Simple arrangement for liquid-droplet experiments due to morphology-dependent resonances

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The arrangement for simple and inexpensive experiments based on the morphology-dependent resonances (MDR) is presented which allows a method of making liquid droplets with stable diameters and efficient light emission from the droplets. The lasing and Raman scattering characteristics due to MDR with this arrangement are also described.

I. INTRODUCTION

Recently there has been a good deal of interest in investigations of the light-scattering properties of dielectric spheres. Dielectric spheres possess natural modes of oscillation at characteristic frequencies corresponding to specific size-to-wavelength ratios defined as a size parameter. The advantages of the spherical resonator have been expected because of the very high quality factor (Q value) of the electromagnetic whispering-gallery mode (WGM)¹ or morphology-dependent resonances (MDR) in the dielectric sphere with micrometer-sized droplets^{2,3} or solid particles.^{4,5} In addition, the high Q value of spherical cavities has been shown to provide very low-threshold lasers.^{2,3}

A number of recent papers have reported lasing and/or stimulated Raman scattering (SRS) from dye-doped and/or nondoped microdroplets, using this high Q value due to MDR.⁶⁻¹¹

Previous investigations of dye-doped *solid* microsphere lasers due to WGM (or MDR)^{5,12-15} have demonstrated their usefulness for practical application and for theory/experimental comparisons in the WGM laser. The use of solid microspheres for dye lasers, however, should produce other problems such as destruction of the unique Q value in the sphere-surrounding media interface and/or essential features due to adsorption of dye molecules onto the sphere surface,⁹⁻¹¹ unless special attention is incorporated into experiments. These problems seem to be the similar ones in SRS experiments.

In this paper, we report the observation of the multi-order Raman spectra (MRS) and dye-fluorescence-assisted MRS from a liquid droplet, of glycerol and glycerol-nile blue (NB) dye mixture, due to MDRs, using low-power cw He-Ne laser pumping. A method of making droplets for the MRS due to MDRs and a variety of droplet sizes are presented.

II. APPARATUS

A. Dye laser and Raman generation system due to MDR

A schematic diagram of an arrangement for the present MDR experiments is shown in Fig. 1, which is similar to those described previously,¹²⁻¹⁵ however, the present experiment employs substituting alternately either a pulsed UV N_2 laser (337.1 nm) or a low-power cw He-Ne laser (632.8 nm), for the experiments of dye laser pumping or Raman scattering, respectively.

A droplet for lasing or Raman generation due to MDR (MDR-D) (of the order of the 10–100 μm diameter), which is suspended twinning round a fine nylon fiber (NF) (with the $\sim 5 \mu\text{m}$ diameter) strung horizontally (in just contrast to “on a glass plate” in previous studies¹²⁻¹⁵), is adjusted finely in the vicinity of the focal point of the spherical quartz lens (SQL) with 5 cm focal length, in the microscope field.

B. Construction of droplet generator

We use as the droplet generator (shown in Fig. 2) modifying a commercial atomizer with an ultrasonic vibrator component (VIB) (TDK TU-26X vibrator element with driving circuit board NB-513S-01). A sample liquid for droplets (SL) in the sample vessel is isolated from the water for cooling of the vibrator element.

Fine aerosol (of the order of a few micrometers) of sample liquids successively ejected from the modified atomizer, by introducing the air, spherically grows to proper-sized droplets (up to the order of a few hundred micrometers by a surface tension of sample material itself) around the horizontal fine nylon fiber.

C. Preparation of liquid droplets for MDR

The sample of glycerol droplet for Raman generation due to MDR was directly prepared by ejecting from the glycerol liquid in the SL vessel, whereas the dye-doped droplets for lasing were preliminarily prepared from dye-

FIG. 1. Experimental arrangement of laser/Raman scattering due to MDR. LASER: pulsed N_2 laser/cw He-Ne laser; SQL: spherical quartz lens; MDR-D: liquid droplet for MDR; NF: fine nylon fiber strung horizontally.

doped ethanol solutions ($\sim 10^{-5}$ – 10^{-3} M); however, the droplets changed their size too fast to enable one to run reliable experiments due to the high evaporation rate of ethanol in air. In order to increase droplet size stability, we then prepared dye-doped droplets from an ethanol-glycerol mixture with a composition ratio of $\sim 5:1$ – $3:1$, thereby the droplet size was held constant throughout experiments. Thus MDR-D was excited by the pulsed or cw laser.

III. EXPERIMENTS AND RESULTS

The present experiment employs substituting alternately either a pulsed UV N_2 laser (337.1 nm), which is the same as that in previous studies,^{12–15} having a 1.3 ns full width at half maximum (FWHM) and a 3 kW peak power, or a low-power cw He-Ne laser (632.8 nm), which has ~ 7 mW power output and 1.5 mrad beam divergence of TEM₀₀ mode, for the experiments of dye laser pumping or Raman scattering, respectively.

Spectra and spatial images of light emitted from the MDR-D droplet irradiated by the laser are analyzed with an optical multichannel analyzer (OMA; Hamamatsu Photonics PMA-10 system) and recorded with a color CCD camera/videotape recorder system, respectively, mounted on a microscope.

A. Spatial images of MDR-D

By finely adjusting the position of the dye-doped MDR-D in the vicinity of the focal point of the SQL (by N_2 laser pumping), it was observed that the rim of the dye-doped droplet above the threshold is brighter than the center, while the image of the emission pattern is rather

FIG. 2. Construction of liquid droplet generator. VIB: ultrasonic vibrator component; SL: sample liquid for droplets; FA: fine aerosol; NF: fine nylon fiber strung horizontally.

FIG. 3. Comparison of the spatial images inside the NA-F dye-doped droplet; above threshold in the upper and below threshold in the lower. Liquid sphere in (a) and solid sphere in (b).

uniform below the threshold. These spatial images of emission suggest features of lasing due to MDR with high Q values, which are similar to results in previous studies.^{12–15}

Figure 3(a) shows the comparison of these spatial images inside the dye-doped droplet; above the threshold in the upper and below the threshold in the lower, which are the cases containing the fluorescein (sodium salt) (NA-F) dye alone as the lasing medium (but, with $\sim 3:1$ ratios of dye-ethanol solution and glycerol). The exposure in the upper is about 1/200 times that in the lower. The upper in Fig. 3(a) (above threshold) clearly shows the brighter rim compared to the rather uniform pattern of lower emission intensity in the lower in Fig. 3(a) (below threshold). Similar figures, which are the cases of dye-doped solid polystyrene sphere, to those of the liquid sphere are shown in Fig. 3(b); the upper and lower correspond to above and below threshold, respectively.

Our findings in the experiments of the use of the liquid spheres (droplets), compared to those of the use of the solid spheres,^{12–15} are as follows: It is observed, from liquid MDR-D, that (1) intensity of emitted light (both fluorescence and lasing emission) is higher, (2) the spectral bandwidth of emitted light is wider [probably identical with the observation of (1)], and (3) lasing occurs in relatively wider dye-concentration regions. These findings suggest that more effective laser performance due to MDR is possible in liquid spheres than in solid spheres. We believe, at present, that this is due to the lower optical losses in liquid than those in solid, such as adsorption losses onto the solid spheres of dye molecules. It seems also from comparison of (a) (liquid) and (b) (solid) in Fig. 3 that the brighter rim of the dye-doped MDR-D above threshold in (a) is in

FIG. 4. Lasing spectra obtained from a NA-F dye-doped sphere with $\sim 60 \mu\text{m}$ diameter by N_2 laser pumping. Liquid or solid-polystyrene sphere corresponds to (a) or (b), respectively.

better order than that in (b). This seems due to better surface of sphere in liquid than in solid.

It was also observed in the Raman experiments of glycerol liquid droplets by He-Ne laser pumping that the rim of the MDR-D sphere shone in a deeper red than the center, although the Raman experiments of solid spheres were not carried out.

B. Spectral characteristics of emitted light from MDR-D

1. Dye laser spectra by N_2 laser pumping

Figure 4 shows the lasing spectra obtained from NA-F dye-doped droplet with $\sim 60 \mu\text{m}$ diameter by N_2 laser pumping, where the multiple structural peaks are seen in the green spectral region; the cases of the spheres of the liquid and solid polystyrene correspond to Figs. 4(a) and 4(b), respectively. It is found from this result that the liquid laser operates in the wider green spectral region than the solid one.

In successive experiments based on the above-mentioned findings, the dye-doped solutions were prepared by mixing rhodamine 101 (RH-101), fluorescein (sodium salt) (NA-F), and dimethyl-POPOP (D-POPOP) ethanol solutions (10^{-5} – 10^{-3} mol/l), for orange, green, and blue spectral regions (three colors), respectively. Figure 5 shows the lasing spectra obtained from three-dye-doped droplet with $\sim 10 \mu\text{m}$ diameter, where the multiple peaks are seen in the three-color regions, which exist around the center wavelengths of ~ 430 (blue), 530 (green), and 620

FIG. 5. Lasing spectra obtained from a three-dye-doped droplet of D-POPOP/NA-F/RH-101 with $\sim 10 \mu\text{m}$ diameter.

FIG. 6. Comparison of the Raman spectra obtained from a variety of diameters of the glycerol droplets. The background levels are arbitrarily shifted, for easy comparison.

nm (orange region), respectively. In the preliminary work using the three-dye-doped polystyrene sphere, we could not achieve a three-color laser using such as dyes.

2. Raman spectra by He-Ne laser pumping

Figure 6 shows a variety of the Raman spectra obtained from the glycerol droplets with about 15 – $120 \mu\text{m}$ diameters, where the background levels are arbitrarily shifted, for the sake of easy comparison. The spectra of (1)–(7) labeled in Fig. 6, which are (1) ~ 653 , (2) 661 , (3) 669 , (4) 674 , (5) 679 , (6) 687 , and (7) 697 nm, correspond to the well-known Raman shifts of 486.2 , 674.1^* , 850.4 , $\sim 960^*$, 1056.9 , $\sim 1250^*$, and 1464.5 cm^{-1} in glycerol, respectively (the shifts marked with asterisk correspond to the spectra with relatively lower intensity in the bulk glycerol). These assignments support that the spectra shown in Fig. 6 are due to the glycerol droplet. It is considered that the periodic spectra in the longer wavelength region occur resulting from the gain coupled between the Raman spectra of glycerol and MDR of droplets.

Figure 7 shows Raman spectra obtained from the droplet of glycerol containing nile blue (NB) fluorescent dye, with $\sim 70 \mu\text{m}$ diameter, where both the upper and lower traces are obtained from the excitation of the higher and lower pumping intensities, by neutral density filters, respectively. It is observed that the enhanced spectra corresponding to both glycerol with relatively larger Raman

FIG. 7. Enhanced Raman spectra obtained from a droplet of glycerol containing NB fluorescent dye, with $\sim 70 \mu\text{m}$ diameter.

shifts and NB with much longer wavelength region around 710 nm spectral peak appear. It is previously exhibited^{16,17} that the fluorescence from the NB dye exists over the spectral range of ~635–780 nm. It is considered that the spectra shown in Fig. 7 might be obtained with enhancements from a result of coupling between the NB fluorescence/glycerol Raman spectra and droplet gain due to MDR of the sphere.

IV. DISCUSSION

We have demonstrated a technique of making liquid droplets for simple and inexpensive experiments based on morphology-dependent resonances (MDR).

We have shown the fundamental characteristics of lasing and Raman scattering due to MDR, where the three-color laser from a three-dye mixture of D-POPOP/NA-F/RH-101 by pulsed N₂ laser pumping and Raman spectra from glycerol/glycerol-NB dye mixture by cw He-Ne laser pumping, have been exhibited using a liquid droplet, according to MDR.

The present technique of making a free-sized liquid sphere and fundamental characteristics of lasing or Raman scattering may be useful for various experiments of laser and SRS based on MDR.

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