

NONLINEAR OPTICAL PHENOMENA

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Stimulated Raman scattering in crystals pumped by phase-modulated picosecond pulses

T.T. Basiev, P.G. Zverev, A.Ya. Karasik, D.S. Chunaev

***Abstract*. The appearance of hysteresis in the dependence of the efficiency of transient SRS on the energy of picosecond laser excitation is studied. The hysteresis appears due to the frequency-phase modulation of laser radiation which changes the SRS gain with changing the degree of modulation.**

Keywords: *stimulated Raman scattering, self-phase modulation, picosecond pulses, chirped laser pulses.*

Passively mode-locked solid-state lasers emit, as a rule, phase-modulated pulses [1]. Self-phase modulation (SPM) leads to the frequency sweep, resulting in the broadening of the laser pulse spectrum and thereby affecting the characteristics of nonlinear optical processes initiated by these lasers. The influence of phase- (frequency)-modulated (chirped) pump radiation on the SRS process was studied theoretically, for example, in papers [2–4]. A review of experimental studies of SRS in this field is presented in Ref. [1].

In particular, it was found in Ref. [5] that upon excitation of various liquids by a train of picosecond pulses from a neodymium glass laser, the SRS gain decreases at the end of the pulse train tentatively due to the broadening of the laser pulse spectrum caused by SPM or due to an increase in the pulse duration. In Ref. [6], the difference between the efficiencies of conversion of the pump to Raman scattering of picosecond pulses from the front and rear parts of the pulse train was explained by the inertial effect of thermal self-focusing in liquids. Nonlinear SRS in conjunction with SPM under conditions when the initial range of the pump frequency sweep is extended upon SRS [7] can cause the transformation of spectral and temporal parameters of SRS and pump radiation [4, 8]. In inorganic crystals, whose thermal and spectral properties differ from those of liquids and gases, the role of SPM in SRS is not adequately studied, and further systematic studies in this field are needed.

By pumping the $BaWO_4$, $SrMoO_4$, $KGd(WO_4)_2$, and $Ca_3(VO_4)_2$ oxide crystals by trains of strongly chirped picosecond laser pulses, the authors of paper [9] found hysteresis in the dependence of the efficiency of transient SRS on the pump energy, which was manifested in the change in the efficiency upon pumping by pulses of the same amplitude from the front and rear parts of the train with the half-width ~ 300 ns. The hysteresis was tentatively treated as the material bistability. However, to reveal the true mechanism of this effect, it is necessary first of all to find the role of the laser pump parameters in SRS.

We used for pumping passively mode-locked lasers on disordered CaLiNbGa garnet (CLNGG) and $YLiF_4$ (YLF) crystals doped with Nd^{3+} ions. We studied earlier [9, 10] the disordered garnet laser with a saturable absorber (dye No. 3274) and showed that the spectrum of picosecond pulses ($\lambda = 1060$ nm) was broadened up to 50 cm^{-1} due to SPM. By replacing the Nd^{3+}: CLNGG active element in the same resonator by the Nd^{3+}: YLF active element, we measured the laser radiation parameters.

Figure 1 shows a 1047-nm train of pulses from the YLF laser whose duration is close to that for the CLNGG laser. Note that the output energy of both lasers was ~ 1.5 mJ. The duration τ of 1047-nm pulses of the YLF laser measured with an IMACON-501 streak camera was 11 ps and did not virtually change at the 523.5-nm second-harmonic wavelength. The width of the second-harmonic emission spectrum of this laser, measured with a PGS-2 polychromator equipped with a CCD array, was $\Delta\lambda = 0.062$ nm ($\Delta\nu = 2.26$ cm^{-1}). The product $\Delta\nu\tau = 0.74$ exceeds the value 0.44 inherent in transform-limited Gaussian pulses [1] and indicates that pulses from the YLF laser

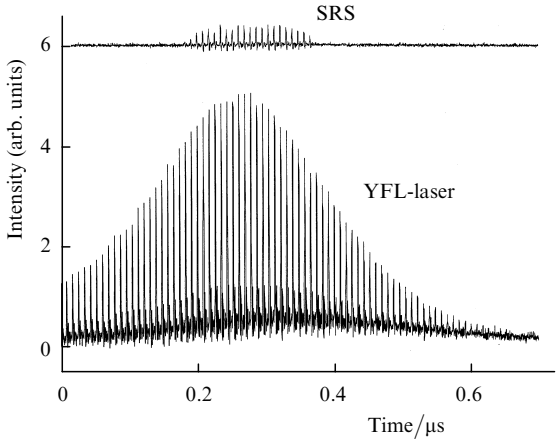

Figure 1. Oscillograms of radiation of the pump YLF laser and SRS in the $PbMO_4$ crystal.

T.T. Basiev, P.G. Zverev, A.Ya. Karasik, D.S. Chunaev Laser Materials and Technologies Research Center, A.M. Prokhorov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia; e-mail: karasik@lst.gpi.ru

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were not completely transform-limited because of some broadening of their spectrum. Compared to the CLNGG laser, in which the spectrum of pulses of close duration (15 ps) was broadened 15–20 times greater ($\Delta\nu = 30 - 50$ cm^{-1}), the SPM effect in the YLF laser is small, which is obviously explained by the relatively small nonlinear refractive index n_2 of fluoride crystals.

The method for measuring the parameters of transient SRS is described in Ref. [9]. Laser radiation, focused into a sample with the help of a concave mirror, and radiation scattered by the sample and propagated through a collimating lens and a dichroic filter rejecting the pump radiation were incident on calibrated fast germanium photodiodes. By using a fast digital oscilloscope, we measured synchronously the energy of individual picosecond pulses in the pump train and the train of scattered radiation during one laser shot, which gave the dependence of the efficiency of each SRS pulse on the pump pulse energy as the ratio of their energies.

Figure 2 shows the dependences of the efficiency of transient SRS in a $PbMO_4$ crystal of length 30 mm on the pump pulse energy for the CLNGG and YLF lasers. In both cases, the laser pump energy was varied with the help of neutral filters from the level sufficient to excite the first Stokes SRS component to the level providing the development of cascade generation of higher-order Stokes components and radiation self-focusing. Upon pumping the crystals by the YLF laser, the radiation focusing was tighter, which resulted in a decrease in the threshold pump energy of SRS (Fig. 2b) compared to that upon pumping by the CLNGG laser (Fig. 2a). Upon pumping by both lasers, the SRS efficiency monotonically increased with

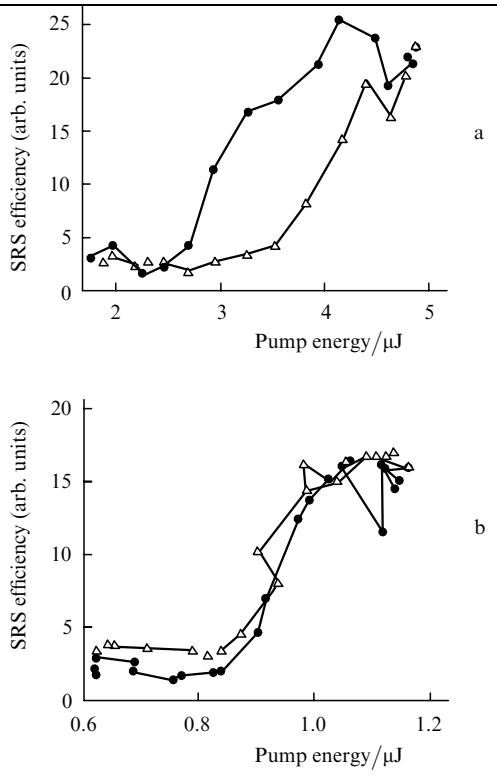

Figure 2. Dependences of the efficiency of transient SRS in the $PbMO_4$ crystal on the pump pulse energy of the CLNGG (a) and YLF (b) lasers upon pumping by pulses from the leading (●) and trailing (△) edges of the train.

increasing energy in the front part of the laser train of pulses and then tended to saturate. However, when crystals were pumped by the CLNGG laser, the hysteresis appeared in the dependence of the SRS efficiency on the pump energy, which was manifested in a decrease in the SRS gain on passing from pumping by pulses in the leading edge of the train to pumping by pulses in the trailing edge of the train (Fig. 2a). A similar dependence for pumping by pulses from the YLF laser revealed no hysteresis (Fig. 2b).

To generalise this effect, we measured similar dependences for a number of nonlinear oxide crystals pumped by the CLNGG and YLF lasers. We studied $Ba(NO_3)_2$, $BaWO_4$, $SrWO_4$, $CaWO_4$, $PbWO_4$, $BaMoO_4$, $SrMoO_4$, $CaMoO_4$, $PbMoO_4$, TeO_2, $NaClO_3$, and $NaBrO_3$ crystals, which have substantially different SRS gains and different molecular vibrational dephasing times ($T_2 = 1.18 - 26.5$ ps) [9]. For all the crystals, these dependences proved to be similar to that for the $PbMO_4$ crystal. Upon pumping by pulses from the CLNGG laser, the hysteresis was observed for all the crystals and was virtually absent upon pumping the crystals by pulses from the YLF laser. A continuous increase in the pump energy, resulting in the cascade SRS generation of higher-order Stokes components, did not cause any controllable change in the hysteresis loop.

Consider the reasons that can cause the appearance of hysteresis upon the SRS amplification. The theory [2] predicts that the gains of transient SRS upon pumping of a nonlinear medium by phase-modulated pulses and pulses without phase modulation should be different. The gains upon non-chirped pumping should exceed the gains upon chirped pumping, all other factors being the same. Then, a change in the degree of phase modulation for different pulses in the laser pump train should result in the corresponding variation in the SRS gain for these pulses and, hence, in the hysteresis observed (Fig. 2).

We studied in Ref. [10] the dynamics of the duration and width of the spectrum of picosecond pulses emitted by the laser based on the neodymium-doped disordered garnet used in this paper. It was shown that, the frequency chirp increases due to increasing SPM in a train of laser pulses, resulting in a monotonic increase in the width of the pulse spectrum in passing from pulses in the leading to trailing edge of the pulse train. Therefore, according to [2], the use of such a laser for exciting SRS should lead to a decrease in the SRS gain or to the hysteresis of the SRS efficiency upon pumping a nonlinear medium by pulses of the same amplitude from the leading and trailing edges of the train (Fig. 2a). The use of a train of nearly transform-limited pulses from the YLF laser for exciting SRS does not result in a noticeable hysteresis in a similar dependence (Fig. 2b). The pump pulse energy in our experiments was changed with the help of neutral filters within a broad range. However, when the pump energy exceeded the threshold energy by two-three times, the cascade SRS generation and parametric four-photon Stokes–anti-Stokes generation appeared [1]. These effects prevent the study of a change in the hysteresis loop at higher pump energies.

The Stokes wave intensity is determined not only by the dependence of the SRS gain on the pump SPM, but also should depend, as predicted by the theory [3], on the width $\Delta\nu$ of the laser pulse spectrum upon frequency-modulated pumping. Then, the Stokes wave intensity in the transient case should be proportional to $(\Delta\nu T_2)^{-1}$, which is caused by a decrease in the peak intensity of initial spontaneous

scattering due to the broadening of the pump radiation spectrum.

To elucidate the influence of the width of a chirped pump pulse on the intensity of spontaneous Raman scattering and, hence, on the SRS intensity, we compared the SRS process in $Ba(NO_3)_2$ ($T_2 = 26.5$ ps) and $PbMO_4$ ($T_2 = 1.77$ ps) crystals. The 15-ps pump pulses from the CLNGG laser were shorter than $T_2 = 26.5$ ps for the $Ba(NO_3)_2$ crystal and much longer than $T_2 = 1.77$ ps for the $PbMO_4$ crystal. Nevertheless, although the values of T_2 and threshold SRS energies for these crystals are substantially different, the type of the hysteresis loop in the SRS efficiency for these crystals was the same (Fig. 2).

Our comparison of the two mechanisms, which can cause the hysteresis in the SRS efficiency, suggests that the frequency-phase modulation of radiation from the CLNGG laser, which is responsible for a change in the transient SRS gain, results in the hysteresis in the dependence of the SRS efficiency on the pump energy.

Note in conclusion that the possibility of changing the frequency-phase modulation of laser radiation allows the control of the SRS gain, in particular, up to a complete SRS suppression. The method for studying SRS parameters demonstrated above can be used to investigate the parameters of laser radiation itself, namely, the dynamics of frequency-phase modulation during generation of ultrashort pulses.

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