Tunable Raman laser using oblique polaritons

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Observation of stimulated Raman scattering (SRS) of light by oblique polaritons is reported for the first time. It is shown that the use of SRS by oblique polaritons in a lithium-iodate crystal permits the frequency of the first Stokes component to be tuned in an interval of 100 cm^{-1}.

Interest in Raman scattering (RS) of light by polaritons, which is one of the possible methods of producing sources of coherent-emission with continuously tunable frequency, has increased recently. In addition, the results of such investigations contain information on the dispersion of the refractive index of the medium on the lattice vibration spectrum, and on transverse RS. Stimulated Raman scattering (SRS) by normal polaritons in a lithium niobate crystal was used in[1] to obtain a tunable frequency. We shall show below that the use of oblique polaritons makes it possible to simplify greatly the experimental setup and extend simultaneously the frequency tuning range.[2]

To tune the coherent-emission frequency we have used in the present study, for the first time, SRS by oblique polaritons. The experiments were performed with a lithium iodate ($LiIO_3$) crystal. Lithium iodate is a uniaxial optical crystal belonging to the hexagonal symmetry group, with two molecules per unit cell.[3] The vibrational spectrum of lithium iodate contains polar vibrations of two types of symmetry—nondegenerate of symmetry $A(z)$, and doubly-degenerate of symmetry (x,y), which are active in Raman scattering.[4,5] Oblique polaritons are produced in lithium iodate by simultaneous coupling of the photons with elastic transverse waves of the polar oscillations of both indicated type of symmetry, with propagation at an acute angle θ to the optical axis z (see the vector diagram in Fig. 2).[2]

Figure 1 shows a diagram of the experimental setup. The pump was the second harmonic of a mode-locked neodymium-glass laser (1). The intensity of the unfocused second-harmonic beam reached 10^8–10^9 W/cm^2 at a spectral width 70–80 cm^{-1} at half-intensity level. The harmonic was focused into the $LiIO_3$ crystal by lens L_1 having $F = 10$ cm. The scattered light was projected by lens L_2 on the slit of an ISP-51 spectrograph.

Under the conditions of our experiment, the SRS developed collinearly with the pump wave in a single pass through the crystal. The vector diagram shown in Fig. 2 corresponds to the momentum-conservation law in this case. Taking the energy-conservation law into account, the wave vector k_n of the elementary excitation responsible for the SRS is equal to

$$k_n = k_p - k_s = 2\pi\nu_n n_o(\nu_p), \tag{1}$$

where k_p and k_s are the wave vectors of the pump and scattered-light photons, respectively, ν_p and ν_n are the frequencies of the pump and of the scattering polariton, respectively, and $n_o(n_e)$ is the refractive index of the medium for the ordinary (extraordinary) ray. In the experiments were used five elements with plane-parallel end faces, in which the pump wave, when normally incident on the entrance phase, propagated at angles $\theta = 90$, 60, 45, 30, and 0° to the z axis in the xz plane.

When the pump wave propagated along the z axis, the SRS developed on the normal polaritons with frequency 750 cm^{-1}, coupled only with the oscillations of the 795 cm^{-1} mode of symmetry A. This result agrees with the data of[6,7]. The coefficient for conversion into the tunable first Stokes component was ~20%.

When obliquely-cut elements were used, the SRS developed on the oblique polaritons coupled with the two upper transverse polar oscillations—the mode $\nu_A^{TO} = 795$ cm^{-1} of symmetry A, and of the mode $\nu_E^{TO} = 769$ cm^{-1} of symmetry E_1. It should be noted that the coefficient of the conversion of the pump energy into a tunable component of SRS by oblique polaritons decreased with decreasing θ, and was only 1–2% at

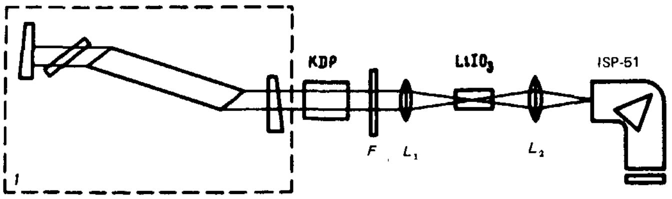

FIG. 1. Diagram of experimental installation: F—SZS-21 optical filter (to separate the 530-nm second harmonic).

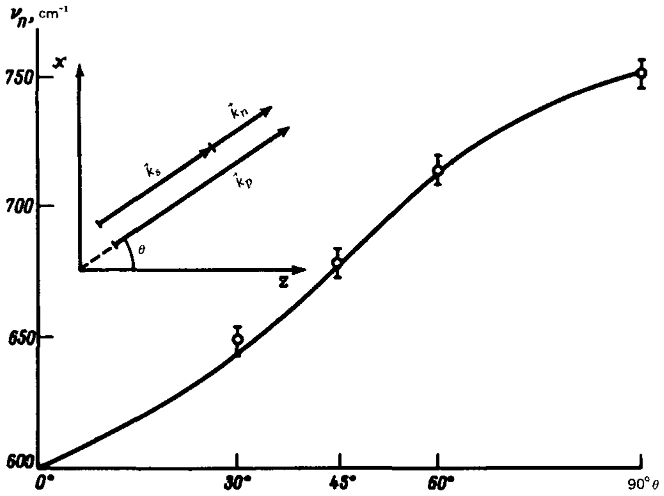

FIG. 2. Plot of frequency tuning of Raman laser with oblique polaritons of a lithium-iodate crystal.

$\theta = 30°$. Furthermore, at $\theta = 20°$ and $\theta = 0°$ no SRS by oblique polaritons developed, owing to the competition on the part of the components of the SRS on the nonpolar phonons of the 824 cm^{-1} mode of symmetry E_2.[4] A tendency was also observed for the spectral width of the tunable radiation to decrease with decreasing angle θ. When the angle changed from 90 to 30° the spectral width decreased from 20 to 10 cm^{-1}.

Figure 2 shows a plot of the frequency shift of the Stokes radiation against the angle θ. As seen from the figure, the experimentally obtained values of the frequency shifts ν_n are well approximated by the curve calculated from formula (2) for inclined polaritons, with allowance for only the upper branches of the polar oscillations[2]:

$$\mathrm{tg}^2\theta = \frac{\left(\dfrac{2\pi\nu_n}{k_n}\right)^2 - \dfrac{1}{n_0^2(\nu_p)}\,\dfrac{(\nu_E^{TO})^2 - \nu_n^2}{(\nu_E^{LO})^2 - \nu_n^2}}{\left(\dfrac{2\pi\nu_n}{k_n}\right)^2 - \dfrac{1}{n_e^2(\nu_p)}\,\dfrac{(\nu_A^{TO})^2 - \nu_n^2}{(\nu_A^{LO})^2 - \nu_n^2}}, \qquad (2)$$

where ν_A^{LO} and ν_E^{LO} are the frequencies of the longitudinal optical phonons of symmetry A and E_1, respectively.

It is seen from the presented data that the attained tuning range is 100 cm^{-1}. Obviously, all the intermediate values of ν_n between 650 and 750 cm^{-1} can be obtained by continuously varying the angle θ. This can be easily done by using a cylindrical sample placed in an immersion liquid.

We note also that the use of oblique cuts makes it possible to construct a tunable source of coherent optical radiation using also longitudinal phonon polar oscillations.

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