# THE EFFECT OF SRS THRESHOLD DECREASE IN WEAKLY ABSORBING AEROSOL PARTICLES: NUMERICAL CALCULATIONS

**Yu.E. Geints, A.A. Zemlyanov, and E.K. Chistyakova**

*Institute of Atmospheric Optics,*
*Siberian Branch of the Russian Academy of Sciences, Tomsk*
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*We present here some results of numerical calculations for the effect of stimulated Raman scattering (SRS) threshold decrease in weakly absorbing ethanol drops under the action of Nd:YAG laser radiation at* $\lambda = 0.532$ μm. *We analyze here physical causes of this effect in spherical particles. The results obtained are compared with the experimental data.*

The effect of stimulated Raman scattering (SRS) in aerosol particles has some peculiarities as compared to that in continuous media. They are caused by both the ability of a spherical particle to concentrate incident electromagnetic radiation in its volume and the presence of natural high-quality electromagnetic resonance modes in a drop. These resonances are observed at certain values of the diffraction parameter of a particle $x = 2\pi a/\lambda$, ($a$ is the radius of a drop, $\lambda$ is the laser radiation wavelength) and characterized by the order $l$ and the number $n$ of a mode of the partial electromagnetic wave causing the resonance.

It is well-known that spontaneous Raman scattering appears in the whole volume of a particle interacting with radiation; however, it is most intense in the range of focusing of incident radiation near the shaded surface. Some waves from the spectrum of spontaneous Raman scattering leave the drop, while the other portion travel along the spherical surface due to the total internal reflection. If the frequency $\omega_s$ of a wave from the spectrum of spontaneous Raman scattering coincides with the frequency of a natural resonance $\omega_{ln}$ of a drop, amplification predominates over absorption and stimulated scattering takes place (the condition of output resonance). The input resonance is achieved when the incident radiation frequency $\omega_L$ is also tuned to a natural resonance of the drop. If both conditions are fulfilled in the drop we have double SRS resonance.

The energy threshold of the SRS process in water and ethanol micron–size drops under irradiation by the second harmonic of Nd:YAG laser ($\lambda = 0.532$ μm) was studied experimentally[1–6] and theoretically.[7] It was demonstrated that the threshold intensity of laser radiation at which SRS radiation ($\lambda = 0.63$ μm) is experimentally observed in drops is about $10^7$–$10^9$ W/cm$^2$ for drops of 10 to 3 μm in diameter. However, an additional 3–5 fold reduction of the threshold intensity was observed in some cases.[4] The authors of this paper assign this effect to the double electromagnetic resonance in particles. In connection with this fact, theoretical study enabling one to estimate the SRS threshold in this case and provide some practical recommendations on low-threshold SRS excitation in micron-size particles is rather interesting. This paper is devoted to this problem.

Since the fulfillment of input resonance conditions requires additional tuning of the drop size or incident radiation wavelength that is complicated from the viewpoint of experimental technique, there are experimental works in which only the output resonance was considered. A later paper[8] describes the observation of double resonance in levitating glycerin drops of 5–7 μm radius irradiated with Nd:YAG laser radiation at $\lambda = 0.532$ μm. It should be noted that the situation when both resonances are observed is rather difficult to create due to small width of their lines (the order of 3 cm$^{-1}$).

We performed a numerical experiment which made it possible to simulate the double resonance in an ethanol drop. The order and number of the output resonance were constant, and the order of input resonance varied. The number of the input resonance mode was determined in correspondence with the value of the resonance radius of the drop. We considered the following cases: $\mathrm{TE}_{km} \to \mathrm{TE}_{ln}$, $\mathrm{TM}_{km} \to \mathrm{TE}_{ln}$, $\mathrm{TM}_{km} \to \mathrm{TM}_{ln}$, where $\mathrm{TE}_{ln}$ ($\mathrm{TM}_{ln}$) is the transversal electric (magnetic) mode with mode number $n$ and mode order $l$. It supports the output resonance (SRS) and $\mathrm{TE}_{km}$ is the designation for the mode feeding input resonance. It was established that several resonance modes with different combination of the parameters $k$ and $m$ can simultaneously exist in a drop and support the output resonance what was shown experimentally. Although the problem of competition among resonance modes of different orders in the process of SRS initiation is complicated and poorly studied, it is evident that lower orders modes have essentially higher $Q$-factor. The advantage of high order modes is their greater extension within the drop; i.e., larger overlapping with the pumping field.[7]

In the stationary case, the threshold of the SRS generation is defined by equalizing the combination wave gain to its total losses in the volume of a particle

$$\Sigma + R = P_g,$$

where

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$$\Sigma = \frac{\sigma}{2} \int\limits_V \mathbf{E}_S(\mathbf{r}, t)\, \mathbf{E}_S^*(\mathbf{r}, t)\, \mathrm{d}V'$$

is the average power of thermal losses inside the particle,

$$R = \frac{c}{8\pi} \int\limits_S \mathbf{E}_S(\mathbf{r}, t) \times \mathbf{H}_S^*(\mathbf{r}, t)]\, \mathbf{n}\mathrm{d}S$$

is the average power of radiative losses through the surface of a particle,

$$P_g = \frac{c}{8\pi} \int\limits_V G_R(I_i(\mathbf{r}, t))\, \mathbf{E}_S(\mathbf{r}, t)\, \mathbf{E}_S^*(\mathbf{r}, t)\, \mathrm{d}V'$$

is the average power of SRS field sources connected with the gain of the Stokes wave in the particle.

Here $\sigma$ is the conductivity of the drop material, $c$ is the speed of light, $\mathbf{E}_S$ and $\mathbf{H}_S$ are the amplitude values of electric and magnetic oscillations of the Stokes wave. $V$ and $S$ are volume and surface of drop, $\mathbf{n}$ – outer normal.

In the stationary regime, the amplification factor for the Stokes wave is $G_R = g_S\, I_i(\mathbf{r})$, where $g_S$ is the Raman amplification coefficient, $I_i(\mathbf{r})$ is the intensity of the pumping field (at the frequency of incident radiation $\omega_i$). The latter can be expressed by the factor of field inhomogeneity inside the particle $B_i(\mathbf{r})$: $I_i(\mathbf{r}) = I_i^0 B_i(\mathbf{r})$, where $I_i^0$ is the intensity of radiation incident on the drop.

Then, the threshold SRS intensity is expressed as

$$I_{SRS} = \frac{2\pi n_a}{Q_{ln}\, g_S \lambda} \times \left[ \int\limits_V \mathbf{E}_S(\mathbf{r},t)\mathbf{E}_S^*(\mathbf{r},t)\mathrm{d}V' \Big/ \left( \int\limits_V B_i(\mathbf{r})\mathbf{E}_S(\mathbf{r},t)\mathbf{E}_S^*(\mathbf{r},t)\mathrm{d}V' \right) \right], \tag{1}$$

where $n_a$ is the refractive index of the material of the drop.

Since for the SRS to occur in a drop the output resonance condition is necessary, the spatial distribution of the SRS field must correspond to the spatial structure of the field of a given resonance mode. Then, in the stationary regime, the electric vector of the Stokes wave $\mathbf{E}_S(\mathbf{r})$ can be represented as a product of an amplitude $\mathbf{E}_S^0$ by a coefficient which is a function of only spatial coordinates:

$$\mathbf{E}_S(\mathbf{r}) = \mathbf{E}_S^0 \sqrt{B_S(\mathbf{r})}\ .$$

Then the ratio of integrals in the right–hand side of Eq. (1) takes the form

$$\int\limits_V B_i(\mathbf{r})\, B_S(\mathbf{r})\, \mathrm{d}V' \Big/ \left( \int\limits_V B_S(\mathbf{r})\, \mathrm{d}V' \right) \equiv B_c. \tag{2}$$

Taking into account Eq. (2), let us write Eq. (1) as

$$I_{SRS} = 2\pi\, n_a/(Q_{ln}\, g_S\, \lambda\, B_c), \tag{3}$$

where $Q_{ln}$ is the $Q$-factor of the resonance mode initiating the SRS process in a drop.

As follows from Eq. (3), the stimulated Raman scattering threshold is determined when varying the dimension of a particle only by the coefficient $B_c$ which, in fact is the coefficient of overlapping for pumping field and SRS field inside the drop. The $I_{SRS}$ threshold values are lower for better overlapping.

The results of numerical calculations of the coefficient $B_c$ for different combinations of input and output resonance modes are presented in Fig. 1. The results obtained under double resonance condition in a drop enable one to arrive at the following conclusions:

1. The values of $B_c$ in the case $TE_{km} \to TE_{ln}$ are larger than in the case $TM_{km} \to TE_{ln}$ (Figs. 1*a*, 1*b*).
2. The increase of drop dimensions leads to excitation of modes with higher values of $n$ (Fig. 2).
3. The value of the coefficient $B_c$ increases with the increase of the diffraction parameter $x$.
4. The values of $B_c$ in the case of double resonance are considerably larger as compared with the case when only the output resonance occurs.
5. The value of $B_c$ increases with the increase of the mode order $l$ under fixed value of the mode number $n$ of the output resonance.
6. The value $B_c$ falls with the increase of the input resonance order under fixed value of the number and order of the output resonance mode. The function $B_c$ of different input resonance orders is presented in Fig. 1*a* for the case of excitation of the mode $TE_{l70}$ supporting the output resonance. The anomalous function for the first input resonance order is explained by the fact that the accuracy of calculations available was not sufficient for exact tuning to the given resonance because low order resonances have very high $Q$-factor and, small width with respect to the $x$ scale.

Figure 3 presents the values of the threshold SRS intensity in ethanol drops of different size in the case of single and double resonances. In the latter case, the calculation was performed for the situation $TE_{km} \to TE_{2n}$. As follows from Fig. 3, the threshold intensity sharply increases with the decrease of the drop dimensions. This is connected with a similar decrease of radiative $Q$-factor for small particles. Since the $Q$-factor is restricted by losses connected with absorption in a liquid, $I_{SRS}$ in fact does not depend on the radius of liquid particles for $x \ge 100$. For $x \le 20$–$40$, the SRS effect can be suppressed by the optical breakdown arising inside the particle. The SRS threshold considerably decreases in the case of double resonance as compared with the non-resonance case that is a consequence of redistribution of the resonance mode field. This decrease is stronger as the $Q$-factor of the corresponding input resonance increases.

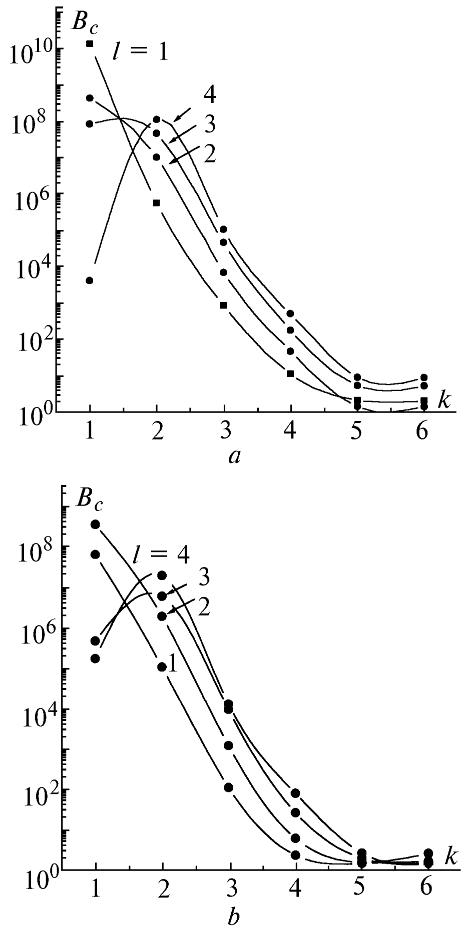


*FIG. 1. The coefficient $B_0$ of overlapping of pumping and SRS fields in an ethanol drop as a function of the input resonance order k in the case of* $TE_{km}$–$TE_{l70}$ *(a) and* $TM_{km}$–$TE_{l70}$ *(b) generation. The calculation was performed for different output resonance order l.*

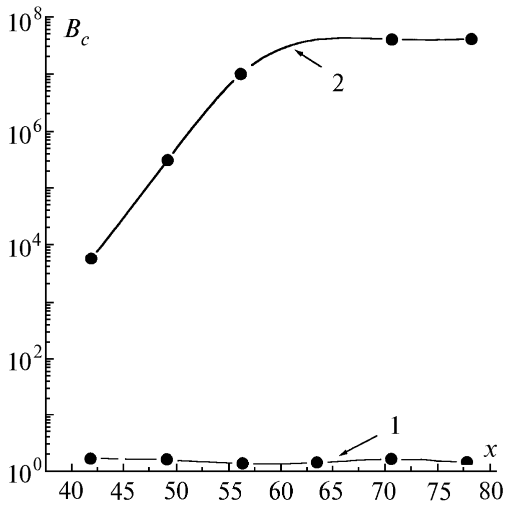


*FIG. 2. The coefficient $B_c$ as a function of the drop diffraction parameter x in the case of single (1) and double (2) resonance.*

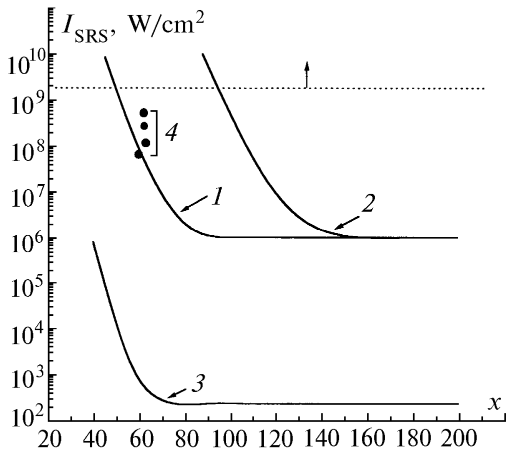


*FIG. 3. Theoretical dependence of the SRS intensity in the ethanol drops on their diffraction parameter. The curves 1 and 2 correspond to lower and higher order resonances respectively in the case of single resonance. The curve 3 is double resonance for the case* $TE_{km}$–$TE_{2n}$. *Dashed line is the optical breakdown threshold of ethanol drops. Experimental data[6] are shown by curve 4.*

Note in conclusion, that there are experimental works[1,2–9] which demonstrate that the stimulated Mandel'stam–Brillouin scattering (SMBS) can arise in a drop simultaneously with the stimulated Raman scattering. The SMBS wave arises earlier than the SRS process because it has higher gain, and then it can be a pumping field for the SRS wave. This case can also be considered within the frames of our model as double resonance where SMBS and SRS play the part of the input and output resonances respectively because SRS and SMBS are in resonance with natural resonance modes of a drop. This fact allows one to explain an additional decrease in the SRS threshold what has been observed in some experiments.

## REFERENCES

1. J.–Z. Zhang and R.K. Chang, J. Opt. Soc. Am. B. **6**, No. 27, 151–153 (1989).
2. J.–Z. Zhang, G. Chen, and R.K. Chang, J. Opt. Soc. Am. B. **7**, No. 1, 108–115 (1990).
3. A.S. Serpenguzer, G. Chen, and R.K. Chang, J. Opt. Soc. Am. B. **9**, No. 6, 871–873 (1992).
4. R.G. Pinnick, A. Biswas, J. Pendleton, and R.L. Armstrong, Appl. Opt. **31**, (1992).
5. H.–B. Lin, J.D. Eversole, and A.J. Camillo, Opt. Lett. **17**, No. 11, 828–830 (1992).
6. P. Chylek, M.A. Jarzembski, V. Srivastava et al., Appl. Opt. **26**, No. 5, 760–762 (1987).
7. Yu.E. Geints, A.A. Zemlyanov, and E.K. Chistyakova, Atmos. Oceanic Opt. **8**, No. 10, 803–807 (1995).
8. A. Biswas, H. Latifi, R.L. Armstrong, and R.G. Pinnick, Phys. Review **A40**, No. 12, 7413–7416 (1989).
9. A.S. Serpenguzer, J.C. Swindal, R.K. Chang, and W.P. Acker, Appl. Opt. **31**, No. 18, 3543–3551 (1992).