

## Decay of excited molecules in absorbing planar cavities

M. S. Tomáš<sup>1</sup> and Z. Lenac<sup>2</sup>

<sup>1</sup>*Rudjer Bošković Institute, P.O. Box 1016, 10001 Zagreb, Croatia*

<sup>2</sup>*Pedagogical Faculty, University of Rijeka, 51000 Rijeka, Croatia*

(Received 16 June 1997)

The decay rate of an excited molecule (atom) embedded in a dispersive and absorbing planar cavity is derived by using a recently obtained compact form of the Green's function for a multilayer. As a by-product, a hint is provided for a straightforward extension of the results obtained for lossless cavities of other shapes to the corresponding absorbing cavities. The decay rate in an absorbing cavity consists of the spontaneous emission rate and of the nonradiative rates caused by the near-field interaction of the molecule with the cavity medium and, for nearby molecules, with the cavity mirrors. Only the spontaneous emission rate is satisfactorily described in the macroscopic approach adopted. The theory is applied to an analysis of the effects of the weak cavity absorption on the decay rate in a dielectric microcavity formed by two metallic mirrors. As expected, dissipation in the cavity medium spoils the conditions for controlled spontaneous emission and strongly suppresses the intensity of spontaneous emission. However, its effect on the spontaneous emission rate is much less pronounced. [S1050-2947(97)03411-2]

PACS number(s): 42.50.Lc, 42.60.Da, 33.50.-j

### I. INTRODUCTION

The development of various light-emitting microdevices based on controlled spontaneous emission (SE) has raised the problem of a theoretical description of spontaneous relaxation of excited molecules (atoms) in realistic, i.e., dispersive and absorbing cavities. Macroscopic electrodynamics is clearly a natural framework in which to deal with such systems. However, it is only very recently that, based on the result of the canonical field quantization for a microscopic model of a dielectric [1,2], a scheme has been proposed for quantization of the macroscopic field in dispersive and absorbing inhomogeneous systems [3–5]. So far, this program has been fulfilled only for waves propagating along the normal to simple multilayers, i.e., effectively one-dimensional systems [4,5]. Since the coupling of the molecule to all possible waves supported by the system has to be considered to describe its decay correctly, the appropriate macroscopic approach to this goal is therefore quantum-mechanical linear-response theory in conjunction with the fluctuation-dissipation theorem, familiar from the theory of spontaneous emission in the presence of (absorbing) boundaries [6,7], or the classical theory developed in the context of molecular fluorescence and energy transfer at interfaces [8–11]. At zero temperature these methods give the same result for the (normalized) molecular decay rate and can be easily extended to fully absorptive systems including also the cavity interior [12–14].

So far, only the SE rate [12] and the total decay rate [14,15] of the molecule in an infinite absorbing cavity (medium) have been considered in more detail. In our previous work [13] we pointed out that a straightforward extension of a classical result for the molecular decay rate in a transparent layer [10,11] should describe the molecular decay rate in a fully absorbing multilayered system. In the subsequent development, however, we have restricted our attention to a transparent cavity case. Recently, Lee and Yamanishi [16] presented a theory on the SE rate in absorbing inhomoge-

neous dielectrics based on a photon Green's-function formalism. In addition to not accounting for the near-field interactions of the molecule, their approach rests heavily on the calculation of a Green's function of the system. In a very recent related work, Nha and Jhe [17] generalized the theory of molecular decay by Wylie and Sipe [7] to finite but transparent (empty) planar cavities. In this paper, using a recently obtained compact form of the Green's function for a multilayer [18], we reconsider and complement our approach in [13] and develop a theory of the molecular decay in an absorbing planar cavity or, generally, a multilayer. In this way, we simultaneously extend the theory of Wylie and Sipe [7] to finite and absorbing cavities, as well as the theory of Barnett *et al.* [12,14] to finite planar cavities.

The paper is organized as follows. In Sec. II we establish a relationship between the classical theory of [13] and the QED approach described and define the normalized rate. Throughout the paper we exploit this equivalence of the (normalized) rate in the two approaches and use (mainly) the classical language to identify various contributions to the total rate. In order to clarify the method of calculation employed in Sec. IV, in Sec. III we briefly rederive a few basic results concerning the decay of a molecule embedded in an absorbing dielectric host. In Sec. IV we derive the molecular decay rate in absorbing planar cavities and discuss the relationship between this result and the results obtained previously. As an application of the theory, in Sec. V we consider the decay of an excited molecule embedded in a dielectric microcavity formed by two metallic mirrors under the circumstances of controlled SE and discuss the effects of cavity absorption on various contributions to the total molecular decay rate. Our conclusions are summarized in Sec. VI.

### II. PRELIMINARIES

Consider an excited molecule at a position  $\mathbf{r}_0$  in a cavity. In the classical approach, the molecule is simulated by a

point dipole  $\mathbf{p}$  oscillating at the frequency of the transition  $\omega$ . The associated current density  $\mathbf{j}(\mathbf{r}, t) = -i\omega\mathbf{p}\delta(\mathbf{r} - \mathbf{r}_0)\exp(-i\omega t)$  gives rise to the electric field in the system of the form  $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}, \mathbf{r}_0; \omega)\exp(-i\omega t)$ . The molecular decay rate  $\Gamma$  is then related via  $\Gamma = W/\hbar\omega$  to the power

$$W(\mathbf{r}_0) = \frac{\omega}{2} \text{Im}\mathbf{p}^* \cdot \mathbf{E}(\mathbf{r}_0, \mathbf{r}_0; \omega) \quad (2.1)$$

lost by the dipole in supporting its own field. Introducing the Green's function of the system through [19]

$$\mathbf{E}(\mathbf{r}, \mathbf{r}_0; \omega) = \frac{\omega^2}{c^2} \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0; \omega) \cdot \mathbf{p}, \quad (2.2)$$

we have

$$\Gamma(\mathbf{r}_0) = \frac{\omega^2}{2\hbar c^2} \mathbf{p}^* \cdot \text{Im}\vec{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}_0; \omega) \cdot \mathbf{p}, \quad (2.3)$$

where we have assumed that  $\vec{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}_0; \omega)$  is the diagonal dyadic.

The quantum-mechanical decay rate is obtained in the usual way starting from the molecule- (total) field interaction Hamiltonian of the standard form [14]  $\hat{H}_{\text{int}} = -\hat{\mathbf{p}} \cdot \hat{\mathbf{E}}(\mathbf{r}_0)$ , using the Fermi golden rule, and employing the fluctuation-dissipation theorem. At zero temperature, one arrives at Eq. (2.3) with  $\mathbf{p} \rightarrow 2\mathbf{p}_{fi}$ , where  $\mathbf{p}_{fi}$  is the corresponding transition dipole matrix element [20]. Therefore, with this replacement in mind, we refer to Eq. (2.3) as the QED rate as well.

In general,  $\Gamma$  consists of the rate  $\Gamma_S$  associated with losses  $W_S$  owing to the quasistatic interaction of the molecule with its environment and the rate  $\Gamma_T$  related to molecular losses  $W_T$  due to its transverse (retarded) interaction with the surroundings. The quantities  $W_S$  ( $\Gamma_S$ ) and  $W_T$  ( $\Gamma_T$ ) can be obtained separately from the above formulas through the quasistatic and transverse parts, respectively, of the total dipole field (Green's function). As usual, we refer to  $\Gamma_T$  as the spontaneous emission rate  $\Gamma_{\text{SE}}$  and define the normalized molecular decay rate  $\hat{\Gamma}$  with respect to the SE rate in the corresponding infinite cavity  $\Gamma_{\text{SE}}^0$ , i.e.,  $\hat{\Gamma} = \Gamma/\Gamma_{\text{SE}}^0 = W/W_T^0$ . Clearly, both the classical and the QED approach lead to the same result for  $\hat{\Gamma}$  given by

$$\hat{\Gamma}(\mathbf{r}_0) = \frac{\hat{\mathbf{p}} \cdot \text{Im}\vec{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}_0; \omega) \cdot \hat{\mathbf{p}}}{\hat{\mathbf{p}} \cdot \text{Im}\vec{\mathbf{G}}_T^0(\mathbf{r}_0, \mathbf{r}_0; \omega) \cdot \hat{\mathbf{p}}}, \quad (2.4)$$

where now  $\hat{\mathbf{p}}$  describes the direction of the transition. Note that eventual local-field corrections do not affect  $\hat{\Gamma}$  for centrosymmetric cavity media and in this work, therefore, we ignore the difference between the local field actually acting on the molecule and the macroscopic field used in the above derivation.

A cavity represents an inhomogeneous system that can be described by the position-dependent complex dielectric function  $\varepsilon(\mathbf{r}, \omega)$  defined in a piecewise fashion. One usually solves for the full dipole field  $\mathbf{E}$  (Green's function  $\mathbf{G}$ ) in such a system. The dipole quasistatic field  $\mathbf{E}_S$  is then obtained by

letting  $c \rightarrow \infty$  in this solution, i.e.,  $\mathbf{E}_S = \lim_{c \rightarrow \infty} \mathbf{E}$ . Obviously, the difference  $\mathbf{E}_T = \mathbf{E} - \mathbf{E}_S$  represents the dipole transverse (retarded) field. Indeed, since Gauss's law holds for both  $\mathbf{E}$  and  $\mathbf{E}_S$ , the field  $\mathbf{E}_T$  obeys the generalized transversality condition

$$\nabla \cdot \varepsilon(\mathbf{r}, \omega) \mathbf{E}_T(\mathbf{r}, \mathbf{r}_0; \omega) = 0, \quad (2.5)$$

as appropriate for inhomogeneous systems [21]. According to Eq. (2.2), the Green's function for the transverse field  $\vec{\mathbf{G}}_T$  is therefore given by [18]

$$\vec{\mathbf{G}}_T(\mathbf{r}, \mathbf{r}_0; \omega) = \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0; \omega) - \frac{c^2}{\omega^2} \lim_{c \rightarrow \infty} \frac{\omega^2}{c^2} \vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0; \omega), \quad (2.6)$$

where the last term represents the Green's function for the quasistatic field  $\vec{\mathbf{G}}_S$ . By definition,  $\vec{\mathbf{G}}_T$  comprises the contributions of all polaritonic (retarded) modes in the system.

### III. DECAY RATE IN AN INFINITE CAVITY

The Green's function  $\vec{\mathbf{G}}^0$  for an infinite medium is obtained by a straightforward generalization of the free-space Green's function and reads [22]

$$\vec{\mathbf{G}}^0(\mathbf{r}, \mathbf{r}_0; \omega) = \frac{1}{k^2} \left[ \frac{3\hat{\mathbf{R}}\hat{\mathbf{R}} - \vec{\mathbf{I}}}{R^3} (1 - ikR) - \frac{4\pi}{3} \vec{\mathbf{I}}\delta(\mathbf{R}) + k^2 \frac{\vec{\mathbf{I}} - \hat{\mathbf{R}}\hat{\mathbf{R}}}{R} \right] e^{ikR}, \quad (3.1)$$

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

$$\tilde{k}(\omega) = \tilde{k}'(\omega) + i\tilde{k}''(\omega) \equiv \sqrt{\varepsilon(\omega)} \frac{\omega}{c} = [\eta(\omega) + i\kappa(\omega)] \frac{\omega}{c}, \quad (3.2)$$

with  $\eta$  and  $\kappa$  being, respectively, the refractive index and the extinction coefficient of the medium. Since  $\vec{\mathbf{G}}^0(\mathbf{r}_0, \mathbf{r}_0; \omega)$  is the isotropic tensor, Eq. (3.1) has to be averaged over  $\hat{\mathbf{R}}$  to find its limit for  $R \rightarrow 0$  correctly. With  $\langle \hat{\mathbf{R}}\hat{\mathbf{R}} \rangle = (1/3)\vec{\mathbf{I}}$ , one therefore has

$$\vec{\mathbf{G}}^0(\mathbf{r}_0, \mathbf{r}_0; \omega) = \lim_{R \rightarrow 0} \left[ -\frac{4\pi}{3\tilde{k}^2} \delta(\mathbf{R}) + \frac{2}{3} \frac{e^{i\tilde{k}R}}{R} \right] \vec{\mathbf{I}}. \quad (3.3)$$

In the spirit of the macroscopic field approach, we remove the singularity that appears in the quasistatic component of the dipole field by letting  $\delta_{R \rightarrow 0}(\mathbf{R}) \rightarrow 1/V_m$ , where  $V_m = (4\pi/3)R_m^3$  is an appropriately chosen spherical volume around the molecule. It is clear from Eq. (3.1) that making this substitution is equivalent to averaging the quasistatic dipole field (Green's function) over  $V_m$ , as suggested by Barnett *et al.* [14]. The total dipole power loss  $W^0$  is therefore given through Eq. (2.1) by

$$W^0 = W_S^0 + W_T^0 \equiv \text{Im} \left[ -\frac{1}{\varepsilon(\omega)} \right] \frac{\omega |\mathbf{p}|^2}{2R_m^3} + \eta(\omega) \frac{\omega^4 |\mathbf{p}|^2}{3c^3}, \quad (3.4)$$

where the first term describes the molecular quasistatic (near-field) loss in the medium, whereas the second one gives the familiar transverse (far-field) loss of the molecule, i.e., the spontaneous emission power. The quantity  $R_m$  can be considered as an effective molecule-medium distance [14]; this interpretation is also suggested by a very recent microscopic result for the molecular near-field loss [15]. Clearly,  $W_S^0$  is due to the excitation of longitudinal modes in the medium and vanishes for nonabsorbing ( $\varepsilon''=0$ ) media. Since  $\hat{\mathbf{p}}$  and  $\hat{\mathbf{R}}$  enter symmetrically into the expression for  $W^0$ , note that Eq. (3.4) could have been obtained by averaging  $W^0$  over  $\hat{\mathbf{p}}$  instead of averaging  $\vec{\mathbf{G}}^0$  over  $\hat{\mathbf{R}}$ . This method of calculating  $W^0$  will prove useful in Sec. IV.

We stress that the need of averaging the Green's function over  $\hat{\mathbf{R}}$  concerns its quasistatic (longitudinal) component and therefore the above procedure is unnecessary if solely the SE rate is to be obtained. Indeed, the application of the recipe (2.6) to Eq. (3.1) leads to

$$\vec{\mathbf{G}}_T^0(\mathbf{r}, \mathbf{r}_0; \omega) = \frac{1}{k^2} \frac{3\hat{\mathbf{R}}\hat{\mathbf{R}} - \hat{\mathbf{I}}}{R^3} [(1 - ikR)e^{ikR} - 1] + \frac{\hat{\mathbf{I}} - \hat{\mathbf{R}}\hat{\mathbf{R}}}{R} e^{ikR}. \quad (3.5)$$

Expanding this for small  $R$ , one directly finds

$$\text{Im} \vec{\mathbf{G}}_T^0(\mathbf{r}_0, \mathbf{r}_0; \omega) = \tilde{k}'(\omega) \frac{2}{3} \hat{\mathbf{I}}. \quad (3.6)$$

Through Eq. (2.3) this gives for the SE rate in absorbing media

$$\Gamma_{\text{SE}}^0 = \eta(\omega) \frac{4\omega^3 |\mathbf{p}_{fi}|^2}{3\hbar c^3}, \quad (3.7)$$

as obtained by Barnett *et al.* [12].

#### IV. DECAY RATE IN A PLANAR CAVITY

Consider an absorbing planar cavity ( $j$ ) formed by two generally multilayered lossy mirrors with an excited molecule embedded in it, as depicted in Fig. 1. A detailed derivation of a convenient plane-wave expansion of the Green's function for such a multilayer was presented in Ref. [18]. Here we quote only the Green's function element relevant to the present problem.

##### A. Green's function

Denoting the (conserved) wave vector parallel to the system surfaces by  $\mathbf{k}=(k_x, k_y)$ , we write the wave vector of an upward (downward) propagating wave in an  $l$ th layer as  $\mathbf{K}_l^\pm = \mathbf{k} \pm \beta_l \hat{\mathbf{z}}$ , where

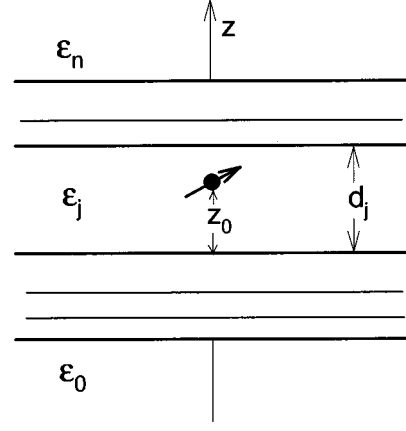


FIG. 1. System considered schematically.  $\varepsilon_l = n_l^2$  are the dielectric functions of the layers.

$$\beta_l = \sqrt{k_l^2 - k^2} = \beta_l' + i\beta_l'', \quad \beta_l' \geq 0, \quad \beta_l'' \geq 0. \quad (4.1)$$

The Green's-function element relating an observation point  $\mathbf{r}$  in the cavity to a source point  $\mathbf{r}_0$  in the cavity then reads

$$\vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0; \omega) = -\frac{4\pi}{k_j^2} \hat{\mathbf{z}}\hat{\mathbf{z}}\delta(\mathbf{r}-\mathbf{r}_0) + \sum_{q=p,s} \vec{\mathbf{G}}_q'(\mathbf{r}, \mathbf{r}_0; \omega), \quad 0 < z, z_0 < d_j$$

$$\begin{aligned} \vec{\mathbf{G}}_q'(\mathbf{r}, \mathbf{r}_0; \omega) &= \frac{i}{2\pi} \int \frac{d^2\mathbf{k}}{\beta_j} \xi_q \frac{e^{i\beta_j d_j}}{D_{qj}} \\ &\times [\mathcal{E}_{qj}^>(\mathbf{k}, \omega; z) \mathcal{E}_{qj}^<(-\mathbf{k}, \omega; z_0) \theta(z - z_0) \\ &+ \mathcal{E}_{qj}^<(\mathbf{k}, \omega; z) \mathcal{E}_{qj}^>(-\mathbf{k}, \omega; z_0) \theta(z_0 - z)] \\ &\times e^{i\mathbf{k}\cdot(\boldsymbol{\rho}-\boldsymbol{\rho}_0)}. \end{aligned} \quad (4.2)$$

Here  $\mathbf{r} \equiv (\boldsymbol{\rho}, z)$ ,  $\xi_p = 1$ ,  $\xi_s = -1$ , and

$$D_{qj} = 1 - r_{j-}^q r_{j+}^q e^{2i\beta_j d_j}, \quad (4.3)$$

with  $r_{j\pm}^q \equiv r_{j/n(0)}^q$  being the reflection coefficients of the upper (lower) cavity mirror. These coefficients obey the usual recurrence relations [18]. The functions  $\mathcal{E}_{qj}^<$  and  $\mathcal{E}_{qj}^>$  describe the  $z$  dependence of the electric field in the cavity of a  $q = p$  polarized or a  $q = s$  polarized plane wave of unit strength incident on the system from its upper (downward) and lower (upward) side, respectively. They are given by

$$\mathcal{E}_{qj}^{\pm}(\mathbf{k}, \omega; z) = \hat{\mathbf{e}}_{qj}^{\mp}(\mathbf{k}) e^{-i\beta_j z^{\mp}} + r_{j\mp}^q \hat{\mathbf{e}}_{qj}^{\pm}(\mathbf{k}) e^{i\beta_j z^{\mp}},$$

$$z^- \equiv z, \quad z^+ \equiv d_j - z$$

$$\hat{\mathbf{e}}_{pj}^{\pm}(\mathbf{k}) = \frac{1}{k_j} (\pm \beta_j \hat{\mathbf{k}} + k \hat{\mathbf{z}}), \quad \hat{\mathbf{e}}_{sj}^{\pm}(\mathbf{k}) = \hat{\mathbf{k}} \times \hat{\mathbf{z}}, \quad (4.4)$$

where  $\hat{\mathbf{e}}_{qj}^\mp$  are the orthonormal (complex) polarization vectors associated with the downward (upward) propagating wave in the cavity.

### B. Molecular decay rate

We insert the Green's function (4.2) into Eq. (2.1) [or Eq. (2.3)] and let  $\mathbf{r} \rightarrow \mathbf{r}_0$ , remembering, however, that the terms in  $W$  corresponding to the infinite-cavity case must be averaged over  $\hat{\mathbf{p}}$ . Then, as before, the singular term in the Green's function gives the molecular quasistatic bulk loss  $W_S^0$ . One can therefore write

$$W(z_0) = W_S^0 + \bar{W}(z_0), \quad (4.5)$$

where  $\bar{W}$  is given through the nonsingular part of the Green's function and, in most cases, gives the only contribution to the molecular loss.

Omitting hereafter the index  $j$  denoting the cavity, we have

$$\begin{aligned} \bar{W}(z_0) = & W_T^0 \text{Re} \frac{3}{4\pi} \int \frac{d^2\mathbf{k}}{k'\beta} \sum_{q=p,s} \xi_q \frac{e^{i\beta d}}{D_q} \\ & \times \mathcal{E}_q^>(\mathbf{k}, \omega; z_0) \cdot \hat{\mathbf{p}} \mathcal{E}_q^<(-\mathbf{k}, \omega; z_0) \cdot \hat{\mathbf{p}}, \end{aligned} \quad (4.6)$$

where  $W_T^0$  is classically given by Eq. (3.4) or quantum mechanically by  $\hbar\omega\Gamma_{\text{SE}}^0$  of Eq. (3.7). Developing  $\mathcal{E}$ 's along  $\hat{\mathbf{k}}$ ,  $\hat{\mathbf{z}}$ , and  $\hat{\mathbf{k}} \times \hat{\mathbf{z}}$  and performing the angular integration ( $\hat{\mathbf{k}} = \cos\varphi\hat{\mathbf{x}} + \sin\varphi\hat{\mathbf{y}}$ ), we obtain

$$\begin{aligned} \bar{W}(z_0) = & W_T^0 \text{Re} \frac{3}{4} \int_0^\infty \frac{dkk}{k'\beta} \left\{ \left[ \frac{1}{D_p} \frac{\beta^2}{k^2} (1 - r_-^p e^{2i\beta z_0^-}) \right. \right. \\ & \times (1 - r_+^p e^{2i\beta z_0^+}) \\ & \left. \left. + \frac{1}{D_s} (1 + r_-^s e^{2i\beta z_0^-}) (1 + r_+^s e^{2i\beta z_0^+}) \right] \hat{\mathbf{p}}_{\parallel}^2 \right. \\ & \left. + \frac{1}{D_p} \frac{2k^2}{k^2} (1 + r_-^p e^{2i\beta z_0^-}) (1 + r_+^p e^{2i\beta z_0^+}) \hat{\mathbf{p}}_{\perp}^2 \right\}, \end{aligned} \quad (4.7)$$

where  $\hat{\mathbf{p}}_{\parallel}$  and  $\hat{\mathbf{p}}_{\perp}$  describe the orientation of the transition dipole relative to the cavity mirrors. To extract the remaining infinite-cavity part  $\bar{W}^0$  from  $\bar{W}$ , we set  $r_{\pm}^q = 0$  in Eq. (4.7). This gives

$$\bar{W}^0 = W_T^0 \text{Re} \frac{3}{4} \int_0^\infty \frac{dkk}{k'\beta} \left[ \left( \frac{\beta^2}{k^2} + 1 \right) \hat{\mathbf{p}}_{\parallel}^2 + \frac{2k^2}{k^2} \hat{\mathbf{p}}_{\perp}^2 \right]. \quad (4.8)$$

Of course, upon prescribed averaging [ $\langle \hat{\mathbf{p}}_{\parallel}^2 \rangle = 2/3$  and  $\langle \hat{\mathbf{p}}_{\perp}^2 \rangle = 1/3$ ], the quantity  $\bar{W}^0$  becomes  $W_T^0$ , as expected. Note that  $\bar{W}^0$ , as it stands, diverges for absorbing cavities owing to the appearance of a nonphysical quasistatic contribution at the upper limit of integration in Eq. (4.8). Therefore, taking its transverse part  $\bar{W}^0 - \lim_{c \rightarrow \infty} \bar{W}^0$  also leads to the correct result. For a transparent ( $k$  is real) cavity, however,  $\bar{W}^0 = W_T^0$

explicitly. This can be easily verified by noting that, in this case, the operation  $\text{Re}$  cuts the above integral at  $\tilde{k}$ .

Equation (4.7) is of a form well known from the theory of molecular fluorescence and energy transfer in transparent layers [10,11]. In [13] we obtained it by making a direct generalization of these classical results for simple systems and conjectured that it also described the molecular power loss in an arbitrary absorbing multilayered system. The present derivation justifies this approach provided, however, that the infinite-cavity part of the total loss is handled more carefully. Actually, according to the above discussion, Eq. (4.7) correctly describes the molecular loss not only for transparent cavities but (i) for a freely rotating (unoriented) molecule, where averaging  $\bar{W}$  over  $\hat{\mathbf{p}}$  is needed, and (ii) if solely the SE power is considered, in which case  $\bar{W} - \lim_{c \rightarrow \infty} \bar{W}$  is to be calculated. A generally valid result is, however,

$$\bar{W}(z_0) = W_T^0 + W^{\text{sc}}(z_0), \quad (4.9)$$

where  $W^{\text{sc}}$  comes from the molecular (dipole) field scattered from the cavity walls and is obtained by subtracting Eq. (4.8) from Eq. (4.7). This finally leads to the normalized molecular decay rate  $\hat{\Gamma} = W/W_T^0$  in the cavity:

$$\hat{\Gamma}(z_0) = \hat{\Gamma}_S^0 + 1 + \hat{\Gamma}^{\text{sc}}(z_0), \quad (4.10a)$$

where

$$\hat{\Gamma}_S^0(z_0) = \frac{3\kappa(\omega)}{|\varepsilon(\omega)|^2} \left( \frac{c}{\omega R_m} \right)^3 \quad (4.10b)$$

is the normalized molecular decay rate due to the quasistatic losses in the bulk of the cavity [14] and

$$\begin{aligned} \hat{\Gamma}^{\text{sc}}(z_0) = & \text{Re} \frac{3}{4} \int_0^\infty \frac{dkk}{k'\beta} \left\{ \left[ \frac{\beta^2}{k^2} \frac{1}{D_p} (2r_-^p r_+^p e^{2i\beta d} - r_-^p e^{2i\beta z_0^-} \right. \right. \\ & - r_+^p e^{2i\beta z_0^+}) + \frac{1}{D_s} (2r_-^s r_+^s e^{2i\beta d} + r_-^s e^{2i\beta z_0^-} \\ & \left. \left. + r_+^s e^{2i\beta z_0^+}) \right] \hat{\mathbf{p}}_{\parallel}^2 + \frac{2k^2}{k^2} \frac{1}{D_p} (2r_-^p r_+^p e^{2i\beta d} + r_-^p e^{2i\beta z_0^-} \right. \\ & \left. \left. + r_+^p e^{2i\beta z_0^+}) \hat{\mathbf{p}}_{\perp}^2 \right\} \end{aligned} \quad (4.10c)$$

is the normalized total cavity-induced decay rate of the molecule.

Clearly,  $W^{\text{sc}} = W_T^0 \hat{\Gamma}^{\text{sc}}$  fully describes the effect of field confinement in the cavity on the decay rate as well as molecular losses due to its interaction with the cavity mirrors. The corresponding quasistatic losses  $W_S^{\text{sc}}$  are obtained from Eq. (4.10c) in the  $c \rightarrow \infty$  limit. As seen, these losses are due to the excitation of  $p$ -polarized waves with the large wave vectors  $k$  ( $\beta_l = ik$ ) and, according to Eq. (4.1), cannot be explicitly separated from the retarded losses in  $W^{\text{sc}}$ . Therefore, further extraction of the SE contribution to  $\hat{\Gamma}$  can be

done only numerically for realistic cavities. This can be performed, e.g., by calculating the transverse part of Eq. (4.10) numerically:

$$\hat{\Gamma}_{\text{SE}}(z_0) = 1 + \hat{\Gamma}^{\text{sc}}(z_0) - \frac{c^3}{\omega^3} \lim_{c \rightarrow \infty} \frac{\omega^3}{c^3} \hat{\Gamma}^{\text{sc}}(z_0), \quad (4.11)$$

where the last term on the right-hand side represents the normalized molecular quasistatic rate  $\hat{\Gamma}_S^{\text{sc}} = W_S^{\text{sc}}/W_T^0$  (see the Appendix). Alternatively, one may simply omit  $\Gamma_S^0$  and cut the integral in Eq. (4.10c) at a wave vector  $K$  that is larger than the wave vector of any polaritonic mode in the system. We note, however, that  $W_S^{\text{sc}}$  is significant only for molecules close ( $\tilde{k}'z_0 < 1$ ) to a lossy mirror. It has been discussed in a number of papers concerning molecular energy transfer at interfaces, e.g., in the context of molecular fluorescence near a mirror [10,11] or in surface physics [23], and its proper account demands a microscopic description of the corresponding mirror (interface). For molecules at larger distances from the mirrors,  $W_S^{\text{sc}}$  may, in a very good approximation, be neglected and for a lossless system it vanishes.

Equation (4.10) is the main result of this work. It extends the early classical results for the molecular decay rate in a transparent layers [10,11] to fully absorbing multilayered systems. Also, it provides a generalization of the corresponding QED result ( $r_{\pm}^q = 0$  and  $\varepsilon = 1$ ) derived by Wylie and Sipe [7] to finite and absorbing cavities as well as that ( $r_{\pm}^q = 0$ ) of Barnett *et al.* [14] to finite planar cavities. To make contact with other QED theories, we return to Eq. (4.7) and, as in [13], use the identity

$$D_q = \frac{1}{2} [(1 - r_-^q e^{2i\beta z_0^-})(1 + r_+^q e^{2i\beta z_0^+}) + (1 + r_-^q e^{2i\beta z_0^-}) \times (1 - r_+^q e^{2i\beta z_0^+})]$$

to rewrite Eq. (4.7) in terms of  $\hat{W} = \bar{W}/W_T^0$  as

$$\hat{W}(z_0) = \frac{3}{4} \text{Re} \int_0^\infty \frac{dk}{\tilde{k}'\beta} k \left\{ \left[ \frac{\beta^2}{k^2} F_p^{\parallel*}(k, z_0) + F_s(k, z_0) \right] \hat{\mathbf{p}}_{\parallel}^2 + \frac{2k^2}{k^2} F_p^{\perp}(k, z_0) \hat{\mathbf{p}}_{\perp}^2 \right\},$$

$$F_p^{\parallel}(k, z_0) = \frac{1}{2|D_p|^2} [I_p^+(k, z_0) |1 - r_-^p e^{2i\beta z_0^-}|^2 + \{\pm \rightarrow \mp\}],$$

$$F_p^{\perp}(k, z_0) = \frac{1}{2|D_p|^2} [I_p^+(k, z_0) |1 + r_-^p e^{2i\beta z_0^-}|^2 + \{\pm \rightarrow \mp\}],$$

$$F_s(k, z_0) = \frac{1}{2|D_s|^2} [I_s^+(k, z_0) |1 + r_-^s e^{2i\beta z_0^-}|^2 + \{\pm \rightarrow \mp\}], \quad (4.12)$$

where

$$I_{\pm}^{\pm}(k, z) = 1 - |r_{\pm}^q|^2 e^{-4\beta''z^{\pm}} + 2i \text{Im} r_{\pm}^q e^{2i\beta z^{\pm}} \quad (4.13)$$

and the symbol  $\{\pm \rightarrow \mp\}$  denotes the expression in the corresponding bracket with changed subscripts and superscripts.

Since for a transparent cavity we have, for example,

$$\text{Re} \frac{1}{\beta} I_{\pm}^{\pm}(k, z_0) = \frac{1}{|\beta|} \times \begin{cases} 1 - |r_{\pm}^q|^2, & k \leq \tilde{k} \\ 2 \text{Im} r_{\pm}^q e^{-2\beta''z_0^{\pm}}, & k > \tilde{k}, \end{cases}$$

in this case  $\hat{W}$  naturally splits into a part  $\hat{W}'$  describing the molecular decay into the propagating ( $\beta$  is real) waves in the cavity and a part  $\hat{W}''$  coming from the molecular interaction with the evanescent ( $\beta$  is imaginary) waves in the cavity.  $\hat{\Gamma}' = \hat{W}'$  precisely reproduces the normalized SE rate as derived by De Martini *et al.* by explicitly employing the quantized radiation field in an empty ( $\varepsilon = 1$ ) cavity with lossless mirrors [24]. De Martini *et al.* have demonstrated that  $\hat{\Gamma}'$  correctly reproduces the SE rate in a number of special cases considered previously. In a dielectric ( $\varepsilon > 1$ ) cavity, however, in addition to the decay rate due to the coupling of the molecule to the unbound (photonlike) waves,  $\hat{\Gamma}'$  also gives the molecular decay rate due to its coupling to the waveguide modes of the cavity.  $\hat{\Gamma}'' = \hat{W}''$  describes the decay rate of the molecule due to its interaction with the waves guided by the cavity mirrors and, for a nearby molecule, due to its quasistatic interaction with the mirrors. Clearly, this picture remains basically valid also in (weakly) absorbing cavities, although the mode pattern is smeared since  $\beta$  is complex for all waves and, accordingly, a sharp distinction between propagating and evanescent waves is lost.

We end this section with a remark concerning the molecular decay rate in absorbing cavities of other shapes. First, we note that generally, according to Eq. (2.3),

$$\hat{\Gamma}^{\text{sc}}(\mathbf{r}_0) = \frac{3}{2\tilde{k}'(\omega)} \hat{\mathbf{p}} \cdot \text{Im} \vec{\mathbf{G}}^{\text{sc}}(\mathbf{r}_0, \mathbf{r}_0; \omega) \cdot \hat{\mathbf{p}}, \quad (4.14)$$

where  $\vec{\mathbf{G}}^{\text{sc}}$  is the Green's function for the field scattered from the cavity walls. For an absorbing planar cavity,  $\vec{\mathbf{G}}^{\text{sc}}(\mathbf{r}_0, \mathbf{r}_0; \omega)$  is easily recognized in Eq. (4.10c) using  $\hat{\mathbf{p}}_{\parallel}^2 = \hat{\mathbf{p}} \cdot (\hat{\mathbf{x}}\hat{\mathbf{x}} + \hat{\mathbf{y}}\hat{\mathbf{y}}) \cdot \hat{\mathbf{p}}$  and  $\hat{\mathbf{p}}_{\perp}^2 = \hat{\mathbf{p}} \cdot \hat{\mathbf{z}}\hat{\mathbf{z}} \cdot \hat{\mathbf{p}}$ . It differs from the corresponding Green's function element for a transparent, or empty, cavity only in that this time  $\tilde{k}$  is complex [18]. This is a general property of the Green's function. Indeed, since material parameters of a system enter the Green's function only through  $\tilde{k}$ 's for various regions,  $\vec{\mathbf{G}}$  obtained for a lossless system is extended to describe the corresponding dispersive and absorbing system simply by replacing the relevant real  $\tilde{k}$ 's with their complex counterparts. Now, numerous considerations of the decay rate in transparent cavities of various shapes ended with Eq. (4.10a) and the form Eq. (4.14) for  $\hat{\Gamma}^{\text{sc}}$ . Therefore, with the above replacement in  $\vec{\mathbf{G}}^{\text{sc}}$  and an obvious modification of the prefactor in Eq. (4.14), these results describe the decay rate in the corresponding absorbing systems as well [25].

## V. DISCUSSION

In this section we analyze the effects of cavity absorption on various contributions to the total decay rate in a dielectric cavity. As is the case in practical applications of controlled SE, we assume a weakly absorbing cavity ( $\kappa \ll \eta$ ), i.e., we consider a molecular transition not coinciding with an absorption resonance in the cavity. In this case, one may still differentiate between the (dominantly) propagating ( $\beta' \geq \beta''$ ) and the (dominantly) evanescent ( $\beta'' > \beta'$ ) waves in the cavity. According to Eq. (4.1), these two classes of waves are described by the wave vectors  $k \leq k'$  and  $k > k'$ , respectively, where  $k' = \sqrt{\epsilon'} \omega / c$ . In the configurations with  $\sqrt{\epsilon'} > \eta_E$ , where  $E=0$  or  $E=n$  denotes the external layer with a larger index of refraction, propagating waves can be further recognized as photonlike (unbound externally) waves with the wave vectors  $0 \leq k \leq k_E$  and cavity-guided waves (evanescent externally) described by  $k_E < k \leq k'$ . In such systems, the evanescent waves actually represent the waves guided by the cavity mirrors since they are also bound externally. Referring to Eq. (4.10), the decay rate can therefore be written as

$$\hat{\Gamma}(z_0) = \hat{\Gamma}_{\text{ph}}(z_0) + \hat{\Gamma}_{\text{CGW}}(z_0) + \hat{\Gamma}_{\text{MGW}}(z_0), \quad (5.1)$$

where the separate contributions of photonlike (ph), cavity-guided waves (CGW), and mirror-guided waves (MGW) are determined by the integrals in Eq. (4.10c) over the corresponding wave vector ranges. In writing Eq. (5.1) we have ignored  $\hat{\Gamma}_S^0$  as it is not completely specified in the present approach. Note also that  $\hat{\Gamma}_{\text{MGW}}$  incorporates the decay rate  $\hat{\Gamma}_S^{\text{sc}}$  owing to the quasistatic molecule-mirror interaction.

All modes supported by the system show up as resonances in  $|D_p|^{-2}$  and  $|D_s|^{-2}$  considered as functions of the wave vector  $k$ . For photonlike waves, we set

$$r_{\pm}^q(k) = \xi_q |r_{\pm}^q(k)| e^{2i\phi_{\pm}^q(k)}, \quad \xi_p = 1, \quad \xi_s = -1. \quad (5.2)$$

This transforms  $|D_q|^{-2}$  into a familiar (Airy) form [26] generalized, however, for absorption in the system:

$$\frac{1}{|D_q|^2} = \frac{(1 - |r_{-}^q r_{+}^q| e^{-\alpha d})^{-2}}{1 + F_q \sin^2(\beta' d + \phi_q)}, \quad (5.3a)$$

where

$$F_q = \frac{4|r_{-}^q r_{+}^q| e^{-\alpha d}}{(1 - |r_{-}^q r_{+}^q| e^{-\alpha d})^2} \quad (5.3b)$$

determines the cavity finesse  $f_q = (\pi/2) \sqrt{F_q}$  for a wave. Here  $\alpha = 2\beta'' = (\omega/c)^2 \epsilon'' / \beta'$  is the absorption coefficient of the cavity for the wave and  $\phi_q = \phi_{-}^q + \phi_{+}^q$  is the cumulative phase half shift of the wave due to two successive reflections in the cavity.

The effects of the cavity absorption on  $\hat{\Gamma}_{\text{ph}}$  can be deduced from the properties of the above generalized Airy function. Thus the wave vectors  $k_{qm}$  ( $\beta'_{qm}$ ) of the cavity resonant (antiresonant) modes are found as solutions of

$$\beta' d = m\pi - \phi_{-}^q(k) - \phi_{+}^q(k), \quad k \leq k_E, \quad (5.4)$$

with  $m$  being an integer (half integer). Owing to the appearance of  $\phi'$ 's, dissipation in the system affects the mode positions. Specially, for an  $M$ th-order SE enhancement (inhibition) cavity, one has  $k_M = 0$  and  $\beta'_M = \tilde{k}'$ . This gives the tuning condition for the cavity  $d_M = [M - \phi(0)/\pi] \lambda / 2\eta$ , so that  $d_M$ 's are shifted from the standard half- or quarter-wavelength values in an absorbing system. The mode widths are found in a standard way [26]. Owing to the decrease of the average reflectivity of the cavity mirrors by the factor  $\exp(-\alpha_{qm}d)$ , they are increased in an absorbing cavity. Since the peak values of the corresponding resonances are simultaneously decreased, the dissipation in the cavity spoils conditions for either enhanced or suppressed SE.

The wave vectors of guided modes and their widths are given by zeros  $\tilde{k}_{qm}$  of  $D_q$  in the corresponding regions of the complex- $k$  plane. For waveguide modes it is convenient to use the representation  $r_{\pm}^q = \exp(-2i\tilde{\phi}_{\pm}^q)$ , as appropriate for the total reflection of the waves in the cavity. Therefore, owing to the dissipation in the system, the phases  $\tilde{\phi}_{\pm}^q$  are complex. This leads to the usual mode equation of a planar waveguide

$$\beta d = m\pi + \tilde{\phi}_{-}^q(k) + \tilde{\phi}_{+}^q(k), \quad k > k_E, \quad (5.5)$$

where  $m$  is an integer. Using the recurrence relations for reflection coefficients [18], the solutions for CGW's and MGW's can be found using standard methods of integrated optics in the regions  $\beta' > \beta''$  and  $\beta'' > \beta'$ , respectively. For  $k$  close to a  $\tilde{k}_{qm}$ , one can use  $D_q(k) \approx D'_q(\tilde{k}_{qm})(k - \tilde{k}_{qm})$ , where  $D'_q = dD_q/dk$ . Accordingly,

$$\frac{1}{|D_q|^2} = \frac{|D'_q(\tilde{k}_{qm})|^{-2}}{(k - \tilde{k}_{qm})^2 + (\tilde{k}_{qm}'')^2}, \quad (5.6)$$

so that the waveguide-mode part of the SE spectrum consists of a series of Lorentzian peaks. As before, since absorption causes increase of  $\tilde{k}_{qm}''$ , guided-wave resonances are suppressed in absorbing cavities.

We illustrate these considerations by numerically analyzing the decay rate in a symmetric dielectric cavity formed by two metallic (Ag) mirrors and surrounded by air (see the inset in Fig. 2) taken as a simple system that supports all kinds of modes. Figure 2 illustrates the mode pattern of such a transparent and an absorbing cavity. It is obtained from Eq. (4.12) by calculating the normalized molecular power distribution over  $k$  defined as [13]

$$\hat{W}(k, z_0) = \frac{\tilde{k}' |\beta|}{k} \frac{d\hat{W}(z_0)}{dk}. \quad (5.7)$$

Since for propagating waves in a transparent cavity one may set  $k = \tilde{k} \eta \sin \vartheta$ , this definition leads to the usual SE intensity distribution  $\hat{W}(\vartheta, z_0)$  over the polar angles  $\vartheta \leq \pi/2$  in the cavity. In general, however, it gives a suitably weighted  $k$ -space power spectrum of the molecule. In order to pick up all modes, we have assumed a freely rotating molecule lo-

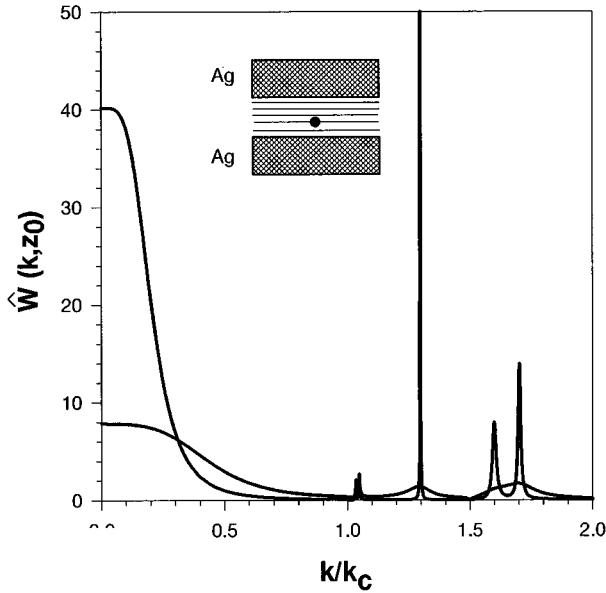


FIG. 2. Normalized power loss distribution of a freely rotating molecule in the cavity shown in the inset. The upper and lower curves correspond to the  $n=1.5$  (transparent) and  $n=1.5+i0.05$  (absorbing) cavities, respectively. With  $n_{\text{Ag}}=0.05+i5.858$  at the SE vacuum wavelength  $\lambda=582$  nm [27] and the thickness of the mirrors  $l_m=0.06\lambda$ , the transparent cavity finesse is  $f(0)\approx 31$ . In terms of the SE wavelength  $\bar{\lambda}=\lambda/1.5$  in the cavity, the cavity length is  $d=0.872\bar{\lambda}$  and the molecule position is  $z_0=0.186\bar{\lambda}$ . The wave vector  $k$  is expressed in units of  $k_c=\omega/c$ .

ated at the first maximum (antinode) of  $\text{Re}F_{p+}^{\parallel}(0, z_0) = \text{Re}F_{s+}(0, z_0)$  in the second-order enhancement ( $\lambda$ ) cavity. Since, for a freely rotating molecule,  $\hat{W}(k, z_0) = \hat{\Gamma}(k, z_0)$ , the curves actually represent the corresponding probabilities of the decay into a  $k$  mode of the system.

Three different regimes in  $\hat{W}(k, z_0)$  anticipated in Eq. (5.1) are clearly resolved in the figure. In the region  $0 \leq k/k_c \leq 1$ , one deals with  $\hat{W}_{\text{ph}}(k, z_0)$ . As mentioned above, in the case of the transparent cavity, this part of the spectrum corresponds to the total (above and below the molecule) intensity distribution  $\hat{W}(\vartheta, z_0)$  over the radiation modes in the cavity expressed in the figure as a function of  $\eta \sin \vartheta$ . The system is adjusted so as to obtain enhanced SE in the normal direction. As seen, only the fundamental cavity mode exists for this cavity length. Note that  $\hat{W}_{\text{ph}}(0, z_0)$  is 80 times larger than the SE intensity of the freely rotating molecule in the infinite cavity [ $\hat{W}_{\text{rad}}^0(\vartheta) = 1/2$ ]. For a transparent cavity, Eq. (4.12) correctly describes the SE spectrum for an oriented molecule too. Accordingly, for an oriented molecule, this part of the spectrum should be multiplied by  $(3/2)\mathbf{p}_{\parallel}^2$  since it comes only from the parallel dipole transition moment. The effect of absorption in the cavity on  $\hat{W}_{\text{ph}}(k, z_0)$  is seen in the increase of the fundamental-mode width and a strong suppression of SE intensity.

The waveguide-mode part of the spectrum can be understood on the basis of the properties of a symmetric metal-clad planar waveguide [28] taking into account the finite thickness of the cladding (mirrors). It is dominated by the two TM modes in the region  $k/k_c > 1.5$  formed by the anti-

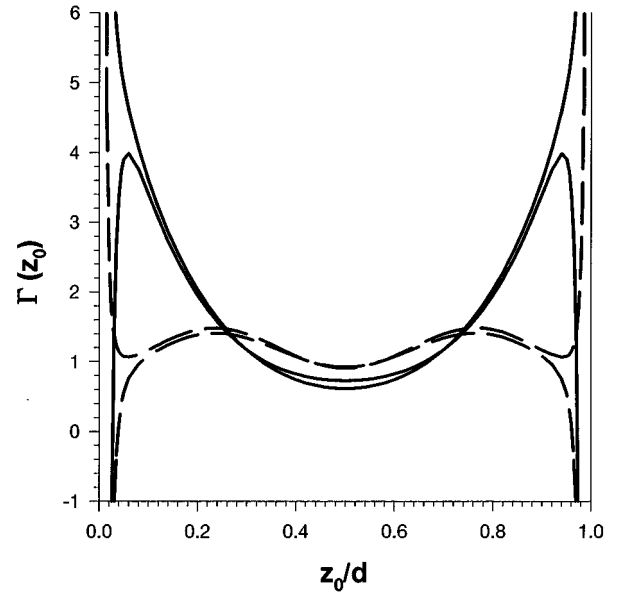


FIG. 3. Dependence of the decay rates  $\hat{\Gamma}_{\perp}$  (solid line) and  $\hat{\Gamma}_{\parallel}$  (dashed line) on the molecule position  $z_0$  in the transparent (upper line) and the absorbing (lower line) cavity described in Fig. 2. The cavity length is  $d=0.872\bar{\lambda}$ .

symmetric and symmetric coupling of surface plasmon-polariton (SP) modes of the mirrors [29]. We denote these modes by  $\text{SP}_{-}$  ( $\text{TM}_1$ ) and  $\text{SP}_{+}$  ( $\text{TM}_0$ ) according to the symmetry of their fields across the system. Since the  $z$  component of the SP field dominates, this part of the spectrum is mainly due to the (average) perpendicular dipole transition moment. The  $\text{SP}_{-}$  mode has a smaller wave vector and is cut off at small cavity lengths. For large  $d$ , the  $\text{SP}_{-}$  and  $\text{SP}_{+}$  modes become degenerate as the SP of the mirrors become decoupled. Accordingly, the  $\text{SP}_{+}$  mode persists for all cavity lengths and, as discussed previously [13,18], provides an efficient decay channel for molecules close to a mirror that may spoil the conditions for controlled SE. In this respect, one must not be confused by Fig. 2 in judging the contribution of the SP modes to the total SE rate since the weighting factor ( $\beta$ ) in Eq. (5.7) is very small in this  $k$  region. Actually, for the system presented, nearly half the SE rate is due to the coupling of the molecule to the SP modes. As before, the effect of the cavity absorption on this part of the spectrum is seen in the increased widths of the modes and an overall (strong) suppression of SE intensity.

The role of various terms in Eq. (5.1) is best seen in Fig. 3, where we have plotted the position dependence of the decay rate in the transparent and the absorbing cavity of Fig. 2 for the perpendicular and the parallel transition dipole orientations. Whereas  $\hat{\Gamma}_{\text{ph}}^{\perp}(z_0)$  is the largest in the middle of the cavity, the total decay rate  $\hat{\Gamma}_{\perp}(z_0)$  is actually the smallest at this position. This is due to the increasing coupling of the molecule to the SP modes as it moves from the cavity center. On the other hand, since the molecule-SP coupling is weak for this transition dipole orientation,  $\hat{\Gamma}_{\parallel}(z_0)$  is the largest around the usual  $z_0=\lambda/4$  and  $z_0=3\lambda/4$  molecule positions. At short molecule-mirror distances, the rates are governed by the quasistatic molecule-mirror interaction inadequately accounted for in the point-dipole model for the molecule and

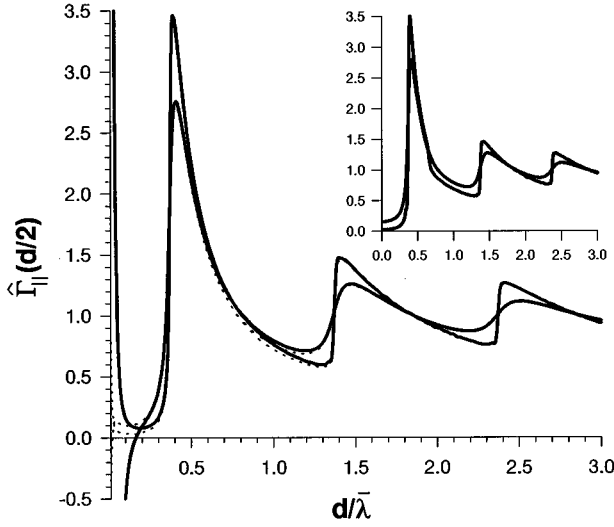


FIG. 4. Dependence of the total decay rate  $\hat{\Gamma}_{\parallel}$  (solid line) and the SE rate  $\hat{\Gamma}_{SE}^{\parallel}$  (dotted line) on the cavity length  $d$  for the systems described in Fig. 2. The inset shows the rate  $\hat{\Gamma}'_{\parallel}$  for the decay into propagating waves in the cavity only. The upper and lower curves refer to the transparent and absorbing cavities, respectively. The molecule position is  $z_0 = d/2$ .

the macroscopic approach to the cavity mirrors [7]. For this reason,  $\hat{\Gamma}_{\perp}(z_0)$  and  $\hat{\Gamma}_{\parallel}(z_0)$  in the transparent cavity diverge when  $z_0$  approaches a mirror. The corresponding curves for the absorbing cavity tend to negative values owing to our omission of the quasistatic bulk rate  $\Gamma_S^0$ . Using Eq. (4.10b), in the Appendix we show that the total quasistatic rate  $\Gamma_S^0 + \hat{\Gamma}_S^{\text{sc}}(z_0)$  is, of course, positive. However, as one may conclude from the figure ( $d \approx \bar{\lambda}$ ), the effective range of the quasistatic molecule-mirror interaction is of the order of  $\bar{\lambda}/10$ . Accordingly, this theory adequately describes the molecular decay in the whole cavity space excluding the layers of the thickness  $\sim 1/\bar{k}'$  at the mirrors. As seen in the figure, the effect of the cavity absorption on  $\hat{\Gamma}_{\perp}(z_0)$  and  $\hat{\Gamma}_{\parallel}(z_0)$  is very small through this cavity region, except, of course, for the enhancement by  $\hat{\Gamma}_S^0$ .

The dependence of the decay rate on the cavity length is illustrated in Figs. 4 and 5, where we have plotted  $\hat{\Gamma}(d/2)$  for the parallel and perpendicular transition dipole orientations, respectively, in the two cavities of Fig. 2. One again observes that, disregarding the appearance of  $\hat{\Gamma}_S^0$ , the cavity absorption affects  $\hat{\Gamma}_{\parallel}$  and  $\hat{\Gamma}_{\perp}$  only slightly and, as concluded earlier, tends to spoil the conditions for controlled SE. At larger  $d$ 's, the curves exhibit the familiar enhanced or suppressed SE behavior characteristic for a cavity with realistic mirrors [24,13]. However, it is worth noticing that  $\hat{\Gamma}_{\parallel}(d/2) > 3$  at the first peak, i.e., the predicted rate is larger than the rate in the corresponding cavity with ideal ( $r_{\pm}^q = \xi_q$ ) mirrors. A more detailed analysis reveals that both  $\hat{\Gamma}_{\text{ph}}(d/2)$  and  $\hat{\Gamma}_{\text{CGW}}(d/2)$  contribute to this peak and that, because of  $\hat{\Gamma}_{\text{CGW}}(d/2)$ , it increases rather strongly with the cavity refraction index, e.g., for  $n = 3\hat{\Gamma}_{\parallel}(d/2) \approx 10$  at the first peak. Therefore, this somewhat surprising result is a combined medium-cavity effect that perhaps, in view of technological

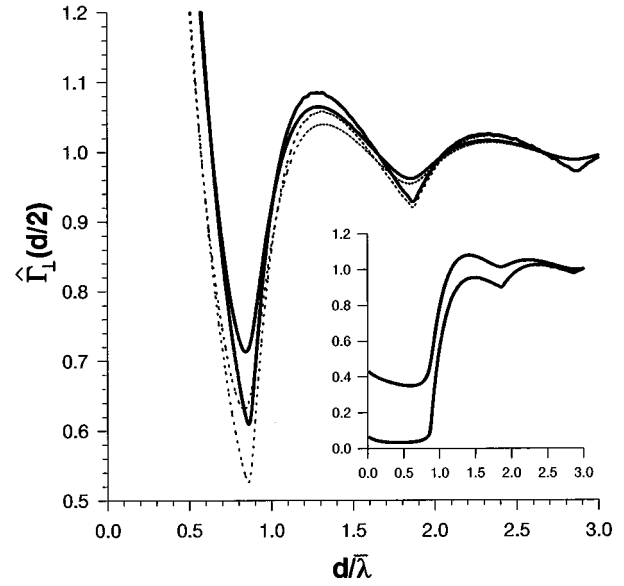


FIG. 5. Same as in Fig. 4, but for the perpendicular dipole orientation.

applications, deserves further exploration.

The small- $d$  behavior of  $\hat{\Gamma}_{\parallel}(d/2)$  and  $\hat{\Gamma}_{\perp}(d/2)$  can be understood by invoking the mirror symmetry of the system and the properties of the SP modes. Owing to the central position of the molecule, the parallel dipole couples only to the  $\text{SP}_{-}$  mode whose parallel (small) field component is symmetric across the system. Since this mode is cut off at small  $d$ , the rate  $\hat{\Gamma}_{\parallel}(d/2)$  is weakly affected by the cavity mirrors for almost all relevant cavity lengths. This is clearly seen in Fig. 4, where the total rate  $\hat{\Gamma}_{\parallel}(d/2)$  is slightly larger than the rate  $\hat{\Gamma}'_{\parallel}(d/2) = \hat{\Gamma}_{\text{ph}}^{\parallel}(d/2) + \hat{\Gamma}_{\text{CGW}}^{\parallel}(d/2)$  for the decay into propagating waves in the cavity only (see the inset). Contrary to this,  $\hat{\Gamma}_{\perp}(d/2)$  increases sharply at small ( $d < \bar{\lambda}$ ) cavity lengths, while  $\hat{\Gamma}'_{\perp}(d/2)$  demonstrates the usual suppressed SE behavior (see the inset in Fig. 5). This behavior of  $\hat{\Gamma}_{\perp}(d/2)$  is a consequence of the sharp increase of  $\hat{\Gamma}_{\text{MGW}}^{\perp}(d/2)$  at small molecule-mirror distances caused by the strong coupling of the perpendicular dipole to the  $\text{SP}_{+}$  mode with the symmetric perpendicular (large) field component.

As before, for  $d \rightarrow 0$ , the rate  $\hat{\Gamma}_{\parallel}(d/2)$  becomes inadequately described in the present approach owing to the prevailing quasistatic molecule-mirror interaction. In addition to this interaction, the perpendicular dipole is strongly coupled to the surface plasmon mode of the system, i.e., to the quasistatic counterpart of the (remaining)  $\text{SP}_{+}$  mode. This causes continuous increase of  $\hat{\Gamma}_{\perp}(d/2)$  in the range presented in Fig. 5. Of course, the corresponding SE rates should always remain finite and positive. To check this, we have used Eq. (4.11) to calculate  $\hat{\Gamma}_{SE}^{\parallel}(d/2)$  and  $\hat{\Gamma}_{SE}^{\perp}(d/2)$ . This method of calculating the SE rate becomes numerically unstable for  $d \rightarrow 0$  as one has to subtract the two uncorrelated divergent terms. Nevertheless, the dotted curves in Figs. 4 and 5 clearly indicate the proper behavior of  $\hat{\Gamma}_{SE}^{\parallel}(d/2)$  and  $\hat{\Gamma}_{SE}^{\perp}(d/2)$ . In turn, this calculation proves that, indeed, to determine the SE rate in a system one may avoid the involved



calculation of the corresponding transverse Green's function by using instead the full Green's function and, when necessary, by extracting its transverse part numerically, as proposed in this work.

## VI. SUMMARY

Using a recently obtained compact form of the Green's function for an absorbing multilayer, in this work we have derived the decay rate of an excited molecule (atom) embedded in a planar cavity (or, generally, a multilayer) by rigorously taking into account cavity dispersion and absorption. In this respect, the presented theory generalized numerous previous considerations of the molecular decay rate in lossless or empty planar cavities. From comparison with these works, a hint emerged for a straightforward extension of the results obtained for lossless cavities of various shapes to the corresponding absorbing cavities.

In addition to the spontaneous emission rate, the total decay rate in an absorbing cavity consists of the contributions coming from the quasistatic (near-field) interaction of the molecule with the cavity medium and, for nearby molecules, also with the cavity mirrors. These contributions were inadequately described in the point-dipole model for the molecule and the macroscopic approach to the mirrors adopted. However, this approach suffices to determine the spontaneous emission rate of the molecule correctly and we have proposed (and checked) a simple method for numerical extraction, when necessary, of the spontaneous emission rate from the otherwise inadequate total decay rate. In this way, the spontaneous emission rate in an absorbing inhomogeneous system can be calculated without having the transverse field quantized or, equivalently, without knowing the corresponding transverse Green's function.

The theory was applied to an analysis of the effects of the weak cavity absorption on the decay rate in a dielectric microcavity formed by two metallic mirrors. A detailed discussion of the various contributions to the decay rate was presented. Dissipation in the cavity strongly reduced the molecular power distribution over the available retarded modes, i.e., the SE intensity. However, except for an overall enhancement by the quasistatic bulk rate, the total decay rate away from the mirrors was weakly affected by the dissipative processes in the cavity. The same holds for the SE rate. As the strong dissipation in the cavity may effectively destroy its finesse, this may no longer be true for the molecular transitions in resonance with those of the cavity medium and

we plan to address this situation in a forthcoming paper along with the frequency dependence of the SE rate in a dispersive and absorbing microcavity.

## APPENDIX: QUASISTATIC RATE

From Eqs. (4.10c) and (4.11) we have

$$\hat{\Gamma}_S^{\text{sc}}(z_0) = \frac{3}{4\eta} \text{Im} \frac{1}{\varepsilon} \int_0^\infty \frac{dkk^2}{(\omega/c)^3} \frac{1}{D} [(r_- e^{-2kz_0^-} + r_+ e^{-2kz_0^+} - 2r_- r_+ e^{-2kd}) \hat{\mathbf{p}}_{\parallel}^2 + 2(r_- e^{-2kz_0^-} + r_+ e^{-2kz_0^+} + 2r_- r_+ e^{-2kd}) \hat{\mathbf{p}}_{\perp}^2], \quad (\text{A1})$$

where  $D = 1 - r_- r_+ \exp(-2kd)$  and  $r_{\pm}$  are obtained from  $r_{\pm}^p$  by setting  $\beta_i = ik$  for all relevant layers. These reflection coefficients, therefore, obey the recurrence relation [18]

$$r_{ij/k} = \frac{r_{ij} + r_{j/k} e^{-2kd_j}}{1 + r_{ij} r_{j/k} e^{-2kd_j}}, \quad r_{ij} = \frac{\varepsilon_j - \varepsilon_i}{\varepsilon_j + \varepsilon_i}. \quad (\text{A2})$$

The above result is valid for small ( $\tilde{k}'d < 1$ ) cavity lengths. For larger cavity lengths and for a molecule close ( $\tilde{k}'z_0 < 1$ ) to a mirror, only that mirror matters. In this case, Eq. (A1) simplifies to

$$\hat{\Gamma}_S^{\text{sc}}(z_0) = \frac{3}{4\eta} \text{Im} \frac{1}{\varepsilon} \int_0^\infty \frac{dkk^2}{(\omega/c)^3} r(k, \omega) e^{-2kz_0} (\hat{\mathbf{p}}_{\parallel}^2 + 2\hat{\mathbf{p}}_{\perp}^2), \quad (\text{A3})$$

where  $r$  and  $z_0$  are the reflection coefficients of the corresponding mirror and the molecule-mirror distance, respectively. A microscopic approach to  $\Gamma_S^{\text{sc}}$  leads to this result with a nonlocal expression for  $r$  [23]. With Eq. (4.10b), the total quasistatic rate is therefore

$$\hat{\Gamma}_S(z_0) = \frac{3}{2} \text{Im} \left\{ -\frac{\eta^2}{\varepsilon} \left[ \frac{1}{(\tilde{k}'R_m)^3} - \frac{(\hat{\mathbf{p}}_{\parallel}^2 + 2\hat{\mathbf{p}}_{\perp}^2)}{(2\tilde{k}'z_0)^3} \right] \times \frac{1}{2} \int_0^\infty dy y^2 r(y/2z_0, \omega) e^{-y} \right\}. \quad (\text{A4})$$

Since  $R_m$  is necessarily smaller than  $z_0$ , in an absorbing cavity  $\hat{\Gamma}_S$  is mainly determined by the bulk rate  $\hat{\Gamma}_S^0$ .

[1] B. Huttner and S. M. Barnett, *Europhys. Lett.* **18**, 487 (1992); *Phys. Rev. A* **46**, 4306 (1992).  
 [2] S.-T. Ho and P. Kumar, *J. Opt. Soc. Am. B* **10**, 1620 (1993).  
 [3] T. Gruner and D.-G. Welsch, *Phys. Rev. A* **51**, 3246 (1995).  
 [4] T. Gruner and D.-G. Welsch, *Phys. Rev. A* **53**, 1818 (1996).  
 [5] 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).  
 [6] G. S. Agarwal, *Phys. Rev. A* **11**, 230 (1975); **12**, 1475 (1975).

[7] J. M. Wylie and J. E. Sipe, *Phys. Rev. A* **30**, 1185 (1984); **32**, 2030 (1985).  
 [8] H. Kuhn, *J. Chem. Phys.* **53**, 101 (1970).  
 [9] K. H. Drexhage, in *Progress in Optics XII*, edited by E. Wolf (North-Holland, Amsterdam, 1974), p. 165.  
 [10] R. R. Chance, A. Prock, and R. Silbey, *Adv. Chem. Phys.* **37**, 1 (1978).  
 [11] G. W. Ford and W. H. Weber, *Phys. Rep.* **113**, 195 (1984).  
 [12] S. M. Barnett, B. Huttner, and R. Loudon, *Phys. Rev. Lett.* **68**,

- 3698 (1992); S. M. Barnett and R. Loudon, *ibid.* **77**, 2444 (1996).
- [13] M. S. Tomaš and Z. Lenac, *Opt. Commun.* **100**, 259 (1993).
- [14] S. M. Barnett, B. Huttner, R. Loudon, and R. Matloob, *J. Phys. B* **29**, 3763 (1996).
- [15] G. Juzeliūnas, *Phys. Rev. A* **55**, R4015 (1997).
- [16] Y. Lee and M. Yamanishi, *Phys. Rev. A* **52**, 2312 (1995).
- [17] H. Nha and W. Jhe, *Phys. Rev. A* **54**, 3505 (1996).
- [18] M. S. Tomaš, *Phys. Rev. A* **51**, 2545 (1995).
- [19]  $G_{\alpha\beta}(\mathbf{r}, \mathbf{r}_0; \omega)$  is, by definition, related to the retarded electromagnetic Green's function  $D_{\alpha\beta}^R(\mathbf{r}, \mathbf{r}_0; \omega)$  in the  $\varphi=0$  gauge via  $G_{\alpha\beta}(\mathbf{r}, \mathbf{r}_0; \omega) = -(1/\hbar)D_{\alpha\beta}^R(\mathbf{r}, \mathbf{r}_0; \omega)$ . See E. M. Lifshitz and L. P. Pitaevskii, *Statistical Physics* (Pergamon, Oxford, 1991), Pt. 2, Chap. 8.
- [20] See Ref. [14]. In this reference, the Green's function is defined as  $(\omega^2/c^2)\vec{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0; \omega)$ . Also, the classical time-varying quantities are defined as twice those in the present work, so that the classical and QED rates coincide provided that  $\mathbf{p} \rightarrow \mathbf{p}_{fi}$ .
- [21] R. J. Glauber and M. Lewenstein, *Phys. Rev. A* **43**, 467 (1991).
- [22] One lets  $\omega/c \rightarrow \tilde{k}$  in the free-space Green's function. More rigorously, Eq. (3.1) is obtained from [19] as  $\vec{\mathbf{G}}^0(\mathbf{r}, \mathbf{r}_0; \omega) = (\tilde{k}^{-2}\nabla\nabla + \vec{\mathbf{I}})e^{i\tilde{k}R}/R$  and using  $\nabla\nabla R^{-1} = (3\hat{\mathbf{R}}\hat{\mathbf{R}} - \vec{\mathbf{I}})R^{-3} - (4\pi/3)\vec{\mathbf{I}}\delta(\mathbf{R})$ .
- [23] See, for example, B. N. J. Persson and W. L. Schaich, *J. Phys. C* **14**, 5583 (1981).
- [24] F. De Martini, M. Marocco, P. Mataloni, L. Crescentini, and R. Loudon, *Phys. Rev. A* **43**, 2480 (1991); F. De Martini, F. Cairo, P. Mataloni, and F. Verzeqnessi, *ibid.* **46**, 4220 (1992); F. De Martini, M. Marocco, P. Mataloni, D. Murra, and R. Loudon, *J. Opt. Soc. Am. B* **10**, 360 (1993).
- [25] For example, the decay rate in an absorbing spherical cavity can be deduced from, e. g., W. Jhe and K. Jang, *Phys. Rev. A* **53**, 1126 (1996) and that in an absorbing system with cylindrical symmetry from T. Erdogan, K. G. Sullivan, and D. G. Hall, *J. Opt. Soc. Am. B* **10**, 391 (1993).
- [26] M. Born and E. Wolf, *Principles of Optics* (Pergamon, New York, 1980), Chap. 7.
- [27] P. B. Johnson and R. W. Christy, *Phys. Rev. B* **6**, 4370 (1972).
- [28] See, for example, I. P. Kaminow, W. L. Mammel, and H. P. Weber, *Appl. Opt.* **13**, 396 (1974).
- [29] For this combination of the surrounding dielectrics ( $\epsilon$ ), each mirror supports one SP at the frequency considered.