Lasing threshold for whispering-gallery-mode microsphere lasers

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The analytical expression for the lasing threshold for whispering-gallery-mode microsphere lasers under the strong-coupling condition is obtained by solving a quasi-three-level quantized model. It explicitly displays the dependence of the lasing threshold on the cavity radius, the homogeneous linewidth, the quality factors and linewidths of the relevant cavity modes. [S1050-2947(99)06107-7]

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I. INTRODUCTION

It is well known that the presence of microstructure electromagnetic resonances may greatly enhance or inhibit optical emissions occuring in microcavities depending on whether or not they spectrally coincide with cavity resonance modes, the so-called whispering-gallery-modes (WGM's). Since the microcavity-modified effects on the optical processes such as fluorescence, lasing, and Raman scattering in microcavities are not only of fundamental theoretical importance but also have a host of applications, there has been intensive study on the subject and great progress has been achieved over the last decade [1–4]. Thresholdless lasing is achieved in the low-*Q* cavity quantum electrodynamics (CQED) regime [4]. Small Fabry-Perot cavities involving special supermirrors and atomic beams have been used both for high-*Q* CQED experiments and for single-atom laser action [5]. Lasing has also been reported in high-*Q* microdroplets [1] and in high-*Q* microspheres [6,7]. A very low lasing threshold is observed in high-*Q* Nd-doped silica whispering-gallery-mode microsphere lasers [7]. The *Q* factors of WGM's in both liquid and solid microcavities have reached very high values (over 10^8) [7,8], enabling one to observe nonlinear optical effects with only a few photons in the mode [8]. In addition, an effort has been made towards achieving a thresholdless operation in Nd-doped silica whispering-gallery-mode microsphere lasers by cooling them at liquid helium temperature so as to decrease the homogeneous linewidth of the transitions to the MHz range [9]. In that case one should be able to achieve, in a Nd-doped silica microsphere, the strong coupling regime between a few particles and a few hundred photons stored in the WGM's [9]. It is therefore important and desirable to develop a theory for obtaining the lasing threshold for whispering-gallery-mode microsphere lasers under the strong coupling condition, which will be undertaken in this article. It is emphasized that no one has been able to obtain, even numerically not to mention analytically, the dependence of this threshold on the various physical quantities (such as the radius of the microsphere, the homogeneous linewidth, the quality factors and linewidths of the relevant cavity modes) under the strong-coupling condition due to the difficulties mentioned in the next section although such dependence has already been obtained previously in the weak-coupling regime [1]. In this article, we shall derive such dependence under the strong-coupling condition. The paper is organized as follows. In Sec. II, we first describe the more realistic quasi-three-level system, and then briefly discuss the difficulties to deal with such a system. In Sec. III, we shall develop an approach to overcome these difficulties. In Sec. IV, we derive the analytical expression of the lasing threshold for whispering-gallery-mode microsphere lasers under the strong coupling condition, and discuss, by means of the analytical expression, its dependence on the radius of the microsphere, the homogeneous linewidth, and the quality factors and linewidths of the relevant cavity modes. Section V concludes the article with a summary.

II. DESCRIPTION OF THE SYSTEM

We consider the system of a particle (molecule, atom, or ion) placed in a microcavity. The particle is supposed to have a quasi-three-level structure, i.e., two sharp levels 1 and 2 as well as a level-band $3'$ consisting of a series of closely spaced sublevels of a total width Γ_h, as shown in Fig. 1. The level 1 (2) and level-band $3'$ are coupled by a beam of light $p(l)$ with its frequency $\omega_p(\omega_l)$, which is assumed to coincide with the central frequency of one (another) cavity resonance mode. The beam p serves as a pump and the beam l accounts for a produced lasing light. In proposing such a system, we have taken into account the fact that two laser beams with different frequencies (one is the pump and another the lasing light) are usually involved in the normal experimental arrangements for investigating lasing phenomenon in microspheres [1,9] and hence at least three particle's levels should be considered. The width Γ_h of the level-band $3'$ accounts for either the intrinsic molecular wide-band structure frequently occurring in the optical processes in the microdroplets [1] or the level (homogeneous and nonhomogeneous) width due to all the possible radiative and nonradiative relaxation processes. This width is usually relatively large for typical lasing and/or fluorescence experiments in microspheres (typically, Γ_h/c is in the range of 1 to 100 cm^{-1}) [1,9] and one of important tasks for achieving efficient whispering-gallery-mode microsphere lasers nowa-

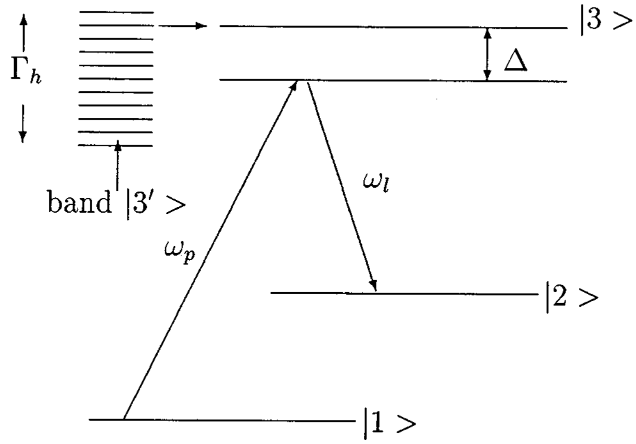

FIG. 1. Molecule's quasi-three-level structure: two sharp levels 1 and 2 as well as a level-band $3'$ consisting of a series of closely spaced sublevels of a total width Γ_h. The combination of two beams of light (pump p and lasing light l) with levels 1, 2, and 3 (one typical sublevel of band $3'$) forms a sharp three-level system in the Λ configuration.

days is to decrease this width by various possible measures [9].

Although the system considered here is a more realistic one than other previous two- or three-level models for typical whispering-gallery-mode microsphere lasers, it adds considerable difficulties to handle with such a complicated quasi-three-level system. First of all, it appears that no systemmatical master-equation formalism is available at present to handle such a problem. Even if exists, it is most likely too mathematically complicated to be handled with such a problem. Second, the quasi-three-level system is not a weak-coupling system for high-Q microcavity and hence the frequently used Fermi golden rule or the like is not suitable. At the same time, it may not be a strong coupling system if we consider the level-band $3'$ as a single level, particularly when the bandwidth Γ_h is large and therefore the strong-coupling theory is not directly applicable. Therefore, we need to develop a new approach to deal with such a system.

First we will first briefly go over some of the work of Lai and co-workers on the transition rate of a two-level atom in a microcavity [10,11]. The time-varying rates of the probability amplitudes for level occupation read [10] $p=-\gamma/4 \pm i\sqrt{K_\gamma\Gamma_0-(\gamma/2)^2}/2$ where K_γ is originally denoted as $K\gamma$ [10] and here it represents a shorthand notation for the integral $\int[\rho_c(\omega)/\rho_0]d\omega$, ρ_c and ρ_0 are the density of states in cavity and in free space respectively, γ is the linewidth of the cavity mode, $\Gamma_0=\tau_0^{-1}$, and τ_0 denotes the corresponding fluorescence decay time in bulk species. In the weak coupling region ($\gamma^2 \gg 4K_\gamma\Gamma_0$), both p's are negative real numbers (bi-exponential decay) and one of them determines the fluorescence decay rate in the microcavity which roughly equals the well-known Purcell enhancement formula obtained by using the Fermi golden rule with the effect of the cavity on the density of states considered [10]. Under the strong-coupling condition $\gamma^2 \ll 4K_\gamma\Gamma_0$ (note that this relation for the sharp levels is termed the strong-coupling condition throughout this article), $p\approx-\gamma/4\pm i\sqrt{K_\gamma\Gamma_0}/2$, and hence there occur so-called slowly decayed Rabi oscillations with the frequency $\sqrt{\Gamma_0 K_\gamma}$ and a single very small decay rate $\gamma/2$, implying that one can express the Rabi frequency as $\Omega \approx \sqrt{\Gamma_0 K_\gamma}$ and neglects the very small decay rate ($\gamma/2 \ll \Omega$) in using the Hamiltonian formalism in strong coupling regime. The discussion also connects dipole-coupling constants in Eq. (1) below with the parameters of the molecule and the two cavity resonance modes, i.e., $2g=\sqrt{\Gamma_0 K_\gamma}$.

III. APPROACH TO DEAL WITH THE QUASI-THREE-LEVEL SYSTEM

In this section, we develop an approach to obtain the lasing threshold for the quasi-three-level system. The approach is outlined as follows. (i) The wide-band quasi-three-level system is not a strong-coupling system if the band $3'$ is considered as a level when Γ_h^2 is not much less than $4K_\gamma\Gamma_0$. However, we can first consider an atom with three sharp levels, levels 1 and 2 as well as a typical sublevel 3 of the level-band $3'$, as shown in Fig. 1, where Δ denotes the detuning parameter and calculate the corresponding Rabi frequency $\Omega(\Delta)$ by using the Hamiltonian formalism under the strong coupling condition $\gamma^2 \ll 4K_\gamma\Gamma_0$ for any two of the three *sharp* levels involved. Note that this strong-coupling condition here and hereafter is defined for sharp levels involved. (ii) The band structure of ''level'' $3'$ is taken into account by averaging $\Omega(\Delta)$ over the detuning parameter Δ according to the integration $\langle\Omega\rangle=\int\Omega(\Delta)h(\Delta)d\Delta$ over a Lorentzian spectrum $h(\Delta)$ of the width Γ_h. (iii) The averaged Rabi frequency is related to the lasing threshold.

We begin with the first step. The Hamiltonian for the typical three-level system composed of sharp levels 1, 2, and 3 can be written as [12]

$$H=\sum_{i=1}^{3} E_i\sigma_{ii}+\hbar\omega_p a_p^\dagger a_p+\hbar\omega_l a_l^\dagger a_l+\hbar g_p(a_p\sigma_{31}+a_p^\dagger\sigma_{13}) +\hbar g_l(a_l\sigma_{32}+a_l^\dagger\sigma_{23}), \tag{1}$$

where a_j represents the photonic annihilation operators of the light j, $\sigma_{ii}=|i\rangle\langle i|$ are the level occupation numbers and $\sigma_{ij}=|i\rangle\langle j|$ $(i\neq j)$ are the transition operators from levels j to i, dipole-coupling constants g_j, as explained in the last paragraph, can be expressed as $2g_j=\sqrt{\Gamma_{0j}K\gamma_j}$ $j=p,l$. This three-level system can be exactly turned into an effective two-level problem for an arbitrary detuning Δ [12], and the corresponding explicit effective two-level Hamiltonian H_{eff} in the interaction picture reads [12]

$$H_{eff}=-\hbar g(a_p^\dagger a_l\sigma_{12}+a_f^\dagger a_p\sigma_{21})+\frac{1}{2}\hbar\delta(\sigma_{22}-\sigma_{11}), \tag{2}$$

where $\delta=\tau(\Delta)(\overline{g_p^2}-\overline{g_l^2})/(\overline{g_p^2}+\overline{g_l^2})$, $g=\tau(\Delta)g_p g_l/(\overline{g_p^2}+\overline{g_l^2})$, $\tau(\Delta)=(\sqrt{(\Delta/2)^2+\overline{g_p^2}+\overline{g_l^2}}-|\Delta|/2)\mathrm{sgn}(\Delta)$, Δ denotes the detuning as shown in Fig. 1, $\overline{g_j}=g_j\sqrt{N_j}$, $j=p,l$. Here $N_p=n_p+\sigma_{22}$ and $N_l=n_l+\sigma_{11}$ ($n_j=a_j^\dagger a_j$ denotes photon number of the jth mode) are two conserved quantities whose values can be determined by initial conditions. In studying the lasing problem, we can assume that initially the molecule is in level 1 and there exist n_p photons in the pump light p and no photon in the produced lasing light l. This initial condition makes the two conserved quantities take the values of $N_p = n_p$ and $N_l = 1$, respectively. The Rabi frequency for this effective two-level system and hence for the typical three-level system is [13] $\Omega(\Delta) = \sqrt{\delta^2 + 4g^2 N_p N_l}$, which after some manipulation becomes

$$\Omega(\Delta) = \sqrt{\left(\frac{\Delta}{2}\right)^2 + \overline{g_p^2} + \overline{g_l^2}} - \frac{|\Delta|}{2}. \tag{3}$$

Since we have obtained the Rabi frequency $\Omega(\Delta)$ for the typical sharp three-level system, the band characteristic of the ''level'' $3'$ is taken into account by avaraging $\Omega(\Delta)$ over the detuning parameter Δ by the integration $\langle\Omega\rangle = \int \Omega(\Delta) h(\Delta) d\Delta$. Here $h(\Delta)$ denotes a Lorentzian spectrum of the width Γ_h.

In order to obtain a simple analytical expression for the average Rabi frequency, the Lorentzian spectrum $h(\Delta) = \pi^{-1}(\Gamma_h/2)/[(\Gamma_h/2)^2 + (\Delta)^2]$ in $\langle\Omega\rangle = \int \Omega(\Delta) h(\Delta) d\Delta$ is approximately taken as $h(\Delta) \approx \overline{h}(\Delta)$ with $\overline{h}(\Delta) = 2/(\pi\Gamma_h)$ for $|\Delta| < \pi\Gamma_h/4 \equiv \Gamma'_h$ and $\overline{h}(\Delta) = 0$ otherwise. Note that Γ'_h is the half-width of the approximated rectilinear spectrum $\overline{h}(\Delta)$. It is then straightforward to yield

$$\frac{2\langle\Omega\rangle}{\Gamma'_h} = [\sqrt{1+W} - 1] + W \ln\left[\frac{1+\sqrt{1+W}}{\sqrt{W}}\right], \tag{4}$$

with

$$W \approx \frac{(n_p+1)}{\Gamma_h'^2 \tau_0} \int_{\omega_p - \Gamma'_h}^{\omega_p + \Gamma'_h} \frac{\rho_c(\omega)}{\rho_0} d\omega, \tag{5}$$

where $\Gamma'_h \equiv \pi\Gamma_h/4$, n_p is the initial photon number of the pump, and $\rho_0 = \Gamma_0^{-1}$ is the fluorescence lifetime in bulk species involving level 1 and the level band $3'$. In writing the expression $W = [N_p(2g_p)^2 + N_l(2g_l)^2]/\Gamma'_h$, we have made use of $N_p = n_p$, $N_l = 1$, and $(2g_l)^2 \approx (2g_p)^2 = \Gamma_0 \int [\rho_c(\omega)/\rho_0] d\omega$.

The expression (4) with its W defined by Eq. (5) is a very general expression for the average Rabi frequency for the quasi-three-level particle placed in a microcavity with an abitrary geometrical shape, and hence is suitable for discussing the corresponding lasing threshold in that cavity by establishing the relation between the lasing threshold and the average Rabi frequency for that cavity. From now on, we shall concentrate on the spherical microcavities and microspheres, to establish such a relation and leave the discussions of other geometrcal microcavities elsewhere.

IV. LASING THRESHOLD FOR MICROSPHERE LASERS

In this section, we shall first derive the concrete expression of the average Rabi frequency in Eq. (4) by evaluating its W for microspheres, and then relating it to the corresponding lasing threshold. Finally, we shall obtain the analytical expression of the lasing threshold which explicitly demonstrates its dependence on the radius of the microsphere, the width of the particle's upper level-band, and the quality factor Q of the cavity modes.

By separating the density of states in the microcavity into resonant and background contributions [14] $\rho_c = \rho_r + \rho_b$, and then ultilizing $\int [\rho_r(\omega)/\rho_0] d\omega = \eta\gamma$ [10] and the sum rule [14] to obtain $\rho_b \approx \rho_0(1 - \eta\gamma/\Delta\omega)$, where $\Delta\omega \approx c/(na)$ is the separation of the two adjacent cavity modes [14], n is the refractive index, a is the radius of the microsphere, η and γ are the Purcell enhancement factor and the linewidth for the cavity resonance mode with its central frequency ω_p. It is then easy to put W in Eq. (5) into another form,

$$W = \frac{2(n_p+1)}{\Gamma'_h \tau_0}\left[1 + \left(\frac{a_c}{a}\right)^2 - \frac{a_b}{a}\right], \tag{6}$$

where $a_c = \sqrt{2a^2\eta\gamma/(\pi\Gamma_h)}$, $a_b = na^2\eta\gamma/c$, and n is the refractive index. The product $\eta\gamma$ in Eq. (5) can be calculated by the formulas [10] $\eta\gamma = 3c\lambda^2/(2\pi V_m)$, and $V_m \approx 3.4\pi^{3/2}(\lambda/2n\pi)^3(2\pi a/\lambda)^2$ (also see p. 183 in Ref. [1]).

The threshold pump photon number $n_{p,th}$ for the lasing under the strong-coupling condition can be calculated by the relation $p_{av}\langle\Omega\rangle_{n_p \to n_{p,th}} = \gamma$, where p_{av} is a numerical factor of the order unity and γ is the decay rate of the cavity resonant mode [2]. Let us explain this relation. The particle jumps from the level 1 up to the level 2 in one Rabi cycle by simultaneously absorbing a pump photon ω_p and emitting a photon ω_l. But, the emitted photon ω_l only has the chance p_{av}, on average, to contribute to the cavity resonance mode with the central frequency ω_l. Lasing occurs when the photon production rate $\langle\Omega\rangle p_{av}$ is equal to or greater than the decay rate of the cavity resonance mode γ. The average probability p_{av} can be estimated as follows. Considering the isotropic feature of the photon ω_l emission, and noting the fact that the emitted photon contributes the cavity resonance mode only if it hits the cavity surface of radius a with its incident angle greater than the critical angle $\theta_c = \arcsin(1/n)$, and it leaks out of the cavity otherwise, one easily finds $p = 0$ for a particle within the sphere of the radius a/n, where p denotes the emitted (by that particle) photon's probability to contribute the cavity mode, and hence the photon ω_l emitted by the particle does not contribute the lasing, while $p = \sqrt{1-(a/nr)^2}$ if the particle locates at radius r within the shell bounded by the critical surface of the radius a/n and the cavity surface. A mean probability p_{av} is obtained by averaging p over the cavity volume by taking into account the spatial distributions of the cavity resonance modes and the active particles involved, or over only the shell due to the strongly peaked feature of the cavity resonance modes within that shell. Here we simply take p at $r = (a + a/n)/2$ as a rough estimation of the mean probability p_{av}, i.e., $p_{av} \approx \sqrt{1-[2/(n+1)]^2}$ or $p_{av} \approx 0.515$ for $n = 4/3$. If M active molecules is involved, we can treat each of them by the one-molecule model, and the corresponding fraction of the emitted photon to contribute the lasing mode is, on average, $p_{av}\langle\Omega\rangle$. Lasing occurs if the ratio of the $Mp_{av}\langle\Omega\rangle_{n_p \to n_{p,th}}$ lasing photons to the total M photons emitted is equal to or greater than the decay rate γ of the cavity resonant mode, i.e., $p_{av}\langle\Omega\rangle_{n_p \to n_{p,th}} = \gamma$ mentioned at the beginning of this paragraph, and cited from Ref. [2].

Consequently, the threshold pump photon number $n_{p,th}$ for the lasing can be calculated by

$$\frac{2\gamma}{p_{av}\Gamma'_h} = [\sqrt{1+W} - 1] + W \ln\left[\frac{1+\sqrt{1+W}}{\sqrt{W}}\right], \tag{7}$$

where W is given by Eq. (6) with the replacement $n_p \to n_{p,th}$, and $p_{av} \approx 0.5$ represents the efficiency of the lasing mechanism.

For the most interesting situations of $\gamma/\Gamma_h' \leqslant 0.1$ satisfied for the typical experiments nowadays on whispering-gallery-mode microsphere lasers, Eq. (7) can be greatly simplified to read $W/4e \approx \gamma/(p_{av} e \Gamma_h')/\ln[(e p_{av}\Gamma_h'/\gamma)\ln(e p_{av}\Gamma_h'/\gamma)]$. Ultilizing this form and Eq. (6) after neglecting its very small a_b term ($a_b \approx 0.2$ μm for the experiment in Ref. [7]), we can explicitly express the threshold pump photon number for $\gamma/\Gamma_h' \leqslant 0.1$ as follows:

$$\frac{n_{p,th}}{n_{p,th}^{(0)}} \approx \frac{a^2}{a^2+a_c^2} \equiv \frac{\Gamma_h}{\Gamma_h+\Gamma_{ha}}, \tag{8a}$$

$$n_{p,th}^{(0)} = \frac{\beta\tau_0}{\ln[(e\Gamma_h\pi/2\beta)\ln(e\Gamma_h\pi/2\beta)]}, \tag{8b}$$

where a is the radius of the microsphere; τ_0 is the fluorescence lifetime in bulk species; $a_c^2 \approx 1.76\lambda c n^3/(\pi^{2.5}\Gamma_h)$; $\Gamma_{ha} = 1.76\lambda c n^3/(\pi^{2.5}a^2)$; $\beta \equiv 2\gamma/p_{av} = 4\pi c/(\lambda Q p_{av})$; n is the refractive index; and λ, γ, and Q are the wavelength, linewidth, and quality factor of the cavity resonance mode serving as the pump. Once the threshold photon number $n_{p,th}$ is obtained by Eq. (7), the lasing threshold pump intensity for whispering-gallery-mode microsphere laser is simply given by the relation $I_{p,th} = \hbar\omega_p n_{p,th}$.

Let us have some discussion on Eq. (8), which explicitly expresses how the lasing threshold for whispering-gallery-mode microsphere lasers depends on the various physical quantities of the relevant whispering-gallery-mode and the active particles which contribute the lasing light. Due to the slowly varying feature of the logarithm function in Eq. (8b), $n_{p,th}$ is approximately proportional to both the cavity loss ($\propto Q^{-1}$) and the fluorescence lifetime τ_0 in bulk species, and its dependence on the radius a and the homogeneous linewidth Γ_h is approximately described, as shown in Eq. (8a), by the factors $a^2/(a^2+a_c^2)$ and $\Gamma_h/(\Gamma_h+\Gamma_{ha})$ respectively. Consequently, the lasing threshold $n_{p,th} \propto a^2\Gamma_h$ as $a \ll a_c$, and it depends only weakly on both the radius a and the homogeneous linewidth Γ_h as $a \gg a_c$. It is pointed out that the nonhomogeneous linewidth is easily seen to be able to include in our approach, and the outcome is the same so long as Γ_h is reinterpreted as the total linewidth that originated from both homogeneous and nonhomogeneous linewidths.

V. CONCLUSION

In summary, we have developed an approach to dealing with a quasi-three-level system in which one of the ''levels'' is actually a continuous band. Such a complicated system appears to be more realistic than the previous models for the lasing phenomenon in both liquid and solid spherical microcavities. In addition, it is very difficult to handle by the previous methods, due to the reasons mentioned in Sec. II. We have derived the analytical expression (8) of the lasing threshold for a whispering-gallery-mode microsphere laser under the strong-coupling condition by solving a quasi-three-level quantized model. Note that the strong-coupling condition throughout this article is defined for the sharp levels involved. Our final result explicitly displays the dependence of the lasing threshold on the cavity radius, the active molecular homogeneous linewidth, the bulk fluorescence lifetime, and the quality factors and linewidths of the relevant cavity modes.

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