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Cavity-*Q*-driven spectral shift in a cylindrical whispering-gallery-mode microcavity laser

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Cavity-*Q*-driven spectral shift of lasing was observed in a cylindrical microcavity formed by rhodamine6G-doped quinoline in a capillary. The envelope of lasing spectrum showed a blueshift induced by the decreasing cavity *Q* of whispering gallery modes as the pump fluence increases. The thermally induced refractive index changes were measured from the shifts of individual lasing modes. The observed cavity-*Q*-driven spectral shift was well described by a simple dye laser model, which accounts for the dependence of cavity *Q* on the thermally induced refractive index change. © *2000 American Institute of Physics.* [S0003-6951(00)01925-2]

A microsphere or a microcylinder can be a high-*Q* optical cavity due to the trapping of light by total internal reflections at the cavity boundary. The resonance modes in a circular microcavity are called whispering gallery modes (WGMs). A WGM is specified by the azimuthal mode number *n*, the radial mode order *l*, and the polarization transverse magnetic (TM) or transverse electric (TE).[1] The cavity *Q* of WGM depends weakly on *n* and strongly on *l* for a given cavity. Due to the existence of high-*Q* WGMs and the gain enhancement of the cavity quantum electrodynamics origin, the threshold pump power for lasing can be reduced substantially, which is desirable for optical devices such as low-threshold microcavity lasers.[2] The microcavity lasing has been studied extensively in systems of semiconductor disk, dye-doped microdroplets, and liquid jet.[3–6]

A microcavity laser consisting of a fused-silica capillary filled with dye-doped liquid is an interesting system, where a coupling exists between the core WGM and the reflected light from the outer surface. Observation of interference modulation from a layered microcylinder was first performed by Knight *et al.*[7,8] Moreover, we recently performed a precise dynamic measurement of the interference period by controlling the refractive index of the liquid continuously.[9] Spectrum of microcavity laser is also important in that it reveals inner works of lasing process in the microcavity. Related to this, blueshift of lasing in microcavity have been observed by introducing scatterer or absorbers for enhancing the scattering and absorption losses, respectively.[10,11] However, the effect of the cavity-*Q* change on the spectral profile has not been successfully isolated in microcavity lasers. In this letter, we report the observation of cavity-*Q*-driven spectral shift resulted from the refractive index changes of a cylindrical microcavity.

Cavity *Q* of a WGM is strongly dependent on the refractive index of the cavity medium. In a dye doped liquid microdroplet or a jet generated from an orifice, the refractive index is difficult to control independently of its size. On the other hand, the temperature of the dye-doped liquid filled in a capillary is easy to control by adjusting the fluence of a pump beam as demonstrated previously.[9] Because the expansion coefficient of fused silica is negligibly small compared to that of the liquid, the cavity size is independent of the liquid temperature, which permits the control of refractive index isolatedly from other factors.

Our experimental scheme is as follows: As the pump fluence increases, the effective cavity length, which is proportional to the refractive index of the cavity medium, will decrease due to the decrease of the refractive index. The wavelength of a given WGM will decrease (or shift to the blue) because it is proportional to the effective cavity length. Thus, the refractive index changes can be deduced from the wavelength shifts without the knowledge of actual pump fluences in this *in situ* experiment. The cavity *Q* of a given WGM also decreases (or its intrinsic leakage loss increases) with the decrease of the refractive index. So the wavelength, around which WGMs of given *l* undergo lasing, will be shifted to the higher gain region in order to overcome the increased cavity loss, which will result in the blue shift of the spectral profile.

The experimental setup, shown in Fig. 1, is similar to that of Ref. 9. The liquid sample was 1.8 mM/L Rh6G-doped

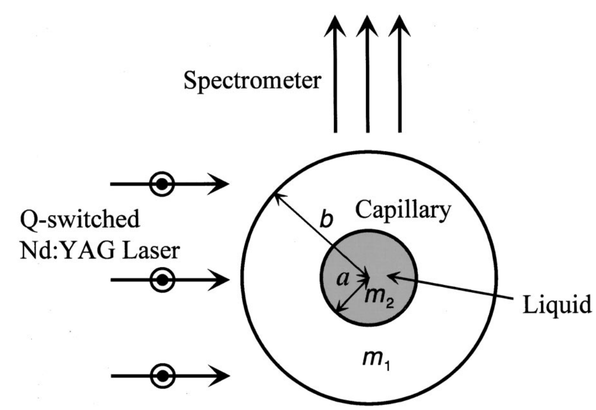

FIG. 1. Our experimental setup. A capillary tube of inner diameter of *a* and outer diameter *b* contains a dye solution, which is excited by a frequency-doubled Nd:YAG laser. The lasing spectrum is recorded by a spectrometer.

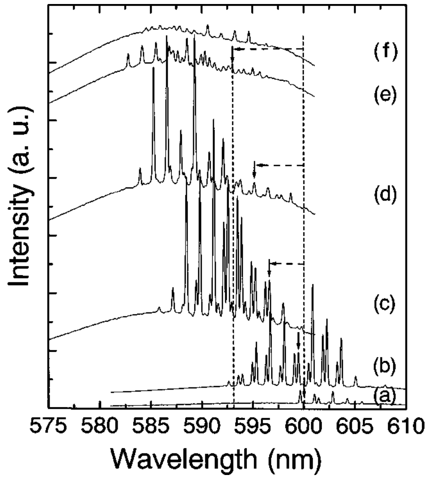

FIG. 2. Typical single-shot lasing spectra from a cylindrical microcavity filled with Rh6G-doped quinoline solution for various pump fluence. Changes of the refractive index of the liquid were thermally induced by the pump. The wavelength shifts of the TM mode denoted by the arrow were measured for various average pump power density I_{ave}: (a) $I_{ave} \sim 0.5\,W/cm^2$, slightly above threshold; (b) $I_{ave} \sim 2\,W/cm^2$ with 0.55 nm shift; (c) $I_{ave} \sim 12\,W/cm^2$ with 3.55 nm shift; (d) $I_{ave} \sim 18\,W/cm^2$ with 4.95 nm shift; (e) $I_{ave} \sim 25\,W/cm^2$ with 7.05 nm shift; and (f) $I_{ave} \sim 30\,W/cm^2$. For this the indicated mode cannot be identified due to the fluctuations of WGM peaks from shot to shot.

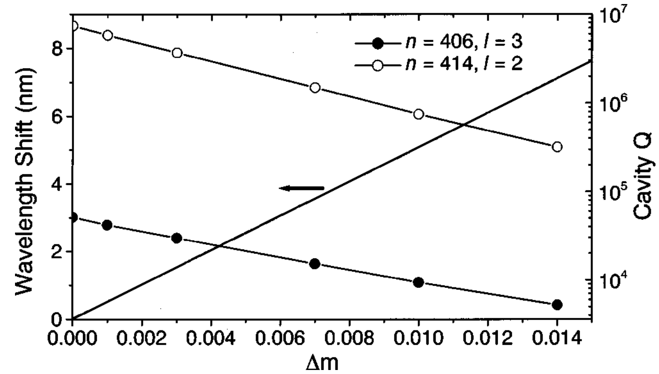

FIG. 3. Calculated amount of the blueshift of the WGM peaks and the cavity Q for two specific modes ($l = 2$ and 3 mode) with respect to the change of relative refractive index.

quinoline (refractive index $m_2 \simeq 1.626$ at 20 °C), which flowed through a fused-silica capillary (inner radius $a \sim 25\,\mu m$, outer radius $b \sim 160\,\mu m$, $m_1 \simeq 1.458$ at $\lambda = 600\,nm$). The absorption spectrum of the liquid sample was measured with a spectrophotometer and the fluorescence spectrum was taken with a spectrometer excited by a frequency-doubled continuous wave (cw) Nd:yttrium–aluminum–garnet (YAG) laser (wavelength: 532 nm). The emission cross section σ_e, deduced from the measured spectra, showed its maximum at about 570 nm. The absorption cross section, σ_a, rapidly decreased in the range of 580–630 nm. A diode-pumped Nd:YAG laser, both acousto-optically (AO) Q-switched and frequency-doubled (wavelength: 532 nm, pulse width: 200 ns, Q-switching frequency: 10 kHz), was loosely focused perpendicular to the capillary axis. The Q-switching on-time was externally controlled from 1 to 10 ms at repetition rate of 20 Hz. Hence, each pulse train contained a few tens of Q-switched pulses. Fine adjustment of the pump fluence was achieved by varying the Q-switching on-time or the pumping diode current.

Because of the strong absorption of the dye-doped liquid at the pump wavelength, the temperature, and consequently the refractive index, of the liquid can be controlled by the pump fluence. Our pumping scheme makes possible not only sufficiently high peak intensity for lasing, but also high average pump power over sufficiently wide range of refractive index change. The lased light from the capillary was collected on the entrance slit of a spectrometer. The spectrum was then recorded with a photodiode array attached on the image plane of the spectrometer.

Figure 2 shows the typical single-shot lasing spectra from the cylindrical microcavity. At slightly above the laser threshold [Fig. 2(a)], two groups of peaks with the same mode spacing of about 1.4 nm appears around 602 nm, slightly displaced from each other. Polarization analysis shows that the right peak of each pair is in a TM mode (one of them is denoted by an arrow) and the left peak, a TE mode. As the pump fluence increases, all of the WGM peaks show continuous blue shift due to the decreased refractive index of the liquid, m_2. In Fig. 2, the gradual shift of the TM peak marked by the arrow can be traced as the pump fluence increases. The shifts in Figs. 2(b)–2(e) are measured to be 0.55, 3.55, 4.95, and 7.05 nm, respectively. For high pump fluence complex mode structure is observed as shown in Figs. 2(e) and 2(f), in which the location of WGM peaks fluctuates in time so that it is difficult to determine the amounts of shift. This complex mode structure might be due to the nonuniform temperature distribution near the cavity boundary. Hence, our analysis, assuming uniform refractive index change, should be limited for the small wavelength shifts, as in the figures from Fig. 2(a) possibly to Fig. 2(e).

The profile of lasing spectrum also changes as the pump fluence increases. The central wavelength of the profile also shows a blueshift, as individual WGM peaks do, because the cavity Q of given order decreases with the decrease of relative refractive index $m = m_2/m_1$. From the amount of the blueshift of the marked TM peak, we can estimate the relative refractive index change $\Delta m \equiv |\Delta m_2|/m_1$. For $b \gg a$, the core WGM resonances are determined by the core size parameter $x_c \equiv 2\pi a m_1/\lambda$, and the associated cavity Q is that of a homogeneous fiber with the same diameter and of a relative refractive index $m = m_2/m_1$.[7] Figure 3 shows the calculated blueshift of a WGM peak and its cavity Q with respect to Δm. The initial value of m was chosen to be 1.117 and a was fitted to 25.5 μm based on the measured mode spacing.[9] The cavity Q was calculated for two modes with different mode orders, $n = 414$, $l = 2$ and $n = 406$, $l = 3$, which corresponds to the resonance wavelengths of about 600 nm. It shows that the wavelength shift is approximately linear to Δm with a slope of about 507 nm whereas the cavity Q decreases exponentially with respect to Δm. With the calculated slope, Δm was estimated to be 0.001, 0.007, 0.01, and 0.014 for Figs. 2(b)–2(e), respectively. The cavity Q for the $l = 2$ mode decreases from 7.4×10^6 to 3.2×10^5 as Δm increases from 0 to 0.014. At $\Delta m = 0.007$, the cavity Q reaches about 1.5×10^6, which is fairly larger than $Q_{abs} \equiv 2\pi m/\lambda\alpha(\lambda) \simeq 2 \times 10^5$ at $\lambda = 600\,nm$, where $\alpha(\lambda)$ is the absorption coefficient of the liquid at λ. On the other hand, the cavity Q for the $l = 3$ mode varies from 5.1×10^4 to 5.2

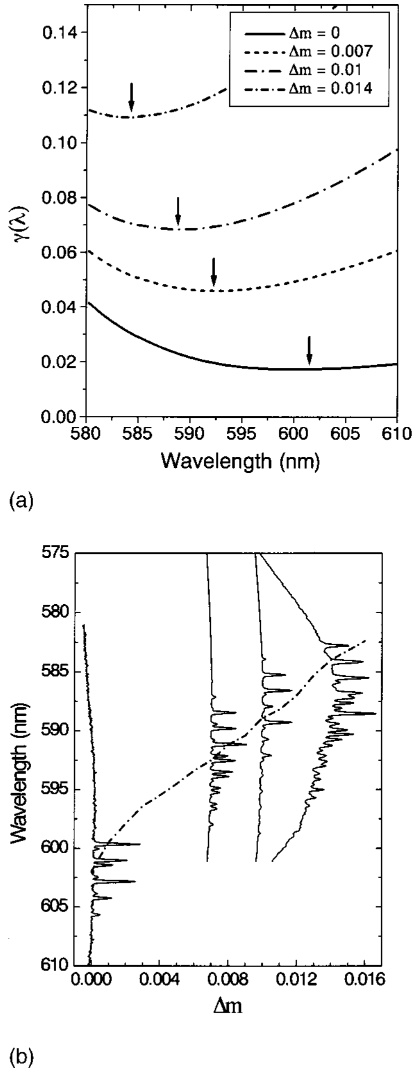

FIG. 4. (a) Calculated $\gamma(\lambda)$ at $\Delta m = 0$, 0.007, 0.01, and 0.014. As Δm increases, the minimum of $\gamma(\lambda)$, which is indicated by an arrow, shows a blueshift. (b) The comparison of the measured spectra (lines) and the calculated wavelength (dash dots) at which $\gamma(\lambda)$ is the minimum.

$\times 10^3$ in the range of $\Delta m = 0-0.014$. Hence, the mode coupling efficiency[4] (or mode visibility) $Q_{\mathrm{abs}}/(Q+Q_{\mathrm{abs}})$ is nearly 0 for the $l=2$ mode and nearly 1 for $l=3$ mode, in all range of Δm. Therefore, the continuously shifting peaks in Fig. 2 appear to be $l=3$ modes.

To analyze the observed spectral change, we apply a model of four-level dye lasing in a Fabry–Perot cavity.[11,12] Let n_1 and n_0 be the number densities of molecules in the first excited and the ground electronic singlet state, respectively, with $n_t \equiv n_1 + n_0$, the total number density of dye molecules. For lasing to occur, the single-pass gain should exceed the single-pass loss, which can be expressed as

$$n_1 \sigma_e \geqslant 2\pi m/\lambda Q + n_0 \sigma_a, \tag{1}$$

and then $\gamma(\lambda) \equiv n_1/n_t$, the minimum fraction of molecules to be excited for lasing at λ, is given by

$$\gamma(\lambda) = \frac{2\pi m/\lambda Q n_t + \sigma_a}{\sigma_a + \sigma_e} \approx \frac{\sigma_a(\lambda)}{\sigma_e(\lambda)}\left[1 + \frac{Q_{\mathrm{abs}}(\lambda)}{Q(\lambda)}\right]. \tag{2}$$

Calculated $\gamma(\lambda)$ at various Δm for the mode with $n = 406$ and $l=3$ are shown in Fig. 4(a). For comparison, the measured spectral profile (Fig. 2) with the calculated wavelengths for the minimum of $\gamma(\lambda)$ is shown in Fig. 4(b). When $\Delta m = 0, \gamma(\lambda)$ curve is nearly flat in the range of 595–610 nm and its minimum occurs at about $\lambda \sim 602$ nm. This curve well describes the observed spectrum in Figs. 2(a) and 2(b) at small Δm. As Δm increases to 0.007, the minimum of $\gamma(\lambda)$ shifts near to 592 nm, which is consistent with the observed spectrum in Fig. 2(c). When Δm increases more to 0.01, 0.014, the minimum of $\gamma(\lambda)$ shifts more to the blue (586 and 583 nm), which describes the spectra in Figs. 2(d) and 2(e). As can be seen in Fig. 4(b), the analysis from the simple model explains well the observed spectral shift at $\Delta m \leqslant 0.01$. When we apply the analysis for the $l=2$ mode, the minimum of $\gamma(\lambda)$ is located well above 600 nm at all values of Δm. This confirms the previous assertion, deduced from the mode visibility consideration, that the observed WGM peaks correspond to $l=3$ rather than $l=2$.

In conclusion, we demonstrated the effect of refractive index change on the cavity Q of WGM of a microcylinder by continuously controlling the refractive index of the cavity medium. By using a simple dye laser model, we could quantitatively analyze the observed shift of the spectral profile. Present scheme can be improved by using two different laser sources, one pulsed laser for pumping and a cw laser for inducing the refractive index changes. Our results also demonstrates that the pump process itself can cause the decrease of cavity Q by thermally induced refractive index change, which will in turn increase the laser threshold. For ultrahigh Q microcylinder/microsphere the cavity Q can be substantially lowered even with small decrease of refractive index of the cavity medium. Our technique provides a clever way of quantitatively assessing such effects induced by the refractive index changes, and therefore it can serve as an important tool in the development of the ultralow-threshold microcavity lasers.

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