Volume 9, Number 8
1009-1963/2000/09(08)/0590-09
CHINESE PHYSICS
August, 2000
© 2000 Chin. Phys. Soc.

LINEAR AND NONLINEAR SEMICLASSICAL THEORY OF WHISPERING-GALLERY-MODE MICROSPHERE LASER*

CHAI JIN-HUA(柴金华)[a)b)], LU YI-QUN (路轶群)[b)], and LEUNG PUI-TANG(梁培)[a)]
[a)] *Department of Physics, Chinese University of Hong Kong, Hong Kong*
[b)] *Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Hefei* 230031, *China*

(Received 30 May 1999; revised manuscript received 4 March 2000)

A complete whispering-gallery-mode microsphere laser theory is developed, which combines usual laser semiclassical theory with Lorenz-Mie theory. The linear and nonlinear problems of the microsphere lasing are dealt with in the universal mode picture. The threshold condition, average amplitude of the steady state and frequency effect are obtained. A comparison between the theory and experiments is also made.

Keywords: whispering-gallery-mode, microsphere laser, cavity enhancement factor, threshold condition
PACC: 4255B, 4250, 3280

I. INTRODUCTION

The optical resonances of a dielectric microsphere are termed morphology-dependent resonances (MDR's)[1] to emphasize the fact that the resonance frequencies depend on the structure (size, shape and refractive index) of the microsphere, and also termed the whispering-gallery modes (WGM's) because of the similarity to whispering-gallery acoustic waves. WGM's have been studied extensively[2] for some reasons, for example, WGM's have extremely high Q value, lead to decreased thresholds for lasing and several types of nonlinear optical processes, give rise to the effect of cavity quantum electrodynamics (QED), and have important applications in photonics or optoelectronics.[3,4]

The lasing from microspheres occurring at discrete wavelengths corresponding to WGM's is referred to as whispering-gallery-mode microsphere laser. WGM laser was observed first in a spherical millimetric crystal sample of CaF_2:Sm^{++} by Garrett, Kaiser and Bond.[5] Later, in the past decades, there have been a number of experimental papers on WGM laser from liquid or solid microspheres, such as ethanal 30-μm-radius droplets doped with rhodamine,[6−8] dye-doped polystyrene solid spheres with diameters ranging from 10 to 92 μm[9] and 40–800μm,[10] microdroplets seeded fluorescent sol[11] or undoped polystyrene latex spheres[12] or nanoparticles,[13] or highly scattering intralipid,[14] double-layered dye doping microdroplet consisting of a spherical dye-doped solid inner core covered with a dyedoped liquid-solution outer shell,[15] three-dye mixed doped liquid microspheres,[16] microdroplets containing an absorber of concentration change,[17] Nd: YAG spheres[18] with about 200 μm-diameter, Nd: glass sphere with a radius of 40–60μm,[19] pure silica sphere of 25-50 μm-radius,[20] and so on. In these experiments, observations of threshold behavior, emission wavelength dependence and spatial distribution of emission have provided strong evidence for the presence of laser emission linked with high-Q WGM's. An important result is that the threshold pump level of the laser in the microsphere is much lower than that for a conventional laser.

From the theoretical viewpoint, WGM lasers can operate in a manner analogous to the conventional linear Fabry-Perot cavity lasers except that the cavity modes provided by the mirror cavity of a linear cavity laser are replaced by WGM's. The gain of lasing is provided by the feedback coming from microspheres. Therefore, several authors have directly applied one-dimensional laser theory in many laser texts[21,22] to the WGM laser. For example, Lin *et al.*[23] have examined the effect of spatial hole burning on the lasing of 15.3-μm-diameter R6G-water solution droplets, and indicated that although the lasing gain in R6G is nominally homogeneously broadened on a nanosecond

*Project supported by the Foundation of Chinese University of Hong Kong

time scale, the presence of spatial hole burning permits multimode oscillations simultaneously on many spherical cavity mode resonances. Mazumder *al et.*[17] have used the standard dye-laser theory[24] to explain the absorption-dependent blue shift of dye lasing in microdroplets. However, although there is an analogy between WGM laser and usual linear cavity laser, a detailed comparison of WGM laser experiments and theory requires a complete WGM laser theory which combines usual laser theory with the Lorenz-Mie theory. Such a WGM laser theory, to our knowledge, has not been made to date, in spite of several related basic work appeared.[25−27] This is just the motivation of our study.

The purpose of this paper is to develop a semiclassical theory of WGM laser and apply it to the related experiments. Our work is the extension of the traditional laser theory of Haken[28] and Lamb[29] and the one-dimensional output coupling laser theory of Ujihara[30,31] and Baseia,[32] and emphasize the lasing action associated with WGM in the microsphere. There are some differences between the two theories: In Haken's and Lamb's theory, an ideally reflecting optical cavity is assumed, i.e., no leakage cavity, an ohmic loss homogeneously distributed over the cavity is introduced, and a set of discrete spectrum mode is used in the ideal mode picture. In Ujihara's and Basiea's theory, the practical cavity is a part of the whole cavity, the coupling loss can be naturally considered, derived automatically and no loss term is phenomenologically added to the field equation. The continuous spectrum is used in the universal mode picture. In the present work, the microsphere is regarded as complete homogeneous and symmetrical, so Mie formalism is entirely adequate for an analysis of the field and Q value of WGM's which give rise to lasing, and the three-dimensional cavity (spherical cavity) is of one-dimensional simplicity. Our study is also done in the universal mode picture similar to Ujihara's and Baseia's theory, but a main difference is that a multimode WGM lasing theory can be formed based on the linear and nonlinear lasing theory of the single mode WGM laser.

This paper is structured as follows: In Sec. II, we give the Maxwell-Bloch equations of microsphere cavity. In Sec. III and Sec. IV, we deal with the linear and nonlinear laser problems respectively, and obtain the threshold and the average amplitude of the laser field. The comparison between theory and experimental results is also performed. In this paper we do not consider the linewidth of lasing mode and the fluctuation effect of the laser field, as done in usual semiclassical laser theory. In Sec. V, we give the conclusions.

Ⅱ. MAXWELL-BLOTCH EQUATION

We consider the system consisting of a uniform dielectric microsphere of radius a and dielectric constant n^2 placed inside a large concentric hollow ideally conducting sphere of radius $\Lambda(\Lambda \to \infty$ eventually).

From Maxwell equations (c.g.s.), the electric field $\boldsymbol{E}(\boldsymbol{r}, t)$ and the macroscopic polarization $\boldsymbol{P}(\boldsymbol{r}, t)$ satisfy

$$-\bigtriangledown^2 \boldsymbol{E}(\boldsymbol{r},t) + \frac{1}{c^2}\frac{\mathrm{d}^2}{\mathrm{d}t^2}\boldsymbol{E}(\boldsymbol{r},t) = -\frac{4\pi}{c^2}\frac{\mathrm{d}^2}{\mathrm{d}t^2}\boldsymbol{P}(\boldsymbol{r},t). \quad (1)$$

In this universal mode picture, $\boldsymbol{E}(\boldsymbol{r}$,t) and $\boldsymbol{P}(\boldsymbol{r}$,t) can be decomposed into eigenfunctions $\boldsymbol{e}_s(\boldsymbol{r})$ of the microsphere cavity:

$$\boldsymbol{E}(\boldsymbol{r},t) = \sum_s E_s(t)\boldsymbol{e}_s(\boldsymbol{r}), \quad (2)$$

$$\boldsymbol{P}(\boldsymbol{r},t) = \sum_s P_s(t)\boldsymbol{e}_s(\boldsymbol{r}), \quad (3)$$

where $\boldsymbol{e}_s(\boldsymbol{r})$ satisfies[25]

$$\bigtriangledown \times \bigtriangledown \times \boldsymbol{e}_s(\boldsymbol{r}) = \frac{\omega_s^2}{c^2} n^2(\boldsymbol{r})\boldsymbol{e}_s(\boldsymbol{r}). \quad (4)$$

Here $n^2(\boldsymbol{r})$ equals the constant n^2 inside the microsphere and unity outside the microsphere.

We choose in this paper

$$\int_{(r\le\Lambda)} \mathrm{d}^3 r n^2(\boldsymbol{r})\boldsymbol{e}_s(\boldsymbol{r})\cdot\boldsymbol{e}_{s'}(\boldsymbol{r}) = 4\pi\hbar\omega_s\delta_{ss'}. \quad (5)$$

From Eqs. (1), (2) (3) and (4) we easily obtain

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}E_s(t) + \omega_s^2 n^2(\boldsymbol{r})E_s(t) = -4\pi\frac{\mathrm{d}^2}{\mathrm{d}t^2}P_s(t). \quad (6)$$

The polarization $\boldsymbol{P}(\boldsymbol{r}$,$t)$ can also be decomposed into single atom contributions,

$$\boldsymbol{P}(\boldsymbol{r},t) = \sum_\mu \delta(\boldsymbol{r}-\boldsymbol{r}_\mu)\boldsymbol{p}_\mu(t), \quad (7)$$

where the summation runs over the laser-active atoms (for simplicity, we use "atoms" as a general term for practical atoms or molecules or ions).

Assuming $\boldsymbol{e}_s(\boldsymbol{r})$, $\boldsymbol{E}(\boldsymbol{r}$,$t)$ and $\boldsymbol{p}_\mu(t)$ are parallel to one another, we obtain from Eqs. (2) and (5)

$$\sum_s \frac{1}{4\pi\hbar\omega_s} n^2(\boldsymbol{r})e_s(\boldsymbol{r}_\mu)\cdot e_s(\boldsymbol{r}) = \delta(\boldsymbol{r}-\boldsymbol{r}_\mu), \quad (8)$$

and

$$P_s(t) = \sum_\mu \frac{1}{4\pi\hbar\omega_s} n^2(\boldsymbol{r}_\mu)e_s(\boldsymbol{r}_\mu)p_\mu(t). \quad (9)$$

Then Eq.(6) can be written as

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}E_s(t)+\omega_s^2 n^2(\boldsymbol{r})E_s(t)= -\frac{1}{\hbar\omega_s}\sum_{\mu} n^2(\boldsymbol{r}_\mu)e_s(\boldsymbol{r}_\mu)\frac{\mathrm{d}^2}{\mathrm{d}t^2}p_\mu(t). \quad (10)$$

Next, we only consider the negative frequency part $E_s^-(t)$ and $p_\mu^-(t)$ of $E_s(t)$ and $p_\mu(t)$. We take rotating wave approximation and slowly varying amplitude approximation ($\frac{\mathrm{d}^2}{\mathrm{d}t^2}\tilde{E}_s \ll \omega_s\frac{\mathrm{d}}{\mathrm{d}t}\tilde{E}_s, \frac{\mathrm{d}^2}{\mathrm{d}t^2}\tilde{P}_s \ll \overline{\omega}_\mu\frac{\mathrm{d}}{\mathrm{d}t}\tilde{P}_s \ll \overline{\omega}_\mu^2\tilde{P}_s$, where $\overline{\omega}_\mu$ is the transition frequency of the laser atom μ), and from Eq. (10) we obtain

$$2i\omega_s\frac{\mathrm{d}}{\mathrm{d}t}\tilde{E}_s^-(t)+[n^2(\boldsymbol{r})-1]\omega_s^2\tilde{E}_s^-(t)=\frac{1}{\hbar\omega_s} \sum_{\mu} n^2(\boldsymbol{r}_\mu)\overline{\omega}_\mu^2 e_s(\boldsymbol{r}_\mu)\tilde{p}_\mu^-(t)\exp[i(\overline{\omega}_\mu-\omega_s)t], \quad (11)$$

where

$$\tilde{E}_s^-(t)=\exp(-i\omega_s t)E_s^-(t),\qquad \tilde{p}_\mu^-(t)=\exp(-i\overline{\omega}_\mu t)p_\mu^-(t). \quad (12)$$

Thus we finally obtain the field equation

$$\frac{\mathrm{d}}{\mathrm{d}t}E_s^-(t)=i\omega_s' E_s^-(t)-\frac{i}{2\hbar\omega_s^2} \sum_{\mu} n^2(\boldsymbol{r}_\mu)\overline{\omega}_\mu^2 e_s(\boldsymbol{r}_\mu)p_\mu^-(t), \quad (13)$$

where $\omega_s'$is defined as $\omega_s'=\frac{1}{2}[1+n^2(\boldsymbol{r})]\omega_s$.

On the other hand, according to the density matrix theory[29] and adopting twolevel model, with upper level a and lower b, we can derive the Bloch equations.

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_{ba}=(i\overline{\omega}_\mu-\gamma_\perp)\rho_{ba}+\frac{1}{i\hbar}E(\boldsymbol{r}_\mu,t)\theta_{ba}d_\mu, \quad (14)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}d_\mu=-\gamma_\parallel(d_\mu-d_{\mu,0}) +\frac{2}{i\hbar}E(\boldsymbol{r}_\mu,t)(\theta_{ab}\rho_{ab}^*-\theta_{ba}\rho_{ab}), \quad (15)$$

where θ_{ab} and θ_{ba} are the atomic dipole moment matrix elements ($\theta_{ab}=\theta_{ba}^*$); d_μ is the population probability difference between level a and level b ($d_\mu=\rho_{aa}-\rho_{bb}$); the nondiagonal density matrix elements ρ_{ab} and ρ_{ba} ($\rho_{ab}=\rho_{ba}^*$) stand for, respectively, the positive frequency part and negative freqency part of the polarization $p_\mu(t)$, with $p_\mu^+(t)=-\rho_{ab}\theta_{ba}, p_\mu^-(t)=-\rho_{ba}\theta_{ab}$; $d_{\mu,0}$ is the population inversion probability without coherent interaction, only determined by the pumping and relaxation processes; $\gamma_\perp$ and $\gamma_\parallel$ are the decay constants of the atom, representing the transverse relaxation and longitudinal relaxation behavior, respectively.

Under rotating approximation, Eqs.(14) and (15) become

$$\frac{\mathrm{d}}{\mathrm{d}t}p_\mu^-(t)=(i\overline{\omega}_\mu-\gamma_\perp)p_\mu^-(t) -\frac{1}{i\hbar}\theta_{ab}^2\sum_s E_s^-(t)e_s(\boldsymbol{r}_\mu)d_\mu, \quad (16)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}d_\mu=-\gamma_\parallel(d_\mu-d_{\mu,0})+\frac{2i}{\hbar}\sum_s [E_s^+(t)e_s(\boldsymbol{r}_\mu)p_\mu^-(t)-E_s^-(t)e_s(\boldsymbol{r}_\mu)p_\mu^+(t)], \quad (17)$$

where we have assumed that $\theta_{ab}, \boldsymbol{p}_\mu(t)$ and $\boldsymbol{E}(\boldsymbol{r}_\mu$,t) are parallel.

Equations 13, (16) and (17) are Maxwell-Bloch equations of the system, the basic equations for solving WGM laser problems in the next sections.

Ⅲ. LINEAR THEORY

We assume that the population inversion probability d_μ is kept constant, that is, we make a linear approximation.

Letting t=0, the atoms are coupled to the field. From Eqs. 13 and (16), noting Eq.(2), we obtain

$$E^-(\boldsymbol{r},t)=\sum_s E_s^-(0)e_s(\boldsymbol{r})\exp(i\omega_s' t) -\frac{i}{2\hbar}\sum_s e_s(\boldsymbol{r})\exp(i\omega_s' t) \times\int_0^t \exp(-i\omega_s' t')\frac{1}{\omega_s^2} \times\sum_\mu n^2(\boldsymbol{r}_\mu)\overline{\omega}_\mu^2 e_s(\boldsymbol{r}_\mu)p_\mu^-(t')\mathrm{d}t', \quad (18)$$

$$p_\mu^-(t)=p_\mu^-(0)\exp(i\overline{\omega}_\mu-\gamma_\perp)t+\frac{i}{\hbar}\theta_{ab}^2\exp(i\overline{\omega}_\mu-\gamma_\perp)t \times\int_0^t \exp[(-i\overline{\omega}_\mu-\gamma_\perp)]t']E^-(\boldsymbol{r}_\mu,t')d_\mu\mathrm{d}t' \quad (19)$$

Substituting Eq.(19) into Eq. (18), then it becomes

$$E^-(\boldsymbol{r},t)=\sum_s E_s^-(0)e_s(\boldsymbol{r})\exp(i\omega_s' t)-\frac{i}{2\hbar}\sum_s e_s(\boldsymbol{r})\exp(i\omega_s' t)$$
$$\frac{1}{\omega_s^2}\sum_\mu n^2(\boldsymbol{r}_\mu)\overline{\omega}_\mu^2 e_s(\boldsymbol{r}_\mu)p_\mu^-(0)\frac{\exp[i(\overline{\omega}_\mu-\omega_s')-\gamma_\perp]t-1}{i(\overline{\omega}_\mu-\omega_s')-\gamma_\perp}$$
$$+\frac{1}{2\hbar^2}\sum_s\sum_\mu\theta_{ab}^2 n^2(\boldsymbol{r}_\mu)e_s(\boldsymbol{r}_\mu)d_\mu\frac{\overline{\omega}_\mu^2}{\omega_s^2}\int_0^t\exp[i\omega_s'(t-t')]$$
$$\int_0^{t'}\exp(i\overline{\omega}_\mu-\gamma_\perp)(t'-t'')e_s(\boldsymbol{r}_\mu)E^-(\boldsymbol{r}_\mu,t'')\mathrm{d}t''\mathrm{d}t', \tag{20}$$

which is the basic equation of the laser field.

In the following, we consider (i) $\gamma_\perp t\gg 1$, i.e. neglecting the term of $p_\mu^-(0)$ in Eq. (20); (ii) adiabatic hypothesis, that is $\gamma_\perp\gg\gamma$, here γ is the decay rate of the field.

We assume that the slowly varying amplitude of the field is

$$\tilde{E}^-(\boldsymbol{r},t)=\exp(-i\Omega t)E^-(\boldsymbol{r},t), \tag{21}$$

where Ω is the frequency of the laser field. Substituting Eq.(21) into Eq.(20), we finally obtain

$$\tilde{E}^-(\boldsymbol{r},t)=\sum_s E_s^-(0)e_s(\boldsymbol{r})\exp[i(\omega_s'-\Omega)t]+\frac{1}{2\hbar^2}\sum_s\sum_\mu\frac{\overline{\omega}_\mu^2}{\omega_s^2}\theta_{ab}^2 n^2(\boldsymbol{r}_\mu)e_s(\boldsymbol{r}_\mu)d_\mu$$
$$\int_0^t e_s(\boldsymbol{r}_\mu)\tilde{E}^-(\boldsymbol{r}_\mu,t')\frac{\exp[i(\omega_s'-\Omega)(t-t')]}{i(\Omega-\overline{\omega}_\mu)+\gamma_\perp}. \tag{22}$$

By Laplace transform, Eq.(22) becomes

$$\tilde{E}^-(\boldsymbol{r},p)=\sum_s E_s^-(0)e_s(\boldsymbol{r})\frac{1}{p-i(\omega_s'-\Omega)}+\frac{1}{2\hbar^2}\sum_s\sum_\mu\frac{\overline{\omega}_\mu^2}{\omega_s^2}\theta_{ab}^2 n^2(\boldsymbol{r}_\mu)e_s(\boldsymbol{r}_\mu)d_\mu$$
$$\times\tilde{E}^-(\boldsymbol{r}_\mu,p)\frac{1}{i(\Omega-\overline{\omega}_\mu)+\gamma_\perp}\frac{1}{p-i(\omega_s'-\Omega)}. \tag{23}$$

In Eq.(23), letting $\boldsymbol{r}=\boldsymbol{r}_\mu$, then replacing $\boldsymbol{r}_\mu$ by $\boldsymbol{r}$, (that is, now $\boldsymbol{r}$ represents the region of the atoms and $\boldsymbol{E}(\boldsymbol{r}$,t) is the field inside the microsphere), we obtain

$$\tilde{E}^-(\boldsymbol{r},p)=\frac{\sum_s E_s^-(0)e_s(\boldsymbol{r})[p-i(\omega_s'-\Omega)]^{-1}}{1-\frac{n^2}{2\hbar^2}\sum_s\sum_\mu\theta_{ab}^2|e_s(\boldsymbol{r})|^2 d_\mu\frac{\overline{\omega}_\mu^2}{\omega_s^2}[i(\Omega-\overline{\omega}_\mu)+\gamma_\perp]^{-1}[p-i(\omega_s'-\Omega)]^{-1}}. \tag{24}$$

Next we only account for TE mode, since there is a similar discussion for TM mode. According to Ref.[25], $\boldsymbol{e}_s(\boldsymbol{r})$ can be expressed as

$$\begin{aligned}\boldsymbol{e}_s(\boldsymbol{r})&=\beta j_l(nkr)\boldsymbol{X}_{lm}(\theta,\varphi),\ r<a,\\&=[\gamma^{(1)}h_l^{(1)}(kr)+\gamma^{(2)}h_l^{(2)}(kr)]\boldsymbol{X}_{lm}(\theta,\varphi),\ r>a\end{aligned} \tag{25}$$

and the local density of states $\rho(\omega_s,\boldsymbol{r})$ can be written as

$$\rho(\omega_s,\boldsymbol{r})\propto\varepsilon(\omega_s,\boldsymbol{r})=\frac{1}{4\pi\hbar\omega_s}\sum_{s'}|e_{s'}(\boldsymbol{r})|^2\delta(\omega_s-\omega_{s'}). \tag{26}$$

Here $\omega_s\varepsilon(\omega_s,\boldsymbol{r})$ is near narrow Lorentzian with width $\frac{\gamma}{2}$, centered at ω, therefore we assume

$$\omega_s\varepsilon(\omega_s,\boldsymbol{r})=\frac{\omega^3}{\pi^2c^3}K\frac{(\frac{\gamma}{2})^2}{(\omega_s-\omega)^2+(\frac{\gamma}{2})^2}|f(\omega,\boldsymbol{r})|^2, \tag{27}$$

where

$$|f(\omega,\boldsymbol{r})|^2=|j_l(nkr)\boldsymbol{X}_{lm}(\theta,\varphi)|^2, \tag{28}$$

and $\frac{\omega^3}{\pi^2c^3}$ is the value of $\omega\varepsilon(\omega)$ for a vacuum of infinite extent, K is the cavity enhancement factor of $\varepsilon(\omega_s,\boldsymbol{r})$ in resonant peak $\omega_s=\omega$.

We note that

$$\sum_s \to \sum_{\omega_s}\sum_{lm} \to \frac{\Lambda}{\pi c}\int d\omega_s, \tag{29}$$

where ω_s labels radial quantum numbers, the sum over l and m is dropped due to considering the single mode with fixed l and m. Thus we can express $|e_s\boldsymbol{r})|^2$ as

$$|e_s(\boldsymbol{r})|^2 = \frac{\pi c}{\Lambda}K'\omega^3\frac{(\frac{\gamma}{2})^2}{(\omega_s-\omega)^2+(\frac{\gamma}{2})^2}|f(\omega,\boldsymbol{r})|^2, \tag{30}$$

where $K' = \dfrac{4\hbar}{\pi c^3}K$.

Hence from Eqs.(24) and (30), we obtain

$$\tilde{E}^-(\boldsymbol{r},p) = \frac{\sum_s E_s^-(0)e_s(\boldsymbol{r})[p-i(\omega_s'-\Omega)]^{-1}}{1-\dfrac{\pi n^2}{2\hbar^2}\omega K'\gamma\sum_\mu\theta_{ab}^2|f(\omega,\boldsymbol{r})|^2 d_\mu\overline{\omega}_\mu^2[i(\Omega-\overline{\omega}_\mu+\gamma_\perp]^{-1}\left[p+\dfrac{\gamma'}{2}-i(\omega'-\Omega)\right]^{-1}}. \tag{31}$$

where $\gamma' = \dfrac{1+n^2}{2}\gamma, \omega' = \dfrac{1+n^2}{2}\omega$.

Now we discuss the case of homogeneously broadened atomic line. We assume that θ_{ab}^2 are the same for atoms. In this case the lasing transition frequency $\overline{\omega}_\mu$ does not depend on the index μ, i.e., $\overline{\omega}_\mu = \overline{\omega}$. The sum over the random atomic position $\boldsymbol{r}_\mu$ can be performed by an integration, i.e., $\sum_\mu \to \rho\int_{(r<a)}\mathrm{d}V$, here ρ is the density of active atoms.

Equation (31) has two poles. One is $i(\omega_s'-\Omega)$, the other is

$$p_0 = -\frac{\gamma'}{2}+i(\omega'-\Omega)+\frac{A\gamma}{i(\omega-\overline{\omega})+\gamma_\perp}, \tag{32}$$

where

$$A = \frac{\pi n^2}{4\hbar^2}\theta_{ab}^2\omega K'\overline{\omega}^2\rho\int|f(\omega,\boldsymbol{r})|^2 d_\mu\mathrm{d}V, \tag{33}$$

and d_μ usually depends on spatial position, so next we assume that the average population inversion probability is approximately expressed as

$$\overline{d} \approx \frac{\int|f(\omega,\boldsymbol{r})|^2 d_\mu\mathrm{d}V}{\int|f(\omega,\boldsymbol{r})|^2\mathrm{d}V}. \tag{34}$$

Then using the residue technique, we obtain

$$\begin{aligned}\tilde{E}^-(\boldsymbol{r},t) =& A\gamma\exp(p_0t)\sum_s E_s^-(0)e_s(\boldsymbol{r})[p_0-i(\omega_s'-\Omega)]^{-1}[i\Omega-\overline{\omega})+\gamma_\perp]^{-1}\\ &+\sum_s E_s^-(0)e_s(\boldsymbol{r})\exp[i(\omega_s'-\Omega)t]\frac{\frac{\gamma'}{2}+i(\omega_s'-\omega')}{i(\omega_s'-\Omega)-p_0}.\end{aligned} \tag{35}$$

Letting p_0=0, we obtain the threshold condition of laser oscillation. The threshold population in version probability is

$$\overline{d}_t = \frac{(1+n^2)\hbar^2\gamma_\perp}{\pi n^2\theta_{ab}^2\omega K'\overline{\omega}^2\rho\int|f(\omega,\boldsymbol{r})|^2\mathrm{d}V}\left[1+(\frac{\omega'-\overline{\omega}}{\dfrac{\gamma'}{2}+\gamma_\perp})^2\right]. \tag{36}$$

The frequency at the threshold is given by

$$\Omega = \frac{\dfrac{\gamma'}{2}\overline{\omega}+\gamma_\perp\omega'}{\dfrac{\gamma'}{2}+\gamma_\perp}, \tag{37}$$

which is similar to the expression of frequency in one-dimensional usual laser, and means that there exists a frequency pulling effect, that is, the laser frequency Ω is shifted from the cavity mode ω to the center frequency $\overline{\omega}$ of the atoms.

According to Eq.(36), we can see the following main features of the threshold $\overline{d}_t$:

(1) $\overline{d}_t \propto K^{-1}$. The order of enhancement factor K can be evaluated[26] as $K \sim \dfrac{DQ\lambda^3}{8\pi V}$, where D is degeneracy, Q is the quality factor, and V is the volume of the microsphere. For optical wavelengths, $\lambda = 0.5\mu$m, $Q = 10^6$, $a = 1-100\mu$m, the typical values would be $K \sim 10^4$–10. Therefore, we see that WGM laser has a very low threshold due to the larger enhancement factor compared to the ordinary laser. For example, the threshold reported in the experiment[20] of Nd-doped silica microspheres are smaller than that for conventional Nd-doped fiber lasers by more than

three orders of magnitude.

(2) $\overline{d}_t$ is increased as $(\omega' - \overline{\omega})^2$, where $(\omega' - \overline{\omega})$ stands for the difference between the cavity mode and the atomic center line. In addition, $\overline{d}_t \propto \rho$, and $\overline{d}_t \propto Q^{-1}$. These agree with the ordinary lasing case.

IV.NONLINEAR THEORY

In nonlinear theory of laser, the population inversion is not constant, we will take into account the saturation of the population inversion and give the average field amplitude in steady state and analyze the effect of frequency.

We assume that the inversion $d_{\mu,0}$ is established by the pumping and by relaxation processes, and that the laser field with frequency Ω is

$$E_s^-(t) = \tilde{E}_s^-(t)\exp(i\Omega t). \tag{38}$$

In Eq.(16), letting $d_\mu = d_{\mu,0}$ (zeroth approximation) and considering $|\frac{\mathrm{d}}{\mathrm{d}t}\tilde{P}_\mu(t)| \ll |\gamma_\perp \tilde{P}_\mu(t)|$, we obtain the first order polarization $P_\mu^{(1)}(t)$, then substitute it into Eq.(17), and get the first order population inversion $d_\mu^{(1)}$, again in Eq.(16) putting $d_\mu = d_\mu^{(1)}$, we finally derive the polarization under the third order approximation, i.e., the second order polarization $P_\mu^{(2)}(t)$:

$$\begin{aligned} P_\mu^-(t) = P_\mu^{(2)}(t) =& \frac{i}{\hbar}\theta_{ab}^2 d_{\mu,0}\sum_s \tilde{E}_s^-(t)e_s(\boldsymbol{r}_\mu)\frac{\exp(i\Omega t)}{i(\Omega-\overline{\omega}_\mu)+\gamma_\perp} + \frac{2i}{\hbar^3}\theta_{ab}^4 d_{\mu,0} \\ & \sum_s\sum_{s'}\sum_{s''}\tilde{E}_s^+(t)e_s(\boldsymbol{r}_\mu)\tilde{E}_{s'}^-(t)e_{s'}(\boldsymbol{r}_\mu)\tilde{E}_{s''}^-(t)e_{s''}(\boldsymbol{r}_\mu) \\ & \frac{\exp(i\Omega t)}{\gamma_\parallel[i(\Omega-\overline{\omega}_\mu)+\gamma_\perp]}\left(\frac{1}{i(\overline{\omega}_\mu-\Omega)-\gamma_\perp}+\frac{1}{i(\Omega-\overline{\omega}_\mu)-\gamma_\perp}\right), \end{aligned} \tag{39}$$

where the slowly varying amplitude $\tilde{E}_s^-(t)$ of the field can be given by Eq.(13) as

$$\frac{\mathrm{d}}{\mathrm{d}t}\tilde{E}_s^-(t) = i(\omega_s'-\Omega)\tilde{E}_s^-(t) - \frac{i}{2\hbar\omega_s^2}\sum_\mu n^2(\boldsymbol{r}_\mu)\overline{\omega}_\mu^2 e_s(\boldsymbol{r}_\mu)\tilde{p}_\mu^-(t), \tag{40}$$

and by integrating this equation, we obtain

$$\tilde{E}_s^-(t) = E_s^-(0)\exp[i(\overline{\omega}_s-\Omega)t] - \frac{i}{2\hbar}\int_0^t \exp[i(\Omega-\omega_s')(t'-t)]\frac{1}{\omega_s^2}\sum_\mu n^2(\boldsymbol{r}_\mu)\overline{\omega}_\mu^2 e_s(\boldsymbol{r}_\mu)\tilde{p}_\mu^-(t')\mathrm{d}t'. \tag{41}$$

Substituting Eq.(41) into Eq.(40), making a summation $\sum_s e_s(\boldsymbol{r}_\mu)$ in Eq.(40), using Eq.(30), and defining calculation sign

$$[\mu] = -\frac{i\pi}{4\hbar}\omega K'\gamma\sum_\mu n^2(\boldsymbol{r}_\mu)\overline{\omega}_\mu^2|f(\omega,\boldsymbol{r}_\mu)|^2, \tag{42}$$

we can derive the following equation

$$\begin{aligned} \sum_s \frac{\mathrm{d}}{\mathrm{d}t}\tilde{E}_s^-(t)e_s(\boldsymbol{r}_\mu) =& \left[-\frac{\gamma'}{2}+i(\omega'-\Omega)\right]\int_0^t \mathrm{d}t'[\mu]\tilde{p}_\mu^-(t')\exp\left[-\frac{\gamma'}{2}+i(\omega'-\Omega)\right](t-t') \\ & + [\mu]\tilde{p}_\mu^-(t) + \sum_s[i(\omega_s'-\Omega)]\tilde{E}_s^-(0)e_s(\boldsymbol{r}_\mu)\exp[i(\omega_s'-\Omega)t]. \end{aligned} \tag{43}$$

We express Eq.(30) as $|e_s(\boldsymbol{r})|^2 = M_s^2|f(\omega,\boldsymbol{r})|^2$, where M_s^2 is written as

$$M_s^2 = \frac{\pi c}{\Lambda}K'\omega^3\frac{(\frac{\gamma}{2})^2}{(\omega_s-\omega)^2+(\frac{\gamma}{2})^2}, \tag{44}$$

so that we can define a collective variable $A(t)$, as done in Ref.[32], that is

$$A(t) = \sum_s \tilde{E}_s^-(t)M_s, \tag{45}$$

which stands for the amplitude associated with a single quasimode, thus from Eqs. (39) and (43), we obtain

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}A(t) =& \left[-\frac{\gamma'}{2}+i(\omega'-\Omega)\right]\int_0^t \mathrm{d}t'[\mu]\tilde{p}_\mu^-(t')\exp\left[-\frac{\gamma'}{2}+i(\omega'-\Omega)\right](t-t') \\ & + [\mu]\tilde{p}_\mu^-(t) + \sum_s[i(\omega_s'-\Omega)]\tilde{E}_s^-(0)M_s\exp[i(\omega_s'-\Omega)t]. \end{aligned} \tag{46}$$

and

$$\tilde{p}_{\mu}^{-}(t) = \alpha A(t) + \beta |A(t)|^2 A(t). \tag{47}$$

Here, for simplicity, α and β are defined as

$$\alpha = \frac{i}{\hbar}\theta_{ab}^2 d_{\mu,0} \frac{1}{i(\Omega - \overline{\omega}_{\mu}) + \gamma_{\perp}}, \tag{48}$$

$$\beta = \frac{2i}{\hbar^3}\theta_{ab}^4 d_{\mu,0} |f(\omega, \boldsymbol{r}_{\mu})|^2 \frac{1}{\gamma_{\parallel}[i(\Omega - \overline{\omega}_{\mu}) + \gamma_{\perp}]} \left(\frac{1}{i(\overline{\omega}_{\mu} - \Omega) - \gamma_{\perp}} + \frac{1}{i(\Omega - \overline{\omega}_{\mu}) - \gamma_{\perp}} \right). \tag{49}$$

By mathematical transformation for Eq.(46), we finally have

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}A(t) = &\left[-\frac{\gamma'}{2} + i(\omega' - \Omega) \right] A(t) + [\mu][\alpha A(t) + \beta |A(t)|^2 A(t)] \\ &+ \sum_s \left[\frac{\gamma'}{2} + i(\omega_s' - \omega') \right] \tilde{E}_s^{-}(0) M_s \exp[i(\omega_s' - \Omega)t], \end{aligned} \tag{50}$$

which is the expression of the laser field in nonlinear case. In comparison with the linear case, in Eq.(50) there is a saturation term (proportional to $|A(t)|^2$), when $|A(t)|^2$=0, from Eq.(50) we can give the result in linear case, i.e., Eq.(35). This can be checked easily.

In Eq.(50), the last term depends on the initial value $\tilde{E}_s^{-}(0)$ and stands for the fluctuation contributing to the fluctuation of the laser field. In this paper we do not discuss the fluctuation effects and will study the problem in other paper. We now take account of the average amplitude $\overline{A}$ of the field in the steady state and the frequency Ω of the field. From Eq.(50), $\overline{A}$ and Ω satisfy, respectively, the following equations:

$$-\frac{\gamma'}{2} + \mathrm{Re}[\mu]\alpha + \mathrm{Re}[\mu]\beta \overline{A}^2 = 0, \tag{51}$$

and

$$\Omega = \omega' + \mathrm{Im}[\mu]\alpha + \mathrm{Im}[\mu]\beta \overline{A}^2. \tag{52}$$

In Eq.(51), the first term presents loss, the second one is gain and the third one is saturation. Equations (51) and (52) are similar to Lamb laser equations.[29] Next, we use these equations to discuss lasing action in homogeneously broadened atomic line case (available for liquid and solid microspheres in experiments reported).

Equation (42) and (48)–(52) can give the following results:

(1) Gain for small signal

$$G = \mathrm{Re}[\mu]\alpha = A_0 \gamma \frac{\gamma_{\perp}}{\gamma_{\perp}^2 + (\omega - \overline{\omega})^2}, \tag{53}$$

where

$$A_0 = \frac{\pi n^2}{4\hbar^2}\theta_{ab}^2 \omega K' \overline{\omega}^2 \rho \int |f(\omega, \boldsymbol{r})|^2 d_{\mu,0} \mathrm{d}V. \tag{54}$$

The expression of A_0 has the similar form to Eq.(33) except that d_{μ} is replaced by $d_{\mu,0}$. We note that $d_{\mu,0}$ generally depends on spatial position, but for simplicity we assume that there is a spatially homogeneous pumping, i.e., $d_{\mu,0} = d_0$ (constant). We see that

(i) $G \propto K$, that is, the gain is proportional to the enhancement factor of the microsphere;

(ii) $G \propto d_0$, here d_0 stands for the pumping level;

(2) Average photon number $\overline{n}(\overline{n} \equiv \overline{A}^2)$ in stationary state

$$\begin{aligned} \overline{n} = &\frac{\hbar^2}{4\theta_{ab}^2}\gamma_{\perp}\gamma_{\parallel} \left[1 + \left(\frac{\omega' - \overline{\omega}}{\frac{\gamma'}{2} + \gamma_{\perp}} \right)^2 \right] \\ &\frac{\int |f(\omega, \boldsymbol{r})|^2 \mathrm{d}V}{\int |f(\omega, \boldsymbol{r})|^4 \mathrm{d}V} \left(1 - \frac{\overline{d}_t}{d_0} \right). \end{aligned} \tag{55}$$

This means that (i) the average lasing intensity increases as pumping level d_0 in certain region is above threshold, which has been proved by many experimental results, such as those in Refs.[6], [13] and [14]; (ii) the average lasing intensity decreases with increasing size parameter x for the fixed lasing mode, higher Q value and pumping level near the threshold. The reason is that, from Appendix A, $[\int |f(\omega, \boldsymbol{r})|^2 \mathrm{d}V][\int |f(\omega, \boldsymbol{r})|^4 \mathrm{d}V]^{-1} \sim [x^{4/3} - x_0^{4/3}][x_0^{-1/3} - x^{-1/3}]^{-1}$, when $\overline{d}_t \propto x^{5/3} \sim d_0$, then Eq.(55) shows that $\overline{n}$ will decrease as x increases, which agrees with the result of the experiment described in Ref.[18].

(3) The frequency shift in stationary state

$$\Omega - \omega' = \frac{\frac{\gamma'}{2}}{\frac{\gamma'}{2} + \gamma_{\perp}} (\overline{\omega} - \omega'), \tag{56}$$

which is consistent with the formula of frequency at the threshold. This equation can be used to explain the wavelength shifts of dye lasing in microdroplets.[17] In the experiment, the lasing spectra of dyes, such as Rhodamine B, are blue shifted as the Nigroson absorption is increased. In fact, the decay rate of the field γ can be regarded as consisting of output loss $\gamma_{\rm out}$ and absorption loss $\gamma_{\rm abs}$. Generally, $\gamma_{\rm abs} \ll \gamma_{\rm out}$ if there is no absorber. When the concentration of the absorber is increased, i.e., $\gamma_{\rm abs}$ increased, the frequency shift (Ω–ω') is increased, that is, the lasing spectra is blue shifted. But for the lasing spectra of dye DCM, the blue shift is very small, because for DCM in ethanol there is a small overlap between the spectra of the absorption and emission, corresponding to the case that $\gamma_{\rm abs}$ is very small, thus no apparent wavelength shift is observed as the absorber concentration is increased. In addition, the red shift of lasing spectra as the lasing dye concentration increases can be understood using Eq.(56). The reason is that, when the dye concentration is increased, that is, the decay rate of the atom dipole moment $\gamma_\perp$ increases, the lasing frequency shift ($\Omega - \omega'$) is decreased, i.e., the red shift appears.

(4) The frequency in nonstationary state is

$$\Omega = \omega' - A_0\gamma\frac{\Omega-\overline{\omega}}{\gamma_\perp^2+(\omega-\overline{\omega})^2} + \frac{4\theta_{ab}^2}{\hbar^2}\frac{\gamma_\perp}{\gamma_\parallel} A_0\gamma\frac{\int|f(\omega,\boldsymbol{r})|^4{\rm d}V}{\int|f(\omega,\boldsymbol{r})|^2{\rm d}V}\frac{\Omega-\overline{\omega}}{[\gamma_\perp^2+(\omega-\overline{\omega})^2]^2}\overline{A}^2, \quad (57)$$

where the second term on the right side stands for frequency pulling and the third term leads to frequency pushing. In Lamb's book,[29] these two terms are, respectively, expressed by the frequency pulling coefficient and frequency pushing coefficient.

V. CONCLUSION

We have developed a semiclassical theory of WGM microsphere laser based on traditional laser theory. We have solved many lasing problems in microspheres, such as the threshold, the average amplitude and the frequency shift. The theoretical formulas can be used to explain lasing actions in liquid and solid microspheres. In our theory, light field is assumed to consist of a set of WGM's which have Lorentzian linewidth, one-dimensional output coupling laser theory is extended to microsphere cavity. From this semiclassical theory a complete quantum theory and a mulitimode theory of WGM microsphere laser will be easily established and the linewidth and multimode problems of WGM lasing can be solved, which will be discussed in other papers.

APPENDIX A

From Eq.(28), we now evaluate the ratio in Eq.(53), i.e., $[\int|f(\omega,\boldsymbol{r})|^2{\rm d}V][\int|f(\omega,\boldsymbol{r})|^4{\rm d}V]^{-1}$.

From

$$\boldsymbol{X}_{lm}(\theta,\varphi) = \frac{1}{\sqrt{l(l+1)}}\boldsymbol{L}Y_{lm}(\theta,\varphi), \quad (A.1)$$

$$\int \boldsymbol{X}^*_{l'm'}(\theta,\varphi)\boldsymbol{X}_{lm}(\theta,\varphi)\sin\theta{\rm d}\theta{\rm d}\varphi = \delta_{ll'}\delta mm', \quad (A.2)$$

we get

$$\int|f(\omega,\boldsymbol{r})|^2{\rm d}V = \int r^2|j_l(nkr)|^2{\rm d}r, \quad (A.3)$$

where

$$j_l(x) = \sqrt{\frac{\pi}{2x}}J_{l+\frac{1}{2}}(x), \quad x = nkr. \quad (A4)$$

The lasing modes observed in experiments usually correspond to large l values, so we only concern the region $x = nkr \sim l$, that is, $r \in (r_0, a)$, here $a - r_0$ implies the radial depth of the whispering gallery field distribution. Using the asymptotic expression[33] of $J_{l+\frac{1}{2}(x)}$ for larger l and x and $x \sim l$, we obtain

$$j_l(x) \sim \sqrt{\frac{\pi}{2x}}\frac{1}{3\pi}\left[\sin\frac{\pi}{3}\Gamma(\frac{1}{3})(\frac{x}{6})^{-1/3}\right], \quad (A.5)$$

and

$$\int_{r_0}^a r^2|j_l(nkr)|^2{\rm d}r \sim \frac{1}{16\pi}\sin^2\frac{\pi}{3} \Gamma^2(\frac{1}{3})(\frac{1}{6})^{-2/3}(a^{4/3}-r_0^{4/3})(nk)^{-5/3}. \quad (A.6)$$

Similarly,

$$\int|f(\omega,\boldsymbol{r})|^4{\rm d}V = \int r^2|j_l(nkr)|^4{\rm d}r \int|\boldsymbol{X}_{lm}(\theta,\varphi)|^4\sin\theta{\rm d}\theta{\rm d}\varphi. \quad (A.7)$$

We derive that

$$\int_{r_0}^a r^2|j_l(nkr)|^4{\rm d}r \sim 3C^4 (r_0^{-1/3}-a^{-1/3})(nk)^{-10/3}, \quad (A.8)$$

where

$$C = \sqrt{\frac{\pi}{2}}\frac{1}{3\pi}\sin\frac{\pi}{3}\Gamma(\frac{1}{3})(\frac{1}{6})^{-1/3}. \quad (A.9)$$

Finally we obtain

$$[\int|f(\omega,\boldsymbol{r})|^2{\rm d}V][\int|f(\omega,\boldsymbol{r})|^4{\rm d}V]^{-1} \sim [x^{4/3}-x_0^{4/3}][x_0^{-1/3}-x^{-1/3}]^{-1}. \quad (A.10)$$

REFERENCES

[1] S. C. Hill and R. E. Benner, "Morphology-dependent resonances," in P. W. Baber and R. K. Chang, eds., Optical Effects Associated with Small Particales, (World Scientific, Singapore, 1988).

[2] R. K. Chang and A. J. Campillo, eds, Optical Processes in Microcavities, (World Scientific, Singapore, 1996).

[3] A. Serpenguzel, D. Anold, G. Griffel, *Opt. Lett.,* **20**(1995), 654.

[4] N. Dubreuil, J. C. Knight, D. K. Leventhal, V. Sandoghdar, J. Hare and V. Lefevre, *Opt. Lett.,* **20**(1995),813.

[5] C. G. B. Garrett, W. Kaiser and W. L. Bond, *Phys Rev.,* **124**(1961),1807.

[6] H. M. Tzeng, K. F. Wall, M. B. Long and R. K. Chang, *Opt. Lett.,* **9**(1984),499.

[7] S. X. Qian, J. B. Snow, H. M. Tzeng and R. K. Chang, *Science* **231**(1986),486.

[8] H. B. Lin, A. L. Huston, B. J. Justus and A. J. Campillo, *Opt, Lett.,* **11**(1986),614.

[9] M. Kuwata. Gonokami, K. Takeda, H. Yasuda and K. Ema, *Jpn. J. Appl. Phys.,* **31**(1992),L99.

[10] H. Taniguchi, H. Yamada, T. Fujiwara, S. Tanosaki, H. Ito, H. Morozumi and M. Baba, *Jpn. J. Appl. Phys.,* **32**(1993),L58.

[11] R. L. Armstrong, J. G. Xie, T. E. Ruekgauer and R. G. Pinnick, *Opt. Lett.,* **17**(1992),943.

[12] R. L. Armstrong, J. G. Xie, T. E. Ruekgauer, J. Gu and R. G. Pinnick, *Opt, Lett.,* **18**(1993),119.

[13] H. Taniguchi, M. Nishiya, S. Tanosaki and H. Inaba, *Opt. Lett.,* **21**(1996),263.

[14] H. Taniguchi, S. Tanosaki, K. Tsujita and H. Inaba, *IEEE J. Q. E.* **32**(1996),1864.

[15] H. Taniguchi, H. Tomisawa and Sarjono, *Opt. Lett.,* **19** (1994),366.

[16] H. Taniguchi and S. Tanosaki, *Jpn. J. Appl. Phys.,* **32** (1993),L1421.

[17] M. M. Mazumder, G. Chen, R. K. Chang and J. B. Gillespie, *Opt. Lett.,* **20**(1995),878.

[18] T. Baer, *Opt. Lett.,* **12**(1987),392.

[19] Y. Z. Wang, B. L. Lu, Y. Q. Li and Y. S. Liu, *Opt. Lett.,* **20**(1995),770.

[20] V. Sandoghdar, F. Treussart, J. Hare, V. Lefevre-Seguin, J. M. Raimond and S. Haroche, *Phys, Rev.,* **A54**(1996),R1777.

[21] A. Yariv, Quantum Electronics, 2nd ed. (Wiley, New York, 1975).

[22] A. E. Siegman, Lasers (University Science, Mill Valley, Calif., 1986).

[23] H. B. Lin, J. D. Eversole and A. J. Campillo, *J. Opt. Soc. Am.,* **B9**(1992),43.

[24] F. P. Schafer, Dye lasers (Springer Verlag, New York, 1977).

[25] S. C. Ching, H. M. Lai and K. Young, *J. Opt, Soc. Am.,* **B4**(1987),1995.

[26] S. C. Ching, H. M. Lai and K. Young, *J. Opt. Soc. Am.,* **B4**(1987),2004.

[27] H. M. Lai, P. T. Leung and K. Young, *Phys, Rev.,* **A37**(1988),1597.

[28] H. Haken, Laser theory (Springer Verlag Berlin, Heidelberg, 1984).

[29] M. Sargent III, M. O. Scully, and W. Lamb, Laser Physics (Addison-Wesley, Reading, 1974).

[30] K. Ujihara, *Jpn. J. Appl. Phys.,* **15**(1976),1529.

[31] K. Ujihara, *Phys. Soc. Jpn.,* **40**(1976),1406.

[32] B. Baseia and H. M. Nussenzveig, *Optica Acta,* **31** (1984),39.

[33] Z. X. Wang and D. R. Guo, Special functions (World Scientific, Singapore, 1989).