a^0(r)=\frac{\omega\varepsilon''}{8\pi}\int_{V_r} d^3\mathbf{r}|\mathbf{E}^0(\mathbf{r};\omega)|^2. \tag{A4}$$

With

$$|\mathbf{E}^0(\mathbf{r};\omega)|^2=\frac{|k|^6|\mathbf{p}|^2}{|\varepsilon|^2}\left[4\left|\frac{h_1^{(1)}(kr)}{kr}\right|^2\cos^2\vartheta+\left|\frac{h_1^{(1)}(kr)}{kr}-h_0^{(1)}(kr)\right|^2\sin^2\vartheta\right]$$

and performing the angular integration, we have

$$W_a^0(r)=\frac{\omega|\mathbf{p}|^2}{3}\frac{\varepsilon''}{|\varepsilon|^2}\int_{R_c}^{r}dr\left[\frac{3}{r^4}|(1-ikr)e^{ikr}|^2-2\frac{k'^2-k''^2-k''|k|^2r}{r^2}e^{-2k''r}+|k|^4e^{-2k''r}\right]. \tag{A5}$$

Partially integrating the first term and noticing that the remaining integrals involving the near field of the dipole cancel, we obtain

$$W_a^0(r)=\frac{\omega|\mathbf{p}|^2}{3}\frac{\varepsilon''}{|\varepsilon|^2}\left[-\frac{|(1-ikr)e^{ikr}|^2}{r^3}\right]_{R_c}^{r}+\eta\frac{\omega k_0^3|\mathbf{p}|^2}{3}(e^{-2k''R_c}-e^{-2k''r}). \tag{A6}$$

This, together with Eq. (A2), gives Eq. (18) for the total dipole power loss $W_f^0(r)+W_a^0(r)$.

APPENDIX B: DIPOLE FIELD IN MULTILAYERED SPHERICAL MEDIA

Generalizing the result for an infinite medium [Eq. (A1b)], the magnetic field of a dipole oscillating along the z axis in the center of an N-layered spherical system [Fig. 1] can be written in the form

$$\mathbf{B}(\mathbf{r};\omega)=\varepsilon_1 k_0^3 p f(r)\sin\vartheta\hat{\boldsymbol{\phi}}. \tag{B1}$$

Through

$$\mathbf{E}(\mathbf{r};\omega)=\frac{i}{k_0\varepsilon(r)}\nabla\times\mathbf{B}(\mathbf{r};\omega),$$

the electric field is therefore given by

$$\mathbf{E}(\mathbf{r};\omega)=ik_0^2p\frac{\varepsilon_1}{\varepsilon(r)}\left[\frac{2f(r)}{r}\cos\vartheta\hat{\mathbf{r}}-\frac{[rf(r)]'}{r}\sin\vartheta\hat{\boldsymbol{\theta}}\right], \tag{B2}$$

with the prime denoting the derivative of the function in the brackets. The general form of the function $f(r)$ in the lth layer is

$$f_l(r)=h_1^{(1)}(k_1r)\,\delta_{l1}+C_{l+}^N h_1^{(1)}(k_lr)+C_{l-}^N h_1^{(2)}(k_lr), \tag{B3}$$

where $h_1^{(i)}(z)$ are spherical Hankel functions. The last two terms here are the solutions of the homogeneous Maxwell's equations [37] and give the scattered field $\mathbf{E}^{\mathrm{sc}}(\mathbf{r};\omega)$ in the system. The regularity of this field at the origin demands that $C_{1+}^N=C_{1-}^N\equiv C_1^N/2$, while the outgoing-wave condition at infinity demands that $C_{N-}^N=0$. The rest of the coefficients $C_{l\pm}^N$ are determined from the boundary conditions

$$f_l(r_l)=f_{l+1}(r_l)\quad\text{and}\quad\frac{1}{\varepsilon_l}[rf_l(r)]'_{r_l}=\frac{1}{\varepsilon_{l+1}}[rf_{l+1}(r)]'_{r_l}$$

at the layer interfaces. The scattered field in the central layer is, therefore, generally given by

$$\mathbf{E}_1^{\mathrm{sc}}(\mathbf{r};\omega)=ik_1k_0^2pC_1^N\left[\frac{2j_1(k_1r)}{k_1r}\cos\vartheta\hat{\mathbf{r}}-\frac{[k_1rj_1(k_1r)]'}{k_1r}\sin\vartheta\hat{\boldsymbol{\theta}}\right], \tag{B4}$$

where $j_1(z)$ is the spherical Bessel function. In the other layers it is given by ($\mathbf{E}_l=\mathbf{E}_l^{\mathrm{sc}}$)

$$\mathbf{E}_l(\mathbf{r};\omega)=ik_1k_0^2p\sqrt{\frac{\varepsilon_1}{\varepsilon_l}}\left[C_{l+}^N\left\{\frac{2h_1^{(1)}(k_lr)}{k_lr}\cos\vartheta\hat{\mathbf{r}}-\frac{[k_lrh_1^{(1)}(k_lr)]'}{k_lr}\sin\vartheta\hat{\boldsymbol{\theta}}\right\}+C_{l-}^N\left\{\frac{2h_1^{(2)}(k_lr)}{k_lr}\cos\vartheta\hat{\mathbf{r}}-\frac{[k_lrh_1^{(2)}(k_lr)]'}{k_lr}\sin\vartheta\hat{\boldsymbol{\theta}}\right\}\right], \tag{B5}$$

with $C_{N-}^N=0$. Specially, since $j_1(z)/z\to 1/3$ and $[zj_1(z)]'/z\to 2/3$ for small z, the scattered field in the center of the system is given by

$$\mathbf{E}_1^{\mathrm{sc}}(\mathbf{r};\omega)\big|_{r\to 0}=ik_1k_0^2C_1^N\frac{2}{3}\mathbf{p}. \tag{B6}$$

For $N=2$, we find ($\rho_i=k_ir_1$)

$$C_1^2(\varepsilon_1,\varepsilon_2;r_1)=\frac{1}{D}\{\varepsilon_2h_1^{(1)}(\rho_2)[\rho_1h_1^{(1)}(\rho_1)]'-\varepsilon_1h_1^{(1)}(\rho_1)\times[\rho_2h_1^{(1)}(\rho_2)]'\}, \tag{B7}$$

$$C_{2+}^2(\varepsilon_1,\varepsilon_2;r_1)=\frac{i\varepsilon_2}{\rho_1D}, \tag{B8}$$

where

$$D=\varepsilon_1 j_1(\rho_1)[\rho_2 h_1^{(1)}(\rho_2)]'-\varepsilon_2 h_1^{(1)}(\rho_2)[\rho_1 j_1(\rho_1)]'. \tag{B9}$$

For $N=3$, we have ($\rho_{ij}=k_i r_j$)

$$C_1^3(\varepsilon_1,\varepsilon_2,\varepsilon_3;r_1,r_2) = \frac{1}{j_1(\rho_{11})} \times\left[\frac{\beta_2 h_1^{(1)}(\rho_{21})-\beta_1 h_1^{(2)}(\rho_{21})}{\alpha_1\beta_2-\alpha_2\beta_1}-h_1^{(1)}(\rho_{11})\right], \tag{B10}$$

$$C_{2+}^3(\varepsilon_1,\varepsilon_2,\varepsilon_3;r_1,r_2)=\frac{\beta_2}{\alpha_1\beta_2-\alpha_2\beta_1}, \tag{B11a}$$

$$C_{2-}^3(\varepsilon_1,\varepsilon_2,\varepsilon_3;r_1,r_2)=\frac{-\beta_1}{\alpha_1\beta_2-\alpha_2\beta_1}, \tag{B11b}$$

$$C_{3+}^3(\varepsilon_1,\varepsilon_2,\varepsilon_3;r_1,r_2)=-\frac{i\varepsilon_3}{\rho_{22}}\frac{2}{\alpha_1\beta_2-\alpha_2\beta_1}, \tag{B12}$$

where

$$\alpha_j=-\frac{i\rho_{11}}{\varepsilon_2}\{\varepsilon_1 j_1(\rho_{11})[\rho_{21}h_1^{(j)}(\rho_{21})]'-\varepsilon_2 h_1^{(j)}(\rho_{21}) \times[\rho_{11}j_1(\rho_{11})]'\}, \tag{B13}$$

and

$$\beta_j=\varepsilon_3 h_1^{(1)}(\rho_{32})[\rho_{22}h_1^{(j)}(\rho_{22})]'-\varepsilon_2 h_1^{(j)}(\rho_{22}) \times[\rho_{32}h_1^{(1)}(\rho_{32})]'. \tag{B14}$$

[1] See, for example, R. R. Chance, A. Prock, and R. Silbey, Adv. Chem. Phys. **37**, 1 (1978); G. W. Ford and W. H. Weber, Phys. Rep. **113**, 195 (1984).

[2] M. S. Tomaš and Z. Lenac, Phys. Rev. A **56**, 4197 (1997); M. S. Tomaš, *ibid.* **51**, 2545 (1995).

[3] T. Gruner and D.-G. Welsch, Phys. Rev. A **53**, 1818 (1996); H. T. Dung, L. Knöll, and D.-G. Welsch, *ibid.* **57**, 3931 (1998); S. Scheel, L. Knöll, and D.-G. Welsch, *ibid.* **58**, 700 (1998).

[4] R. Matloob, R. Loudon, S. M. Barnett, and J. Jeffers, Phys. Rev. A **52**, 4823 (1995); R. Matloob and R. Loudon, *ibid.* **53**, 4567 (1996); R. Matloob, *ibid.* **59**, 1 (1999).

[5] A. Tip, Phys. Rev. A **56**, 5022 (1997); **57**, 4818 (1998).

[6] S. M. Barnett, B. Huttner, and R. Loudon, Phys. Rev. Lett. **68**, 3698 (1992).

[7] S. M. Barnett, B. Huttner, R. Loudon, and R. Matloob, J. Phys. B **29**, 3763 (1996).

[8] S. Scheel, L. Knöll, and D.-G. Welsch, Phys. Rev. A **60**, 4094 (1999); **61**, 069901(E) (2000). There is an obvious misprint in Eq. (53) of this reference: in the second term on the rhs of this equation $16\epsilon_R(\omega_A)$ should stay instead of $12\epsilon_R(\omega_A)$.

[9] H. T. Dung, L. Knöll, and D.-G. Welsch, Phys. Rev. A **62**, 053804 (2000).

[10] Y. Lee and M. Yamanishi, Phys. Rev. A **52**, 2312 (1995).

[11] G. Juzeliūnas, Phys. Rev. A **55**, R4015 (1997).

[12] M. S. Tomaš and Z. Lenac, Phys. Rev. A **60**, 2431 (1999).

[13] H. A. Lorentz, Ann. Phys. (Leipzig) **9**, 641 (1880).

[14] L. Lorenz, Ann. Phys. (Leipzig) **11**, 70 (1881).

[15] R. H. Pantell and H. E. Puthoff, *Fundamentals of Quantum Electronics* (Wiley, New York, 1962), Chap. 2.

[16] J. E. Sipe and J. Van Kranendonk, Phys. Rev. A **9**, 1806 (1974).

[17] J. Knoester and S. Mukamel, Phys. Rev. A **40**, 7065 (1989).

[18] L. Onsager, J. Am. Chem. Soc. **58**, 1486 (1936).

[19] E. Yablonovitch, T. J. Gmitter, and R. Bhat, Phys. Rev. Lett. **61**, 2546 (1988).

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

[21] Chang-qi Cao, W. Long, and H. Cao, Phys. Lett. A **232**, 15 (1997).

[22] S. Scheel, L. Knöll, D.-G. Welsch, and S. M. Barnett, Phys. Rev. A **60**, 1590 (1999).

[23] M. Fleischhauer, Phys. Rev. A **60**, 2534 (1999).

[24] See, e.g., O. J. F. Martin, C. Girard, and A. Dereux, Phys. Rev. Lett. **74**, 526 (1995); A. Rahmani, P. C. Chaumet, F. de Fornel, and C. Girard, Phys. Rev. A **56**, 3245 (1997).

[25] P. de Vries, D. V. van Coevorden, and A. Lagendijk, Rev. Mod. Phys. **70**, 447 (1998).

[26] See *Optical Processes in Microcavities*, edited by R. K. Chang and A. J. Campillo (World Scientific, Singapore, 1996).

[27] H. Chew, J. Chem. Phys. **87**, 1355 (1987); Phys. Rev. A **38**, 3410 (1988).

[28] K. G. Sullivan and D. G. Hall, Phys. Rev. A **50**, 2708 (1994).

[29] W. Jhe and K. Jang, Phys. Rev. A **53**, 1126 (1996).

[30] V. V. Klimov, M. Ducloy, and V. S. Lethokov, J. Mod. Opt. **43**, 2251 (1996); **43**, 549 (1996).

[31] Fam Le Kien, Nguyen Hong Quang, and K. Hakuta, Opt. Commun. **178**, 151 (2000).

[32] V. V. Klimov, V. S. Letokhov, and M. Ducloy, Phys. Rev. A **56**, 2308 (1997); V. V. Klimov, M. Ducloy, and V. S. Letokhov, J. Mod. Opt. **44**, 1081 (1997); Phys. Rev. A **59**, 2996 (1999).

[33] H.-B. Lin, J. D. Eversole, C. D. Merritt, and A. J. Campillo, Phys. Rev. A **45**, 6756 (1992).

[34] M. D. Barnes, W. B. Whitten, S. Arnold, and J. M. Ramsey, J. Chem. Phys. **97**, 7842 (1992); M. D. Barnes, C-Y. Kung, W. B. Whitten, J. M. Ramsey, S. Arnold, and S. Holler, Phys. Rev. Lett. **76**, 3931 (1996).

[35] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill, New York, 1968), Chap. 11.

[36] Using the reaction-force type of arguments (see, e.g., Ref.

[28]), the classical cavity-induced frequency shift $\hat{\Delta}^{sc} = \Delta\omega^{sc}/\gamma_0$ of the molecule normalized to its (free-space) radiative damping rate γ_0 can be given as

$$\hat{\Delta}^{sc} \approx -(3/4k_0^3|\mathbf{p}|^2)\mathrm{Re}\,\mathbf{p}^* \cdot \mathbf{E}^{sc}(\mathbf{r}_0,\mathbf{r}_0;\omega).$$

With $\mathbf{E}^{sc}(\mathbf{r}_0,\mathbf{r}_0;\omega)$ from Eq. (B6), this gives Eq. (34) for $\hat{\Delta}^{sc}$, while using Eq. (10) one obtains the general result given in Eq. (45).

[37] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975), Chap. 16.