Multicomponent doubled-frequency scattering excited in ZnSe by a single picosecond laser pulse

V. A. Zyul'kov and V. P. Gribkovskiĭ
Institute of Physics, Academy of Sciences of the Belorussian SSR

(Submitted 10 January 1983)
Pis'ma Zh. Eksp. Teor. Fiz. **37**, No. 4, 179–182 (20 February 1983)

Multicomponent scattering at twice the laser frequency has been observed, for the first time, when ZnSe single crystals are pumped by a single picosecond first-harmonic pulse from a neodymium laser.

PACS numbers: 79.20.Ds, 78.45. + h, 42.65.Cq, 42.60.He

Stimulated Raman scattering by polaritons and second-harmonic generation can occur in crystals lacking an inversion center at high pump levels.[1] It is thus possible to arrange a situation in which the second harmonic serves as a test wave for probing stimulated Raman scattering by polaritons. This letter reports the first observation of this scattering.

The zinc selenide single crystal, a plane-parallel plate ~ 1 mm thick ($T = 300$ K), is pumped with a single picosecond pulse ($\tau_p \cong 8$ ps) in the first harmonic ($\hbar\omega_1 = 1.17$ eV) of a neodymium laser. The "forward" scattered light at twice the laser frequency is detected photographically on a spectrograph or by means of a photomultiplier and an oscilloscope.

The integrated intensity of the signal in the band near $2\omega_l$ increases quadratically with the pump power level, reaching saturation at a high pump level. At a pump intensity $I_{\omega_0} \cong 8$ GW/cm^2 the emission spectrum consists of a line at the frequency corresponding to $2\omega_l$ and two shifted bands: Stokes and anti-Stokes bands, the latter

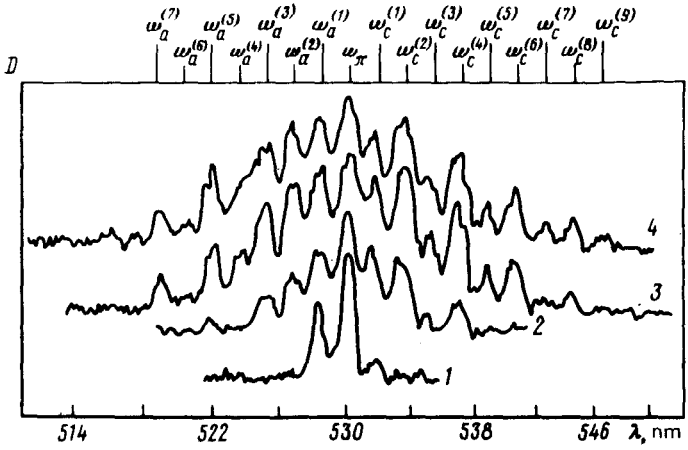

FIG. 1. Photomicrographs of the nonlinear-scattering spectra in ZnSe. 1—$I_\omega = 8$; 2—45; 3—90; 4—220 GW/cm^2.

 0021-3640/83/040213-02$01.00

significantly more intense than the former (1 in Fig. 1). With increasing pump power, the number of shifted bands increases. The spectrum stretches out toward both lower and higher frequencies. The intensities of the Stokes and anti-Stokes components become more nearly equal; the anti-Stokes components are more intense in the odd-numbered bands, while the Stokes components are more intense in the even-numbered bands. The band intensity falls off in a nonmonotonic fashion with increasing band index: $I_a^{(7)} > I_a^{(6)}$, $I_a^{(5)} > I_a^{(4)}$, $I_c^{(2)} > I_{c'}^{(1)}$ etc. (3 and 4 in Fig. 1). In addition to the discrete lines there is a broad structureless band with a maximum near $2\omega_l$ (Fig. 2). At $I_\omega = 28 I_{\omega_0}$, up to nine Stokes and seven anti-Stokes components are detected.

The occurrence of multicomponent emission at twice the laser frequency during intense pumping of a semiconductor by single picosecond pulses can be attributed to the simultaneous occurrence of second-harmonic generation and scattering of the $2\omega_l$ light by coherent polaritons excited in a parametric interaction of three waves coupled by the nonlinearity of the medium: the pump wave (ω_l), the Stokes wave (ω_{lc}), and the polariton wave (ω_p) (three-photon cascade processes). There is also a scattering of the

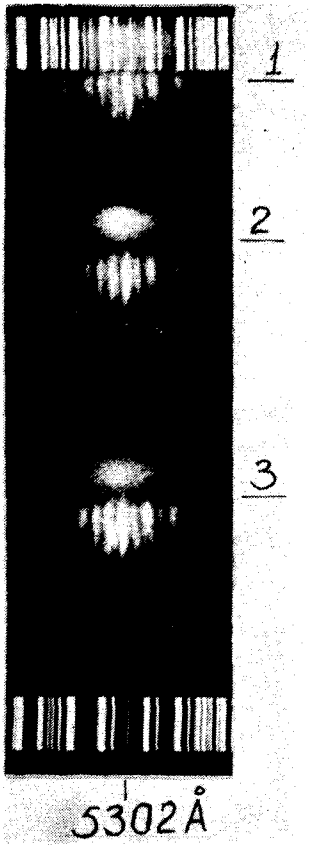

FIG. 2. Photographs of nonlinear-scattering spectra. 1—$I_\omega = 220$; 2—60; 3—90 GW/cm^2. Shown at the top and bottom is the spectrum of iron, used for calibration.

doubled frequency in four-photon direct processes, which do not reflect polariton excitations. The first type of scattering results from the quadratic nonlinear susceptibility of the medium, while the second results from the cubic nonlinear susceptibility ($\chi^{(2)}$ and $\chi^{(3)}$, respectively). The relationships among the frequencies for the interactions responsible for the excitation of the first Stokes and anti-Stokes components are as follows:

$$\omega_l + \omega_l = \omega_\pi \quad \text{(a)}$$
$$\omega_l = \omega_{lc}^{(1)} + \omega_p \quad \text{(b)}$$
$$\omega_l + \omega_p = \omega_{la}^{(1)} \quad \text{(c)}$$
$$2\omega_l = \omega_{lc}^{(1)} + \omega_{la}^{(1)} \quad \text{(d)}$$

$$\omega_\pi = \omega_c^{(1)} + \omega_p \quad \text{(e)}$$
$$\omega_\pi + \omega_p = \omega_a^{(1)} \quad \text{(f)}$$
$$\omega_{lc}^{(1)} + \omega_\pi = \omega_l + \omega_c^{(1)} \quad \text{(g)}$$
$$\omega_l + \omega_\pi = \omega_{lc}^{(1)} + \omega_a^{(1)} \quad \text{(h)}$$
$$\omega_l + \omega_{lc}^{(1)} = \omega_c^{(1)} \quad \text{(i)}$$
$$\omega_l + \omega_{la}^{(1)} = \omega_a^{(1)} \quad \text{(j)}$$

The Stokes emission $\omega_c^{(1)}$ results from three-photon processes (b, e, and i) and four-photon processes (g). The anti-Stokes wave $\omega_a^{(1)}$ is excited primarily by a direct four-photon process (h).

Because of the coherence of the scattering, the interference of the three-photon and four-photon processes can complicate the line structure at high excitation levels.[2]

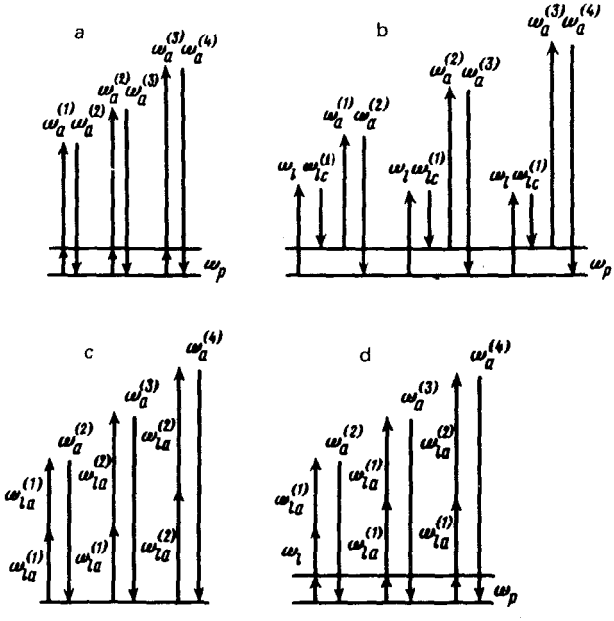

FIG. 3. Transition schemes for the excitation of the higher-order anti-Stokes scattering. a—Raman scattering by polaritons; b—direct four-photon processes; c—second-harmonic generation; d—hyper-Raman scattering. $\omega_p = 60\ \text{cm}^{-1}$.

This effect can be seen well in the spectra (3 and 4 in Fig. 1).

The fact that $\omega_a^{(1)}$ is more intense than $\omega_c^{(1)}$ (1 in Fig. 1) indicates that four-photon process (h) is predominant in the scattering of the test wave, since process (h) can contribute to the wave $\omega_c^{(1)}$ only in combination with the corresponding three-photon process (b). This situation is observed at low pump levels, at which there is the ordinary spontaneous Raman scattering of the test wave by fluctuation polaritons.[3] With increasing pump level, the threshold for stimulated Raman scattering by polaritons is reached; the scattered-light spectrum expands considerably; and the intensities of the Stokes and anti-Stokes components become more nearly equal. As before, however, the scattering caused by direct four-photon processes is stronger, as can be seen in the higher intensity of the anti-Stokes components in the odd-numbered bands and in the disruption of the monotonic decrease in the intensity of the shifted bands with increasing band index, which is typical of ordinary cascade stimulated Raman scattering. The predominance of four-photon processes can be attributed to the resonant nature of interactions g and h, since we have $\omega_\pi + \omega_l = 3.51$ eV $> E_g$ ($E_g = 2.71$ eV is the gap width of ZnSe). Figure 3 shows the transition schemes for processes which result in the excitation of the higher-order coherent anti-Stokes scattering.

The appearance of the structureless band at a high pump intensity is apparently a result of the scattering of the emission of the nonresonant electronic cubic susceptibility.

Hyper-Raman scattering is not significant in the emission at twice the laser frequency, since this type of scattering has not been observed in CdS oriented in a manner which rules out second-harmonic generation (with pump beam propagating along the optic axis of the crystal).

We wish to thank V. L. Strizhevskii for a discussion of the results.

[1]S. A. Akhmanov and N. I. Koroteev, Metody nelineĭnoĭ optiki v spektroskopii rasseyaniya sveta (Methods of Nonlinear Optics in Light-Scattering Spectroscopy) Nauka, Moscow, 1981.
[2]A. M. Panarin and V.L. Strizhevskiĭ, Kvant. Elektron. (Moscow) **5**, 1694 (1978) [Sov. J. Quantum Electron. **8**, 964 (1978)].
[3]V. L. Strizhevskiĭ and Yu. N. Yashkir, Kvant. Elektron. (Moscow) **2**, 995 (1975) [Sov. J. Quantum Electron. **5**, 541 (1975)].

Translated by Dave Parsons
Edited by S. J. Amoretty