NOTIZEN

Stimulated Raman Scattering from a Polariton in $LiJO_3$

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Stimulated Raman scattering from the polariton associated with the phonon of symmetry A at 793 cm^{-1} was observed at 753 cm^{-1} from the ruby exciting line for 0° scattering, together with the second Stokes component at 1506 cm^{-1}.

KURTZ and GIORDMAINE [1] were the first to observe stimulated Raman scattering (SRS) from polaritons in $LiNbO_3$. Another group at Stanford succeeded in tuning the SRS Stokes emission from polaritons in $LiNbO_3$ through 200 cm^{-1} by changing the angle between the incident laser beam and the direction of highest Raman gain within [2] and without [3] a resonator, and also detected the infrared emission of the idler wave [3]. Since then other materials with polariton shifts are being investigated in order to find intense tunable light sources in the visible and infrared.

CLAUS [4–6] has studied the phonon and polariton spectrum of $LiJO_3$ with argon laser excitation. From these data it seemed to be promising to search for stimulated Raman scattering from polaritons also in this crystal.

Experimental

Three crystal samples were used in these investigations. They were plates of a thickness of 5 mm in the direction of the optical z-axis. Two samples with a usable length of 19 mm perpendicular to the z-axis were grown by Haussühl, another sample with a length of 21.5 mm was obtained from a commercial source. Two opposite faces of the crystals were polished approximately parallel.

The spectra were excited with a passively Q-switched 6″-ruby laser giving pulses of 20 nsec duration and a linewidth of ~ 0.02 cm^{-1}. The power was varied by an aperture in the resonator. The laser beam was focussed by a lens with $f = 60$ cm into the crystals. The transmitted light illuminated a diffusing screen, an image of which was projected on the entrance slit of a 1 m Jarrell-Ash Czerny-Turner Spectrograph. Kodak I-N plates were used to photograph the spectra.

Results and Discussion

The most suitable scattering geometry was found by 0° scattering experiments with a 50 mW He-Ne laser. Figure 1 shows the spectra obtained with the divergence of the scattered light limited to less than 2° about the forward direction. They were recorded with a 1 m RSV double monochromator. The upper trace corresponds to $x(yy)x$, the lower to $x(zz)x$ scat-

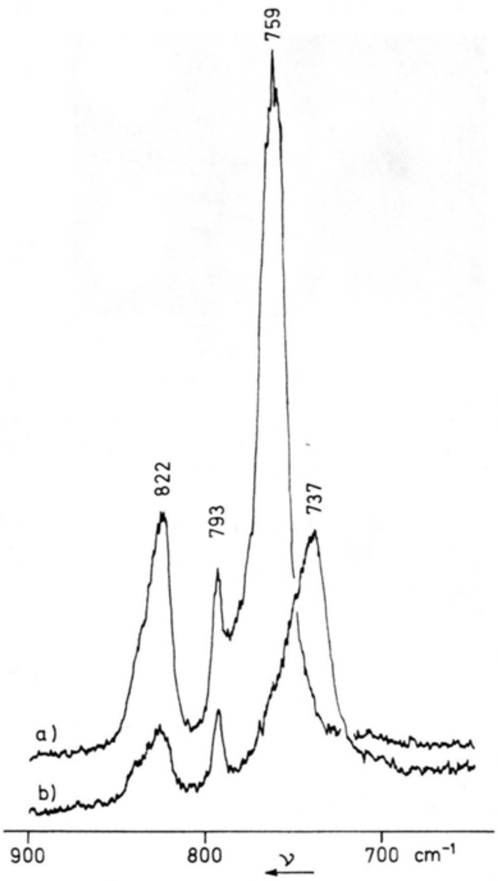

Fig. 1. Raman scattering of $LiJO_3$ into a cone of $< 2°$ around the forward direction with 50 mW He-Ne laser excitation. a) $x(yy)x$ scattering, b) $x(zz)x$ scattering. Slit 3 cm^{-1}, time constant 1 sec, scanning speed 45 cm^{-1}/min.

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tering geometry. The line at 793 cm^{-1} is caused by diffuse or reflected backscattering and indicates the position of the TO phonon line for large angle scattering. The feature with a peak at 822 cm^{-1} has the position of the LO phonon the directional dependence of which was studied by OTAGURO et al. [7]. Because the extraordinary refractive index is smaller for $LiJO_3$ than the ordinary [8], the observable polariton shift is greater for $x(zz)x$ scattering [5]. However, due to the magnitude of the tensor components the scattering cross section is by more than a factor of 2 greater for ordinary incident light and therefore it should be easier to observe SRS from the polariton in the $x(yy)x$ geometry.

This expectation was confirmed by the experiments. While attempts to observe SRS for extraordinary incident light were not successful, the spectra shown in Fig. 2 were obtained with ordinary incident light. Due to the high gain for 180° scattering [1, 9] the line with the lowest threshold of approximately 5 MW is the phonon line appearing at 793 cm^{-1} from the ruby exciting line at 694.3 nm in the upper spectrum. With a threshold of approximately 10 MW stimulated scattering from the polariton was observed at 753 cm^{-1} as shown in the lower spectrum. In three spectra the threshold for observation of the second Stokes component of the polariton line at 1506 cm^{-1} was reached. It is faintly visible in the reproduction of the lower spectrum.

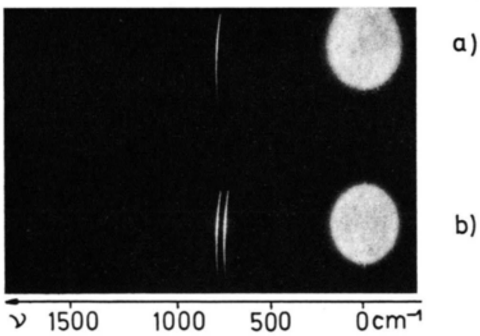

Fig. 2. Stimulated Raman spectra of $LiJO_3$ excited with a giant pulse ruby laser in $x(y, y+z)x$ scattering geometry. a) 8 MW, 180° phonon scattering, b) 20 MW, 0° polariton +180° phonon scattering. The ruby line was suppressed by a Schott RG 10 filter.

A more thorough quantitative investigation was precluded by the heavy optical damage occurring in the crystals at every laser shot. A new light path through the crystal had to be chosen for each spectrum. This had a detrimental effect on the reproducibility of the measurements. The thresholds were slightly higher for the shorter crystals, but otherwise no difference in the spectra and in the damage behaviour was found between the crystals from different sources.

The small range of 40 cm^{-1} for the polariton shift and the heavy optical damage at the power densities required make stimulated polariton scattering in $LiJO_3$ not very promising as tunable light source. In order to make use of the greater shifts expected for $x(zz)x$ scattering even higher laser power would be necessary.

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