Properties of a laser based on evanescent-wave amplification

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The criteria for lasing in a dielectric sphere surrounded by an active medium are given. They are presented in simple analytical form and confirmed by numeric calculations of the amplitudes of the morphology-dependent resonances in spontaneous emission spectra. The determined lasing threshold of this novel-type laser is compared with that of a conventional spherical laser with an optically active internal layer. The diverse advantages of a laser based on the gain of the evanescent part of a whispering-gallery mode are discussed. © 2005 Optical Society of America

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1. INTRODUCTION

A laser can be considered as a photonic device that consists of an active medium and an optical cavity. It is commonly understood that the active medium should be located inside of the optical cavity.[1] Recent years have shown an increased interest in spherical microlasers.[1–8] Their modes, called whispering-gallery modes (WGMs), are commonly imagined as closed-trajectory rays confined within the cavity by almost total internal reflection from the spherical surface. Because of these modes, morphology-dependent resonances (MDRs) appear in the spectra of elastic light scattering, fluorescence, Raman scattering, and lasing. The WGM microcavities are characterized by (i) extremely high-quality factors, (ii) small volumes of excitation, and (iii) the surface character of the electromagnetic modes. Owing to these properties, a novel WGM laser could be realized in which the active medium is located outside of the sphere.

New optical effects in WGM microlasers have emerged through recent research, and such discoveries are listed here. A microsphere laser pumped by the evanescent field of a prism by way of the evanescent field of a microsphere was developed.[2,5] Coupling between the lasing particle and a glass plate[3] or other particle[8,9] was observed. A theory of coupling of high-Q WGMs in a microsphere with different coupler devices was developed.[10] Theoretical calculations showed that the evanescent part of the WGM of a transparent microsphere should dramatically increase the efficiency of inelastic light scattering (Raman scattering and fluorescence) in vapor.[11] The radiative coupling of individual atoms in a dilute vapor to the external evanescent field of WGM of a fused-silica microsphere was reported.[12] It was demonstrated that a laser threshold condition could be satisfied in a microlaser based on light emission from a single quantum dot in the evanescent part of a microsphere WGM.[13,14] Lasing and quantum electrodynamic (QED) effects in the composite microsphere–quantum-dot optical cavities were detected.[15–17] Schemes of microlasers with optically active dye molecules[1] or ions[18] located within a surface monomolecular layer[1] or a shell surrounding a microspherical cavity[18] were investigated. Recently, laser generation due to pumping of dye-doped liquid surrounding a fused-silica spherical microcavity was demonstrated.[19,20]

In this paper, the criteria for lasing in a sphere with an active internal layer and in a spherical WGM laser with evanescent-wave amplification (EWA) are derived. We predict also that the concept of EWA will enable new effects, e.g., a high saturation level of the emission intensity and realization of strong coupling between cavity modes and quantum-dot-type emitters. The size of this laser could be as small as a few tens of micrometers.

2. DEFINITION OF THE LASING THRESHOLD CONDITIONS

A. Einstein Coefficient for Spontaneous Emission in an Active Cavity

Recently, a quantum theory for spontaneous emission in absorbing and dispersive media has been developed.[21,22] This theory expresses the Einstein coefficient $\mathcal{A}$ for spontaneous emission through the Green tensor of a classical wave equation, the Maxwell equation for vector potential. In particular, the rate of electric dipole emission in a multilayered spherical structure with arbitrary complex refractive indices could be determined. For brevity's sake, the formulas for $\mathcal{A}$ that we apply in this paper are presented in Appendix A.[23]

Even in the first calculations of the Einstein coefficient for spontaneous emission in a micrometer-sized sphere, sharp spikes in the dependence of $\mathcal{A}$ on angular frequency ω of light appeared.[24,25] Below, we show that the amplitudes of such MDRs in spontaneous emission spectra increase if the light is amplified whether (i) within the sphere or (ii) in its surroundings.

Figure 1 shows a simplified scheme of a spherical laser. Its basic element is a silica or polymer, e.g., polymethyl-

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methacrylate (PMMA), sphere with a refractive index of $n_{3r}=1.5$. First, let us investigate the properties of an optically active homogeneous sphere placed in water (with $n_{1r}=1.33$). In this case, layers 1 and 2 shown in Fig. 1(a) are, in fact, one layer. A value of $\mathcal{A}$ can be found readily using the formulas of Eqs. (A1)–(A6) given in Appendix A. The solid curves denoted as 1 and 2 in Fig. 2 present the calculated ratios A/A_h, where $\mathcal{A}\equiv\langle\mathcal{A}\rangle$ denotes the rate averaged over the orientation of the dipole and its location in volume 3 [see Fig. 1(a)]; A_h is the rate of spontaneous emission in a homogeneous boundless volume with refractive index n_{3r}; n_f and $k_f=(\omega/c)n_f$ are the refractive index and the wave vector of light in layer f, respectively; and c is the speed of light in a vacuum. The calculations we executed for definite values of the parameter $x=k_1a_2$ are $x=221.8734588$ and $x=221.5292874$ for Figs. 2(a) and 2(b), respectively, at which the particular transverse magnetic (TM_l^q) and transverse electric (TE_l^q) modes with the number $l=240$ and order $q=1$ are dominant in the spontaneous emission process.

Figure 2 demonstrates that the local characteristics of spontaneous emission of the homogeneous sphere

$$A\rightarrow\infty \tag{1}$$

under certain conditions. To specify these conditions we added the left vertical lines in Figs. 2(a) and 2(b). These were found analytically from a consideration of the balance between the rates of WGM energy dissipation and amplification. Details of this analytical calculation are given in Subsection 2.B.

According to the executed numeric and analytical calculations, relation (1) takes place if (i) the angular frequency of radiation ω is equal to the resonant frequency ω_{s0} of a WGM,

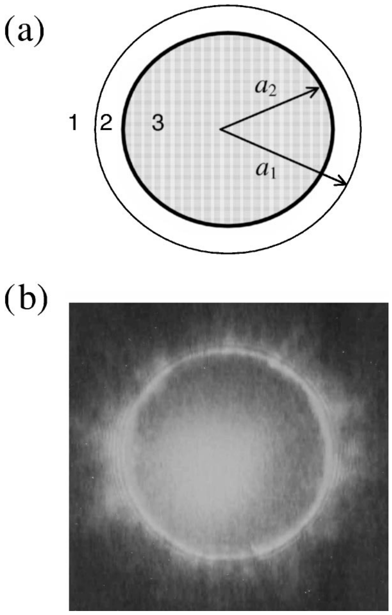

Fig. 1. (a) Scheme of the spherical laser and (b) negative photo of its prototype. (a) Index $f=3$ denotes a transparent dielectric, 1 and 2 are the same surrounding medium with possible gain in layer 2, $a_1=a_2n_{3r}/n_{1r}$ is the radius of the external caustic.[26] (b) Fluorescence of a 12 μm radius dye-doped PMMA sphere immersed in water and pumped by a 440 mW cw laser at a 532 nm wavelength.

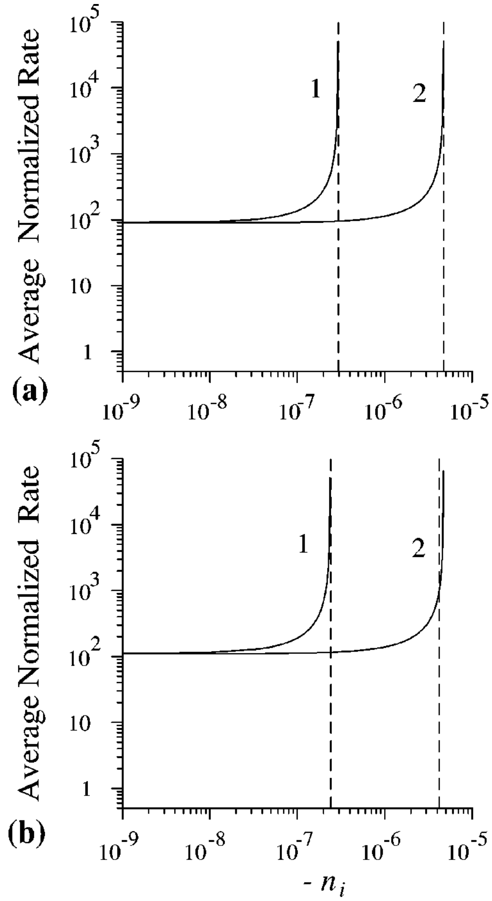

Fig. 2. Average rate A/A_h versus $-n_i$ for emission with the MDR frequencies of the (a) TM_{240}^1 and (b) TE_{240}^1 WGMs (see text). The solid curves were calculated by use of the formulas of Eqs. (A1)–(A8). Curves 1 describe the emission of an active sphere in water, $n_3=1.5+\imath\, n_i$; curves 2 relate to the emission of a passive sphere surrounded by an active water layer, $n_3=1.5+\imath\, 10^{-9}$, $n_2=1.33+\imath\, n_i$, $a_1-a_2\simeq 0.13\, a_2$. The vertical dashed lines show the lasing threshold values given by Eq. (16) at $a=a_2$, $y=m_r$ with factors of Eq. (9) [(a), right vertical], Eq. (10) [(b), right vertical], and Eq. (11) (left verticals).

$$\omega=\omega_{s0}, \tag{2}$$

and if (ii) energy losses in the cavity are compensated by the light gain. In this paper we name these two conditions the lasing threshold conditions. It is worth noting that there is an alternate for relation (1). Namely, the increases in the MDR amplitudes are concomitant with a narrowing of the resonances. Therefore, instead of relation (1), one could state that the width of a MDR $\Delta\omega_s$ in a calculated spectral dependence of A approaches zero. This trend can be described quantitatively by the equation

$$\Delta\omega_s=C_s/A(\omega_{s0})\rightarrow 0, \tag{3}$$

where C_s is a constant. Note that Eqs. (1) and (3) occur when a sphere is optically active but is not lasing sphere. Therefore the nonlinear growth of $A(\omega_{s0})$ and nonlinear narrowing of the MDRs are not caused by a lasing process.

Strictly speaking, relation (1) indicates that the approximation of weak coupling of an electric dipole and electromagnetic field, and hence the concept of the Einstein coefficients, is no longer valid.[24,25,27] However, we do not examine the regime of strong coupling leading to the Rabi oscillations[22,27,28] but rather consider the relation (1) as a sign that the threshold for lasing has been achieved.

Although the above consideration has not, to our knowledge, been presented elsewhere, it does not differ from the common laser concept. The novelty of this paper consists in the exploration of lasing in a three-layered spherical structure of innovative design. We propose to place the passive sphere 3 into a dye-doped liquid (see Fig. 1). The liquid layer 2 is presumed to be optically excited and has a complex refractive index $n_2=1.33+\imath\, n_i$.[23] In the particular calculations, we added an imaginary value of $\imath\times 10^{-9}$ to n_{3r} to take into account the natural light absorption in the dielectric. The solid curves 2 in Fig. 2 show the change of the averaged normalized rate of spontaneous emission inside sphere 3 caused by light gain in the evanescent-wave region of WGM. The singularities of A/A_h found for this scheme clearly show that lasing can be achieved when the active medium is located outside of the resonator volume. Of course, the correspondent lasing threshold values of $-n_i$ from 2×10^{-7} to 4×10^{-6} are higher than those of the conventional WGM laser. However, they are well below the value of $-n_i\simeq 2.5\times 10^{-3}$, which can be obtained by illumination of dye-doped liquid.[29] Hence, such an EWA laser is experimentally viable.

B. Quality Factor of a Whispering-Gallery Mode

The above-formulated conditions of the lasing threshold can be found in the framework of classical electromagnetic theory. Furthermore, the problem can be much simplified if one considers layer 2 in Fig. 1(a) as a perturbation of medium 1 (for an EWA laser) and sphere 3 (for a conventional WGM laser). In this case, the characteristics of a homogeneous sphere have to be calculated. The right-hand side of Eq. (2) can be found both numerically and analytically.[6,7,30] The second lasing threshold condition can be written as follows:

$$1/Q_s = 0, \tag{4}$$

where Q_s is the quality factor of the mode s with frequency ω_s.

The value of Q_s should account for various processes of energy loss and the process of light amplification, and it can be found from the following equation:[6,7,30–32]

$$\frac{1}{Q_s}=\frac{1}{Q_{s0}}+\frac{1}{Q_a},\quad \frac{1}{Q_{s0}}\equiv\sum_i\frac{1}{Q_i}, \tag{5}$$

where Q_i is the partial quality factor pertinent to process i. The processes that affect the quantity Q_{s0} are the following: light radiation by an homogeneous sphere of ideal spherical shape,[7,33] light scattering on inhomogenieties of the cavity surface[6,7] or on submicrometer-sized inclusions,[34] or the sum of light absorption and amplification by the resonator material (described by the factor Q_a). A minor role can also be played by the coupling of the cavity modes with other laser device components. In solid cavities with extremely high Q factors, allowance should be given for light absorption by a surface water nanolayer.[7]

In practice, Q values as large as $10^9–10^{10}$ were obtained.[7,35] We chose a value of a_2 so that a quantity of $Q_{s0}\simeq 2\times 10^6$ is determined by the radiative energy loss.

In Eq. (5) we separated the factor Q_a since we paid special attention to the amplification of radiation. Neglecting the influence of n_i on the mode structure and intermode energy exchange, the factor Q_a could be calculated from the following equation:

$$\frac{1}{Q_a}=\frac{1}{4\pi N_s}\int\int\int \epsilon_i(\mathbf{r}),|\mathbf{E}_s(\mathbf{r})|^2\mathrm{d}V, \tag{6}$$

with

$$N_s=\frac{1}{4\pi}\int\int\int \epsilon_r(\mathbf{r})|\mathbf{E}_s(\mathbf{r})|^2\mathrm{d}V, \tag{7}$$

where ϵ_i and ϵ_r are the imaginary and real part of the permittivity, respectively, and $\mathbf{E}_s(\mathbf{r})$ is the electric field of the resonant mode. Integrations in the right-hand sides of Eqs. (6) and (7) are over the resonator volume. For the WGM, this volume is defined by inequality $r\leqslant m_r a$, where $m_r\equiv n_{3r}/n_{1r}$ is the relative refractive index.[7,33] The estimate $r_c=m_r a_2$ of the open-cavity radius is in accordance with the ray-optics studies.[11,26]

The normalization of the electric field by Eq. (7) is rigorous for a closed cavity and is approximate for an open one. Methods of normalization of the electromagnetic modes of an open cavity allowing for radiative energy losses were discussed elsewhere.[28,33,36]

From the formula of Eq. (6) we can find factors that express the effects of light absorption or amplification inside $(0\leqslant r\leqslant a)$ or outside $(a\leqslant r\leqslant ay)$ a homogeneous sphere with radius a:

$$\frac{1}{Q_a}=\frac{2n_{3i}}{n_{3r}}\theta_{\mathrm{in}},\quad \frac{1}{Q_a}=\frac{2n_{3i}}{n_{3r}}\theta_{\mathrm{ex}}(y), \tag{8}$$

where quantity y can, in principle, lie in the interval from 1 to infinity. In this paper we define, for the first time to our knowledge, the following formulas for the parameters of Eqs. (8) $\theta_{\mathrm{ex}}(y)$ and θ_{in}. The factor $\theta_{\mathrm{ex}}(y)$ is equal to

$$\theta_{\mathrm{ex}}=\frac{m_r^2}{\eta_{\mathrm{in}}+\eta_{\mathrm{ex}}(m_{\mathrm{r}})}\left\{\eta_{\mathrm{ex}}(y)+\frac{1}{x}\mathrm{Re}\left[\frac{\zeta^*(y)\zeta'(y)}{|\zeta(x)|^2}-\frac{\zeta'(x)}{\zeta(x)}\right]\right\} \tag{9}$$

for a TM_l mode and

$$\theta_{\mathrm{ex}}=\frac{m_r^2\eta_{\mathrm{ex}}(y)}{m_r^2\eta_{\mathrm{in}}+\eta_{\mathrm{ex}}(m_r)} \tag{10}$$

for a TE_l mode. Here, for simplicity, we set the magnetic permeability of all layers equal to unity. The factors θ_{in} can be expressed through correspondent factors $\theta_{\mathrm{ex}}(m_r)$ as follows:

$$\theta_{\mathrm{in}}=1-\frac{1}{m_r^2}\theta_{\mathrm{ex}}(m_r). \tag{11}$$

In Eqs. (9) and (10), the term η_{in} is defined by Eq. (A8) and

$$\eta_{ex}(y) \equiv \int_1^y \left| \frac{\zeta_l(x\xi)}{\zeta_l(x)} \right|^2 d\xi. \quad (12)$$

The factors of Eqs. (9) and (10) that allow for the influence of the evanescent electric field located in a layer of width $d = y\ a_2$ on the mode energy dynamics are presented in Fig. 3. Figure 3 shows the results of the calculations executed for the TM^1_{240}, TE^1_{240}, and TE^2_{232} modes of a silica or PMMA microsphere immersed in water at $x = 221.8734588$, $x = 221.5292874$, and $x = 221.96325$, correspondingly.

In the literature a more simple formula for the fraction K of the TE-mode energy circulating outside of a spherical cavity is found:[35]

$$K = [k_1 a_2 n_{3r}^2 (m_r^2 - 1)^{3/2}]^{-1}. \quad (13)$$

The quantity K, that was called the coefficient of inclusion,[35] gives the ratio of the TE-mode energy in a layer $a_2 < r < a_2 + d$ to the total TE-mode energy at

$$d = [k_1 (m_r^2 - 1)^{1/2}]^{-1}. \quad (14)$$

A value of $\theta_{ex}(d)$ can be found by multiplication of the factor K by m_r^2:

$$\theta_{ex}(d) = K\ m_r^2. \quad (15)$$

According to Fig. 3, Eqs. (13)–(15) do not give correct values for either the full width of the layer filled by the evanescent field or for the quantity $\theta_{ex}(d)$. On the other hand, Fig. 3 shows that the evanescent field energy is localized in a layer with a width much narrower than $(m_r - 1)a_2 = r_c - a_2$.

Equations (8)–(12) allow us to calculate the lasing threshold value of n_i for a WGM laser:

$$n_i = -\frac{n_r}{2Q_{s0}\theta}. \quad (16)$$

The vertical dashed lines in Fig. 2 show the equivalence of the latter condition with relation (1). Values of Q_{s0} in Eq. (16) were found by calculations of the widths of the MDRs in the spectral dependence of A.

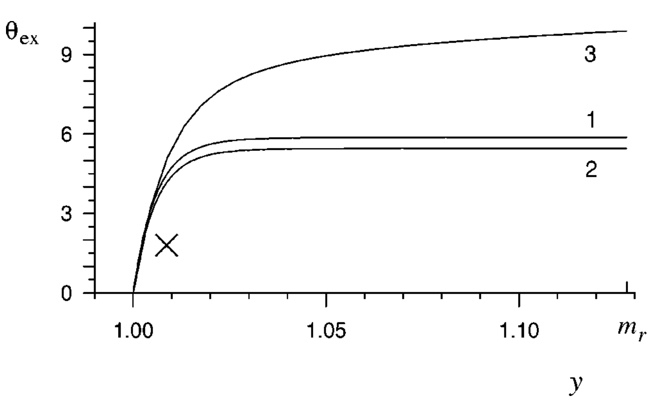

Fig. 3. Dependences of factors θ_{ex} on the parameter y calculated for the TM^1_{240} (curve 1), TE^1_{240} (curve 2), and TE^2_{232} (curve 3) modes of the silica microsphere immersed in water. The cross indicates the coordinates calculated with Eqs. (13)–(15) for the TE^1_{240} mode.

Note that even in the case of a homogeneous distribution of the absorption inside a spherical cavity, the quantity of Eq. (6) differs from the value $1/Q_a = 2n_{3i}/n_{3r}$ known from the literature.[6,7,30,31] The ratio θ_{in} of these quantities is less than unity because the evanescent wave gives a small but noticeable contribution to the integral in the right-hand side of Eq. (7).

In this paper we consider only a uniform angular distribution for the gain and a similar distribution for the light absorption. It is hard to achieve such a gain distribution experimentally. It seems that a three-dimensional problem should be solved to simulate the realization of this EWA concept. However, the cavity characteristics of interest depend on the integrals of Eq. (6) that can be calculated readily for an arbitrary spatial distribution of n_i. Therefore the problem does have a one-dimensional simplicity if the competition between WGMs with different azimuthal numbers can be neglected. The latter assumption can be valid even though the solid angle Ω of the active region is much less than 4π. In fact, just a few modes with $m \simeq l$ are excited in melted-tip microspherical lasers.[2,5,15,16,18,20] Under the dot excitation of the cavities, lasing threshold values of n_i should be increased by a factor of $4\pi/\Omega$.

3. DISTINCTIONS OF A LASER BASED ON GAIN OF THE EVANESCENT WAVE

A. Removal of Parasitic Effects

Spatial separation of the laser-pumping region from the region of the maximal field density allows one to avoid various undesirable processes. An and Moon[20] have reported the possibility of improving the Q factor of a spherical cavity. According to their opinion, pumping of an external medium can be used to minimize the pumping-induced stress and index of refraction change[37] in the cavity medium. The aim of the following discussion is to show that, in fact, the scope of the evoked phenomena can be much broader.

First, under optical pumping of spherical microcavities, lensing of the incident emission is of great importance. Because of this effect, one or two regions with a highly increased density of the electromagnetic field are formed.[30,38,39] This is accompanied by a few parasitic effects.[30,37] One of them is local heating of the cavity.[30] Under high pumping intensities, ponderomotive forces[30,40] or electrostriction[32] play negative roles. Stimulated Raman scattering (SRS)[30,31,39] and stimulated Brillouin scattering (SBS)[30,38,41] appear in a microsphere that is often concomitant with the broadening of resonant lines and their shift to another wavelength region. Eventually, these effects lead to the suppression of lasing.[42]

Because of the displacement of the active medium from the laser resonator, optical gain decreases. Therefore, to obtain lasing, one should increase a value of n_i by a factor of θ_{in}/θ_{ex} [see Eq. (16)]. Similarly, it should grow the thresholds for the SRS and SBS processes, but the increase should occur to a greater extent. Indeed, in a SRS laser, optical gain is proportional to the product of the inversion of the population of the initial and final energy levels and the intensity of light with the photon energy equal to the difference between the initial and intermediate energy level. This pump light intensity can be decreased in an EWA laser in $X_{\text{in}}/X_{\text{ex}}$ times where X_{in} denotes a factor of focusing of the incident radiation inside a microsphere. The parameter X_{ex} describes concentration of the input radiation in the evanescent-wave region. A typical value of X_{in} is $X_{\text{in}} > 10^2$,[37,39] whereas parameter X_{ex} could be as large as unity. Thus a threshold value for the SRS process in an EWA laser shall be higher by a factor of $(\theta_{\text{in}}/\theta_{\text{ex}})(X_{\text{in}}/X_{\text{ex}})$ in comparison with the conventional laser scheme. Therefore the SRS and SBS processes that under typical conditions usually have higher thresholds can be avoided in an EWA laser.

B. Energetic and Spectral Characteristics

The character of high-power lasing in an EWA laser should differ markedly from that of a conventional microsphere laser. Indeed, the saturation of an active medium during EWA is a less important process since there is no active medium inside the sphere. Saturation of the microlaser should therefore be observed at higher emission intensities.

Another important effect is the burning of holes in the spatial gain distribution.[43] In the experiment of Ref. 43, WGMs of a minimum of three orders with both TE and TM polarization were observed in the lasing spectrum. It was the spatial hole-burning effect that explained the distinction from other experiments in which only WGMs of the first order contributed to lasing.[1,2] In an EWA laser local gain has a similar influence on WGMs of all orders. This peculiarity can be seen from Fig. 3, which compares functions $\theta_{\text{ex}}(y)$ for the WGMs of the first and second orders. Let us examine a dependence of cw lasing on pumping power P in an EWA laser. According to Eq. (16), under an increase of P, lasing first starts on the modes with the highest Q factors, i.e. first-order modes. The cw lasing on these modes will lock the inversion population on a constant level determined by the equality of the rates of light energy amplification and dissipation. This means that lasing on the modes of the first order should prevent lasing on other modes. Hence, in a cw EWA laser, WGMs of several orders cannot be excited simultaneously.

In fact, only TE-polarized WGMs of the first order were observed in the emission spectra of the realized EWA laser.[20] Hence the number of emission lines in a high-power EWA laser can be decreased by several times in comparison with that of a conventional laser. This peculiarity opens new possibilities for cavity QEDs. Indeed, many cavity QED effects depend on the separation between the MDRs in the emission spectra. In particular, an increase of the separation between the MDRs should result in a corresponding increase of the stimulated emission cross section σ, according to Refs. 43 and 44.

C. Potential Applications

Another peculiarity of the EWA-based laser is the opportunity to manipulate the mode's Q factor by changing the environment. In particular, it is possible to reach the regime of strong coupling between the cavity modes and an internal atom or molecule or quantum dot. This peculiarity could be used in basic research in the field of cavity QED.[5,45]

The schemes with pumping energy stored and active particles located in different parts of a volume could be used in the research of donor–acceptor energy transfer processes. The spatial separation of donor and acceptor molecules would provide more refined experimental conditions for the study of the role of WGMs in energy transfer.[4]

Also, excitation of the outer medium of a spherical cavity can enrich spectroscopy based on spherical microsensors.[5–7,19]

Generally, spatial confinement and amplification of light are attracting increased interest from both basic and applied sciences. New concepts of microlaser design are being explored: microdisk, photonic crystal, random-media lasers, and so on. What is actually necessary for lasing is to compensate for round-trip energy losses in the cavity by the gain. Thus the basic concept of the microlaser discussed in this paper can be extended to other laser geometries as well.

D. Experimental Accomplishments

According to Fig. 1(b), in the fluorescence of Rhodamine-doped polymer microspheres, the evanescent escape of emission from the microspheres was considerable. Actually, it was responsible for the visualization of the WGM. Note that the width of the rim in Fig. 1(b) is approximately the chosen width of the evanescent field region in Fig. 1(a).

Lasing in an EWA laser was achieved for the first time to the best of our knowledge by Fujiwara and Sasaki.[19] They showed that the optically active dye solution just outside of a passive glass microsphere can function as a gain medium for the optical cavity. In the experiments, silica glass microspheres of a radius a from 5 to 15 μm and the refractive index of 1.5 were dispersed in a 10^{-2}–10^{-4} mol/l solution of Rhodamine B (RhB) in distilled water. In addition to the glass microspheres, PMMA latex spheres with $n_3 = 1.5$ and $a = 10$–15 μm were used for spectral measurement. The second-harmonic output of a Q-switched Nd:YAG laser was applied to excite the RhB solution within a 5 μm spot just outside of the microsphere. The conclusion about demonstration of lasing was drawn with three arguments. First, excitation of one edge of a sphere ($a = 13$ μm) by the green light at a high pumping power P of 9 mW resulted in an intense red emission appearing on the illuminated side of the sphere. In addition, the red light was radiated by the whole cavity surface with a bright spot on the shadow edge. Second, the emission spectrum of a PMMA latex 12.5 μm radius microsphere had anomalously enhanced MDRs of the TM modes of the first order (with $l = 177$–179). Third, the excitation power dependence of the emission intensity of a resonant peak at $\lambda = 616$ nm of a glass microsphere ($a = 14$ μm) in 10^{-2} mol/l RhB solution was determined. The emission from the pure dye solution exhibited an approximately linear dependence of the red emission intensity on P. In contrast, the intensity of the resonant microsphere emission increased by 2 orders of magnitude when P increased from 1 to 10 mW. The steep growth of the peak in the emission spectrum was observed at $P \geqslant 7$ mW, which was assigned as the lasing threshold pumping power. This threshold power was comparable with values reported previously for RhB-doped PMMA microspheres. This fact allowed for the prediction of a number of applications of the novel laser scheme.

First, Fujiwara and Sasaki noted that dye molecules doped in particles are easily degraded by photochemical destruction or photo-oxidation.[19] These effects should not be important in an EWA laser because of thermal diffusion of the dye molecules in the liquid solution. In addition to this advantage, a lasing microsphere was expected to be utilized as an active precise microsensor. It could be used to visualize dielectric bodies in the near-field zone and to monitor the refractive index or the absorption coefficient of a thin layer of liquid on the microsphere interface.

In subsequent papers,[20,46] experiments with capillary microcavity lasers based on the gain in the evanescent field region of WGMs were described. In these experiments, composite cavities were used. Their basic element was a fused-silica capillary of 200 μm inner and 320 μm outer diameters filled by solutions of Rhodamine 6G (Rh6G) in ethanol or ethanol–glycerol mixtures. The dye molecules were excited by 532 nm emission of the second harmonic of a Nd:YAG laser. Lasing at longer-wavelength regions of the spectrum was observed after insertion of a long segment of a fused-silica optical fiber 123 μm in diameter[46] or a spherical melted tip of a fused-silica fiber.[20] To confirm the effect of lasing on WGMs, the following arguments were used: (i) Bright rims around the fiber and the microsphere were observed; (ii) an increase of the pumping intensity led to a nonlinear growth of the MDRs in emission spectra; and (iii) a decrease of Rh6G concentration lowered the light absorption and consequently resulted in a significant shift of lasing lines toward shorter wavelengths. However, in the experiments, reflection of light from the inner capillary surface was likely to be of importance since lasing was achieved without inserted internal cavities.[46]

Some of the theoretical and empirical findings of Refs. 20 and 46 are similar to the estimates of this paper. To evaluate the lasing threshold in the EWA laser, the authors of Refs. 20 and 46 introduced an occupation factor for the evanescent field of a WGM. This factor is analogous to the factors of Eqs. (9) and (10). Analysis of lasing spectra[20,46] led to the conclusion that only single-order modes could produce lasing, in accordance with the above discussion. As a rule, the number of observed lasing modes was lower than the number of MDRs in the lasing spectra of traditional Rh6G-doped spherical cavities. Moreover, under some experimental arrangements, just one mode was observed in the lasing spectrum.

4. CONCLUSION

In this paper we provided a theoretical background for the new scheme of the WGM laser. In the scheme, lasing is achieved by excitation of the medium surrounding the spherical cavity. In the ray-optics picture, light amplification is obtained when a light ray reflects totally from a cavity surface with an active layer outside of it. In electromagnetic theory, this corresponds to an increment of the mode energy due to light amplification by the evanescent part of the mode. The properties of this laser are quite different from those of conventional WGM lasers.

The theoretical model proposed can be extended easily to simulate experiments with nonuniform distributions of gain.

APPENDIX A: SPONTANEOUS EMISSION IN A THREE-LAYERED SPHERICAL STRUCTURE

The theoretical model of Ref. 22 enables calculation of the rate of spontaneous emission $\mathcal{A}$ in a three-layered spherical structure such as that shown in Fig. 1. A value of $\mathcal{A}$ depends on the distance r_d of the electric dipole from the center of the sphere and dipole orientation. Averaging over all equally possible dipole directions, we obtained the following formula for the normalized rate for emission in layer 3:

$$\frac{\mathcal{A}(\xi)}{A_h} = 1 + \frac{3}{2}\mathrm{Re}\left(\frac{1}{z_5\xi}\sum_{l=1}^{\infty}\{C_{0l}(2l+1)\psi_l^2(z_5\xi) + C_{1l}[(l+1)\psi_{l-1}^2(z_5\xi) + l\psi_{l+1}^2(z_5\xi)]\}\right), \qquad \text{(A1)}$$

where $\xi \equiv r_d/a_2$, $\psi_l(y)$ is the lth-order Ricatti–Bessel function of the argument y, $z_{2f} \equiv k_f a_f$, $z_{2f+1} \equiv k_{f+1} a_f$,

$$C_{pl} = \frac{T_{F2}^{pl}}{T_{P2}^{pl}} \frac{T_{P1}^{pl} R_{P2}^{pl} + T_{F1}^{pl} R_{P1}^{pl}}{T_{P1}^{pl} + T_{F1}^{pl} R_{P1}^{pl} R_{F2}^{pl}}, \qquad \text{(A2)}$$

$$R_{Pf}^{pl} = \frac{k_{f+1-p}\zeta_l(z_{2f})\zeta_l'(z_{2f+1}) - k_{f+p}\zeta_l'(z_{2f})\zeta_l(z_{2f+1})}{k_{f+1-p}\psi_l(z_{2f})\zeta_l'(z_{2f+1}) - k_{f+p}\psi_l'(z_{2f})\zeta_l(z_{2f+1})}, \qquad \text{(A3)}$$

$$R_{Ff}^{pl} = \frac{k_{f+1-p}\psi_l(z_{2f})\psi_l'(z_{2f+1}) - k_{f+p}\psi_l'(z_{2f})\psi_l(z_{2f+1})}{k_{f+1-p}\zeta_l(z_{2f})\psi_l'(z_{2f+1}) - k_{f+p}\zeta_l'(z_{2f})\psi_l(z_{2f+1})}, \qquad \text{(A4)}$$

$$T_{Pf}^{pl} = \frac{k_f[\psi_l(z_{2f+1})\zeta_l'(z_{2f+1}) - \psi_l'(z_{2f+1})\zeta_l(z_{2f+1})]}{k_{f+1-p}\psi_l(z_{2f})\zeta_l'(z_{2f+1}) - k_{f+p}\psi_l'(z_{2f})\zeta_l(z_{2f+1})}, \qquad \text{(A5)}$$

$$T_{Ff}^{pl} = \frac{k_f[\psi_l(z_{2f+1})\zeta_l'(z_{2f+1}) - \psi_l'(z_{2f+1})\zeta_l(z_{2f+1})]}{k_{f+p}\psi_l(z_{2f+1})\zeta_l'(z_{2f}) - k_{f+1-p}\psi_l'(z_{2f+1})\zeta_l(z_{2f})}, \qquad \text{(A6)}$$

where indices $p=0$ and $p=1$ denote the transverse electric (TE) and transverse magnetic (TM) modes, respectively; derivatives of the functions with respect to their argument are designated by the prime; $\zeta_l(y) \equiv (\pi y/2)^{1/2} \times H_{l+1/2}^{(1)}(y)$ where $H_{l+1/2}^{(1)}$ is a Hankel function of the first kind.[23]

To reduce a number of arbitrary input parameters, one can average $\mathcal{A}$ over ξ:

$$A = 3\int_0^1 \mathcal{A}(\xi)\xi^2 \mathrm{d}\xi. \qquad \text{(A7)}$$

Note that the calculated rate A is a characteristic of emission inside the sphere, whereas the active medium can be located outside of it. The integral on the right-hand side of Eq. (A7) can be calculated by the following relation:

$$\int_0^1 \left| \frac{\psi_l(z\xi)}{\psi_l(z)} \right|^2 \mathrm{d}\xi \equiv \eta_{\mathrm{in}} = \frac{1}{2} \begin{cases} 1 + \frac{1}{p_l^2} - \frac{2l+1}{zp_l} & \mathrm{Im}z = 0 \\ \frac{\mathrm{Im}(zp_{l+1})}{\mathrm{Im}(z)\mathrm{Re}(z)} & \mathrm{Im}z \neq 0 \end{cases}, \qquad \text{(A8)}$$

where $p_l \equiv \psi_l(z)/\psi_{l-1}(z)$.

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