Ultimate enhancement of the local density of electromagnetic states outside an absorbing sphere

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The local density of states (LDOS) of the electromagnetic field on the surface of an absorbing dielectric microsphere at frequencies of the whispering-gallery modes (WGMs) is evaluated using the dyadic Green tensors of the electric and magnetic fields. In the calculation, the morphology-dependent resonances in the frequency dependence of the Mie coefficients are described analytically in terms of the resonant frequency and the partial quality factors which allow for light radiation and absorption. The Purcell factor of LDOS enhancement by WGMs is calculated to be proportional to the product of the quality factor and the squared ratio of the light wavelength to the sphere diameter. The ultimate values of the electric-field LDOS enhancement are estimated to be of the order of 10^7 for a fused-silica microsphere in air. The efficiency of radiation of the resonant spontaneous emission from electric dipoles located near the surface outside of a sphere is determined. The characteristics of a dielectric microsphere are compared with those of other cavities.

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I. INTRODUCTION

Many experiments in optics study or apply spatial or spectral concentration of the energy of the electromagnetic field. In particular, such a concentration can be described by a parameter commonly called the local density of states (LDOS) [1,2]. This parameter is the number of the field states per unit volume and unit frequency intervals. A variation of the LDOS brings about an alteration of rates of optical transitions. Thus, it is possible to enhance or inhibit various physical processes including spontaneous emission, fluorescence, Raman scattering, and energy transfer.

An enhancement of spontaneous emission in a cavity was predicted by Purcell in far 1946 [3]. In fact, he estimated the density of states related with the Einstein *A* coefficient. According to Purcell, in a cavity with the quality factor Q and the volume V, the density of states on a resonant frequency is higher than in free space by a factor of [3]

$$f = \frac{3Q\lambda^3}{4\pi^2 V}, \quad (1)$$

where λ is the wavelength of the emission in vacuum. This note of Ref. [3] gave an inceptive impulse for diverse optical research and was appreciated in studies of photonic crystals [4], light-emitting diodes [5], micrometer-sized cavities [6] (such as dielectric microspheres [7], toroidal [8] and micropost cavities [9]), nanocavities [10,11], noble-metal nanoparticles [12], complexes of semiconductor nanoparticles and dielectric microspheres [13], 2D-photonic-crystal defect nanocavities [14], dielectric nanowires [15], metal-dielectric interfaces, refractive media, dielectric slabs (see references in Ref. [1]), etc.

In particular, it was shown that a micrometer-sized dielectric sphere can increase the LDOS by orders of magnitude due to the whispering-gallery modes (WGMs) [16–18]. Such an enhancement is of importance in a single-quantum-dot laser [19], complexes of micrometer- and nanometer-sized particles [13], single-molecule sensors [20–22], and cavity quantum electrodynamics (CQED) atom-microsphere systems [23,24].

The next section briefly reviews modifications of the Purcell formula in studies of microspheres. Relevant calculations executed without characterization of the factor f are also referred to. According to Sec. II, it is desirable to gain insight into the Purcell's estimate. This paper aims at a rigorous derivation of the Purcell factor and evaluation of the ultimate value of the normalized LDOS upon the surface of a dielectric microsphere. For that, Sec. III cites known relations between the LDOS and the dyadic Green tensors of the electric and magnetic fields. Section IV defines the LDOS on the surface of a sphere. Section V gives an approximate formula for calculating a Mie coefficient at frequencies in the region of a morphology-dependent resonance in an absorbing dielectric sphere. As a result, an analog of the Purcell formula describing resonant properties of the sphere is obtained. Section VI presents numeric illustrations and their discussion. Finally, a brief conclusion is given in Sec. VII.

II. HISTORICAL BACKGROUND

Chew [16,17], Ching *et al.* [18] were among the first who revealed the phenomenon of LDOS enhancement in micrometer-sized dielectric spheres. They studied emission from electric dipoles embedded in the spheres. The theoretical models applied the classical electromagnetic theory [16], a quantum-physics expression of the Einstein *A* coefficient through the dyadic Green tensor of the electric field [16], and a procedure of quantization of the electromagnetic field [18,25]. In the latter approach, a sphere of an ideal dielectric was considered as a part of a closed cavity of an infinite size.

Later, a quantum formalism for describing spontaneous decay of an excited two-level atom in the presence of dispersing and absorbing dielectric bodies was developed [26–28]. It was established how the Einstein *A* coefficient relates with the classical dyadic Green tensor of the field in a medium with a complex permittivity. The general theory was applied to study spontaneous decay of an excited atom in

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spherical cavities [26,28]. References [16,28] provided formulas for calculating the transition rates of an excited atom inside or outside of a sphere.

Lange and Schweiger showed a role of microsphere WGMs in the process of Raman scattering [29]. They calculated the external Raman-scattering efficiency defined as the ratio of the Raman- or fluorescence-scattered power radiated by molecules in the vicinity of a sphere to the scattered power of the same molecules in the homogeneous space. For instance, this ratio was computed to be 2×10^6 if a sphere with a refractive index $n=1.5$ was placed in air and had a radius $a=10$ μm, Raman-active molecules were located on the surface of the sphere, the input emission was not in resonance with a particle WGM, and the output emission was in resonance with the TM^1_{60} mode (the size parameter $x=2\pi a/\lambda$ was equal to 64.0 for the input emission and 44.877 376 006 for the output one). In the numeric calculations [29,30], some quantities were likely to be in error because light absorption within a particle was neglected. For example, it was implicitly presumed that widths of morphology-dependent resonances (MDRs) can be as narrow as approximately $x/10^{14}$ for the TM^1_{88} and TE^1_{88} modes. (The correspondent external efficiency was calculated to be about 10^{11} for the double-resonance scattering.) The anticipated Q factors of the order of 10^{14} were much larger than the record value 0.8×10^{10} of Q found experimentally for fused-silica spheres in air [31].

None of the preceding calculations gave an estimate for the ultimate value of the LDOS enhancement. In addition, the role of light absorption was not clearly established. Because of the deficiencies of the numeric simulations, it is useful to apply a simple analytical estimate, like Eq. (1). However, for a cavity with surface modes, the Purcell factor should be modified.

As a rule, the electromagnetic field is not uniform within the cavity. Therefore, the value V is commonly replaced by an effective mode volume $V_{\mathbf{m}}$. This parameter is frequently introduced as

$$V_{\mathbf{m}} = \frac{\int\int\int \mathbf{E}^2(\mathbf{r})d\mathbf{r}}{\max \mathbf{E}^2}, \qquad (2)$$

where $\mathbf{E}$ is the electric field of the mode. If the mode is degenerate, components of the multiplet can have different maxima in different points of space. Therefore, a definition and computation of $V_{\mathbf{m}}$ for a degenerate mode is not a trivial exercise. Nevertheless, the right-hand side of Eq. (1) is often merely multiplied by the mode degeneracy g. In addition, the Purcell factor (1) should be decreased three times if one averages over the radiating-dipole direction. Using these speculations, the following formula is achieved:

$$f = gQ\frac{\lambda^3}{4\pi^2 V_{\mathbf{m}}}. \qquad (3)$$

There are several definitions of the parameter $V_{\mathbf{m}}$ for WGMs of a sphere. Braginsky, Gorodetsky, and Ilchenko defined [32]

$$V_{\mathbf{m}} = 3.4\pi^{3/2}\left(\frac{\lambda}{2\pi n}\right)^3 l^{11/6}(l-m+1)^{1/2}, \qquad (4)$$

where l is the angular momentum, or the mode number, m is the azimuthal mode number. This equation was applied at $m=l$ in Ref. [20] and $m=1$ in Ref. [33]. Lin, Eversole, and Campillo [34] also used, with reference to Ref. [35], $V_{\mathbf{m}} \simeq V/(g^{1/2}n^2)$, here and below $V=4\pi a^3/3$ and $g=2l+1$. In Refs. [24,36–38], values $V_{\mathbf{m}}$ were calculated numerically. A generalization of the calculations in a review paper [36] gave $V_{\mathbf{m}\theta}/V=0.24/l^{1.159}$ for TE modes and $V_{\mathbf{m}\phi}/V=0.63/l^{1.142}$ for TM modes. The effective volumes were defined as [36] $V_{\mathbf{m}\kappa}=\int\int\int E^2_{\kappa s}(\mathbf{r})d\mathbf{r}/\max E^2_{\kappa s}$, where κ denotes a field projection, s is a set of the mode indexes. The above cited equations of Ref. [36] imply that $m=\pm l$ and the WGMs are of the first order. In Ref. [37], a dependence similar to Eq. (4) with $m=l$ was adopted,

$$V_{\mathbf{m}} = \begin{cases} 1.02(2a)^{11/6}(\lambda/n)^{7/6} & \text{for TE mode}, \\ 1.08(2a)^{11/6}(\lambda/n)^{7/6} & \text{for TM mode}. \end{cases} \qquad (5)$$

Note that in the strictest sense, the integration on the right-hand side of Eq. (2) or similar definitions should be over the volume of the universe if a cavity is open. A finite value of $V_{\mathbf{m}}$ cannot be obtained without restrictions imposed on the integrals [39].

Thus, there are distinctions in the characterization of the mode volume $V_{\mathbf{m}}$ and, hence, the factor f. Below in this paper, it is shown that the quantity $V_{\mathbf{m}}$ is an elucidative parameter and can be omitted in a rigorous calculation of f. The normalized LDOS upon the surface of a dielectric microsphere, in fact f, is found without calculating and even introducing the modal volume. The factor f is established to be in inverse proportion to the surface area $S=4\pi a^2$ of a spherical cavity,

$$f \simeq Q\frac{2\lambda^2}{\pi S}. \qquad (6)$$

Neither the above-mentioned numeric calculations nor estimates of the quantities $V_{\mathbf{m}}$ and Q have determined the upper limit of the Purcell factor. In addition, the known approaches could hardly propose ways to increase f allowing for a destructive physical process. In the next sections, the ultimate value of f is evaluated and consequences of light absorption are investigated.

III. NORMALIZED LOCAL DENSITY OF STATES

The local density of electromagnetic states $\rho(\mathbf{r},\omega)$ is generally expressed through the dyadic Green tensors of the electric and magnetic fields, $\overleftrightarrow{\mathbf{G}}^{\mathbf{E}}(\mathbf{r},\mathbf{r},\omega)$ and $\overleftrightarrow{\mathbf{G}}^{\mathbf{H}}(\mathbf{r},\mathbf{r},\omega)$, respectively [2],

$$\rho(\mathbf{r},\omega) = \frac{\omega}{\pi c^2}\,\mathrm{Im}\,\mathrm{Tr}[\overleftrightarrow{\mathbf{G}}^{\mathbf{E}}(\mathbf{r},\mathbf{r},\omega) + \overleftrightarrow{\mathbf{G}}^{\mathbf{H}}(\mathbf{r},\mathbf{r},\omega)], \qquad (7)$$

where ω is the angular light frequency, c is the speed of light in vacuum. The density ρ is often normalized to the free-space value $\rho_{\mathbf{v}}=\rho^{\mathbf{E}}_{\mathbf{v}}+\rho^{\mathbf{H}}_{\mathbf{v}}$, where $\rho^{\mathbf{E}}_{\mathbf{v}}=\rho^{\mathbf{H}}_{\mathbf{v}}=\omega^2/(2\pi^2c^3)$.

If one studies electric-dipole transitions, then only the quantity

$$\rho^{\mathbf{E}}(\mathbf{r},\omega) = \frac{\omega}{\pi c^2}\,\mathrm{Im}\,\mathrm{Tr}[\overleftrightarrow{\mathbf{G}}^{\mathbf{E}}(\mathbf{r},\mathbf{r},\omega)] \qquad (8)$$

is of importance. For instance, a calculation of the Einstein A coefficient for spontaneous emission, in accordance with the Fermi's golden rule, yields

$$\frac{\langle A\rangle}{A_{\mathbf{v}}} = \frac{\rho^{\mathbf{E}}}{\rho^{\mathbf{E}}_{\mathbf{v}}}. \qquad (9)$$

Here, $A_{\mathbf{v}}$ is the rate of spontaneous emission in vacuum, $\langle\cdots\rangle$ means averaging of the rate over the dipole direction. It should be mentioned that Eq. (9) is valid if dipole's environment has no influence on the moment of the electric-dipole transition.

If a cavity is examined, factor $f(\mathbf{r}) \equiv \rho^{\mathbf{E}}(\mathbf{r},\omega_s)/\rho^{\mathbf{E}}_{\mathbf{v}}$ can be called the space-dependent Purcell factor, here ω_s is a frequency of a cavity mode s. Note that for a dielectric sphere, quantities averaged over location $\mathbf{r}$ inside the sphere are [18] $\langle\rho^{\mathbf{E}}\rangle_{\mathbf{r}} \simeq \langle\rho^{\mathbf{H}}\rangle_{\mathbf{r}}$ and, consequently, $\langle f\rangle_{\mathbf{r}} \simeq \langle\mathcal{F}\rangle_{\mathbf{r}}$, where $\mathcal{F}(\mathbf{r}) \equiv \rho(\mathbf{r},\omega_s)/\rho_{\mathbf{v}}$ is a generalized Purcell factor.

IV. FIELD ENHANCEMENT ON THE SURFACE OF A SPHERE

Consider a sphere (medium 2) with a complex electric permittivity ϵ and a magnetic permeability $\mu=1$ placed in the free space (medium 1) where $\epsilon=\mu=1$. To study the essence of the problem, let us consider only the LDOS on the external surface of the sphere. In this way, on the one hand, it will be possible to evaluate the field enhancement in a WGM cavity by the order of magnitude. On the other hand, such a specific calculation is of particular importance in processes involving molecules [20–22], atoms [24] or quantum dots [13,19] located on the sphere surface.

Using the formulas for the dyadic electric-field Green tensor of a sphere presented elsewhere [26,40], the normalized LDOS at $r \geqslant a$ are found to be

$$\frac{\rho}{\rho_{\mathbf{v}}} = 1 + \frac{1}{4y^2}\sum_{l=1}^{\infty}(2l+1)\mathrm{Re}\left\{(a_{0l}+a_{1l}) \times\left[\left(1+\frac{l(l+1)}{y^2}\right)\zeta_l^2(y) + [\zeta_l'(y)]^2\right]\right\} \qquad (10)$$

and

$$\frac{\rho^{\mathbf{E}}}{\rho^{\mathbf{E}}_{\mathbf{v}}} = 1 + \frac{1}{2y^2}\sum_{l=1}^{\infty}(2l+1)\mathrm{Re}\left[a_{0l}\zeta_l^2(y) + a_{1l}\left([\zeta_l'(y)]^2 + \frac{l(l+1)}{y^2}\zeta_l^2(y)\right)\right]. \qquad (11)$$

Here,

$$a_{pl} = -\frac{k_{2-p}\psi_l'(z)\psi_l(x) - k_{1+p}\psi_l(z)\psi_l'(x)}{k_{2-p}\psi_l'(z)\zeta_l(x) - k_{1+p}\psi_l(z)\zeta_l'(x)} \qquad (12)$$

are the Mie coefficients for the transverse magnetic (TM, $p=1$) and transverse electric (TE, $p=0$) modes. The function ψ_l is a Riccati-Bessel function of the order l, $\zeta_l(x) \equiv \sqrt{\pi x/2}H^{(1)}_{l+1/2}(x)$ is a Riccati-Hankel function, related with the Hankel function $H^{(1)}_{l+1/2}$ of the first kind. Arguments in Eqs. (10)–(12) are $x \equiv k_1 a$, $y = k_1 r$, and $z \equiv k_2 a$, where k_1 and k_2 are the wave numbers in the surrounding medium and the sphere, respectively.

It is worth noting that Eq. (11) can be obtained from above Eq. (9) and Eqs. (6′), (7′) of Ref. [16] or Eqs. (A1), (A2) of Ref. [28] giving $\langle A\rangle = \frac{1}{3}A^{\perp} + \frac{2}{3}A^{\parallel}$, where $A^{\perp}$ and $A^{\parallel}$ are the rates of transitions with radially and tangentially oriented dipole moments, respectively.

V. MIE SCATTERING RANGE

In the region of the Mie scattering, sharp spikes commonly called MDRs appear in spectra of spontaneous emission, elastic and inelastic (fluorescent and Raman) scattering [7]. The MDRs originate from resonant excitation of WGMs. Under the resonance, just one term in each series of Eqs. (10) and (11) exceeds the sum of the rest by orders of magnitude. The dominant coefficient a_{pl} can be calculated analytically, if one solves Bessel equations using the WKB approximation [41]. The correspondent asymptotic expressions for the Bessel functions applicable at large l read as [42]

$$\psi_l(z) \simeq \sqrt{\pi\beta}\,\mathrm{Ai}(\xi), \quad \psi_l'(z) \simeq -\sqrt{\pi/\beta}\,\mathrm{Ai}'(\xi), \qquad (13)$$

where $z = l + \frac{1}{2} - \xi\beta$, Ai is the Airy function, $\beta \equiv [\frac{1}{2}(l+\frac{1}{2})]^{1/3}$, $|\xi|\beta \ll l$. For the Riccati-Hunkel function the Debye expansion [42] can be applied,

$$\zeta_l(y) \simeq -ie^{T}\left(1 + \frac{i}{2}e^{-2T}\right)(\sinh\eta)^{-1/2}, \qquad (14)$$

$$\zeta_l'(y) \simeq ie^{T}\left(1 - \frac{i}{2}e^{-2T}\right)(\sinh\eta)^{1/2}, \qquad (15)$$

where $y = (l+\frac{1}{2})/\cosh\eta$, $T = (l+\frac{1}{2})(\eta - \tanh\eta)$, $T^{2/3} \geqslant 1$.

If x is about a resonant size parameter x_s, Mie coefficient (12) expressed through functions (13)–(15) can be written in the following form:

$$a_s = -\frac{1}{Q_{s0}}\left(\frac{1}{Q_s} - i2\frac{x - x_s}{x}\right)^{-1}, \qquad (16)$$

where index s designates the set of quantum numbers p, l, and the order q of a WGM,

$$x_s = \frac{1}{n_r}\left(l + \frac{1}{2} - t_q\beta - n_{\mathbf{r}}^{1-2p}(n_{\mathbf{r}}^2 - 1)^{-1/2}\right), \qquad (17)$$

$n_{\mathbf{r}}$ is the real part of the light refractive index, t_q is the qth root of the equation $\mathrm{Ai}(t_q)=0$. The quality factor Q_s of the mode s is expressed through the partial quality factors Q_{s0} and $Q_{\mathbf{a}}$,

$$1/Q_s = 1/Q_{s0} + 1/Q_{\mathbf{a}}. \qquad (18)$$

Factor Q_{s0} is the radiative quality factor [41] that describes radiative energy losses by a sphere of an ideal spherical shape made of an ideal dielectric,

$$Q_{s0} = \frac{1}{2}x_s e^{2T} n_{\mathbf{r}}^{2p}(n_{\mathbf{r}}^2 - 1)^{1/2}, \qquad (19)$$

see a discussion of the accuracy of Eq. (19) in Ref. [24]. The factor $Q_{\mathbf{a}}$ takes into account the process of light absorption, $Q_{\mathbf{a}}=n_{\mathbf{r}}/(2n_{\mathbf{i}})$, where $n_{\mathbf{i}}$ is the imaginary part of n.

Equation (16) is one of the prime original results of this study. It permits to explain many features of the MDRs. For instance, Eq. (16) gives $a_s(x_s)=-Q_s/Q_{s0}$ at $x=x_s$. On substituting the obtained real number into Eqs. (10) and (11) with allowance of Eqs. (14) and (15), the Purcell factors $\mathcal{F}$ and f can be found. Namely, they become

$$\mathcal{F}(a+0) = \frac{2}{x_s^2}Q_s\frac{n_{\mathbf{r}}^{3-2p}}{n_{\mathbf{r}}^2-1}. \qquad (20)$$

$$f(a+0) = \frac{2}{x_s^2}Q_s\left(\frac{n_{\mathbf{r}}}{n_{\mathbf{r}}^2-1} + \frac{p}{n_{\mathbf{r}}}\right), \qquad (21)$$

Disregarding the dependence of the above factors on $n_{\mathbf{r}}$ and p, one gets Eq. (6) for both quantities $\mathcal{F}$ and f.

Formulas (20) and (21) determine LDOS in the near-surface region, at $r=a+0$. If the distance from the surface of the sphere increases, the both quantities $\rho(r)$ and $\rho^{\mathbf{E}}(r)$ decrease approximately by an exponential law,

$$\ln\frac{\rho(r)}{\rho(a+0)} \simeq \ln\frac{\rho^{\mathbf{E}}(r)}{\rho^{\mathbf{E}}(a+0)} \simeq \frac{4\pi}{\lambda}\sqrt{n_{\mathbf{r}}^2-1}(a-r), \qquad (22)$$

where $a \leqslant r \leqslant n_{\mathbf{r}}a$.

VI. NUMERIC EXAMPLES AND THEIR DISCUSSION

To illustrate the obtained relations, factor (21) was calculated for spheres with a refractive index $n=1.5(1+\imath 10^{-10})$. The highest possible value of the Q factor at such n is equal to 5×10^9 close to a value $Q=8\times10^9$ measured for fused-silica microspheres in air [31]. The result of the calculation is presented in Fig. 1(a). The estimated factor $f(a+0)$ can be as large as 10^6–10^7. The effect that hampers the LDOS enhancement is light absorption that is extremely important at $Q_{\mathbf{a}} \leqslant Q_{s0}$.

It should be noted that Eq. (9) describes the acceleration of the spontaneous transition in a local area. Despite the great enhancement of LDOS, some MDRs shall be eliminated from spectra of spontaneous emission, fluorescence, and Raman scattering. If a dipole located outside of a sphere radiates power $\hbar\omega\langle A\rangle$ then only power $\hbar\omega\langle R\rangle$ is detectable in the far-field zone. The rest power shall be absorbed by the sphere. In this paper, the ratio $\phi=\langle R\rangle/\langle A\rangle$ is called the efficiency of radiation of spontaneous emission.

In the framework of the classical electromagnetic theory from Eqs. (6) and (7) of Ref. [16], the normalized rate $\langle R\rangle = \frac{1}{3}R^{\perp}+\frac{2}{3}R^{\parallel}$ is found to be

$$\frac{\langle R\rangle}{A_{\mathbf{v}}} = \frac{1}{2y^2}\sum_{l=0}^{\infty}(2l+1)\Bigg(|\psi_l(y)+a_{0l}\zeta_l(y)|^2 + \frac{l(l+1)}{y^2}|\psi_l(y) + a_{1l}\zeta_l(y)|^2 + |\psi_l'(y)+a_{1l}\zeta_l'(y)|^2\Bigg). \qquad (23)$$

For the resonant emission, terms with a single coefficient a_s are dominant in Eqs. (23) and (11), and the relations $\mathrm{Re}\,\zeta_l^2(y) \simeq -|\zeta_l(y)|^2$ and $\mathrm{Re}[\zeta_l'(y)]^2 \simeq -|\zeta_l'(y)|^2$ take place at y near x_s. The efficiency of radiation of mode s is hence approximately equal to

$$\phi_s = -\frac{|a_s|^2}{\mathrm{Re}\,a_s} = \frac{Q_s}{Q_{s0}}. \qquad (24)$$

Here, the value of ϕ_s is independent of the distance $r \geqslant a$ in the range where approximations (14) and (15) are valid. Thus, at $Q_{\mathbf{a}} \leqslant Q_{s0}$ a weakly absorbing dielectric body captures almost all resonant emission from a neighboring source. The efficiency identical to Eq. (24) has been found for the resonant emission from electric dipoles located inside a sphere, $r<a$ [43]. The ratio Q_s/Q_{s0} was also called the mode efficiency [44].

To show up the importance of light absorption let us calculate ϕ for WGMs of a fused-silica sphere. For the TM_{88}^1 and TE_{88}^1 modes values $Q_{s0}=0.92\times10^{14}$ and $Q_{s0}=1.06\times10^{14}$ predicted by Eq. (19) are close to the ratios $x/\Delta x$ of the values x and Δx computed in Refs. [29,30]. If $n=1.5(1+\imath 10^{-10})$, the efficiency of radiation of these modes is

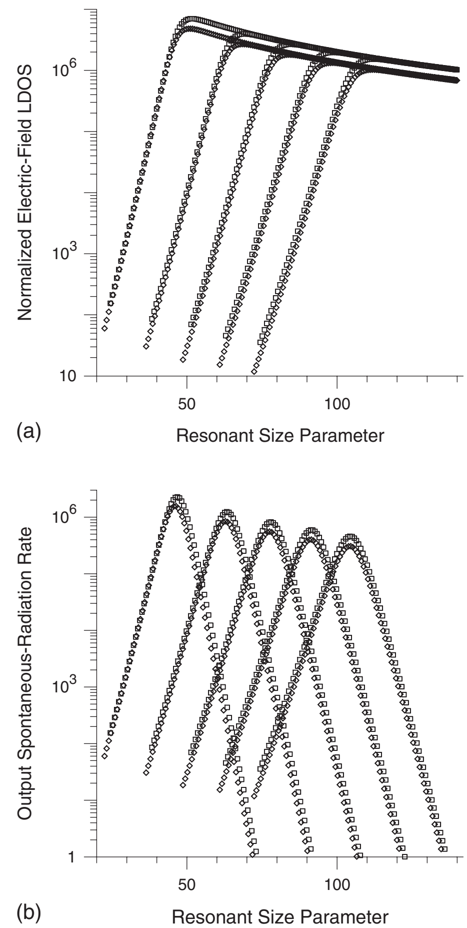

FIG. 1. (a) Peak values of the normalized electric-field LDOS predicted by Eq. (21) and (b) the normalized rate for the output resonant spontaneous emission $\langle R\rangle/R_{\mathbf{v}}$ on the surface of a sphere with a refractive index of 1.5 $(1+\imath 10^{-10})$ versus the resonant size parameter x_s. The TE (◇) and TM (□) whispering-gallery modes of five orders are taken into account.

$\phi \simeq 5\times 10^{-5}$. Such a feature should be allowed for in simulations.

The calculated values of the resonant enhancement of the rate $\langle R \rangle$ is presented in Fig. 1(b). According to Fig. 1, at equal factors f only the mode with higher orders can be seen in spectra of spontaneous emission from dipoles located on sphere surfaces at $x \geqslant 70$. Nevertheless, the lowest-order mode should be dominant in lasing spectra [21].

Let us briefly compare the predicted Purcell enhancement with those found in other media. Current nanotechnologies widely apply optical properties of noble-metal nanoparticles. It is the LDOS enhancement that results in anomalous optical processes such as surface-enhanced Raman scattering and metal-enhanced fluorescence (see, for example, Refs. [12,45,46]). It has been shown that decompositions in forms of Eqs. (10) and (11) turn out to be unphysical for x in the Rayleigh scattering range and $n_{\mathbf{i}} \neq 0$ [47,48]. For example, Eq. (11) predicts $\rho \rightarrow \infty$ on the surface of a noble-metal sphere at $x=0.2$ [49] due to the unlimited contribution from spherical harmonics of unbounded orders. However, a particle cannot be polarized by an electric field if a dimension of the surface angular field oscillation is lower than a certain microscopic value [47,50,51]. A consideration of space dispersion of the permittivity (by means of a dielectric function depending on the light wave vector) has defined a cutoff number l_{c} of the multipole polarizabilities [50]. In practice, only the TM_1 mode sometimes called the Fröhlich mode (giving rise to the dipolar plasmon polariton) was expected to be the most important in single metal spheres at $a \leqslant 20$ nm [52]. For a single noble-metal nanosphere, the ultimate value of the Purcell factor was thought to be 10^3 [45]. (The factor of the electromagnetic SERS enhancement proportional to f^2 was hence in the range of six to seven orders of magnitude [53].) Thus, the Purcell factor characteristic for single noble-metal nanospheres is much less than values $f=10^6-10^7$ found for dielectric microspheres. Moreover, the LDOS increases on the whole microparticle surface which is, of course, much larger than that of a nanoparticle. However, the enhancement occurs in narrow spectral intervals.

Recently, it has been demonstrated that toroidal microcavities with WGMs are promising candidates for the immediate use in strong-coupling CQED studies [8]. Letter [54] reported $Q=4\times 10^8$ measured and $V_{\mathbf{m}} \simeq 180\ \mu\mathrm{m}^3$ calculated for a toroid with a principle diameter of 29 μm and a minor diameter of 6 μm at $\lambda=1.55\ \mu$m. According to Ref. [54], an inferred value of f in excess of 2×10^5 was over values obtained at other cavity geometries.

This paper predicts even higher values of f. For example, $f(a+0)=7.0\times 10^6$ at $p=1$, $l=70$, $q=1$ and $n=1.5\ (1+\imath 10^{-10})$, according to Eq. (21). One could find the Purcell factor numerically directly from Eq. (11). For good convergence of the Mie series it required to take $l_{\max}=1.1\ |z|+50$ terms [55], $l_{\max}=135$ in the considered case. The resonant frequency of the mode should be determined using a sufficient number of the significant decimal digits, $x=51.799\,860\,733\,7525$ for the above example. In a numeric calculation with the above parameters, it was established that one term in the series of Eq. (11) is higher than the sum of the rest in 6.7×10^6 times (this number remained constant if $l_{\max}$ was increased in 100 times). The normalized LDOS computed from Eq. (11) was equal to $\rho^{\mathbf{E}}/\rho^{\mathbf{E}}_{\mathbf{v}}=6.3\times 10^6$. In this case, $Q_s=4.76\times 10^9$ according to the numeric calculation and $Q_s \simeq 4.67\times 10^9$ according to analytic Eqs. (18) and (19). Though the factor f is maximal for the given absorption, the efficiency of radiation (24) is low, $\phi_s=0.067$ according to Eqs. (18) and (19).

The Purcell factor can be thought to consist of a spectral energy density Q_s and a spatial energy density $1/V_{\mathbf{m}}$ [11]. It is the high value of Q_s that causes the large enhancement of the LDOS. In addition, the morphology factor $\lambda^3/(4\pi^2 V)$ of Eq. (1) turns out to be replaced by $2\lambda^2/(\pi S)$ for the surface-mode cavity. It is worth noting that this study has not yet reported a record value of $f(r)$. The normalized LDOS attains even higher maximum inside the sphere.

From Eqs. (3) and (5) it could be expected that f is roughly proportional to the product of the factor $Q\lambda^2/S$ and the mode degeneracy $g=2l+1$. Despite the common Eq. (3), the factor g is not present in the obtained Eq. (21). Thus, the algorithm of the calculation of $f(r)$ proposed in this paper has an advantage over intuitive estimates.

VII. CONCLUSION

This paper has, at first, introduced the space-dependent Purcell factors through the resonant local densities of electromagnetic- and electric-field states. Then, for studying a spherical cavity, the asymptotic formula of the Mie coefficient applicable at resonant excitation of a WGM by an incident plane wave has been derived allowing for light absorption. Owing to this formula, the peak values of LDOS upon the surface of a micrometer-sized dielectric sphere have been analytically calculated. The Purcell factor $f(a+0)$ has been found to be proportional to the product of two factors, the cavity Q factor and the squared ratio of the wavelength to the sphere diameter. The upper limit of $f(a+0)$ of the order of 10^7 has been evaluated for a realistic fused-silica microsphere in air. The LDOS enhancement is constrained by the process of light absorption within the cavity. This process also reduces the efficiency of radiation of the resonant spontaneous emission even for dipoles located in the exterior of the sphere.

The predicted factor $f(a+0)$ is more than one order of magnitude over the recently demonstrated Purcell factor of a toroidal microcavity.

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