ANGULAR CHARACTERISTICS OF THE SRS FIELD FROM TRANSPARENT PARTICLES

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The SRS effect in spherical transparent droplets is considered. The results are compared with the experimental data available. The angular structure of the SRS in the far zone was numerically modeled for a stationary distribution of the SRS field in a droplet. It was shown that angular distribution of Raman scattering has a maximum in the backward and forward directions with respect to the direction of incident radiation and a minimum in the direction normal to it. The SRS directional pattern is shown to be more uniform than that of the elastic scattering. This is explained by the fact that no field component at the Raman shifted frequency is diffracted on the particle contour. The angular structure of the SRS if supported by one resonance mode of a droplet is symmetric with the period multiple to π/2. At multimode regime of the SRS generation, this symmetry is not observed.

Nonlinear optical effects in transparent droplets of the micron size (SRS, SBS, stimulated fluorescence) are now the subject of numerous investigations. The possibility for these effects to occur in small volumes, such as droplets, is caused by the fact that a spherical particle can concentrate, in its volume, the energy of incident electromagnetic field. This property, in its turn, results from presence, in a droplet, of its own high-Q ($Q \sim 10^8$) resonance modes with the spatial structure similar to the mode structure of the “whispering galleryB type in acoustics. From this point of view, a droplet can be considered as an optical cavity, in which the spherical surface plays the role of mirrors. The spectrum of free modes of such a cavity is governed, as known, by the diffraction parameter of a droplet and the refractive index of the liquid.

The physical mechanism of the SRS effect in a micron spherical particle, whose diffraction parameter is far greater than unity, is rather clear now. As known, in such particles the incident field is focused mainly by the front drop surface onto the area being close to its rear surface. Spontaneous Raman scattering occurs everywhere in a droplet, but the strongest amplification is observed just in this area of the focal volume. A portion of waves from spontaneous Raman scattering leaves the droplet, another portion propagates along the spherical surface around a circle due to total internal reflection. When propagating along the particle surface, these waves undergo both amplification at the expense of continuous energy exchange with the pumping field (the laser radiation affecting the droplet) and attenuation due to energy absorption by the droplet substance and radiation losses through the surface. If any frequency of the Raman noise spectrum falls in resonance with the droplet free vibrational modes, the amplification dominates over the absorption, and stimulated scattering occurs. In the experiments SRS is observed, as a rule, as two glowing spots or two arcs on the droplet surface in the forward and backward directions along the droplet main diameter. It is important to consider the angular structure of the SRS field far from a droplet and to study its dependence on the spatial structure of SRS field inside the droplet. In this paper we present the results of theoretical study of the angular characteristics of the SRS field in a spherical droplet exposed to laser radiation.

The mathematical statement of the problem on the emission of a spherical volume with a preset spatial distribution of the electromagnetic field, corresponding to a resonance mode, into the droplet’s environment is based on the system of inhomogeneous Maxwell equations for the field components (fields are believed quasimonochromatic):

$$\begin{aligned}&\mathrm{rot}\,\mathbf{H}_s(\mathbf{r},t) - i\,\omega_s\,\varepsilon\,\mathbf{E}_s(\mathbf{r},t) = \mathbf{J}_a;\ \mathrm{div}\,\mathbf{H}_s(\mathbf{r},t) = 0;\\ &\mathrm{rot}\,\mathbf{E}_s(\mathbf{r},t) + i\,\omega_s\,\varepsilon\,\mathbf{H}_s(\mathbf{r},t) = 0;\ \mathrm{div}\,\mathbf{E}_s(\mathbf{r},t) = 0,\end{aligned} \qquad (1)$$

where ω_s is the Stokes frequency, ε is the dielectric constant, $\mathbf{J}_a = \varepsilon_a\,\partial\mathbf{E}_a(\mathbf{r},t)/\partial t$ is the density of polarization currents induced by the internal field, $\mathbf{E}_a(\mathbf{r},t)$ is the vector of the Stokes electric field strength in the droplet, $\mathbf{r}$ is the radius-vector. Once the vector potential $\mathbf{A}(\mathbf{r},t)$ is introduced in the standard way

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$$\mathbf{H}_\mathrm{s}(\mathbf{r},\, t) = \mathrm{rot}\, \mathbf{A}(\mathbf{r},\, t);\; \mathbf{E}_\mathrm{s}(\mathbf{r},\, t) = -\,\partial \mathbf{A}(\mathbf{r},\, t)/\partial t,$$

the system (1) transforms into the Helmholtz equation

$$\nabla^2\, \mathbf{A}(\mathbf{r},\, t) + (\omega_\mathrm{s}^2/\varepsilon)\, \mathbf{A}(\mathbf{r},\, t) = -\, \mathbf{J}_a(\mathbf{r},\, t) \qquad (2)$$

under the condition that div $\mathbf{A}(\mathbf{r},\, t) = 0$.

Equation (2) has the solution

$$\mathbf{A}(\mathbf{r},\, t) = i\, \omega_\mathrm{s}\, \varepsilon \int\limits_{V_a} \frac{\mathbf{E}_a(\mathbf{r}',\, t)}{4\pi\, R} \exp\,(-\, i\, \mathbf{k}_\mathrm{s}\, \mathbf{R})\; \mathrm{d}\mathbf{r}'.$$

Here $\mathbf{R} = \mathbf{r} - \mathbf{r}'$; $\mathbf{k}_\mathrm{s}$ is the Stokes wave vector. The integral is taken over the droplet volume V_a.

Thus, the SRS field at the observation point with the radius vector $\mathbf{r}$ can be written as

$$\mathbf{E}(\mathbf{r},\, t) \approx \mathrm{rot\, rot} \int\limits_{V_a} \frac{\mathbf{E}_a(\mathbf{r}',\, t)}{4\pi\, R} \exp\,(-\, i\, \mathbf{k}_\mathrm{s}\, \mathbf{R})\; \mathrm{d}\mathbf{r}' \qquad (3)$$

As is seen, the integrand in Eq. (3) is the Stokes electric field strength $\mathbf{E}_a(\mathbf{r},\, t)$ in the droplet. The spatial configuration of the Stokes electric field corresponds to one of the resonance modes $TE_n(TH_n)$. As known,[1,2] this mode can be represented as a standing wave, localized near the droplet surface being produced due to the interference of two waves propagating in opposite directions under the condition of phase synchronism between them.

Then, using the known Mie solution, for example, for TE_n resonance mode of spherical resonator, we derive the following expression:

$$\mathbf{E}_a(\mathbf{r},\, t) = A_E(t) B_n(\omega_\mathrm{s}) \psi_n(k_\mathrm{s}\, r)\, \frac{\mathbf{Y}_{nm}(\theta,\, \varphi) + \mathbf{Y}^*_{nm}(\theta,\, \varphi)}{2}, \qquad (4)$$

where A_E is the amplitude of the SRS wave; B_n is the Mie coefficient[2] at the Raman frequency ω_s, having the meaning of the amplitude of a partial wave in a resonator; $\mathbf{Y}_{nm}$ is the spherical vector–harmonics; ψ_n is the Bessel spherical function; θ and φ are spherical coordinates. Similar expression can be written for TH_n–modes too.

By substituting Eq. (4) into Eq. (3) and expanding the function integrand into a series over spherical harmonics, we finally obtain

$$\mathbf{E}(\mathbf{r},\, t) \cong \frac{k_\mathrm{s}^2}{4\pi\, r} A_E(t) B_n(\omega_\mathrm{s}) \times$$

$$\times \int\limits_{V_a} \sum_{l;m} \frac{2l + 1}{l(l + 1)}\, \psi_n(k_\mathrm{s}\, r')\, \psi_l(k_\mathrm{s}\, r) \times$$

$$\times\, Y_{l1}(\theta,\, \varphi)\, [\mathbf{Y}_{nm}(\theta',\, \varphi') + \mathbf{Y}^*_{nm}(\theta',\, \varphi')]\; \mathrm{d}\mathbf{r}' \qquad (5)$$

Some results following from numerical solution of Eq. (5) are presented below.

Figures 1 and 2 show the angular dependence of SRS relative intensity in the far zone for a droplet with the radius $a_0 = 6.3$ μm. Hereinafter it is believed that water droplets are illuminated with the radiation at $\lambda = 0.53$ μm (second harmonic of a Nd:YAG laser). The Stokes radiation generated in such a way has the wavelength $\lambda_\mathrm{s} \sim 0.65$ μm. The calculation is done for the case when SRS is supported by TE^1_{90} and TE^3_{90} resonance modes of the droplet. The angular distribution of elastic scattering is also presented in these same figures. It can be seen from the figures that, first, the SRS angular structure becomes more uniform when the order of the mode, stimulating the SRS, increases; second, in contrast to the elastic scattering, the SRS directional pattern is practically symmetric in forward and backward directions, that can be explained by the corresponding symmetry of the internal SRS field in the droplet (Fig. 3).

It should be noted that the scattered Stokes field is symmetric only in the case when the SRS process is supported by only one of the droplet's free modes. At the same time, due to high spectral density of free modes in a droplet[2] the situations is possible when the frequencies of several free modes are close to the Stokes frequency ω_s. In that case all of them will contribute into the SRS process inside a droplet. Under such a “multimodeB regime of SRS the angular distribution of the Stokes field in the far zone becomes asymmetric (see Fig. 4).

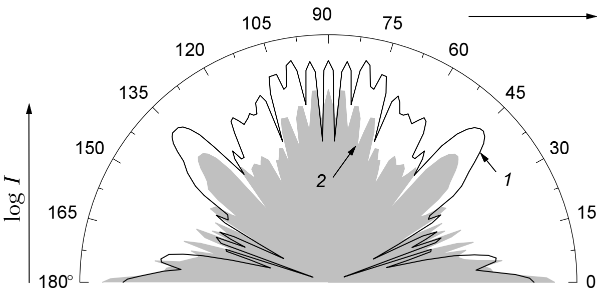

FIG. 1. Angular dependence of the relative intensity, I, of SRS from water droplet for TE^1_{90} (1) and TE^3_{90} (2) resonance modes. Horizontal arrow shows the direction of radiation incidence.

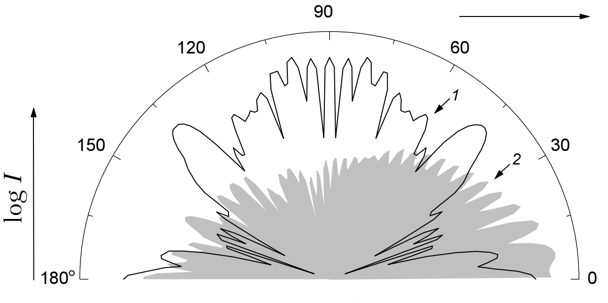

FIG. 2. Angular dependence of the relative intensity, I, of SRS (1) and elastic scattering (2) from water droplet for TE^1_{90} resonance mode. Horizontal arrow shows the direction of radiation incidence.

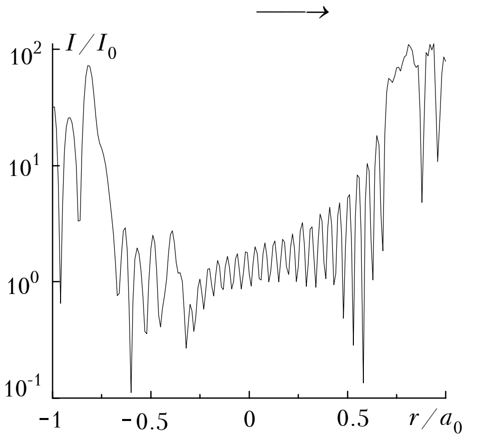

FIG. 3. Distribution of the internal optical field intensity in droplet for TE_{90}^{3} *resonance mode. Horizontal arrow shows the direction of radiation incidence.*

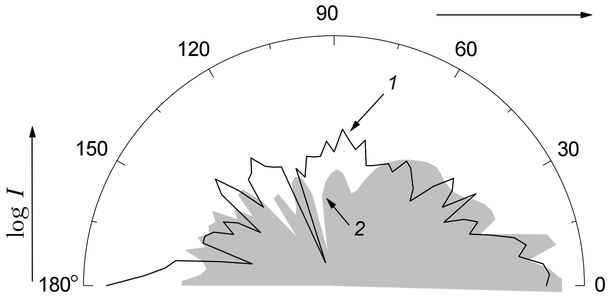

FIG. 4. Comparison of angular dependences of the relative intensity of SRS under multimode regime of generation (1) and elastic scattering (2) from water droplet with a_0 =6.3 μm.

Figure 5 presents a comparison of the results of numerical calculations of the angular structure of SRS from ethanol droplet with the corresponding experimental data published in Ref. 3. The calculated angular dependence of the elastic scattering intensity is also shown in this figure. One can readily see the typical V–shaped angular dependence of the SRS signal. This shape was also observed in Ref. 4. Analysis of these regularities shows that the theoretical model of SRS well fits the regularities of the SRS field formation in the far zone.

Thus, the conclusion can be drawn that on the whole the SRS directional pattern is more uniform than that of the elastic scattering. This is connected with the absence of the Raman-shifted field component, diffracted on the particle contour. In contrast to the elastic scattering, the angular structure of unimode SRS is symmetric with the period multiple to π/2. This is a consequence of the angular symmetry of the internal Stokes field, being in resonance with a droplet's free vibrational mode. In the multimode regime of the SRS in a droplet, such a symmetry breaks.

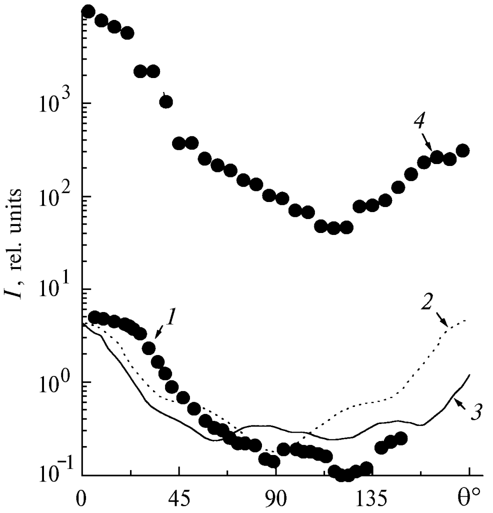

FIG. 5. Experimental data[3] *on the angular structure of SRS (1) and elastic scattering (4) from the ethanol droplet* 15.1 μm *in radius in the far zone. Solid curves are for theoretical calculations of the SRS angular dependence at unimode (2) and multimode (3) regimes of SRS in a droplet; dots are for the experimental data.*

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