# *STIMULATED RAMAN SCATTERING OF NEODYMIUM LASER RADIATION IN LIQUID NITROGEN*

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Submitted July 12, 1968

Zh. Eksp. Teor. Fiz. 56, 430–434 (February, 1969)

Efficient (up to 25%) transformation of the radiation emitted by a Q-switched neodymium laser into the Stokes component was observed during stimulated Raman scattering in liquid nitrogen. The transformation efficiency and threshold pumping intensities were practically independent of the total spectral line width of the exciting radiation which varied between $10^{-3}$ and 50 $cm^{-1}$.

NEODYMIUM lasers are very rarely used in experiments on stimulated Raman scattering. We are aware only of the work of Martin and Thomas[1] and Johnson et al.,[2] in which stimulated Raman scattering was investigated in various organic liquids and in gaseous hydrogen. The need to use a narrow spectral line of the exciting radiation was especially stressed in[2] and therefore excitation (pumping) was provided by a $CaWO_4:Nd^{3+}$ laser, which had a narrow spectra line with $\Delta\omega_L \lesssim 0.5$ $cm^{-1}$.

The block diagram of the apparatus used in the present investigation is shown in Fig. 1. A light beam of $\sim 5 \times 10^{-3}$ divergence, provided by a Q-switched neodymium laser, was focused by a lens L (with a focal length of 75 cm) on the center of a cuvette 30 cm long filled with liquid nitrogen. The pumping energy, reaching the nitrogen-filled cuvette, was varied by means of optical filters F and measured with a calorimeter $C_1$. The pulse duration was monitored with a coaxial photocell. The spectral composition of the radiation was investigated with an STÉ-1 spectrograph or with a Fabry-Perot etalon. A selective filter, SF, was placed at the exit of the nitrogen-filled cuvette; this filter reflected practically completely the neodymium laser radiation but transmitted 80% of the energy of the first Stokes component of the stimulated Raman scattering whose wavelength was $\lambda_{1St} = 1.415$ $\mu$. The energy of this Stokes component was measured with a calorimeter $C_2$.

The pumping was provided by several types of laser and their principal characteristics are listed in the table. The table shows that the duration of pulses

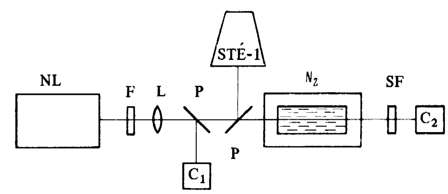


FIG. 1. Block diagram of the apparatus employed. NL is a Q-switched neodymium laser; F represents the neutral optical filters; L is a lens with f = 75 cm; P represents the plane-paralell plates; $C_1$ is a calorimeter for measuring the pumping pulse energy; STÉ-1 is a spectrograph; $N_2$ is a curvette with liquid nitrogin, SF is a selective optical filter, reflecting completely the neodymium laser radiation and transmitting 80% of the energy of the first Stokes component of the stimulated Raman scattering; $C_2$ is a calorimeter for measuring the energy of the Stokes component.

emitted by type I laser varied, from one experiment to another, within the limits 25—50 nsec. The number of light pulses, which were sometimes superimposed to one another, was difficult to determine exactly from the oscillograms. In view of this, estimates of the pumping intensity for type I laser were very approximate. The experimental dependence of the Stokes component yield on the pumping provided by type I laser (cf. the table) is shown in Fig. 4. Line 1 represents this dependence as a function of the pumping energy, while line 2 represents it as a function of the intensity. Although the scatter of the experimental points for line 2 is less than that for line 1, it is still considerably greater than the accuracy of the measurements (approximately 10%). The spectral line emitted by laser I

| Laser No. | Active element | Q-switch of resonator | Total width of spectral line | Pulse duration $\tau$, nsec | Figure showing spectrum |
|---|---|---|---|---|---|
| I | Neodymium glass | Saturable filter (transmission ~ 8–9%) | $\Delta\omega_{L\ I} \approx 10^{-3}$ $cm^{-1}$ | 25÷50 | Fig. 2a Fig. 3d |
| II | $CaWO_4:Nd^{3+}$ | Rotating prism | $\Delta\omega_{L\ II} \lesssim 0{,}5$ $cm^{-1}$ | 30 | Fig. 2a |
| III | Neodymium glass | Kerr cell + saturable filter with 50% transmission | $\Delta\lambda_{L\ III} \approx 30$ Å | 35–40 | Fig. 3a |
| IV | Neodymium glass | Kerr cell | $\Delta\lambda_{L\ IV} \approx 40$ Å | 40 | Fig. 3b |
| V | Neodymium glass | Rotating prism | $\Delta\lambda_{L\ V} \approx 50$ Å | 60 | Fig. 3c |

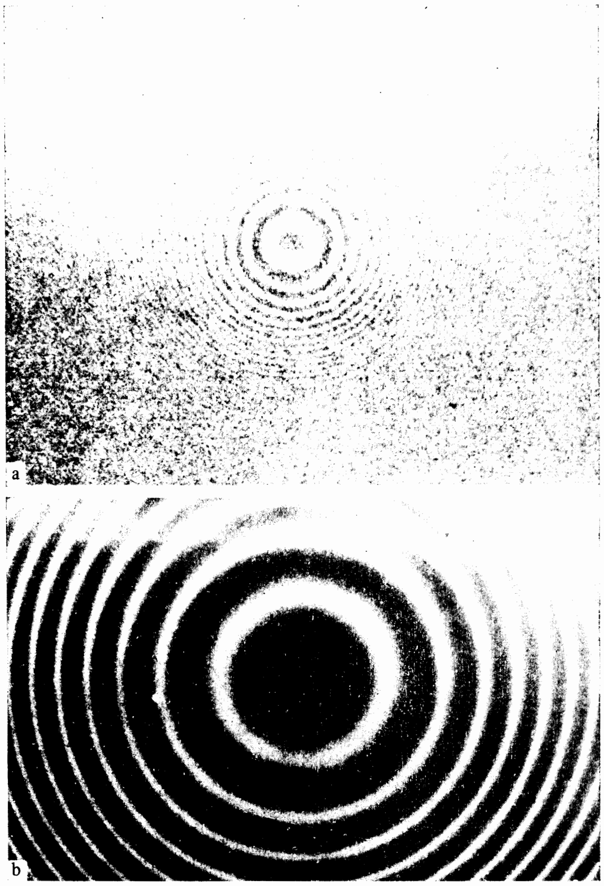


FIG. 2. a) Interferogram of the radiation of a neodymium laser, Q-switched by a saturable filter; the distance between the mirrors in the Fabry-Perot etalon was 15 cm. b) Interferogram of the radiation of the $CaWO_4:Nd^{3+}$ laser, Q-switched by a rotating prism; the distance between the mirrors in the Fabry-Perot etalon was 0.3 cm.

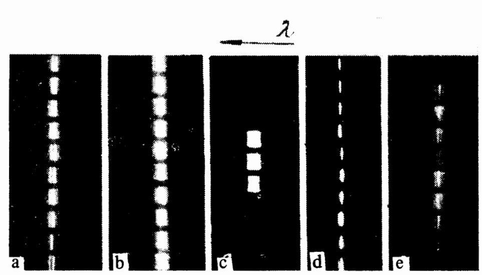


FIG. 3. Spectrograms of the radiation emitted by neodymium glass lasers with different types of Q-switches: a) a Kerr cell and a saturable filter with an initial transmission of 50%; b) a Kerr cell; c) a rotating prism; d) a saturable filter with an initial transmission of 8–9%; e) radiation of the first anti-Stokes component ($\lambda_{1aSt} = 0.85\mu$) of the stimulated Raman scattering, in liquid nitrogen, of the radiation of a type III (cf. table) neodymium laser.

(Q-switched by a saturable filter) could consist of one or several components (Fig. 3d). Analysis of the energy yield of the Stokes component and of the spectral composition of the pumping radiation showed that the scatter of the experimental points was not due to

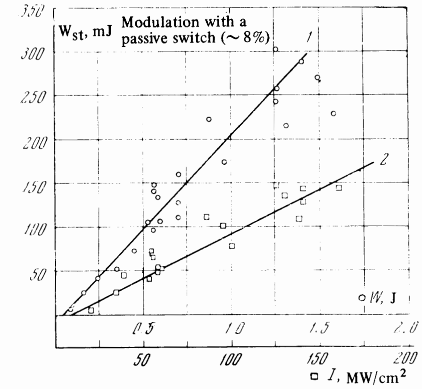


FIG. 4. Dependence of the energy of the Stokes components of the stimulated Raman scattering in liquid nitrogen on the radiation of a neodymium laser Q-switched by a saturable filter on the energy of the pumping pulse (line 1) and on the intensity of the exciting radiation (line 2).

changes in the spectrum of the exciting radiation. Evidently, it was entirely due to variations in the pumping intensity, which were due to changes in the laser pulse duration.

The dependences of the energy of the Stokes component on the pumping energy for other types of laser (Nos. II-V in the table) are given in Fig. 5. The duration of the light pulses provided by these lasers was stable and the scatter of the results was due to the experimental errors.

In a visual study of the spectral lines, a beam emerging from the nitrogen-filled cuvette was passed through a lithium niobate crystal and the STÉ-1 spectrograph. The second harmonic of the neodymium laser at $\lambda_{Nd}^{(2)} = 0.53\ \mu$ and the second harmonic of the first Stokes component at $\lambda_{1St}^{(2)} \approx 0.7\ \mu$ were observed at the output of the spectrograph (using an image

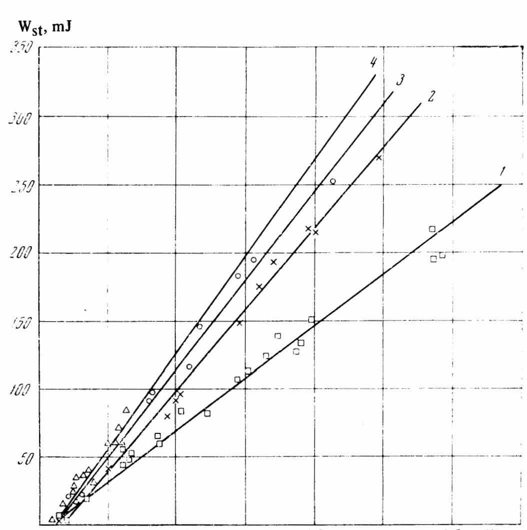


FIG. 5. Dependence of the energy of the Stokes component of the stimulated Raman scattering in liquid nitrogen on the energy of pumping pulses emitted by lasers with different Q-switches: 1) a rotating prism; 2) a Kerr cell; 3) a Kerr cell with a saturable filter; 4) $CaWO_4:Nd^{3+}$, Q-switched by a rotating prism.

converter). The observed spectrum contained also the first anti-Stokes line at $\lambda_{1aSt} = 0.85\ \mu$ (Fig. 3e). The spectral widths of the Stokes and anti-Stokes lines were practically equal to the widths of the exciting radiation lines (Fig. 3d).

Thus, an analysis of the experimental data shows that the efficiency of the transformation (~25%) into the Stokes components and the threshold intensities (~6–10 MW/cm²) for the stimulated Raman scattering of the neodymium laser radiation in liquid nitrogen are practically independent of the total spectral width of the pumping line. The values of the threshold intensities (6–10 MW/cm²) are in agreement with the corresponding intensities found in the excitation of liquid nitrogen with the ruby laser radiation[3] This is quite unexpected and it cannot be interpreted in terms of amplification in the stimulated Raman scattering. In fact, the gain $\gamma_1$ of the first Stokes component is of the form

$$\gamma_1 = \lambda_{1\,st}^2\, \sigma IN/4\Delta\omega,$$

where $\lambda_{1St}$ is the wavelength of the first Stokes component; $\sigma$ is the total Raman scattering cross section; N is the concentration of molecules in the scattering medium; I is the intensity of the pumping radiation (the density of the photon flux);

$$\Delta\omega = \int G(\omega) g(\omega) d\omega ,$$

where $G(\omega)$ is the form-factor of the pumping source line; $g(\omega)$ is the form-factor of the line of the corresponding (vibrational) transition in a molecule. We can show that

$$\Delta\omega = \begin{cases} \Delta\omega_{SPRS} & \text{for} \quad \Delta\omega_{SPRS} \gg \Delta\omega_L, \\ \Delta\omega_L & \text{for} \quad \Delta\omega_{SPRS} \ll \Delta\omega_L, \end{cases}$$

where $\Delta\omega_{SPRS}$ and $\Delta\omega_L$ are, respectively, the widths of the spectral lines of the spontaneous Raman scattering (the vibrational transition in a molecule) and of the pumping source. Since, according to[4], $\Delta\omega_{SPRS}/2 = 0.11\ \text{cm}^{-1}$ for liquid nitrogen, it follows that $\Delta\omega_L \gg \Delta\omega_{SPRS}$ (cf. table) for all the neodymium lasers (except for the laser with a saturable filter) used in the present investigation. The large differences in $\Delta\omega_L$ for different lasers should have resulted in considerable differences in the value of the gain $\gamma_1$ and, consequently, in completely different threshold pumping intensities. In particular, in the case of the Q-switching of a neodymium laser resonator by a saturable filter, when $\Delta\omega_L/2\pi \approx 10^{-3}\ \text{cm}^{-1} \ll \Delta\omega_{SPRS}$, the threshold intensity should be considerably lower than in the other cases of the Q-switching when $\Delta\omega_L/2\pi \approx 50\ \text{cm}^{-1} \gg \Delta\omega_{SPRS}$, but this is not confirmed experimentally. The cause of such behavior of the stimulated Raman scattering of the laser radiation in liquid nitrogen is not yet clear.

We also found that the first Stokes component ($\lambda_{1St} = 1.415\ \mu$) was generated when a cuvette filled with liquid nitrogen was placed in a Fabry-Perot resonator. The reflectivities of the mirrors at the wavelength $\lambda_{1St} = 1.415\ \mu$ were 75 and 30%.

The authors are deeply grateful to Academician N. G. Basov for his interest in this investigation.

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Translated by A. Tybulewicz
54