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OPTICS AND LASER PHYSICS

Anomalous Decrease in the Threshold of Stimulated Raman Scattering near the Surface of Liquid Nitrogen

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An anomalously strong (up to 13 times) reduction of the threshold of the stimulated Raman scattering of 15-ps laser pulses in liquid nitrogen has been found when moving the caustic of a pump beam from the bulk through the free surface. A nonmonotonic N-shaped dependence of the stimulated Raman scattering threshold on the distance of the beam waist to the surface has been recorded for the first time with a minimum reached when the plane front of the beam caustic coincides with the surface. The physical mechanism of the detected phenomenon has been discussed.

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INTRODUCTION

We experimentally study a new phenomenon of an anomalously strong (~13 times) reduction of the threshold energy of the stimulated Raman scattering (SRS) of 15-ps second harmonic (527 nm) pulses of a Nd:YLF laser in the surface layer of liquid nitrogen when moving the beam caustic near the surface.

The threshold of SRS in a homogeneous medium is commonly accepted [1, 2] as the pulse energy at which the "chaos–order" phase transition of spontaneous molecular motions (with an equiprobable scattering to a sphere) to a coherent state occurs with the formation of a coherent Stokes component beam (coaxially to the pump beam). According to [1–5], the frequency ω_S and intensity I_S of the Stokes component are determined by the relations

$$\omega_S = \omega_p - \Omega, \tag{1}$$

$$I_S = I_n \exp G, \tag{2}$$

where ω_p and Ω are the frequencies of pumping and molecular motions, respectively; I_n is the intensity of quantum noise at the Stokes frequency of the component; and

$$G = gI_pL. \tag{3}$$

Here, g is the SRS gain (proportional to the cubic nonlinearity $\chi^{(3)}$ of the medium), I_p is the pump radiation intensity, and L is the gain length. In the case of generation of SRS by parallel pump laser beams, the gain length L is equal to the length of a nonlinear optical sample. In the case of focused pump beams, the SRS gain length in the traveling wave mode is commonly accepted as twice the Rayleigh length (confocal parameter, i.e., the distance from the caustic waist at which the beam area is doubled compared to the cross-sectional area at the waist) [3–6].

It was established experimentally [4, 5] that the SRS threshold (both in the traveling wave mode [4] and at the location of the medium in a laser cavity [5]) is achieved at exponents $G \approx 25$ and a subsequent increase in pumping is accompanied by an exponential increase in the intensity of the Stokes component in accordance with Eq. (2). However, a violation of Eq. (3) was observed in some cases of SRS. For example, the exponential growth of the Stokes component in liquid nitrogen transfers to an anomalous jump by six or seven orders of magnitude [7–9]. Later [10], the jump mechanism was explained by the development of generation of SRS with distributed feedback. The authors of [11] recorded a decrease in the threshold of SRS of picosecond pulses by almost an order of magnitude in water in the pores of globular opal-type crystals and explained the observed phenomenon by an increase in the intensity of the local field near nanoscale globules.

Studying the temperature evolution of Raman spectra in water, we have recently revealed [6] the anomalous reduction of the SRS threshold of 15-ps pulses near the water surface. Taking into account that structural complexes of water can be rearranged at the surface under unsteady pumping conditions and affect the SRS threshold, we decided to start an experimental study of the detected phenomenon with a nonpolar liquid without hydrogen bonds, namely, with liquid nitrogen, which is reported in this work.

EXPERIMENT

The choice of liquid nitrogen is due to several reasons in addition to those mentioned above. Thus, pioneering works on high-power SRS lasers on liquid nitrogen [3] showed that it contains a small amount of impurities and has a high optical homogeneity and, therefore, a high optical breakdown threshold. A high SRS gain of $g = 3.0 \times 10^{-2}$ cm/MW at a wavelength of 527 nm measured in our previous work [9] provides a SRS threshold below the optical breakdown intensity. In addition, the study of four-wave parametric processes of generation of higher Stokes components [12] allowed the correction of the refractive index of liquid nitrogen in the region of 600–830 nm, which includes the 605-nm wavelength of the first Stokes component of the 527-nm SRS pump radiation in our case (the frequency of molecular oscillations $\Omega = 2326$ cm^{-1}). Note also that it is convenient to separate the first Stokes component (605 nm) of SRS in the experiment using glass orange-red light filters (OS-14 or KS-10) and safety spectacles with such glasses owing to a large Stokes shift (2326 cm^{-1}).

The second harmonic of a picosecond Nd:YLF laser [13] with active–passive mode synchronization (527 nm, $M^2 = 1.5$, 15 ps, up to 5 mJ/pulse, 5 Hz, and beam diameter expanded to ~8 mm) was used as pump radiation. The laser beam (see Fig. 1) was deflected downward by a rotary 90° prism toward the position table with a vertical micrometric feed (the maximum displacement is ±20 mm with a step of 0.5 mm). A glass Dewar flask with a mirror coating (its height and diameter were 270 and 55 mm, respectively) with liquid nitrogen was located on the table. The Dewar flask was filled with liquid nitrogen so that the nitrogen surface was below its edge by 25–30 mm. This volume was filled with cold nitrogen vapors, which blocked the contact of the liquid with air and ensured the optical quality of the surface in the absence of boiling traces.

A collecting lens with a focal length of $F = 82$ mm was placed above the Dewar flask so that the beam caustic was in the bulk of liquid nitrogen at a depth of 30 mm when the Dewar flask was moved to the upper position. The dependence of the SRS threshold on the distance between the beam caustic and surface was measured while moving the sample down without changing the rest of the experimental geometry. The exact position of the plane front of the beam on the liquid nitrogen surface (the distance h to the focal plane of the lens in air) was controlled on the screen with the hole (see Fig. 1) by the formation of a parallel beam reflected from the surface behind the lens in the opposite direction (as shown in the inset in Fig. 1). The laser pulse energy was measured by a PE50-DIF-C digital pyroelectric energy meter (Ophir Photonics), on which a part of the beam from the beam splitter was directed. The SRS threshold was determined by measuring the energy of the laser pulse at which the appearance of the Stokes component was observed from the orange-red coloring of the volume of liquid nitrogen in the Dewar flask. Five parallel measurements were made at each Dewar flask position, after which the mean value of the threshold pulse energy and the standard deviation from the mean were calculated.

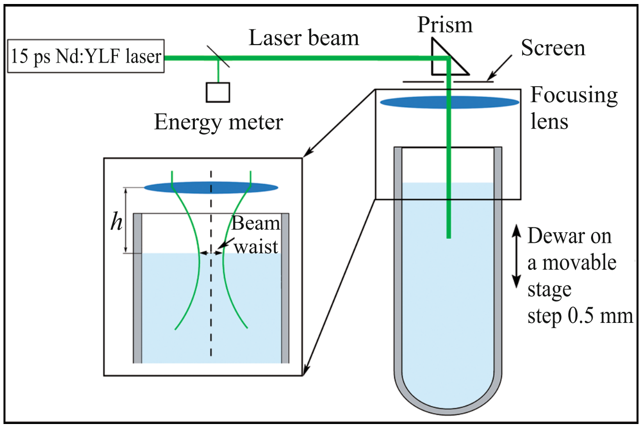

Fig. 1. (Color online) Schematic of the experiment to study the anomalous reduction of the threshold of stimulated Raman scattering of 15-ps pulses (527 nm) in liquid nitrogen when moving the beam caustic through an open surface. The inset shows the schematic of measuring the lens–surface distance.

RESULTS AND DISCUSSION

Figure 2 shows the dependence of the threshold energy of the SRS of 15-ps pulses in liquid nitrogen on the distance h between the liquid surface and lens (F = 82 mm) when the beam caustic moves from the bulk to the surface. The arrow indicates the position of the surface near which an abnormally large decrease in the threshold value was detected. The measurements of the dependence of the SRS threshold in the traveling wave mode (the increment at the gain length reaches a value of 25, see Eq. (3)) were begun when the beam caustic was in the bulk of liquid nitrogen ($h = 50$ mm). The SRS threshold at this caustic position and at its displacement toward the surface by 25 mm (Fig. 3a) remained a constant value of ~130 μJ. Then, the increase in the SRS threshold began after 2 mm of displacement of the Dewar flask down and convergence of the beam caustic with the surface (at $h = 77$ mm, see Fig. 2).

This increase in the threshold is due to a decrease in the SRS gain length L in the traveling wave mode (see Eqs. (2) and (3)) because a part of the beam caustic begins to cross the surface and exit the bulk, as schematically shown in Fig. 3b. Consequently, the plane caustic front (focal plane) is submerged by ~6 mm at this point (the layer thickness is increased by

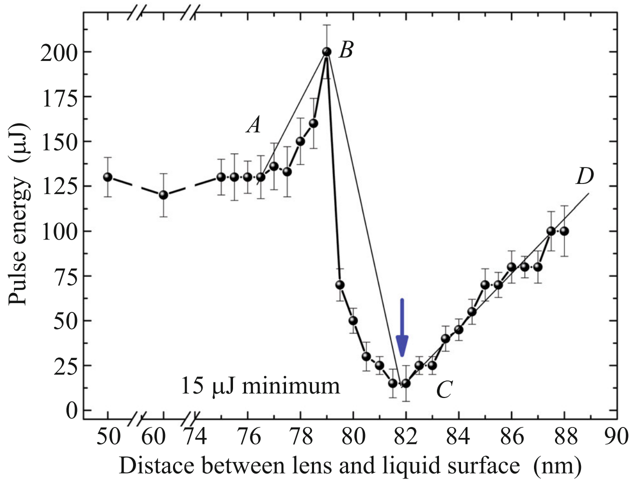

Fig. 2. (Color online) Threshold of stimulated Raman scattering versus the distance between the lens and surface. The points *A*, *B*, and *C* are the points of the beginning of growth, maximum, and minimum of the threshold, respectively.

~1 mm because of the spherical aberration of the lens [14] and the refractive index of liquid nitrogen $n = 1.19$ [12]). Then, the point where the increase in the SRS threshold begins near the surface gives an estimate of ~12 mm (twice the Rayleigh length, $2L_R$ in Fig. 3) for the SRS gain length in the traveling wave mode.

In Fig. 2, the SRS threshold begins to increase at point *A* (~130 μJ at $h = 77$ mm), reaches a maximum at point *B* (~200 μJ at $h = 79$ mm), achieves a minimum at point *C* (~15 μJ at $h = 82$ mm), and increases by several times at point *D* when the waist moves from the surface to the Rayleigh length (see Fig. 3). We connect these points by straight line segments for the convenient subsequent analysis of change in the SRS threshold near the surface. The resulting N-shaped dependence has three stages of the variation of the SRS threshold: increase, significant decrease, and subsequent increase. The threshold increases to ~200 μJ at the first stage, which means that Eq. (3) remains valid at a decrease in the gain length (Fig. 3b). This stage is followed by a sharp (by 13 times) decrease in the SRS threshold to the minimum value, despite the simultaneous decrease in the gain length, since only half of the beam caustic remained under the surface (Fig. 3c). The SRS threshold begins to increase linearly with a gradient of 16 μJ/mm, half that at the first stage at further motion (Fig. 3d) of the second half of the beam caustic from the bulk (the third stage in Fig. 2).

The discovered anomaly indicates a change in the mechanism of the SRS of a traveling wave and a key role of the surface in the new mechanism. The multipass cavity mechanism of generation with an asymmetric cavity is most likely here in contrast to a symmetric cavity with distributed feedback [10], as in the case of the jump in the Stokes component of SRS in liquid nitrogen [7–9] or a decrease in the generation threshold at stimulated Mandelstam–Brillouin scattering in the liquid [15]. It is essential in our case that the flat surface of the liquid–air interface is a lumped element of the feedback and acts as a mirror of the SRS generator with an asymmetric cavity (by the type of feedback).

As shown in Fig. 2, "mirror switching-on" (the largest gradient of decreasing threshold) occurs after the maximum threshold value (at point *B* in Fig. 2) and the next decrease in the gain length by 0.5 mm.

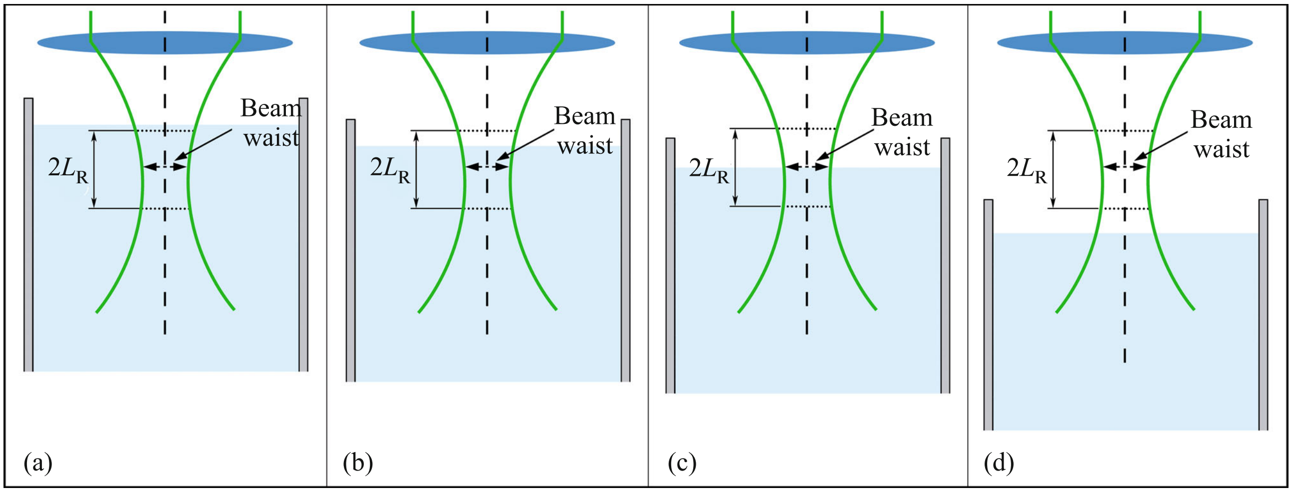

Fig. 3. (Color online) Position of the pump beam caustic (twice the Rayleigh length $2L_R$ or the confocal parameter) relative to the surface of liquid nitrogen when moving the Dewar flask down (disregarding the refractive index of liquid nitrogen $n = 1.19$ [12] and displacement of the beam waist [14] in the nitrogen bulk).

The contribution of two factors is summed up here. The first factor is the decrease in the wavefront curvature at refraction on the surface and along the caustic when approaching its waist. The second factor is the channeling of the beam along the axis and, accordingly, increase in the gain length because of spherical aberration of the lens (elongation of the cylindrical part of the caustic [14]). Note that the self-focusing development or beam filamentation probability [14] is low, since the critical self-focusing power cannot be increased in the falling region (*BC* in Fig. 2). At a subsequent decrease in the gain length (part of the beam caustic) in liquid nitrogen (see Fig. 2), the SRS threshold decreases monotonically to the minimum value (~15 μJ at $h = 82$ mm) as a plane wave front (beam waist) approaches the surface of liquid nitrogen (point *C* in Fig. 2 and Fig. 3c). This fact additionally points to the cavity mechanism of the decrease in the SR threshold because of the reduction of losses at the reflection from a flat mirror (as compared to the distributed feedback [10]) and an increase in the number of passes inside the asymmetric cavity of the SRS laser of a new type.

Undoubtedly, the discovered new cavity mode of SRS generation of picosecond pulses with a minimum threshold near the surface requires a detailed study. The decrease in the SRS threshold by more than an order of magnitude is of particular interest in view of the possibility of ensuring a manyfold pumping excess over the SRS threshold and obtaining simultaneous generation of the Stokes and anti-Stokes components [3, 12]. This biphoton radiation [16] ensures a study of the correlation of fluctuations of the intensity of biphotons at their simultaneous generation in an asymmetric cavity of the SRS laser. The presence of a cavity distinguishes this situation from the spontaneous parametric generation of entangled quantum states [17].

CONCLUSIONS

An anomalous (up to 13 times) decrease in the SRS threshold of 15-ps pulses near the liquid nitrogen surface has been found. The N-shaped dependence of the SRS threshold (increase–decrease–increase) at the displacement of the pump beam caustic through the free interface has been revealed instead of the expected (see Eq. (3)) proportional growth at the reduction of the gain length. A similar anomaly of the SRS threshold was also observed for water in preliminary experiments. The similarity of the detected anomaly in liquid nitrogen without hydrogen bonds indicates a new physical phenomenon: the development of the SRS of pulsed lasers in liquids when focusing pump beams near the surface according to a specific mechanism of generation. Undoubtedly, the proposed mechanism of inclusion of an asymmetric cavity (distributed feedback in the bulk and lumped feedback on the flat surface of liquid nitrogen, a mirror) requires a separate study for the development of new-type SRS lasers on condensed media.

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