Stimulated Raman scattering in a laser glass

V. V. Ivanov, Yu. V. Senatskiĭ, and G. V. Sklizkov
P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow

(Submitted 23 November 1987)
Pis'ma Zh. Eksp. Teor. Fiz. **47**, No. 2, 80–82 (25 January 1988)

Stimulated Raman scattering has been detected during the propagation of a 0.5-ns light pulse at a wavelength of 1.064 μm at a power density $\gtrsim 3$ GW/cm^2 through a neodymium-doped laser phosphate glass. The Stokes frequency shift is ≈ 1180 cm^{-1}. The conversion coefficient ranges up to 5%.

The stimulated Raman scattering which is observed during the propagation of an intense laser beam through a transparent solid insulator is usually observed in crystalline media such as calcite, nonlinear crystals, and double tungstates and molybdates.[1,2] Recently, stimulated Raman scattering in optical fibers has become the subject of active research.[3,4] In the present letter we report the first observation of stimulated Raman scattering in a laser phosphate glass.

In the experiments we use a laser apparatus with Nd:YAG crystals and neodymium glass, which generates a single smooth pulse at the wavelength 1.064 μm with a spectral width <0.2 cm^{-1} and a length $\tau_p = 0.5$–0.7 ns. In the case of stimulated Raman scattering involving subnanosecond pulses in a glass, the competition from stimulated Brillouin scattering is suppressed by virtue of the relations $\tau_{\rm SBS} \gg \tau_{\rm p} \gg \tau_{\rm SRS}$, where $\tau_{\rm SBS} \approx 10$ ns is the time constant of the stimulated Brillouin scattering in the glass, and $\tau_{\rm SRS} = 1/2\pi c\Delta\nu \approx 0.3$ ps is the time constant of the stimulated Raman scattering ($\Delta\nu = 16$ cm^{-1} is the homogeneous Raman linewidth[4]). A collimated beam 2.2 mm in diameter is directed through the polished ends into glass samples 7–15 cm long; the radiant energy density in the sample is 2–5 J/cm^2. We studied the spectral, angular, energy, and temporal characteristics of the light transmitted through the sample.

In the spectral measurements, IR light is focused onto the slit of a UF-90 spectrograph with a diffraction grating with a linear dispersion of 26 Å/mm in the first order. The spectra are recorded on I-3 film (to ≈ 1.3 μm in the Stokes part of the spectrum) and I-1060 film (in the anti-Stokes part) over one to ten laser shots.

At the radiant energy densities used, we observed an intense self-focusing of the laser beam in all of the samples of the phosphate and silicate glasses. This self-focusing was manifested by the appearance of filamentary damage and a scattering of the light through angles $\gtrsim 0.1$ rad. Figure 1 shows the spectra of the light transmitted through the samples. In all of the glasses studied we detected a superbroadening in the Stokes and anti-Stokes regions, caused by a nonlinear interaction of the light with the medium.[2] The superbroadening in K-8 silicate glass amounted to more than 2000 cm^{-1}. In the neodymium-doped phosphate glasses, the anti-Stokes part was significantly shorter than the Stokes part, apparently because of absorption by Nd^{3+} ions, beginning at

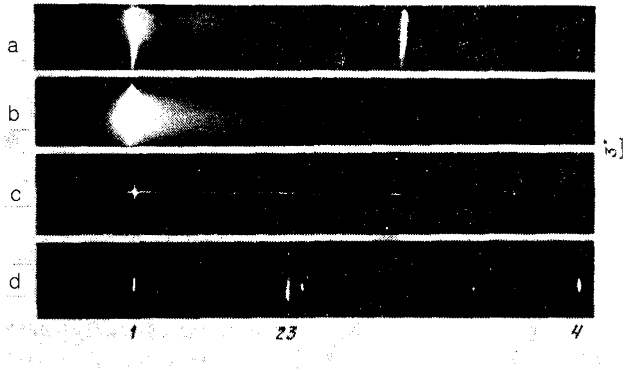

FIG. 1. a, c—Spectra of the light (over one shot) transmitted through a sample of Li-La-Nd-phosphate glass; b—LGS-55 glass; d—calibration lines of (1) an Nd:YAG laser at 1.064 μm and (4) 1.318 μm and (2) from a He-Ne laser at 1.152 μm and (3) 1.160 μm.

wavelengths ≈ 0.9 μm. A superbroadening of this magnitude has been observed previously only in experiments with picosecond pulses.[2]

In a concentrated Li-La-Nd-phosphate glass[5] (the length of the sample was $l = 7$ cm, and the neodymium concentration was $N = 8 \times 10^{20}$ cm^{-3}), at a pulse power density $\gtrsim 3$ GW/cm^2 averaged over the beam cross section, we detected a stimulated-Raman-scattering line at the wavelength ≈ 1.217 μm (Fig. 1a). The Stokes shift ≈ 1180 cm^{-1} corresponds to the phonon peak of valence vibrations of the phosphate tetrahedra of the PO_4 group at this wavelength.[6] The linewidth fell in the range 1.5-5 nm, depending on the power of the pump pulse. We did not detect an anti-Stokes line in the stimulated Raman scattering. Under the same conditions, there was no stimulated Raman scattering in the spectrum of samples of GLS-23P neodymium-doped phosphate glass ($l = 15$ cm, $N = 2.9 \times 10^{20}$ cm^{-3}) or the LGS-55 glass ($l = 13$ cm, $N = 5 \times 10^{20}$ cm^{-3}) (Fig. 1b). We found the same results with the K-8 silicate glass ($l = 13$ cm) in experiments in which we accumulated the results from up to ten laser shots on the photographic film. A possible reason for the lower threshold for stimulated Raman scattering in the Li-La-Nd phosphate glass is a higher degree of structural order.[5]

We studied the spectral and angular distributions of the light transmitted through the sample in experiments in which we focused the light with a cylindrical lens ($F = 10$ cm) perpendicular to the spectrograph slit. We see in Fig. 1c that most of the stimulated Raman scattering is within an angle $\lesssim 4 \times 10^{-3}$ rad (the angular aperture of the optical elements used was 3×10^{-2} rad).

To analyze the temporal profile of the pump pulse and the stimulated Raman pulse, we used a cylindrical lens to focus the light leaving the dispersive prism onto the slit of an Agat-SF-3 image converter. Selective and neutral filters isolated the light exclusively at the wavelength ≈ 1.06 μm, the Stokes component of the stimulated Raman scattering, or the pump and scattering pulses simultaneously (Fig. 2). The

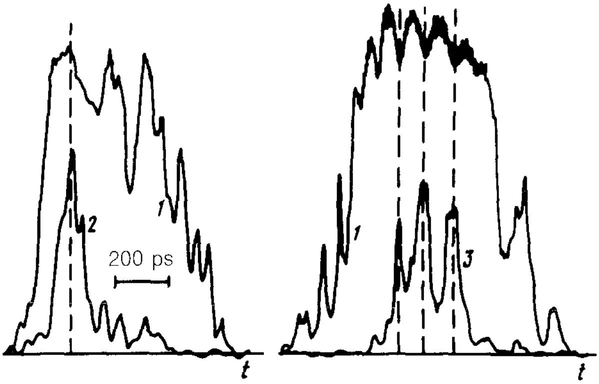

FIG. 2. Densitometer traces of (1) an intense pump pulse which has passed through the sample and the corresponding scattering at the wavelengths (2) 1.217 μm and (3) 1.064 μm.

self-focusing caused intensity dips to form on the profile of the laser pulse transmitted through the sample (because of the scattering of some of the light through large angles) (line 1 in Fig. 2). The modulation of the pulse reached a depth of 30–80% and resulted in the breakup of the pulse into "subpulses" 100–150 ps long. We observed one or two intense pulses of stimulated Raman scattering, 80–120 ps long, at the leading edge of the pump pulse, along with several considerably weaker pulses on the trailing edge. The temporal position of the Stokes pulse of the stimulated Raman scattering coincided with the maxima of the various pump subpulses (Fig. 2a), not with the self-focusing dips, which were correlated with surges in the scattered light at the wavelength 1.064 μm.

The efficiency of the conversion of the pump into the Stokes component of the stimulated Raman scattering was measured by calorimeters with the help of selective dielectric mirrors. At the exit from the sample the energy of the stimulated Raman scattering in light with a divergence $\lesssim 3\times 10^{-2}$ rad was 3–6%. In the light scattered through angles in the interval 0.1–0.4 rad (3–5% of the energy incident on the sample was scattered through such angles by virtue of the self-focusing), the fraction of the stimulated Raman scattering was slightly higher: 5–12%. We did not detect stimulated Raman scattering in the opposite direction. The smaller amplification of the stimulated Raman scattering in the opposite direction is apparently a consequence of the small effective interaction length $l_{\rm eff}$ with a pump subpulse of length $t\approx 10^{-10}$ s: $l_{\rm eff} = ct/2n \approx 1$ cm ($n = 1.55$ is the refractive index).

The observation of stimulated Raman scattering in the glass points out a possible new mechanism for losses during the propagation of a beam through the optical system of a high-power neodymium laser. In our experiments, stimulated Raman scattering was observed during a small-scale self-focusing of the main laser beam. However, the angular directionality of the stimulated Raman scattering and the fact that the stimulated-Raman-scattering pulses do not coincide in time with the scattering of the pump light (Fig. 2) are evidence that most of the stimulated Raman scattering detected is not associated with self-focusing filaments. This experiment also indicates that it might be possible to produce stimulated-Raman-scattering pulses in sources using concentrated phosphate glasses, by analogy with stimulated Raman scattering in lasers using crystals.[1] Furthermore, stimulated Raman scattering may prove to be an effective tool for studying the structure and properties of these glasses, which are now finding widespread applications in laser technology.

We wish to thank N. E. Bykovskii and M. V. Osipov for assistance in this study and P. V. Mamyshev for useful discussions.

[1]K. Andryukas, Yu. Vishchakas, V. Kabelka *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **42**, 333 (1985) [JETP Lett. **42**, 410 (1985)].
[2]S. Shapiro (editor), Ultrashort Light Pulses, Springer-Verlag, New York, 1977.
[3]E. M. Dianov, L. M. Ivanov, A. Ya. Karasik *et al.*, Zh. Eksp. Teor. Fiz. **91**, 2031 (1986) [Sov. Phys. JETP **64**, 1205 (1986)].
[4]T. T. Basiev, E. M. Dianov, Z. A. Zakhidov *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **37**, 192 (1983) [JETP Lett. **37**, 229 (1983)].
[5]B. I. Denker, V. V. Osiko, P. P. Pashinin, and A. M. Prokhorov, Kvant. Elektron. (Moscow) **8**, 469 (1981) [Sov. J. Quantum Electron. **11**, 289 (1981)].
[6]M. E. Zhabotinskiĭ (editor), Lazernye Fosfatnye Stekla (Phosphate Laser Glasses), Nauka, Moscow, 1980.

Translated by Dave Parsons