Ultra-low-threshold europium chelate laser in morphology-dependent resonances

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We show ultra-low-threshold lasing due to morphology-dependent resonances from an europium (Eu)-chelate-doped microdroplet with $\sim 90 \mu\text{m}$ diameter. Spectra and spatial images between lasing and nonlasing are exhibited with comparison. Temporal responses are discussed from the viewpoint of the faster (direct) and slower (indirect) transitions in Eu chelates. © 1995 American Institute of Physics.

Several investigations of rare-earth chelate solutions have reported laser action, at about -100°C and below,¹ at room temperature (25°C),² with lasing thresholds of about 1000 J or above in europium (Eu) chelates, and with low thresholds of about 120 J in neodymium (Nd) systems.³ In such previous studies, special precautions for laser-cavity construction (including pumping volumes), solvent, lasant concentration, and operating temperature, have been exercised in the experiments, because the chelates are strongly absorptive¹ and oxidized to nonfluorescent products,³ for favorable fluorescent efficiency.

In any case in previous chelate lasers, the input energy for laser oscillation needs so high as that for ruby lasers, in consequence, means to avoid over heating were needed. On the other hand, recently there has been a great deal of interest in investigations of the light-scattering properties of dielectric spheres. The spherical resonator have been widely used because of the very high quality factor (Q value) of electromagnetic whispering-gallery mode (WGM)⁴ or morphology-dependent resonances (MDR) in the dielectric sphere with micrometer-sized droplets^{5,6} or solid particles.^{7,8} The high Q value of spherical cavities have been shown to significantly lower the threshold for lasers,^{5,6} which suggested the present investigation of spherical chelate lasers.

An arrangement for the experiments is similar to that described previously.⁹ A transversely excited atmospheric (TEA) ultraviolet (UV) N_2 laser (337 nm) is used to excite a spherical Eu-chelate-doped laser, focusing the pumping UV N_2 -laser beam with a spherical quartz lens (SQL) after beam shaping through a circular aperture. Spectra of light emitted from the laser or spatial images of light in the sphere are analyzed with an optical multichannel analyzer (OMA) or recorded with a color charge coupled device (CCD) camera/videotape recorder system, respectively, mounted on a microscope. A position of spherical laser droplets (with 10–150 μm diameter), which are suspended around a fine tungsten fiber ($\sim 5 \mu\text{m}$ diameter) strung horizontally,⁹ is adjusted

finely in the vicinity of the focal point of the SQL with 5 cm focal length, in the microscope field.

Fine aerosol particles with a 2–3 μm diameter that contain Eu-chelate solutions are successively ejected from a modified atomizer and grow to proper-sized droplets (of the order of 150 μm diameter) around the horizontal fiber. The Eu-chelate solutions were prepared by dissolving $\text{Eu}(\text{DBM})_3(\text{Phen})$ (DBM=dibenzoylmethane, Phen=phenanthroline) in ethanol, and by a mixture of glycerol of them for proper viscosity for making droplets (volume ratio of ethanol:glycerol $\approx$ 3:1). The Eu-doped droplet was excited by the UV N_2 laser, having a 1.3 ns full width at half-maximum (FWHM) and 3 kW peak power ($\sim 4 \mu\text{J}$ pulse energy) at 337 nm.

By finely adjusting the position of the Eu-doped droplet in the vicinity of the focal point of the SQL, it was observed that the rim of the Eu-doped droplet above threshold is brighter than the center, while the image of the emission pattern is rather uniform below threshold. Figure 1 shows the comparison of the spatial images inside the Eu-doped droplet with $\sim 150 \mu\text{m}$ diameter (a) below and (b) above lasing threshold; exposure of (b) is about 1/20 of (a). These spatial images of the emission suggest the features of MDR lasing

FIG. 1. Comparison of the spatial images inside the Eu-doped droplet: (a) below and (b) above lasing threshold; exposure of (b) is about 1/20 of (a).

FIG. 2. Single-shot spectrum obtained from the 10^{-3} mol/l Eu-doped droplet. $\sim 150 \mu\text{m}$ droplet diameter.

with high Q values, which is similar to that in previous studies.⁹

It is known that the Eu-doped system operates at 613 and 615 nm of the Eu-benzoylacetonate (EuB_3),¹ and at 612 nm of Eu-benzoyl-trifluoroacetate [$\text{Eu}(\text{BTF}_4)\text{P}$],² arising from an electric dipole transition between the 5D_0 and 7F_2 levels of Eu^{3+} . In our droplet experiments of the Eu chelates, the higher concentrations up to 10^{-2} mol/l exhibited the higher output intensities (in much higher concentrations beyond the 10^{-2} mol/l, uniform radiation was not obtained in the droplets). The single-shot spectrum obtained from the 10^{-3} mol/l Eu-doped droplet with $\sim 150 \mu\text{m}$ diameter (at $\sim 4 \mu\text{J}$ pumping energy) is shown in Fig. 2, where the additional spectra¹⁰ corresponding to $^5D_0-^7F_0$ (near 580 nm) and $^5D_0-^7F_1$ (590–600 nm) transitions of Eu^{3+} , except for the transition on $^5D_0-^7F_2$ (600–625 nm) previously reported as lasing transition;^{1,2} however, in this concentration (and according to the larger diameter beyond limitation of the OMA resolution, which is approximately 1 nm), we could not confirm to be the laser operation due to MDR. The

FIG. 3. Single-shot MDR-lasing spectrum obtained from the Eu-doped droplet with 10^{-2} mol/l; $\sim 90 \mu\text{m}$ droplet diameter.

FIG. 4. Pulse wave forms of the first (F) and second pulses (S); the second pulse appears or disappears at high (a) or low pumping intensities (b). Horizontal scale is 20 ns/div. The inset shows the dependence on pumping intensity (I_P) of the emission of the second pulse (I_S).

similar spectrum obtained from the Eu-doped droplet with 10^{-2} mol/l and $\sim 90 \mu\text{m}$ diameter, by single shot (at $\sim 4 \mu\text{J}$ pumping energy), is shown in Fig. 3, where the periodic structure characterizing MDR lasing is observed in the $^5D_0-^7F_2$ transition. The observed mode spacing (~ 1 nm) is nearly the same value as expected from mode-separation formula¹¹ due to MDR of spheres, thereby we can confirm that the present Eu-doped droplet operates to be lasing due to MDR. Expected value of mode separation is obtained when the refractive index is ~ 1.4 , by averaging the respective refractive indices by ethanol and glycerol with the mixing ratio of $\sim 3:1$. Furthermore, we examined the lasing threshold from time-varying measurements between pump (337 nm) and emission (613 nm) pulses, by a biplanar phototube (or a photomultiplier tube) and an oscilloscope system. The emission pulse occurred to be consisting of two components (double pulses) at least near 613 nm; one with a few nanosecond delay from the pump pulse (the first pulse) and the other with 100–120 ns delay from the first pulse (the second pulse). We consider that the first (the faster) and second (the slower) pulses correspond to the transition to the upper laser level of Eu ion from excited singlet states of ligand and from excited triplet states via energy transfer within ligand molecules,¹² where the former and latter correspond to the direct and indirect transitions, respectively. The second pulse

corresponding to the slower buildup from the pump exhibited the distinct threshold of lasing, which appears [Fig. 4(a)] or disappears [Fig. 4(b)] at high or low pumping intensities, respectively; however, the lasing threshold of the first pulse remains indistinct to the present. The time response of the N_2 laser pulse (having a 1.3 nm FWHM with biplanar phototube and 275 MHz real-time oscilloscope system) exhibited ~ 5 ns pulse width (FWHM) in the same measuring system (with photomultiplier tube and 100 MHz storage-oscilloscope system) as the 613 nm pulse. From these results, in the second pulse at least, we believe the significantly lowered laser operation (with the threshold of a μJ order) of Eu chelate lasers. Investigations on the lasing threshold of the first pulse are needed for more detailed laser performance. The lasing characteristics with low threshold presented here seem to be due to the high Q value based on MDRs of the spheres. We hope the present technique may be applied to other spherical chelate lasers.

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