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STIMULATED RAMAN SCATTERING BY POLARITONS

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Stimulated Raman scattering from polaritons has been observed in $LiNbO_3$. The measured splitting of 134 cm^{-1} between the 0° and 180° stimulated emission is in good agreement with infrared dispersion of the 628-cm^{-1} A_1 TO phonon. Threshold powers for the forward and backward stimulated emission are in fair agreement with the theory of polariton-stimulated Raman gain.

Coupling between the transverse optical (TO) lattice modes and the electromagnetic field in crystals leads to a strong dependence of the lattice mode frequency on wave vector.[1] The region of strong coupling, in which the lattice excitation (polariton) is of mixed phonon and photon character, has been widely observed in near-forward spontaneous Raman scattering[2-4] in crystals lacking inversion symmetry. We report in this Letter the observation of stimulated Raman scattering (SRS) from polaritons. Forward SRS at the polariton frequency has been generated in $LiNbO_3$, and, at slightly higher threshold power, backward SRS at the TO phonon frequency. Although SRS has been reported earlier[5] in other noncentrosymmetric crystals, the expected polariton character, as shown by the frequency splitting between the 0° and 180° stimulated emission, has not been previously observed.

Our $LiNbO_3$ crystal was cut to 4.4 cm length from an a-axis $[11\bar{2}0]$ boule[6] substantially free of multiple domains, with end faces polished approximately normal to the boule axis. The incident laser beam was polarized in the z direction and travelled in the xy plane at an angle from the boule axis of ~1°, sufficient to decouple surface reflections. The incident light was generated as 40-nsec pulses by an internally apertured 6943-Å ruby laser Q-switched by a Pockels cell, and focused near the crystal by a lens of 1-m focal length. The laser beam was in a single transverse mode with approximately Gaussian distribution; its spectrum consisted mainly of two or three sharp lines of width $\lesssim 0.05$ cm^{-1}, spaced by 0.35 cm^{-1}. At the crystal the beam diameter at half intensity was $(2.9 \pm 0.5) \times 10^{-2}$ cm.

Forward (polariton) SRS appeared at a threshold power of 0.95 ± 0.15 MW, corresponding to a power density at the beam center of $(1.0 \pm 0.2) \times 10^9$ W cm^{-2}. Threshold power was taken to be the laser power at which SRS conversion efficiency reached 0.02%. Backward (phonon) SRS was observed at threshold power of 1.3 ± 0.2 MW. At laser powers $\gtrsim 1.5$ MW, 0° and 180° scattering occurred together with 180° scattering having the higher efficiency. At a laser power of 1.7 MW the phonon SRS conversion efficiency was approximately 3% while the polariton efficiency was 0.1% reproducible to within a factor of 3. The SRS duration was ~20 nsec and coincided with the peak of the peak of the laser pulse.

Figure 1 shows typical SRS spectra. The 180° emission occurs near 631 cm^{-1}, which is in good agreement with the frequency of the A_1-symmetry TO mode of highest peak Raman cross section, as observed[7,8] in large-angle Raman scattering.

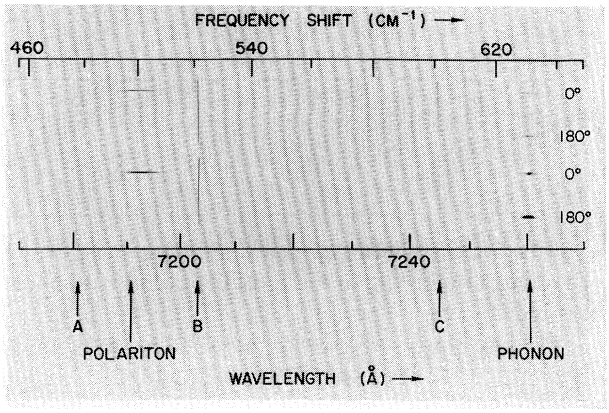

FIG. 1. SRS spectra of $LiNbO_3$, with 6943-Å laser beam near $[11\bar{2}0]$ direction. Upper pair and lower pair of spectra were obtained with single laser pulses. The spectra were obtained with single laser pulses. The spectra were observed in eighth order of a 2-m Harrison grating spectrometer. Calibration lines A,B,C indicate 7180.9 Å (Ne 6383.0 Å in ninth order), 7202.5 Å (Ne 6402.3 Å in ninth order), and 7245.2 Å (Ne eighth order). The optical train attenuated phonon SRS by a factor of 4 relative to polariton SRS. Apparent 0° phonon emission is 180° phonon SRS reflected at the laser (see text).

The measured spectral width of 2-4 cm^{-1} can be compared with the value of 15-20 cm^{-1} observed for spontaneous Raman scattering.[7] The 0° SRS emission occurs near 497 cm^{-1}, in agreement with recent observations[4] of forward spontaneous Raman scattering in $LiNbO_3$. This frequency may be compared with the value 490 cm^{-1} calculated[2,4] from the infrared data of Barker and London, Schaufele and Weber, and Axe and O'Kane[8] for 0° emission and 6943-Å pump. Both 0° and 180° spectra consisted of bands of lines regularly separated by the 0.35-cm^{-1} laser line spacing. The forward emission had complex angular structure, but emerged within a cone of ~0.6° external diameter; its spectral width was about 10 cm^{-1} which may be compared with the width of 1.3 cm^{-1} calculated from the polariton dispersion curve for a 0.6° cone.

No anti-Stokes emission was observed. Stimulated emission with lower efficiency was also detected near 250 cm^{-1}, the frequency of the remaining strongly Raman-active A_1-symmetry mode.[7] A prominent and unexpected feature of the 0° spectra in Fig. 1 is the apparent radiation at the 631-cm^{-1} TO-phonon frequency. This radiation is accounted for by 180° emission reflected at the laser resonator mirrors and retransmitted through the crystal. Relative arrival-time measurements at the spectrometer showed that the 631-cm^{-1} radiation was delayed by 23 nsec with respect to the 497-cm^{-1} radiation, in good agreement with the 7-m round-trip distance between the crystal and the laser.

Crystal damage in the form of filamentary tracks was produced by each laser pulse above SRS threshold, i.e., between 1 and 3 MW. Subsequent illumination of the tracks by a 6328-Å He-Ne laser showed that scattering from the damaged region was confined primarily to the plane containing the z axis and the incident direction; furthermore, the damage was observed to disappear after annealing at 200°C. This behavior resembles that of refractive-index inhomogeneity damage[9] widely observed in $LiNbO_3$ irradiated by low-power cw lasers; irreversible damage of the type frequently observed in solids with high-power lasers was only rarely observed.

Near-field photographs of the exit face of the crystal showed that the SRS emission area had a diameter of 200-400 μ, similar to the laser-beam diameter. Possible evidence for smaller filaments was seen on some photographs; however, the fact that the measured gain described below does not exceed the calculated gain suggests that while some self-focusing[10] may be present, it plays a secondary role in the present experiment.

The theory of polariton-stimulated Raman gain has been discussed by several authors,[11,12] and the 180° gain has been calculated for $LiNbO_3$ from measured Raman cross sections.[13] The geometry of our experiments restricts significant overall Raman gain to the 0° and 180° regions. At room temperature the expected gain[13] at the peak of the 631-cm^{-1} line is 9.4×10^{-9} cm^{-1} $(W\ cm^{-2})^{-1}$ corrected for the 6943-Å pump wavelength. By averaging the gain over the pump-beam profile[14] and using the observed threshold power of 1.3 ±0.2 MW, the effective power gain for the 631-cm^{-1} Stokes wave is found to be 6.4 cm^{-1}, and the total gain 28. The effective quantum noise power entering the crystal in a beam of diameter 2.9×10^{-2} cm, full angular width 0.6°, and spectral width 2 cm^{-1} is 0.1 μW; thus the gain required to reach threshold (~300 W) in our experiments is 22, to be compared with the calculated value of 28. Factors contributing to the difference include induced-refractive-index inhomogeneity and uncertainty in the absolute Raman-scattering cross sections.

The 0°:180° SRS gain ratio can be calculated from the theory of Henry and Garrett.[12] They ob-

tain a ratio of power gains

$$R=\frac{g_0(\omega_L-\omega_1)}{g_{180}(\omega_L-\omega_0)}=\left(\frac{\omega_0}{\omega_1}\right)\left(\frac{\omega_L-\omega_1}{\omega_L-\omega_0}\right)\left[\frac{n(\omega_L-\omega_1)}{n(\omega_L-\omega_0)}\right]\left[1+\left(\frac{\omega_0^2-\omega_1^2}{\omega_P^2}\right)\frac{d_E'}{d_Q'}\right]^2, \quad (1)$$

where ω_L, ω_0, ω_1 are, respectively, the angular frequencies of the laser, the TO phonon, and the polariton giving phase-matched forward scattering; $n(\omega_L-\omega_{0,1})$ are the refractive indices for forward and backward Stokes Raman light; d_E' and d_Q' are nonlinear susceptibilities appearing in the phenomenological energy-density function $-U=(d_E'/4\pi)E(\omega_L)E^*(\omega_L-\omega_{0,1})E^*(\omega_{0,1})+Ned_Q'\times E(\omega_L)E^*(\omega_L-\omega_{0,1})Q^*(\omega_{0,1})+\text{c.c.}$; and ω_P^2 is proportional to the oscillator strength of the lattice absorption. From published data on $LiNbO_3$,[4,7,8] $\omega_0/2\pi c\simeq 628\ \text{cm}^{-1}$, $\omega_1/2\pi c=492\ \text{cm}^{-1}$, $n(\omega_L-\omega_1)\approx n(\omega_L-\omega_0)=2.18$, $\omega_P/2\pi c\simeq 900\ \text{cm}^{-1}$, and $(\omega_0^2-\omega_1^2)/\omega_P^2=0.2$. If local field corrections are neglected,[15] the ratio d_E'/d_Q' can be inferred from the ratio of LO:TO 90° Raman-scattering cross sections.[15] Using the LO:TO data of Kaminow and Johnston[7] for the 631-cm^{-1} mode we obtain $d_E'/d_Q'=+0.3$, and a gain ratio $R=1.5$. An alternative and more direct calculation of d_E'/d_Q' has been carried out using experimental values of the second-harmonic generation coefficient[16] ($\propto d_E'$) and the TO 90° Raman scattering cross section[7] ($\propto d_Q'$). This calculation leads to the value $|d_E'/d_Q'|\simeq 0.4$ and from Eq. (1) a gain ratio $R=1.5$, in fair agreement with the value $R\simeq 1$ estimated from the threshold data in the present experiment. This latter value is subject to possible corrections due to short-term fluctuations in the pump power[17] and residual domain structure.

As predicted by various authors,[11,12,18] we believe that the observed polariton emission in $LiNbO_3$ is accompanied by highly coherent infrared (IR) emission near 20 μ, in spite of the high lattice absorption at the polariton frequency. This IR emission mechanism differs from that of the IR generation reported by Braunstein[19] to accompany SRS in liquid benzene. Because of the macroscopic inversion symmetry, Raman scattering and SRS from polaritons are forbidden in liquid benzene in the electric-dipole approximation. The coherence and mechanism of the IR emission from benzene remain to be established.

In conclusion, we anticipate that polariton SRS in noncentrosymmetric media will yield information on local fields[20] through the ratio d_E'/d_Q', while lending itself well to useful IR and optical oscillators[13] tunable over several hundred cm^{-1} by control of resonator feedback geometry and temperature.

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NEUTRON MULTIPLICITY–SPIN STATE CORRELATIONS FOR ^{239}Pu RESONANCES*

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Measurements were made of the fission-neutron multiplicity associated with eleven ^{239}Pu resonances in the energy range from 20 to 100 eV. It has been found that the multiplicity values are strongly correlated with the spins of the individual resonances and that the average multiplicity for the $J=0$ group is 2.6% higher than for the $J=1$ group.

It has been suggested[1] that the fission-neutron multiplicity ($\bar{\nu}$) might be a function of incident neutron energy in the resolvable resonance regions of the common fissile nuclides. We have measured the energy dependence of $\bar{\nu}$, for a ^{239}Pu sample, for incident neutrons in the range from 20 to 100 eV. The results show that the multiplicity values do vary from resonance to resonance, and fall into two distinct groups that appear to be correlated with the known spins of these levels.

For the experiment reported here the neutrons required for the time-of-flight measurements were produced by the Rensselaer 90-MeV electron linac, and the fission measurements carried out at the end of a 25-m flight path. Fission events were detected by the occurrence of a coincidence between the signals from a fission ionization chamber[2] located in the neutron beam and signals from a gadolinium-loaded liquid scintillation tank that surrounded the chamber. The fission neutrons were detected, after thermalization in the scintillant, by means of the gamma radiation emitted following neutron capture in the gadolinium. The detected neutron multiplicity for each of 256 time-of-flight channels were first stored in a buffer and then transferred to a 256×12 array in the memory of an on-line computer. Thus, if a fission event were recorded at time of flight i with ν neutrons detected, the event would have been stored in the ith time-of flight channel of the νth section of the computer memory.

Many of the techniques used in this experiment are standard and have been described in detail by Mather, Fieldhouse, and Moat,[3] Diven and Hopkins,[4] and others. The principal differences between this experiment and the others can be attributed to the necessity of working with reaction rates associated with a very broad spectrum of neutron energies rather than a monoenergetic beam. The methods used to correct our multiplicity data for time-dependent (a) background events, (b) accidental coincidences, and (c) scaler dead-time effects are briefly described below.

(a) In general, the background multiplicity per fission gate is correlated with the scattering, capture, and fission cross sections of the material present in the fission chamber. A comparison of the upper graph in Fig. 1 (background events per gate) with the lower (fission rate) indicates the character of the background correlations that we observed in this experiment. In order to determine the number of background events per gate, background sampling pulses from a free-running pulse generator were used to initiate simulated fission coincidences. The multiplicity data associated with the sampling pulses were stored in a temporary buffer. If no fission coincidence was detected during the 240-μsec interval centered on the opening of the gate, then the number of the time-of-flight channel and the value of the multiplicity were stored in an additional 256×12 array in the computer.

(b) The fission chamber and associated electronics were designed to minimize the likelihood of detecting alpha particle pile-up events. Nevertheless, there was a reasonable probability that

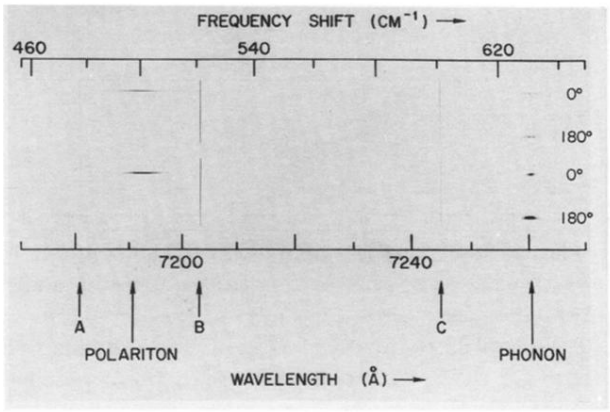

FIG. 1. SRS spectra of $LiNbO_3$, with 6943-Å laser beam near $[11\bar{2}0]$ direction. Upper pair and lower pair of spectra were obtained with single laser pulses. The spectra were obtained with single laser pulses. The spectra were observed in eighth order of a 2-m Harrison grating spectrometer. Calibration lines A, B, C indicate 7180.9 Å (Ne 6383.0 Å in ninth order), 7202.5 Å (Ne 6402.3 Å in ninth order), and 7245.2 Å (Ne eighth order). The optical train attenuated phonon SRS by a factor of 4 relative to polariton SRS. Apparent 0° phonon emission is 180° phonon SRS reflected at the laser (see text).