FLUORESCENCE SPECTRA OF LIQUID DROPLETS CONTAINING A DYE UNDER INTENSE LASER PUMPING

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The fluorescence spectra of the Rhodamine 6G solution were analyzed. Significant difference was revealed between the spectra recorded in a cell and in droplets of millimeter and micron size. The maximum of the fluorescence band recorded in a droplet shifts to shorter waves by 9 nm *with respect to the spectrum recorded in the cell at the power density of laser pumping of* 310 MW/cm^2. *Besides, the relative intensity of the long-wave wing of the fluorescence spectrum decreases. Similar shift of the fluorescence maximum was also detected during the exposure of a jet of micron-size droplets. In the cell containing dye solution, during the exposure in the same range of the pump power density, the shift of the fluorescence spectral maximum was markedly smaller (*4 nm*).*

The possibilities of using liquid spherical particles as new miniature laser facilities and a particular kind of optical devices suitable for solution of the problems in aerosol physics and spectroscopy have been actively studied during the last decade. Optical phenomena in such particles attract considerable interest, because they can act as spherical microresonators with a high Q-factor. This fact fundamentally determines the particular character of nonlinear optical effects in such particles.[1,2]

A spherical particle is capable of concentrating the incident electromagnetic radiation within its volume. This capability, along with the presence of free resonance modes in a particle, results in a substantial local amplification of the amplitude of an optical field in the near-surface layer of a particle. In that case, the amplitude is maximum near the irradiated and dark surfaces of the particle along the direction of the incident wave propagation. The amplification of the internal field in such zones can achieve $10^2 \ldots 10^6$ (Ref. 3) at a certain ratio between the particle radius and the wavelength of incident radiation. This phenomenon significantly lowers the thresholds of a number of nonlinear optical effects, such as SRS, SBS, optical breakdown, optical bistability, and others. This opens further possibilities for studying effects of nonlinear interaction of radiation with the matter. However, this phenomenon manifests itself only at high intensity of laser pumping.

When exposed to a high-power focused laser radiation, dye solutions were found to demonstrate the abnormally high growth of the fluorescence intensity with the increasing intensity of the exciting radiation.[4] The spectrum of dye fluorescence observed in Ref. 4 shifted to shorter waves (by about 3 nm) as the power density of laser pumping changed from 30 kW/cm^2 to 300 MW/cm^2. Such a fluorescence has been called the incoherent superluminescence in Ref. 4.

Similar anomalies in the behavior of luminescence have also been observed in the metal atomic vapor.[5] In Ref. 6, it was proposed that the anomalous behavior of optical centers in the laser pump field is not due to the peculiarities in the interaction of light with complex molecules. Note that, in spite of different hypotheses proposed in Refs. 5–7, the phenomenon of incoherent superluminescence is yet to be explained.

The effect of anomalous fluorescence of a dye is observed in sufficiently strong laser fields. This explains our interest in studying the peculiarities in the fluorescence taking place in spherical particles, because the above-mentioned zones of enhanced power density of the internal optical field exist in such particles. The paper is devoted to consideration of the experimental results on these peculiarities in the fluorescence.

In the experiments, we used pulses of a Nd:YAG laser (the second harmonic wavelength of 532 nm and the pulse duration of 10 ns) to excite the dye fluorescence. Pulse energy was measured with an IMO–2N meter of the average power and energy; pulse duration was measured with a FEK–19KM photocell and a S7–19 oscilloscope. When necessary, laser radiation was attenuated with a Glan polarizing prism and neutral density filters. The power density P of a focused laser radiation varied from 30 kW/cm^2 to 310 MW/cm^2. To determine the spectral characteristics of the dye fluorescence, we used the monochromator built around a DFS–452 spectrograph with the spectral width of the instrumental function of 2 Å.

The focused laser radiation was directed onto a spherical droplet 1.5 mm in radius. The droplet consisted of the Rhodamine 6G solution in dibutylphtalate (DBPh). The concentration of a dye was 10^{-4} mol/liter. The droplet was placed into the rectangular quartz cell filled with the distilled water. To do this, we used the method proposed in Ref. 8 for making liquid optical resonators with the use of interfaces of immersion liquids.

Figure 1 demonstrates the fluorescence spectra of the Rhodamine 6G solution in DBPh (the dye concentration is 10^{-4} mol/liter) in the liquid phase (in the cell). These spectra were recorded at the power density of laser pumping of 1.2 (curve *1*) and 310 MW/cm^2 (curve *2*). The fluorescence spectra recorded at the pump power density of 30 kW/cm^2 and 1.2 MW/cm^2 almost coincide. To prevent lasing in the resonator formed by the cell walls, the dye solution was placed in the cell with non-parallel walls. Fluorescence was recorded in the direction normal to the direction of propagation of the exciting laser beam. As seen from Fig. 1, the maximum of the fluorescence spectrum obtained at laser pumping of 310 MW/cm^2 is shifted to shorter waves. The observed spectral shift (4 nm) is close to similar shift obtained in Ref. 4 at the pump power of the same level.

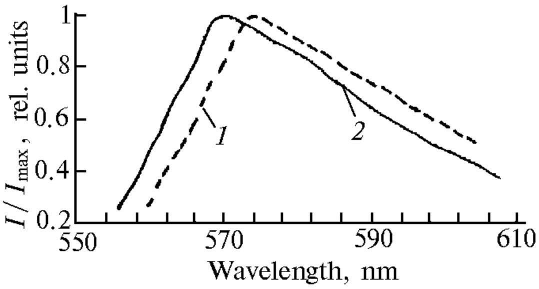

FIG. 1.

Figure 2 shows the fluorescence spectra of a spherical droplet consisting of the Rhodamine 6G solution in DBPh of the same concentration. As seen, the maximum of the fluorescence spectrum recorded at the pump power density of 310 MW/cm^2 (curve *1*) in a droplet is shifted by 9 nm with respect to the position of the maximum corresponding to the pump power of 1.2 MW/cm^2 (curve *2*). The latter, in its turn, is shifted with respect to the maximum of spectrum of the same dye solution at the same pump power, but in the cell (curve *3*). It is also seen that the shape of the fluorescence spectra in a droplet is markedly more narrow as compared to the spectrum of dye solution in the cell.

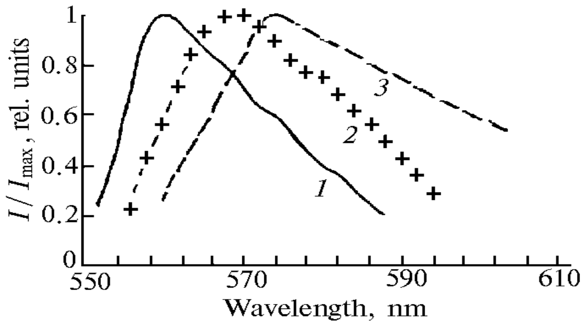

FIG. 2.

Figure 3 shows the dependence of the spectral shift on the pump power.

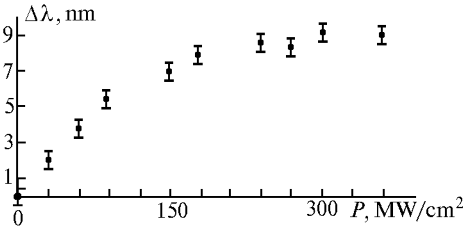

FIG. 3.

The Table I presents the values of the spectral shifts of the fluorescence spectra in a droplet for different concentrations of the Rhodamine 6G in DBPh. As seen, the spectral shift increases with the increasing dye concentration. This fact is in a good qualitative agreement with the experimental data obtained in Ref. 9. It was shown in Ref. 9 that the threshold power of the effect of incoherent superluminescence increases with decreasing dye concentration in the cell.

TABLE I.

Dye concentration in a droplet, mol/liter	Spectral shift, nm
10^{-6}	0
10^{-5}	4
10^{-4}	9

The next series of experiments was aimed at recording the fluorescence spectra of an ensemble of spherical droplets. The droplets consisted of the Rhodamine 6G solution in ethanol and had a micron size. The recording technique and instrumentation were the same as in the above-described experiments. The laser radiation was focused onto a jet of a polydisperse aerosol contained in the dye. The jet was generated by the "Aerozol' U–1B aerosol generator. Droplet radii in the jet varied from 1 to 35 μm, and the particle size-distribution had maximum at about 5 μm. The dye concentration in droplets was $5\cdot10^{-4}$ mol/liter.

The fluorescence spectra of the Rhodamine 6G solution in the cell and the same solution in micron droplets are shown in Fig. 4 for the pump power of 310 MW/cm^2. As seen from Fig. 4, the spectral maximum of the fluorescence from the aerosol jet also shifts toward shorter waves with respect to the spectral maximum recorded in the cell.

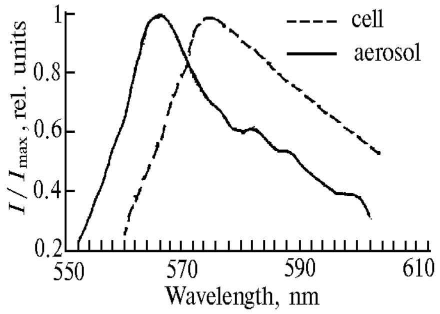

FIG. 4.

As a result, we have revealed a significant difference between the fluorescence spectra of a millimeter-size spherical droplet of the Rhodamine 6G solution in DBPh and the same solution placed in the cell at the intense laser pumping. As the power density of the exciting radiation varies from 1.2 to 310 MW/cm^2, the spectral maximum of dye fluorescence in a droplet shifts toward shorter waves. This shift (~ 9 nm) significantly increases the similar shift obtained in the cell (4 nm) under the same conditions. Besides, the increase in the pumping power initiates lowering of the relative intensity of the long-wave wing of the fluorescence spectrum. Almost the same shift (~ 3 nm) of the fluorescence spectral maximum of the Rhodamine 6G solution in the cell was first observed in Ref. 4 at the pumping power of same level. This shift characterizes the phenomenon referred to as “incoherent superluminescenceB Besides, we have observed similar shift of the spectral maximum of fluorescence during the exposure of micron-size droplets containing a dye.

Indications of the incoherent superluminescence observed in a droplet and a micron aerosol are far more pronounced as compared to similar phenomena observed in dye solutions in the liquid phase. This is apparently caused by a significant amplification of the internal optical field occurring in droplets.

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