Transient response of a thresholdless microdroplet dye laser

Kuei-Huei Lin and Wen-Feng Hsieh

Institute of Electro-Optical Engineering, National Chiao-Tung University, Hsinchu, Taiwan, China

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Lasing in a cavity formed by a dye-doped micrometer-sized liquid droplet showed a new thresholdless steady-state behavior and a time-dependent response. The thresholdless behavior is associated with enhancement and inhibition of the spontaneous emission. The time-dependent response is related to various quality factors of the cavity modes and was determined to be 8×10^8.

Recent observations of below-threshold lasing emission from individual liquid droplets[1] and low-threshold multiple-quantum-well lasers[2] may result from cavity enhancement and inhibition of the spontaneous emission. An enhancement factor F, defined as the ratio of the spontaneous emission rate of atoms in a microcavity to that in free space, A_c/A, quantifies cavity effects on the spontaneous emission rate. The enhancement depends on the location $\mathbf{r}$ of atoms[3,4] inside the cavity and is proportional to the photon density of states $\rho_c(\omega, \mathbf{r})$. The enhancement of the local density of states is theoretically predicted[4] to be 0.2 to 1500 for resonances in a spherical microcavity that has quality factors (Q's) of 10^4 to 10^5. Assuming that the atoms (molecules) are weakly coupled to the electromagnetic field in a single-mode cavity, the spatially averaged[5] density of states $\rho_c(\omega)$ over the entire cavity is proportional to Q. Using time-dependent perturbation theory, we can derive the spatially averaged enhancement factor as[3]

$$F \equiv \frac{A_c}{A} = \frac{g(\omega)}{\pi} \frac{\rho_c(\omega)}{\rho(\omega_0)} \Delta\nu \sim \frac{Q\lambda^3}{V}, \tag{1}$$

where $g(\omega)$ is the line-shape function of atoms (or molecules), ω_0 is the center frequency of the line-shape function, ρ is the photon density of states in free space, $\Delta\nu$ is the spacing between cavity modes, and V is the volume of the microcavity. For a multimode microcavity, F is proportional to the ratio $\int_0^\infty \rho_c(\omega) g(\omega) \mathrm{d}\omega / \int_0^\infty \rho(\omega) g(\omega) \mathrm{d}\omega$. This ratio becomes unity for large cavities (the radius $a \gg \lambda$), e.g., in a conventional laser.

By introducing the cavity-enhanced spontaneous emission rate, we can rewrite the rate equations of four-level lasers as follows[6]:

$$\frac{\mathrm{d}n}{\mathrm{d}t} = p - (1 - \beta)An - \beta A_c(1 + s)n - \Gamma n, \tag{2}$$

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \beta A_c(1 + s)n - \gamma s, \tag{3}$$

where n is the population density of atoms (molecules) in the upper levels, s is the number of photons inside the cavity, p is the rate of excitation of the atoms (molecules) to the upper levels, Γ is the nonradiative depopulation rate, and γ is the leakage rate of photons. The first term of Eq. (3) represents the rate of cavity-enhanced spontaneous and stimulated photons radiated by a fraction β of atoms into cavity modes. The remaining fraction, $1 - \beta$, of atoms radiates at a spontaneous emission rate A into free space. Micrometer-sized liquid droplets are extremely high-Q optical cavities. Q values of these droplets as high as 10^6–10^7 can be obtained from photon lifetime measurements.[7,8] The molecules inside the liquid droplets interact only with internal electromagnetic fields that satisfy certain morphology-dependent resonances (MDR's) that have different Q values. The excited molecules near the rim of the microdroplets interact strongly with higher-Q cavity modes (discrete vacuum field), which have states with large local density $[\rho_c(\omega, \mathbf{r})/\rho(\omega) \gg 1]$ and nearly no coupling to the free-space modes (continuum vacuum field). On the other hand, the excited-state molecules located away from the rim of the droplets radiate photons to the low-Q modes, which have slats with extremely small local density $[\rho_c(\omega, \mathbf{r})/\rho(\omega) \leq 1]$ and are inhibited from emitting photons. For a four-level system with homogeneously broadened emission, the dye molecule emission is coupled mostly into the high-Q cavity modes. The energy corresponding to low-Q mode spontaneous emission is removed to enhance the high-Q mode emission. Consequently the low-Q mode spontaneous emission rate is inhibited, and laser emission occurs only in high-Q cavity modes. This low-leakage microcavity can be considered an isolated system,[3] with β being equal to unity. Furthermore, because of the enhancement of spontaneous emission in high-Q modes, the upper-state molecules can decay only by photon emission ($\Gamma = 0$).

We report observation of thresholdless laser emission of micrometer-sized droplets of Rhodamine 6G in methanol (R6G/MtOH). For a steady-state response we measured a linear dependence of light output versus cw pumping; for a transient response we observed the light output when the droplets were pumped by a Q-switched pulsed laser. The transient response was dependent on cavity-enhanced spontaneous emission, as predicted by solving the modified rate equations with $\beta = 1$ and $\Gamma = 0$.

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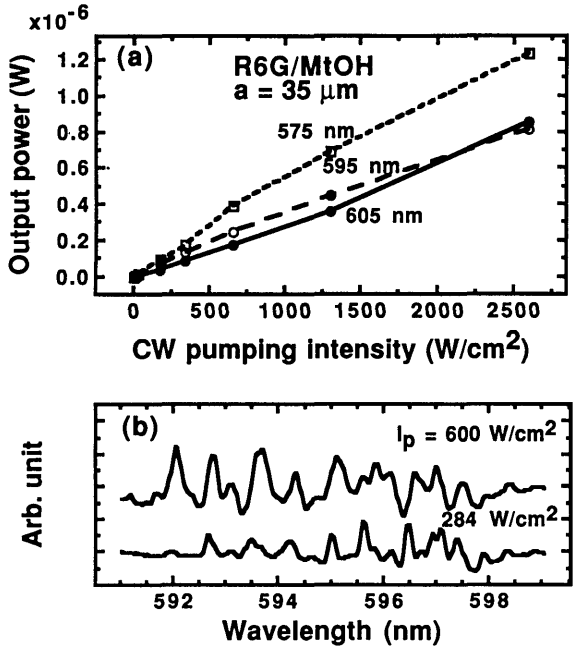

Fig. 1. (a) Measured output power versus the pumping intensity in three spectral ranges centered at 575 nm (dotted curve), 595 nm (dashed curve), and 605 nm (solid curve). (b) Lasing spectra at the center wavelength of 595 nm with pumping intensities of 284 W/cm^2 (lower curve) and 600 W/cm^2 (upper curve).

In the steady-state experiment, a cw argon-ion laser (at 514.5 nm) was focused to a spot size of 150 μm in diameter on a stream of monodispersive R6G/MtOH droplets (with a concentration of 10^{-3} M). The radius droplet size was ~35 μm. The red lasing emission from droplets was collected at 90° by a camera lens through a long-wave-pass filter to block the green elastically scattered pump light and then imaged onto the entrance slit of a Jarrell-Ash 0.3-m spectrometer with a 1200-groove/mm dispersive grating. The integrated spectra were measured by a power meter (United Detector Technology S-550). The pump power was varied by sets of neutral-density filters. No apparent threshold pumping characteristics (linearly proportional to the pumping intensities) were observed for spectral ranges centered at 575, 595, and 605 nm, as shown in Fig. 1(a). The emission spectra centered at 595 nm were recorded by a liquid-nitrogen-cooled charge-coupled device camera (Photometrics CC200). Although the spectra are taken from integration of 50 emitting droplets and may be blurred by a slight difference in individual droplet size, Fig. 1(b) shows clear MDR structure at pump intensities of 284 and 600 W/cm^2. This indicates that laser emission can occur even with pump intensity as low as 284 W/cm^2.

The spontaneous emission, which is mostly coupled into the low-leakage cavity modes, can coherently generate cavity-mode photons without exciting the continuum modes. For a particular high-Q cavity mode having high enhancement, A_c can be much larger than the leakage rate γ. Therefore the laser oscillation condition $(A_c n - \gamma) > 0$ is always satisfied even with weak pumping, where n is small (or $n \sim 1$). In our experiment, oscillation condition is achieved regardless of the size of n for a 35-μm-radius dye droplet, since $A = 2 \times 10^8$ s^{-1}/atom (a fluorescence lifetime of R6G of 4.8 ns), $F = 100$ (estimated according to Ref. 3), $A_c = FA = 2 \times 10^{10}$ s^{-1}/atom, and $\gamma < 2 \times 10^{10}$ s^{-1} (corresponding to $Q > 10^5$). The Q values of 10^5–10^7 have been consistently observed by various groups[1,7,8] in experiments on lasing and stimulated Raman scattering from microdroplets. On the other hand, molecules away from the rim of droplet interact with the continuum vacuum field at $A_c = A < \gamma$. Fluorescence is observed to be linearly proportional to the pumping power and is always much less intense than laser emission from high-Q cavity modes.[1,9]

For transient studies, a second-harmonic beam of a Q-switched pulsed Nd:YAG laser (J. K. Laser System 200) at $\lambda = 532$ nm, with a pulse width of ≈25 ns [see Fig. 2(a)], was used to pump the microdroplet lasers. Both the input laser pulse and the laser emission were detected by a fast photodiode (rise time <1 ns). The photodiode output was

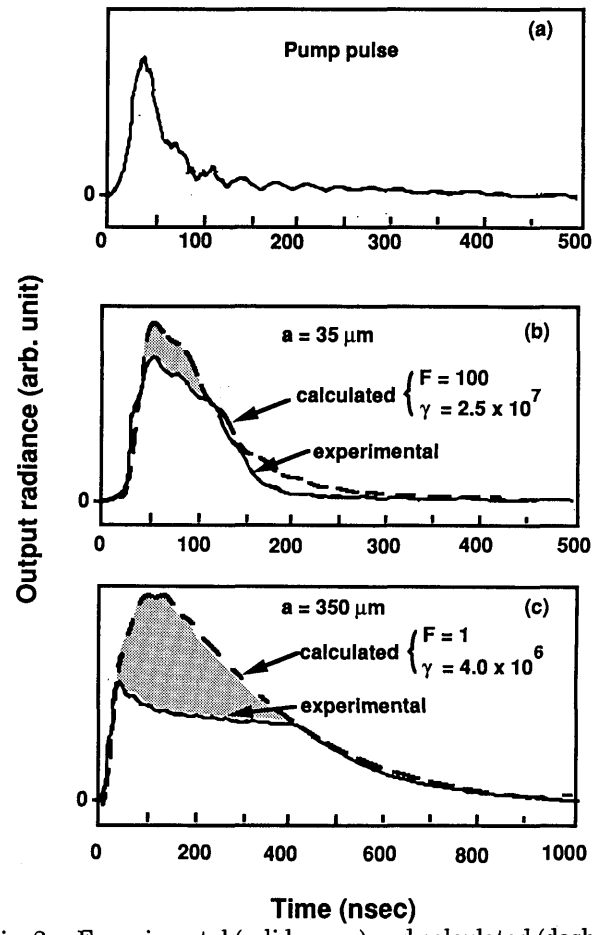

Fig. 2. Experimental (solid curve) and calculated (dashed curve) results of transient response of droplet dye lasers pumped by a Q-switched Nd:YAG laser. (a) The pumping laser pulse; (b) temporal response of $a = 35$ μm droplet dye laser; (c) temporal response of $a = 350$ μm droplet dye laser showing a long decay time of 250 ns, which corresponds to $Q = 8 \times 10^8$. The shaded areas show that the high-Q laser emission has been depleted by pumping stimulated Raman scattering of MtOH. Note that the time scale is changed in (c).

recorded by a storagescope (Iwatsu TS-8123). The pump pulse consisted of a leading high-intensity pulse [FWHM 25 ns; see Fig. 2(a)] followed by decaying relaxation oscillation subpulses of ≈400-ns duration. The emission from a single droplet was again collected at 90° with the same camera lens and color filter, without a spectrometer.

The output pulse of a 35-μm droplet reaches its maximum some 10 ns after the input pulse has reached its maximum [see the solid curve of Fig. 2(b)]. This delay is a result of slowly leaking energy stored in the high-Q modes of the cavity. The later, rippled-structure emission comes mostly from low-Q cavity modes, which have a fast response to input subpulse pumping and quickly leak out of the cavity. The long emission tail of the output pulse is a result of high-Q mode decay. The width of the output pulse width is ~100 ns.

Increasing the droplet size causes the MDR spacing to become closer and, therefore, the enhancement factor eventually to be reduced to unity. The experimental results from a large dye-doped droplet laser ($a \approx 350$ μm) demonstrate two features [see the solid curve of Fig. 2(c)]: (1) a fast response in the output signal resembling the input oscillation structures and (2) a slow exponential decay (with a cavity lifetime of 250 ns) after the pumping [see the tail portion of the output pulse with a time scale twice that of Figs. 2(a) and 2(b)]. This lifetime corresponds to a Q of 8×10^8 (corresponding leakage rate, $\gamma = 4 \times 10^6$ s^{-1}), which is the highest Q factor, to our knowledge, that has been measured in microdroplets.

To calculate the transient response, shown as dashed curves in Fig. 2, we used these parameters: $A = 2 \times 10^8$ s^{-1}/atom and $P = 1$ (normalized pumping rate,[6] which affects only the relative output radiance). We have represented the input pulse by a subpulse structure. For small droplets ($a = 35$ μm) we used $F = 100$ and let γ be an adjustable parameter. The best fit to the experimental curve was achieved with $\gamma = 2.5 \times 10^7$ s^{-1}, which corresponds to $Q = 1.27 \times 10^8$. This result is consistent with experimental observation.[7] The dashed curve of Fig. 2(b) showing a pulse width of 110 ns fits the experimental curve quite well.

For large droplets ($a = 350$ μm), the parameters $F = 1$ and $\gamma = 4 \times 10^6$ s^{-1} (obtained from experimental results) were used. By substituting all the parameters into the rate equations, we obtained the numerical curve shown as a dashed curve in Fig. 2(c). Note that the time scale is twice that of Fig. 2(b). Because of the high cavity Q and little (or no) enhancement of spontaneous emission (owing to the closely spaced MDR's), most of the pump energy is stored in molecules' population inversion or cavity modes, and thus the temporal ripple caused by subpulse pumping is smoothed out.

The differences between our experimental and numerical results for large droplets may come from the fact that the cavity Q factors and the enhancement factor F are quite different for various cavity modes. Low- and high-Q cavity modes can be excited simultaneously in these multimode microcavities. The higher the Q value, the longer the photons stay inside the cavity. Most of the energy of the excited-state molecules is coupled into and stored in high-Q modes during pumping through cavity-enhanced spontaneous emission. Low-Q modes cause a fast response and emit photons only during the pumping period. Therefore a small temporal ripple that is due to low-Q modes can be observed superimposed upon strong, structureless background emission during pumping; after pumping, only the photons in high-Q modes are left, so long decay times are observed. By comparing the calculated (dashed) and experimental (solid) curves, we find that some energy of high-Q laser emission has been removed during pumping [shaded area in Fig. 2(c)]. Recall that the high-Q laser emission photons can be depleted by pumping the stimulated Raman scattering[10] of MtOH at 720 nm, near the rim of the droplets. The same effect of laser emission depleted by pumping stimulated Raman scattering was also observed in a 35-μm-radius droplet [shaded area in Fig. 2(b)].

In summary, we have observed thresholdless behavior in microdroplet lasing cavities resulting from cavity-enhanced spontaneous emission. Because of the enhancement of spontaneous emission, the population of electrons in the excited states can be quickly transferred to stimulated emission photons so that the output response is instantaneous (no buildup time). However, the stimulated photon energy stored in high-Q cavity modes lengthens the output pulse width. Moreover, the high-Q laser emission can be depleted by pumping stimulated Raman scattering of host molecules. Further study of the transient responses of specific cavity modes with particular enhancement factors and Q values will clarify the influence of Q factors on laser emissions from droplets.

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