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(Laser, Atomic and Molecular Physics)

ANOMALOUS LUMINESCENCE OF DISPERSE MEDIA DURING STIMULATED EMISSION INTO WHISPERING GALLERY MODES

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MIRAMARE-TRIESTE

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ANOMALOUS LUMINESCENCE OF DISPERSE MEDIA DURING STIMULATED EMISSION INTO WHISPERING GALLERY MODES

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I. Introduction

Recent years have seen increased interest in the study of gas-liquid disperse media (GLDM) [1,2]. The GLDM permit a wide variation of physical and chemical properties of a medium as well as the use of peculiarities of excited particles both in the condensed and gaseous phases.

There are new effects in chemiluminescence which arise from peculiarities of physical and chemical kinetics in GLDM. Thus, an intensive luminescence arises when oxygen or hydrogen atoms recombine with clusters or microdroplets of a GLDM [3]. In these reactions the microdroplet acts as a third body in recombination and increases the branching ratio into electronically-excited states significantly.

The peculiarities of the kinetics of metastable molecule (MM) radiation and quenching in liquid- and gas-phase bring about new physical phenomena which consist in an intensity enhancement of the MM luminescence in GLDM. In these phenomena a droplet of a GLDM makes an optically forbidden MM transition allowed. The effect should occur both in a stationary case during excitation of an already existing GLDM and in the nonstationary one when droplets quickly appear in a previously excited gas, for example, during its condensation in a supersonic nozzle [6,7].

The possibility of a wide variation of the GLDM properties is an important feature that permits one to create new type of lasers. Comparatively recently investigations of the active GLDM have been started in connection with problems of chemical [4,8-10], gas-dynamic [11,12], and electric-discharge [13] lasers. For example, in Ref. 14 properties of the dye solution aerosols have been studied under optical pumping. In Ref. 15 the laser generation has been achieved with an optical pumping of suspensions.

At the dielectric particle surface the slowly decaying modes of electromagnetic field, or whispering gallery modes (WGM), can be excited [16]. A stimulated emission into the WGM of a spherical millimeter-size sample of CaF_2 with Sm^{++} ions has been achieved, for the first time, in 1961. In the last few years the experiments have been fulfilled in which lasing was observed in optically-pumped dye solution droplets [2,17,18]. For example, some characteristics of a droplet WGM laser have been studied in Ref. 18. The active medium was formed by monodisperse dye-doped water droplets 20 + 30 μm in radius. Stable high-quality lasing spectra have been obtained which consisted from two sets of periodically spaced peaks, corresponding to TM and TE modes generation. The value of microresonator finesse was estimated to be $Q \simeq 10^4$. In the far field, the lasing droplet was observed as a coherent point source emitting uniformly in all directions. In recent papers [19,20] stimulated Raman scattering into the WGM of droplets has been used for determinations of droplet size, index of refraction, and chemical composition.

To this incomplete list of the phenomena in the excited GLDM we add a new effect of the anomalous luminescence. In this paper a theory is derived for the GLDM luminescence during the light generation in the spherical particles which serve as microresonators. It is predicted, for the first time, that there can be a drastic increase of the GLDM luminescence intensity in the case of inverse population of the MM energy levels. The effect consists in the following. As a result of the diffusion, the MM's penetrate from the gaseous phase into the surface layer of a droplet. A region with an inverted population of the MM energy levels forms in this droplet. Under certain conditions the lasing into the WGM's can arise in the droplet that serves as the high-finesse microresonator, so that the photon density becomes considerable. It leads to substantial increase of the quantum yield of the MM emission due to stimulated optical transitions in the droplet. As optical transitions of the MM's are forbidden in gas, the luminescence intensity of the entire GLDM increases by orders of magnitude.

The general outline of this paper is as follows. In Sec. II we present information about the WGM of the spherical resonator. The formula for spherical resonator finesse used in Sec. II is derived in the Appendix. In Sec. III we propose a method for creation of active GLDM that implies the MM diffusion from an electronically-excited gas into droplets. For this method, the condition for the WGM excitation in a droplet is obtained in Sec. IV. In Sec. V we consider the radiative characteristics of the droplet during steady-state stimulated emission into the WGM. Sec. VI is devoted to the investigation of the luminescence of the GLDM with a large number of droplets during the stimulated

emission in the WGM's. Finally, concluding remarks are given in Sec. VII.

II. Decay and Gain of WGM

A dielectric sphere is known to be an open optical cavity, where WGM's can exist [16]. Every such mode is characterized by a set p of indices $(n, k, q, m) \equiv p$; where the mode number n is a large integer, $k = 1$ and $k = 0$ for TM and TE modes, respectively, q is the longitudinal index that is equal to the root number of the equation: $Ai(x) = 0$, where Ai is the Airy function, and m is the azimuthal index ($-n \le m \le n$). The angular frequency ω_p of the p-resonator mode depends on n, k, q and is defined by the formula [16]

$$\omega_p = \frac{c}{M a}\left(n + \frac{1}{2}\right)\left(1 - \Delta_p\right),$$

where $\left(n + \frac{1}{2}\right)\Delta_p = x_q\left[\frac{1}{2}\left(n + \frac{1}{2}\right)\right]^{1/3} + \left(\frac{\varepsilon}{M}\right)^{1-2k}\left(M^2 - 1\right)^{-1/2}$.

c is the velocity of light; M and ε are the relative optical refractive index and the dielectric constant of the resonator material, respectively.

In Ref. 16 a formula is found for the radiative finesse of resonator Q_r that increases exponentially with increasing a:

$$Q_r = \frac{1}{2}\left(n + \frac{1}{2}\right)\left(\frac{\varepsilon}{M}\right)^{2k-1}\left(M^2 - 1\right)^{1/2} \exp\left(2 T_p\right), \quad (1)$$

$$T_p \equiv \left(n + \frac{1}{2}\right)\left(\eta_p - \tanh \eta_p\right), \quad \cosh \eta_p \equiv M\left(1 - \Delta_p\right)^{-1}.$$

For a visible and infrared radiation, formula (1) gives anomalously large values of Q, for example, $Q_r = 10^{48}$ at $a = 100$ μm, $\lambda = 1$ μm, $\varepsilon = 1.5$.

Therefore, we have taken into account not only the purely radiative losses, but also light scattering on deviations of the resonator surface from an ideal sphere. To obtain a general expression for the light losses on these deviations we took the equation of the resonator surface in the following form:

$$r = a + b\,\gamma(\vartheta, \varphi), \qquad b << \lambda, \tag{2}$$

where r, ϑ, φ are the spherical coordinates of points on the resonator surface, $\iint \gamma(\vartheta, \varphi)\, d\Omega = 0$, $\iint \gamma^2(\vartheta, \varphi)\, d\Omega = 1$, $d\Omega \equiv \sin\vartheta\, d\vartheta\, d\varphi$, b is the amplitude of surface deviation from a spherical shape. This equation of the resonator surface is valid for a droplet of incompressible liquid at small deviations of the surface from a spherical one. In the Appendix, we have derived the expression for the resonator finesse Q

$$Q^{-1} = Q_r^{-1} + Q_s^{-1}, \tag{3}$$

where $Q_s \geq \frac{1}{2\pi}\left(n + \frac{1}{2}\right)\left(\frac{\varepsilon}{M}\right)^{2k-1}\left(M^2 - 1\right)^{-1}\left(\frac{\lambda}{b}\right)^2$, $\lambda = 2\pi c/\omega_p$ is the light wavelength. Note that the Q value increases linearly with increasing a (at constant b) starting from a sertain n. For the fixed a the Q value, on the contrary, decreases with the growth of b.

It is known that the vibrational motion of liquid can exist in a droplet. It leads to vibrations of surface:

$$r = a + b_{1m}\, Y_l^m(\vartheta, \varphi)\cos\Omega_l t,$$

where b_{1m}, Ω_l are the amplitude and the eigenfrequency of the capillary vibration, Y_l^m is the spherical function, $l = 2, 3, 4, \ldots$, t is the time. These capillary vibrations are damping. The decay time of the capillary vibrations is equal to:

$$t_l = \frac{a^2\rho}{\eta}\,2\left(l - 1\right)\left(l + \frac{1}{2}\right),$$

where η, ρ are the dynamic viscosity and mass density of the liquid, respectively. For example, for an O_2 droplet at T = 75 K we have $\eta = 3\cdot10^{-4}$ Pa·s, $\rho = 1{,}22$ g/cm^3 and $t_2 = 0.7$ ms. The existence of such damping means that the vibrations arising during a process of the creation of the droplet should vanish by the time $t > t_1$. However, the thermal vibrations exists constantly and they must be taken into consideration. The mean square value of partial vibration amplitudes for a droplet in thermal equilibrium can be expressed by its temperature T [21]

$$\overline{b_l^2} = \frac{k_B T}{\gamma(l-1)(l+2)},$$

where γ is the surface tension and k_B is the Boltzmann constant. After summing of $\overline{b_l^2}$ over l we find the root-mean-square amplitude of thermal capillary vibrations,

$$b \equiv \left(\sum_l \overline{b_l^2}\right)^{\frac{1}{2}} = \left(\frac{11 k_B T}{18\gamma}\right)^{\frac{1}{2}}. \tag{4}$$

It is obvious that Eq. (4) can be used as an estimate for the lowest value of b for the liquid resonator. For example, for the oxygen droplet at T = 75 K the value of b calculated by Eq. (4) is equal to $1.4\cdot10^{-4}$ μm. At $\lambda = 1.6$ μm and $a = 30$ μm the quantities Q_r and Q_s are comparable resulting in $Q \approx 2\cdot10^8$. These numbers illustrate the importance of taking thermal ripples at

the droplet surface into account that was recently noted in Ref. 21. If the droplet contents the molecules with the population inversion of some levels, the WGM can excite due to the high finesse of the microresonator.

The important peculiarity to be stressed is that the sizes of the microresonators under study are small while their Q are high enough (see Eq. (2)). For example, in an open resonator which is an usual pair of mirrors the quality factor $Q \simeq 10^8$ is achieved at a reflectivity of mirrors $R = 0.9$ and a cavity length $L = 10^2$ cm. Therefore, to obtain the lasing in a droplet one has to satisfy the criteria, more weaker than the ordinary ones.

The stimulated emission into the WGM is possible if the increment of the amplitude of the WGM exceeds the one of the decrement, that is

$$< \alpha_p > \; \frac{1}{Q}\,\omega_p, \qquad \alpha_p \equiv \sigma\,\Delta N_{eff}, \qquad \Delta N_{eff} \equiv \frac{\int dV\,\Delta N(r)\,E_p^2}{\int dV\,\varepsilon\,E_p^2}, \qquad (5)$$

where σ is the cross-section for stimulated emission in the droplet, $\Delta N(r)$ is the density of the population inversion of the levels, r is coordinate and E_p is the electric field of the resonator mode p.

For calculation of the excitation criterion of the WGM's it is necessary to determine the distribution of the MM's into the droplets taking to account the transport and the quenching of these MM's in both the gas and liquid phases.

III. Gas-Liquid MM transport in the disperse medium

Let molecules be excited to metastable electron-excited states, so that the population inversion arises between certain

energy levels. A source of MM (for example, $O_2(^1\Delta)$, $NH(a\,^1\Delta)$, $O(^1S)$), may be an electric discharge, an electron beam, chemical reactions, and an optical pumping.

We assume that an external source creates the MM's in gaseous phase of the GLDM. The distribution densities of the MM's in the gaseous (g) phase around one droplet $n_g(r, t)$, and the liquid (l) phase $n_l(r, t)$ are found by solving diffusion equations

$$\frac{\partial\, n_{g(l)}(r,t)}{\partial\, t} = -\vec{\nabla}\cdot\vec{J}_{g(l)} - \frac{n_{g(l)}}{\tau_{qg(l)}} + W_g, \qquad \vec{J}_{g(l)} \equiv -D_{g(l)}\vec{\nabla}\, n_{g(l)},$$

where $D_{g(l)}$ and $\tau_{qg(l)}$ are the diffusion coefficients and the lifetimes of the MM in the gas and the liquid, respectively, (usually $\tau_{qg} >> \tau_{ql}$); W_g is the rate of the MM pumping. The solutions of these equations must satisfy the following boundary conditions. At $r = 0$ the flow J_g must be equal to zero. At $r = a$ the products of the flows $\vec{J}_g$ and $\vec{J}_l$ and the external normal to the liquid surface must be equal and must be equal to the difference between the numbers of the MM's leaving and entering the droplet through unit surface per unit time.

The densities of stationary and nonstationary MM distributions may be found in the g- and l- phases of the GLDM. We will consider the low laser level being empty further on. For the population inversion density inside droplets with radii $a >> L_l \equiv (D_l\,\tau_{ql})^{1/2}$ we have [6,7]

$$\Delta N(r) = y\,N_g\,\frac{D_g}{D_l}\,\frac{L_l}{a}\,\exp\left\{\frac{r-a}{L_l}\right\}, \qquad (6)$$

where y and N_g are the fraction of excited molecules and the total density of molecules in the gaseous phase, respectively. The quantity of y depends on the pumping rate W_g, the gaseous

mixture composition, etc. Thus, if an existing GLDM is excited, we would have $y = y_g^{(0)} \tau_{qd} / \tau_{qg}$, where $y_g^{(0)} = W_g \tau_{qg}$ is the degree of electronic excitation of the gas in the absence of the liquid phase, at the same intensity of the molecules excitation W_g; $\tau_{qd}^{-1} = \tau_{qg}^{-1} + 4\pi a D_g N_d$, where N_d is the concentration of the droplets. If the disperse medium is formed during a rapid injection of the droplets in a previously excited gas, we would have $y = y_o \exp(-t/\tau_{qd})$, where y_o is the initial degree of excitation of the gas.

IV. Condition of the WGM excitation

Inserting the $\Delta N(r)$ value of (4) into (3), we can obtain the condition for the WGM excitation in a droplet. It can be written as a limitation for y:

$$y > y_l \qquad (7)$$

$$y_l \equiv \frac{\pi \varepsilon a^2}{\lambda Q \delta_l \tau_{ql} D_g N_g} \left[\left(2\pi \frac{L_l}{\lambda} \right)^2 M (1 + \delta_k^2) + k \delta_k \right],$$

$$\delta_k \equiv 1 + \left(2\pi \sqrt{M^2 - 1} \; M \left(\frac{\varepsilon}{M} \right)^{2k-1} \right)^{-1} \frac{\lambda}{L_l}.$$

When the condition (7) is met the excitation of high-finesse WGM's takes place. The number of photons in these modes, f, begins to grow and the MM lifetime in the surface layer of the droplet decreases, accordingly: $\tilde{\tau}_{ql}^{-1} = \tau_{ql}^{-1} + \frac{c\delta_l}{V_f} \approx 2$. The mean MM density in the droplet therefore decreases from the value of $\overline{n_l^o}$ [3] to $\overline{n_l} = \tilde{\tau}_{ql} \tau_{ql}^{-1} \overline{n_l^o}$. The number of photons f_p in the mode p reaches a stationary value when the rate of stimulated phototransitions $\sum_p c\alpha_p f_p$ becomes equal to the rate of photon

losses $\sum_p \omega f_p / Q(p)$. From the obvious equation

$$\sum_p c\alpha_p f_p = \frac{V}{\tau_{ql}} \left(\overline{n_l^o} - \overline{n_l} \right)$$

we can find the stationary value of $f = \sum f_p$:

$$f_s = f_o (y - y^*), \qquad (8)$$

where $f_o \equiv 2a Q^e D_g N_g \upsilon^{-1}$, $y^* \equiv \frac{n^{4/3}}{3} \lambda \left(\varepsilon\mu Q^e \delta_l \tau_{ql} D_g N_g \right)^{-1}$,

$Q^e \equiv Q(n, s = 1, q = 1, m)$, υ is the frequency of radiation.

V. Stimulated emission into WGM's of a single droplet

Let us find the parameters of the stimulated emission at steady-state lasing. Using Eq. (8), we determine the power radiated by a single droplet,

$$P_s = h\upsilon \frac{\omega}{Q} f = 4\pi a N_g D_g h\upsilon (y - y^*) \qquad (9)$$

note that when $y >> y^*$ the P_s value does not depend on Q.

Let us define the quantum yield of the stimulated electromagnetic emission for a droplet η as the ratio of the number of photons radiated per second to the number of excited molecules entering the droplet per second. In the above conditions

$$\eta = 1 - \frac{y^*}{y} (1 - \varphi_l) \qquad (10)$$

where $\varphi_l \equiv \tau_{ql} / \tau_{rl}$ is the quantum yield of spontaneous emission in a liquid, τ_{rl} is the radiative lifetime of the MM's in a liquid. If the excitation of the WGM's does not occur, it is obvious that $\eta = \varphi_l$. Usually, this quantity is much smaller than

1. On the contrary, the stimulated emission quantum yield for a droplet can, according to (10), be of the order of unity.

VI. Luminescence of excited disperse media

Let us consider now the luminescence of the GLDM with a large number of droplets under the conditions of the stimulated emission into WGM's. The number of excited molecules diffusing into droplets per second at $a \gg L_l$ does not depend on the MM lifetime in a droplet [6,7]. Therefore, the GLDM luminescence quenching time, τ_{qd} (see Sec. III), does not depend on the lasing and usually τ_{qd} is nearly equal to τ_{qg}. But under the conditions of stimulated emission into WGM's and at $\varphi_l \ll 1$ and $y \gg y^*$ the quantum yield of emission in the droplets increases sharply. Therefore, the GLDM luminescence intensity I_d can be significantly higher than I_g, that is the luminescence intensity of a homogeneous gas at the same MM pumping rate W_g. The calculation of I_d / I_g, with the use of (9), (10), gives

$$\frac{I_d}{I_g} = 4\pi D_g N_d \tau_{rg} \eta + 1 \quad , \qquad (11)$$

where τ_{rg} is the radiative lifetime of MM in a gas.

One can calculate the quantum yield of the GLDM luminescence, φ_d, which is defined as the ratio of the number of radiated photons to the number of excited MM,

$$\varphi_d = \varphi_g \left(4\pi a D_g N_d \tau_{rg} \eta + 1 \right) \cdot \left(1 + 4\pi a D_g N_d \tau_{qg} \right)^{-1}, \qquad (12)$$

where $\varphi_g \equiv \tau_{qg} / \tau_{rg}$ is the quantum yield of spontaneous emission in a gas. Under the conditions of the stimulated emission into WGM's, φ_d can considerably exceed φ_g.

As an example, let us consider the $O_2(^1\Delta)$ MM luminescence in the GLDM. Assume that the GLDM contains droplets of oxygen with $a = 30$ μm, $T = 75$ K, and that the mass fraction of the liquid phase equals 0.1 ($N_d = 10^9$ cm^{-3}). For such droplets we obtain $Q \simeq 2 \cdot 10^8$. We will investigate the possibility of lasing on $O_2(^1\Delta, v' = 0) \rightarrow O(^3\Sigma, v'' = 1)$ transition with $\lambda = 1.58$ μm. There is an inverted population for this transition due to rapid VT- relaxation in liquid. According to calculations by Eq. (7), the WGM lasing in O_2 droplets arises when y, i.e. the $O_2(^1\Delta)$ volume fraction in the gas, exceeds $y_l = 0.02$ ($q = 1$, for TM modes we have $y_l = 0.016$, for TE-modes $y_l = 0.014$). At present gaseous oxygen with $y \leq 0.9$ is being produced [24]. Lasing can consequently be achieved experimentally on $O_2(^1\Delta)$ MM transitions. Note that in accordance with Eq. (8) we have $f = 0$ at $y = y^* = 0.015$. The nearness of y^* and threshold values y_l testifies to a quite good accuracy of approximations made in Sec. IV. The calculations of the $O_2(^1\Delta)$ MM emission in the oxygen droplets give, according to Eqs. (8) + (12), $f_s = 10^{10} \cdot y$, $P_s = 7 \cdot y$ mW, $I_d / I_g = 10^8$, $\varphi_d = 0{,}3$ ($\varphi_g \simeq 3 \cdot 10^{-7}$, $\varphi_l \simeq 10^{-5}$, see Ref. 25). Thus, there would be the stimulated emission into WGM's in 30 μm-radius oxygen droplets located in the gas containing the excited $O_2(^1\Delta)$ molecules, at $y > 0.02$. For this case the quantum yield of singlet oxygen radiation in the GLDM is of the order of unity.

VII. Conclusions

Finally, we predict that, under certain conditions, the presence of a condensed disperse phase in an excited gas bring about a new physical phenomenon - an anomalous increase in the MM

luminescence intensity as a result of the stimulated emission into whispering gallery modes of the droplets. This phenomenon opens a new possibility for an efficient conversion of the energy of long-lived, electronically excited molecules into the energy of the light.

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Appendix. Quality factor of a spherical resonator

Below we calculate the spherical resonator finesse Q in the absence of light absorption, but allowing for the light scattering on inhomogeneities of the resonator surface.

Let $\vec{E}(t, \vec{r}) = \mathrm{Re}(\vec{E}(\omega,\vec{r})\cdot e^{-i\omega t})$, $\vec{H}(t, \vec{r}) = \mathrm{Re}(\vec{H}(\omega,\vec{r})\cdot e^{-i\omega t})$ be the electric and magnetic vectors of an electromagnetic mode in a medium with the electric and the magnetic constant

$$\varepsilon(\vec{r}) = \begin{cases} \varepsilon, & r < a \\ 1, & r > a \end{cases}, \qquad \mu(\vec{r}) = 1,$$

where $\vec{r}$ is the position vector and t is the time. We will find the electric $\vec{E}'$ and magnetic $\vec{H}'$ fields in the medium with ε' and μ' which weakly differ from ε and μ. The $\vec{E}'(\omega, \vec{r})$ and $\vec{H}'(\omega, \vec{r})$ vectors will be determined from the approximate equations

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E}' - k^2 \vec{E}' = \frac{4\pi}{c} ik \vec{J}, \qquad \text{(A.1)}$$

$$\vec{\nabla} \times \vec{\nabla} \times \vec{H}' - k^2 \vec{H}' = \frac{4\pi}{c} \vec{\nabla} \times \vec{J},$$

where $\vec{J} \equiv -i \frac{\omega}{4\pi} (\varepsilon' - 1) \vec{E}$, $k \equiv \omega/c$.

The solution to Eqs (A.1) is known to be [26]

$$\vec{E}'(\omega, \vec{r}') = -i \frac{4\pi}{\omega} \iiint [k^2 \vec{J} + (\vec{J}\vec{\nabla})\vec{\nabla}] \psi \, d\vec{r}, \qquad \text{(A.2)}$$

$$\vec{H}'(\omega, \vec{r}') = -i \frac{4\pi}{\omega} \iiint [-ik \vec{J} \times \vec{\nabla}] \psi \, d\vec{r},$$

where $\psi(\vec{r}, \vec{r}') = -\frac{1}{4\pi} |\vec{r} - \vec{r}'|^{-1} \exp\{ik|\vec{r}-\vec{r}'|\}$,

Let us define the form of ε' in the equation for $\vec{E}$ more explicitly. Let the difference of ε' from ε is caused by deviation of the resonator surface from a spherical shape (2). In this case we shall have for the electric field

$$\vec{E}'(\omega, \vec{r}') = \vec{E}(\omega, \vec{r}') + \iiint (\varepsilon - \varepsilon') k^2 \vec{E} \psi(\vec{r}, \vec{r}')\, d\vec{r}. \quad (A.3)$$

Now we can calculate the resonator finesse Q, that is the ratio of the electromagnetic energy stored in the system, multiplied by 2π, to the energy lost by the resonator during an oscillation period T: $Q = \frac{2\pi}{P\,T}$, where $\overline{\quad} \equiv \frac{1}{8\pi} \iiint (\varepsilon E'^2 + H'^2)\, d\vec{r}$,

$P \equiv \frac{c}{4\pi} \iint_S \overline{[\vec{E}' \times \vec{H}']}\, \vec{e}_r r^2\, d\Omega$. S denotes the sphere with a radius $r = a$, $\vec{e}_1$ is the outward normal to S, $\vec{E}'$ and $\vec{H}'$ depend on t and $\vec{r}$, the line above denotes the time averaging, $d\Omega = \sin\vartheta\, d\vartheta\, d\varphi$. When calculating P for a TE mode we made the following approximations:

$$P = \frac{c}{8\pi} \iint_S |\vec{E}'|^2 r^2 d\Omega \le P_0 + \frac{c}{8\pi} \iint_S |\vec{E}' - \vec{E}|^2 r^2 d\Omega,$$

where P_0 defines the energy loss for an ideal sphere at $b = 0$,

$$|\vec{E}' - \vec{E}|^2 \le \left[(M^2 - 1)\frac{akb}{4\pi r}\right]^2 \iint_S |\vec{E}(\omega, \vec{\alpha})|^2 d\Omega,$$

where $\vec{\alpha}$ is the vector with spherical coordinates a, ϑ, φ. We made similar approximations when calculating P for a TM mode. The equation for Q has been derived as follows:

$$Q^{-1} = Q_r^{-1} + Q_s^{-1},$$

where $Q_s \ge 2\pi (n + \frac{1}{2}) \left(\frac{\varepsilon}{M}\right)^{2k} (\varepsilon\mu - 1)^{-1} (kb)^{-2}$;

Q_r is determined by $\vec{E}$ and $\vec{H}$ and is found in Ref. 17 (See Eq. (1) of Sec. II).

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22. A volume $V_f = 2\pi a^3 |x_q| [\frac{1}{2}(n + \frac{1}{2})]^{-2/3}$ in which the electromagnetic field of WGM is localized is smaller than the droplet volume V.

23. We have $\overline{n_l^o} = 3 \frac{D_g}{a^2} N_g \gamma$ when the gas-phase deactivation of MM's are slow ($\tau_{qg} >> N_d^{2/3} D_g$) and $a >> L_l$.

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