

## AMPLIFICATION OF SRS RADIATION IN A POLYDISPERSION OF DROPLETS WITH A LASER DYE UNDER CONDITIONS OF STIMULATED FLUORESCENCE OF THE DYE

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*Amplification of stimulated Raman scattering (SRS) radiation of ethanol in a polydispersion of microdroplets of ethanol solution of Rhodamine C is obtained experimentally when irradiating the droplets with laser pulses of 0.532  $\mu\text{m}$  wavelength.*

The microdroplet may be a spherical resonator whose natural modes are defined by its radius, radiation wavelength, and complex refractive index of the droplet substance.<sup>1</sup> If a droplet is affected by pumping radiation and one of the wavelengths of the re-emission (for instance, Raman scattering radiation or molecular fluorescence of the dye contained in the droplet) coincides with a natural mode of the droplet resonator, the positive feedback can lead to radiation amplification at this wavelength and appearance of nonlinear effects such as stimulated Raman scattering (SRS) or stimulated fluorescence of the dye (lasing).<sup>2,3</sup>

In Ref. 2, this is called the "output resonance" condition. The physical mechanism of SRS appearance in a spherical droplet is described comprehensively (see, for instance, the review in Ref. 4).

If molecules of optically active dyes are added to a transparent drop, the latter begins to fluoresce actively at irradiating by laser pumping pulses, and when reaching of threshold conditions in the droplet resonator there can occur generation (lasing).<sup>3</sup> Since spontaneous fluorescence spectrum of a dye is rather wide, it may cover many different the natural modes of the drop. The spectral pattern of lasing for a droplet with dye is of a spike structure, and the spectral position of spikes, the distance between them, and the number of spikes within the fluorescence spectrum of the dye are determined by droplet dimensions, physicochemical properties of the droplet substance, and power of the laser pump, that determines the order of natural modes excited in the resonator.

As reported in Refs. 5 and 6, joint manifestation of lasing and SRS effects was observed at irradiating a drop containing the ethanol solution of Rhodamine 6G. The amplification of SRS radiation in ethanol discovered in a droplet with a dye<sup>5,6</sup> takes place when one of the Raman scattering lines of ethanol lies within the fluorescence spectrum of the dye and the conditions of "output resonance" are fulfilled. The fulfillment of all the conditions was achieved by a proper selection of the wavelength of a laser pump. Amplification of SRS

in a droplet was accompanied by partial suppression of the lasing spectral spikes.<sup>6</sup>

In this paper, we present some results of our observations of the ethanol SRS in a system of fluorescent microdroplets of random size. Moreover, fulfillment of the "output resonance" condition for the SRS also occurred randomly.

It is well-known that one of the intense bands of the Raman scattering spectrum of valence CH-oscillations for ethanol molecules can be seen in the vicinity of the wavelength  $\lambda_k = 0.631 \mu\text{m}$  at excitation by radiation with the wavelength  $\lambda = 0.532 \mu\text{m}$  (two other bands of Raman scattering lie in the vicinity of 0.628 and 0.632  $\mu\text{m}$  wavelengths).<sup>7</sup> So, to observe the joint action of ethanol SRS at the wavelength 0.631  $\mu\text{m}$  and dye lasing in the droplet medium, we used a sprayed solution of Rhodamine C whose fluorescence spectrum lies from 0.570 to 0.650  $\mu\text{m}$  with a maximum at  $\lambda = 0.595 \mu\text{m}$  at excitation by laser radiation with the wavelength  $\lambda = 0.532 \mu\text{m}$ .

In the experiment, we used a Nd:YAG laser operating in the Q-switch mode with an active element of 100 mm length and 5 mm diameter. Laser pulses of 10 ns duration (FWHM) were focused in a cloud of a polydisperse aerosol after frequency doubling. The cloud was formed with the aerosol generator "Aerosol' Y-1" and consisted of droplets of Rhodamine C ethanol solution. The droplet radii were distributed in the aerosol cloud from 1 to 35  $\mu\text{m}$  with a maximum near 5  $\mu\text{m}$ . Radiant flux density of the focused laser radiation reached 800 MW/cm<sup>2</sup>. Pulse energy and duration of pulses were measured with an IMO-2N, FEK-19KM and an oscilloscope S7-19. A monochromator with the spectral width of the instrumental function 10 Å and a set of optical filters were used as filters to suppress the elastic scattering from the droplets. The degree of elastic scattering suppression (at 0.630  $\mu\text{m}$  wavelength) was no less than 10<sup>-7</sup>. The spectral pattern of the droplet emission was recorded with the same monochromator with step of 10 Å. Detection of signals was performed

using FEU-79 PMT with a voltage divider selected individually to improve temporal resolution up to 6 ns.

Red emission from droplets was observed visually (through optical filters) at irradiating the cloud of polydisperse pure ethanol droplets with laser pulses of the radiant flux density of 600–800 MW/cm<sup>2</sup>. Figure 1 presents spectral distribution of the averaged signal amplitudes of pure ethanol droplet emission. Every point of the curve is the mean of  $N = 150$  values. One can see that a distinct maximum observed against a background at the wavelength  $\lambda_k \cong 0.631 \mu\text{m}$ ; moreover, the variance of the signal amplitude at this wavelength is higher as compared to that observed in other portions of the spectral range considered. This fact can be explained in the following way. When droplets with dimensions corresponding to the "output resonance" condition are within a laser beam, stimulated Raman scattering with a signal exceeding the background illumination appears in the droplets. The increase of signal amplitude variance at the wavelength of ethanol Raman scattering reflects the random nature of the droplets formation with dimensions satisfying the resonant conditions.

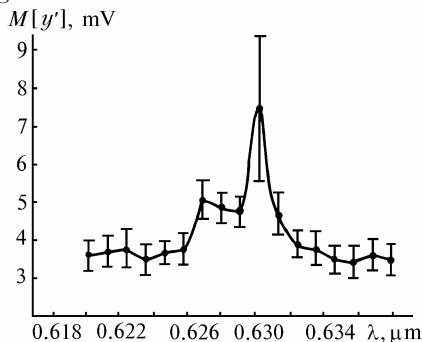


FIG. 1. Spectral distribution of the mean amplitude value  $y'$  of the emission signals from ethanol droplets.

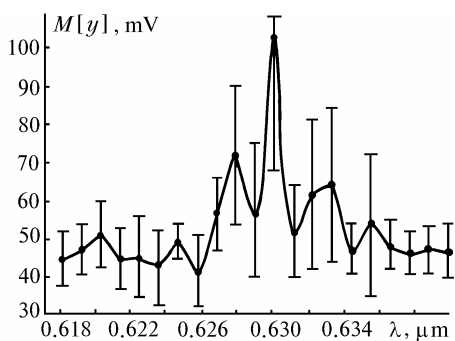


FIG. 2. Spectral distribution of the mean amplitude value  $y$  of fluorescence signals from droplets with Rhodamine C,  $\psi(\lambda = 0.631 \mu\text{m}) = 1$ .

Figure 2 presents a portion of the fluorescence spectrum for a polydisperse system of droplets of Rhodamine C ethanol solution with the dye concentration of  $2 \cdot 10^{-4} \text{g/m}$ . Every point of the curve in Fig. 2 is the average value over the sample size  $N = 150$ . Fluorescence was induced with laser pulses of the radiant flux density of  $P = 800 \text{ MW/cm}^2$ .

Analyzing the papers 2 to 6, one can conclude that lasing of the polydisperse system of droplets is a complicated spiking structure, and the amplitude of spikes and their spectral position and the distance between them over the spectrum change randomly from pulse to pulse, because the number and size of droplets irradiated at each moment change randomly. In Ref. 8 we have demonstrated that the variance and distribution of detected signal values can serve as statistical parameters of fluorescence signals in detecting the lasing effect in this case.

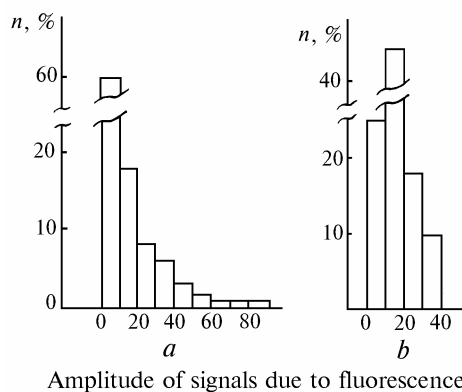


FIG. 3. Histograms of fluorescence signals ( $\lambda = 0.631 \mu\text{m}$ ) from ethanol droplets with Rhodamine C at different radiant flux density of laser pumping  $P$  ( $800 \text{ MW/cm}^2$ , stimulated fluorescence,  $a$ ;  $P = 1.6 \text{ MW/cm}^2$ , spontaneous fluorescence,  $b$ ).

Figure 3 presents histograms of the fluorescence signals detected at the wavelength  $\lambda = 0.621 \mu\text{m}$  for different pumping flux density  $P$ :  $P = 800 \text{ MW/cm}^2$  (Fig. 3a), and  $P = 1.6 \text{ MW/cm}^2$  (Fig. 3b). As was shown in Ref. 8, the presence of the "heavy" right side in the distribution in Fig. 3a evidences the fact that stimulated fluorescence exists in the droplet medium. The values of amplitude variance of fluorescence signals and elastic scattering from ethanol droplets are presented in the Table.

Radiation	Droplet fluorescence, $\lambda = 0.621 \mu\text{m}$		Elastic scattering, $\lambda = 0.532 \mu\text{m}$
Radiant flux density of pumping, $\text{MW/cm}^2$	800	1.6	800
Signal variance	1.68	0.12	0.09

According to Ref. 8, a sharp increase in the fluorescence signal variance at high radiant flux

density of laser pumping is also indicative of the lasing effect.

Let us have a look at Fig. 2. A well pronounced maximum is seen at the wavelength  $\lambda_k \cong 0.631 \mu\text{m}$ . The presence of a maximum at the wavelength corresponding to the line from Raman scattering of ethanol qualitatively agrees with the results from Refs. 5 and 6 on observation of SRS and lasing in a single droplet with a dye. For a comparison, we present the spectrogram obtained in Ref. 6 (Fig. 4).

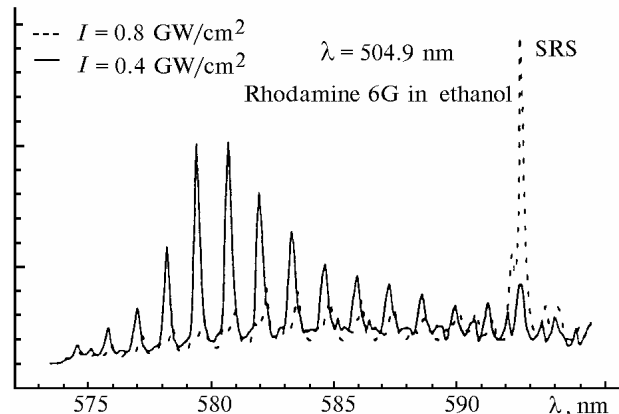


FIG. 4. SRS and lasing spectrograms from a single ethanol droplet with Rhodamine 6G at different radiant flux density of pumping  $I^6$ .

In measurements of the droplet emission signals at the wavelength corresponding to Raman scattering of ethanol, spectral spikes or fragments of spikes due to stimulated fluorescence from droplets whose dimensions do not satisfy the “output resonance” condition for Raman scattering wavelength of ethanol can fall within the spectral interval determined by the instrumental function of the device, together with the spike of amplified SRS radiation. In other words, every act of measurements detects an amplified SRS signal against the noise background which is a superposition of spikes and/or spikes’ fragments of stimulated fluorescence of the droplets for which resonant conditions do not hold at the Raman scattering wavelength. Such a noise (in fact, lasing signals) will be detected at each point of the spectral interval not connected with the Raman scattering wavelength. The value of noise is determined by spectral behavior of the dye fluorescence at every point of the spectral range.

Thus, the signal observed at the wavelength  $\lambda$  in a single test can be defined as follows

$$y(\lambda) = \psi(\lambda) [n^*(\lambda) + CK(\lambda - \lambda_k)], \quad (1)$$

where  $\psi(\lambda)$  is the function describing the dye fluorescence (in our case,  $\psi(\lambda)$  is normalized by the value  $\psi(\lambda_k)$ ,  $\lambda_k = 0.631 \mu\text{m}$ );  $n^*(\lambda)$  is the equivalent noise induced by spike signal not connected with Raman scattering wavelength  $\lambda_k$ ;  $K(\lambda - \lambda_k)$  is the bell-shaped signal defined on the interval  $(\lambda_k - \Delta, \lambda_k + \Delta)$  reaching its maximum  $K(0) = 1$  at

the point  $\lambda_k$ ;  $\Delta$  is the spectral interval defined by the instrumental function of the device;  $C$  is the random amplitude of the spike defined on the interval  $(\lambda_k - \Delta, \lambda_k + \Delta)$ .

Let us then estimate the mean value and variance of the amplitude  $C$  based on the results of independent experiments for different values of  $\lambda$ .

Within the scope of the model (1), the expectation  $M[y(\lambda)]$  of  $y(\lambda)$  is

$$M[y(\lambda)] = \psi(\lambda) [M[n^*(\lambda)] + K(\lambda - \lambda_k) M[C]]. \quad (2)$$

Assuming that the noise  $n^*(\lambda)$  and the random amplitude  $C$  are independent, the variance of  $y(\lambda)$  is written in the form

$$D[y(\lambda)] = \psi^2(\lambda) [D[n^*(\lambda)] + K^2(\lambda - \lambda_k) D[C]], \quad (3)$$

where  $D[n^*(\lambda)]$  and  $D[C]$  are the variance of the noise and amplitude  $C$ , respectively.

Using Eqs. (2) and (3), it is easy to write the moments,  $M[C]$  and  $D[C]$

$$M[C] = [M[y(\lambda)]/\psi(\lambda) - M[n^*(\lambda)]]/K(\lambda - \lambda_k); \quad (4)$$

$$D[C] = [D[y(\lambda)]/\psi^2(\lambda) - D[n^*(\lambda)]]/K^2(\lambda - \lambda_k). \quad (5)$$

Thus, to calculate  $M[C]$  and  $D[C]$ , it is necessary to know the mean value and the variance of the signal  $y$  and the noise  $n^*(\lambda)$ ; beyond the interval  $(\lambda_k - \Delta, \lambda_k + \Delta)$ , these moments are not defined.

For  $\lambda = \lambda_k$ , i.e., at the Raman scattering wavelength, we have

$$M[C] = M[y(\lambda_k)]/\psi(\lambda_k) - M[n^*(\lambda_k)]; \quad (6)$$

$$D[C] = D[y(\lambda_k)]/\psi^2(\lambda_k) - D[n^*(\lambda_k)]. \quad (7)$$

And if  $\lambda_1 \notin (\lambda_k - \Delta, \lambda_k + \Delta)$ , it follows from Eqs. (2) and (3) that (here  $K(\lambda_1 - \lambda_k) = 0$ )

$$M[y(\lambda_1)] = \psi(\lambda_1) M[n^*(\lambda_1)]; \quad (8)$$

$$D[y(\lambda_1)] = \psi^2(\lambda_1) D[n^*(\lambda_1)]. \quad (9)$$

If the noise  $n^*(\lambda)$  is the steady-state process in the broad sense, i. e., in our case,

$$M[n^*(\lambda_k)] = M[n^*(\lambda_1)]; \quad D[n^*(\lambda_k)] = D[n^*(\lambda_1)], \quad (10)$$

the expressions (6) and (7) can be written in the form

$$M[C] = M[y(\lambda_k)]/\psi(\lambda_k) - M[y(\lambda_1)]/\psi(\lambda_1); \quad (11)$$

$$D[C] = D[y(\lambda_k)]/\psi^2(\lambda_k) - D[y(\lambda_1)]/\psi^2(\lambda_1). \quad (12)$$

In the latter expressions, the moments of the random value  $C$  are presented by the moments of signals observed at the wavelengths  $\lambda_k$  and  $\lambda_1$  and can be estimated in a standard way.

The mean value and variance can be estimated similarly for the case of measurements of SRS signals from pure alcohol.

Let us define the amplification SRS signal from ethanol in the system of fluorescing droplets in the lasing regime at the wavelength  $\lambda_k \cong 0.631 \mu\text{m}$  (here  $\psi(\lambda_k) = 1$ ) as follows:

$$G = M[C]/M[C'] , \quad (13)$$

where  $M(C')$  is the expectation of Raman scattering signal amplitude from droplets of pure alcohol.

In our case, from Figs. 1 and 2 we obtain

$$G = 12.8 \pm 2.7.$$

Thus we have demonstrated that significant (by an order of magnitude) amplification of SRS radiation is feasible in a polydisperse system of microdroplets with a dye as a result of interaction between SRS radiation in the droplets and the dye fluorescing in the lasing mode.

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