A/B that can be deduced from Fig. 3. By substituting S and $T_{2e}=9\times10^{-8}$ sec in Eq. (21) we have, however,

$$B/C \approx \tfrac{1}{4} T_{2e}^2 S^2 \approx 250 \to 24 \text{ dB} \quad ,$$

which is greater than the value that can be read from Fig. 3; however, because the harmonic signal at $\omega=\omega_0$ has the same intensity as the noise, the true ratio B/C in Fig. 3 can be much greater than 13 dB. On the other hand, by substituting in the equation $S=\gamma H_z (T_1/T_2)^{1/2}$ the proper values of H_x, T_1, and T_2, we obtain for S, values very close to those obtained from linewidth measurements of the $\omega\sim\omega_0$ line.

Finally, we remark that all of the previous data are taken with the magnetic field geometry of Fig. 2. By using a different magnetic field geometry, we can split the second-harmonic emission signal into a contribution that is due to $M_x(2\omega)$ and one that is due to $M_z(2\omega)$. In particular, if we operate with $\vec{H}_{rf}(\omega)$ perpendicular to $\vec{H}_0$ and $\vec{H}_{rf}(2\omega)$ parallel to $\vec{H}_0$, only the component $M_z(2\omega)$ is different from zero, and, therefore, one single line, near $\omega=\omega_0$, is expected. Actually, by using a cavity with this magnetic field geometry, the detected harmonic spectrum shows only the $\omega\sim\omega_0$ line.

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Scattering from the E_1 Polariton of $LiIO_3$

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Raman scattering from an E_1 mode polariton of $LiIO_3$ is reported. A polariton with energy between 687 and 766 cm^{-1} is observed in near-forward scattering with $y(zx)\bar{y}$ polarization. The polariton is observed only near 764 cm^{-1} in the $y(xz)\bar{y}$ spectra. The results are analyzed. It is also found that the polariton dispersion curve does not change with the orientation of the phonon wave vector.

INTRODUCTION

It was shown by Huang[1] that transverse optical phonons of ionic crystals and photons with nearly the same wave vector and energy can be strongly coupled. The resulting mixed phonon-photon states are now referred to as "polaritons." Raman scattering from the polaritons of GaP was reported for the first time by Henry and Hopfield.[2] Shortly thereafter polariton scattering from anisotropic ZnO was reported by Porto *et al.*[3]

The Raman and polariton spectra of $LiIO_3$ have been examined recently by Claus *et al.*[4] These authors stated that *no* E_1 polariton was "unambiguously" observed [although an A polariton was observed in $x(yy)x$ polarization]. We wish to report the observation of an E_1 polariton in forward scattering in $LiIO_3$ and to clarify possible difficulties in the analysis of the experiments.

$LiIO_3$ belongs to the $P6_3(C_6^6)$ space group.[5] Twen-

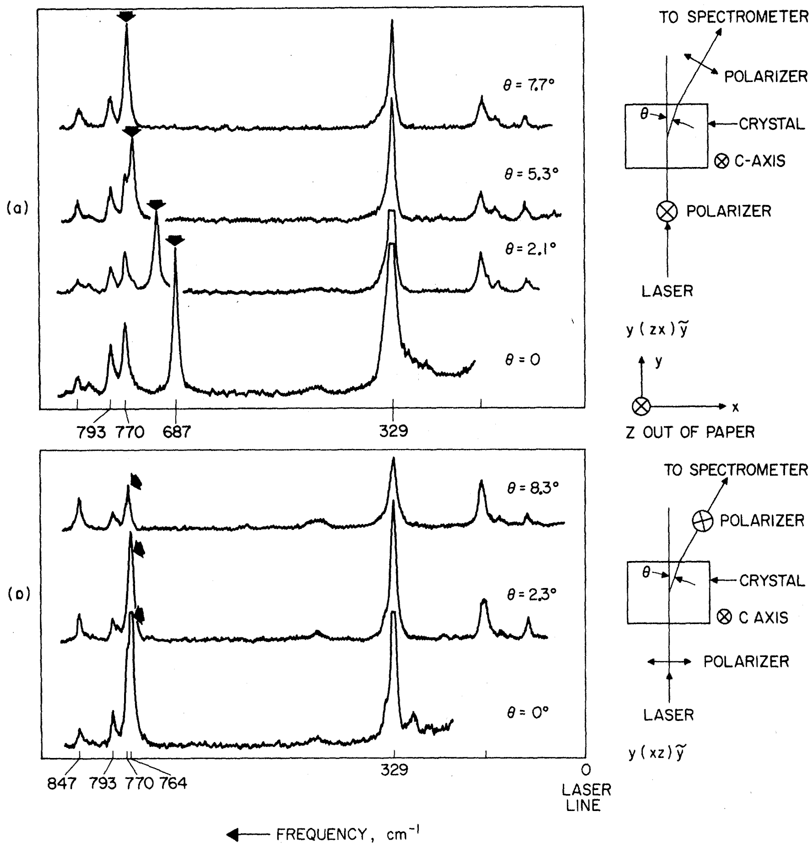

FIG. 1. Near-forward-scattering spectra for the E_1 mode of $LiIO_3$. The arrows point to the polariton line discussed in this paper. (a) $y(zx)\tilde{y}$ polarization, where $\tilde{y}$ lies in the xy plane. θ is the forward-scattering angle, i.e., the small angle between the incident and scattered photon propagation directions. (b) $y(xz)\tilde{y}$ polarization; assymmetry of the two spectra is discussed in the text.

ty-seven optical-phonon modes are allowed: $4A + 5B + 4E_1 + 5E_2$. The $A(z)$, $E_1(x)$, and $E_2(y)$ modes are both infrared and Raman active, permitting possible observation of polariton modes.

RESULTS AND DISCUSSION

The $y(zx)\tilde{y}$ and $y(xz)\tilde{y}$ forward-scattering spectra[6] are shown in Fig. 1. Figure 1(a) shows the polariton line for the $y(zx)\tilde{y}$ polarization for various forward-scattering angles, starting at 687 cm^{-1} at $\theta = 0°$. The polariton line appears to approach and coalesce with the 770-cm^{-1} line as the scattering angle θ is increased. The 5145-Å argon laser line was used.

An intense line at 764 cm^{-1}, with a slight shoulder at 770 cm^{-1}, was observed in the $y(xz)\tilde{y}$ spectra for 0° scattering. This is shown in Fig. 1(b). With increasing forward-scattering angles, the 764-cm^{-1} line decreased in intensity relative to the 770-cm^{-1} line.

The apparent asymmetry in the polarizability components α_{xz} and α_{zx} observed in Figs. 1(a) and 1(b) for the polariton is a result of the difference in refractive indices associated with the two different scattering geometries.

Employing the momentum- and energy-conservation laws and using the small-angle approximation (see Fig. 2), the dispersion relationship for polaritons is easily shown to be

$$k^2 = (1/c^2)\{[(n_i - n_s)\omega_i + n_s\omega]^2 + n_i n_s \omega_i(\omega_i - \omega)\theta^2\},$$

where n_i and n_s are the refractive indices for the incident and scattered photons, respectively, and θ is the scattering angle. The results obtained for the frequency vs wave vector of the polariton is shown in Fig. 3. The open circles were calculated using the $y(zx)\tilde{y}$ scattering geometry. In this case, $n_i = n_e$ (extraordinary wave) and $n_s = n_0$ (ordinary wave). The corresponding scattering angles are shown with the computed points. The open squares were obtained from the $y(xz)\tilde{y}$ scattering geometry. For this configuration, $n_i = n_0$ and $n_s = n_e$. It is noted that each forward-scattering angle corresponds to two different points on the single dispersion curve for the two different scattering geometries. The results shown in Fig. 3 describe the spectra shown in Figs. 1(a) and 1(b).

The phonon directions for the above cases lie in the xy plane since the incident and scattered photons both lie in the plane. Therefore, only the magnitude, not the direction, of the polariton wave vector determine the dispersion curve.

However, if the polariton wave vector lies in the yz plane, mixing of the E_1 and A modes may be expected.[7] Therefore, a number of forward-scattering spectra with the $y(zx)\hat{y}$ geometry were taken, where $\hat{y}$, and therefore $\vec{k}$, lies in the yz plane. The

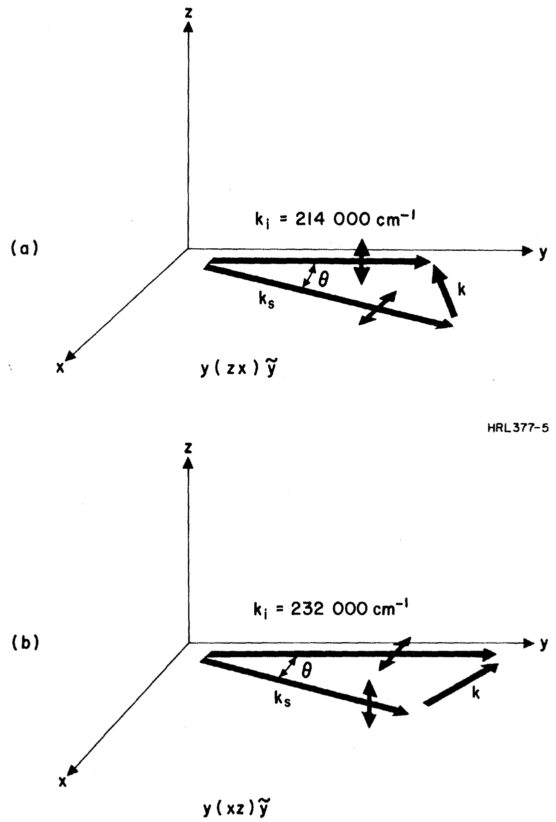

FIG. 2. Momentum-conservation triangles for the two scattering geometries used. (a) $y(zx)\tilde{y}$ polarization with $n_i = n_e$, $n_s = n_0$, and $k_i < k_s$. The range of polariton wave vector is 10 000 cm^{-1} (at $\theta = 0°$) $\leq k_{\text{polariton}} \leq$ 31 000 cm^{-1} (at $\theta = 7.7°$). (b) $y(xz)\tilde{y}$ polarization with $n_i = n_0$, $n_s = n_e$, and $k_i > k_s$. The range of polariton wave vector is 26 000 cm^{-1} (at $\theta = 0°$) $\leq k_{\text{polariton}} \leq$ 36 000 cm^{-1} (at $\theta = 5.7°$), and $n_e = 1.90$, $n_0 = 1.75$.

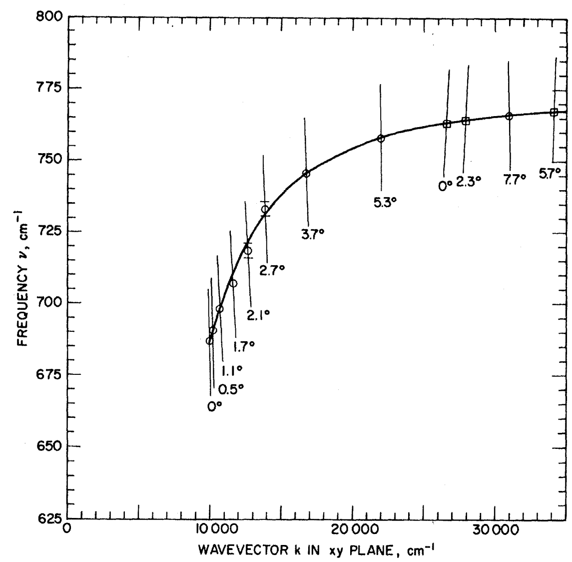

FIG. 3. E_1-mode polariton dispersion curve near $k = 0$ for $LiIO_3$ as determined from near-forward scattering of $y(zx)\tilde{y}$ and $y(xz)\tilde{y}$ polarizations. Points denoted by the open circles were computed from the experimental data for the various forward-scattering angles θ, as indicated, for the $y(zx)\tilde{y}$ polarization. Points denoted by the open squares are those for the $y(xz)\tilde{y}$ polarization.

spectra obtained were compared with the $y(zx)\tilde{y}$ spectra for equal scattering angles. No difference in the polariton frequencies for the two different geometries was observed for scattering angles θ between 0° and 7.7°. For the $y(zx)\hat{y}$ case, the angle between the phonon vector $\vec{k}$ and the xy plane varied from 0 to 76° as θ varied from 0 to 7.7°. This result suggests that the strong coupling between radiation field and lattice vibration, which gives rise to the E_1 polariton mode described here, dominates the lattice anisotropy which mixes the A and E_1 lattice waves.

SUMMARY

Even though the E_1 polariton spectra of $LiIO_3$ shown in Figs. 1(a) and 1(b) appears to be markedly asymmetric, the above analysis shows that this can be accounted for by a single polariton dispersion curve. In many uniaxial and biaxial crystals the indices and frequency differences can be such that a large asymmetry in the polariton spectra can result. This cannot be interpreted as an asymmetry in the polarizability tensor.

In addition, it was found that the E_1 polariton dispersion curve does not change with the orientation of the phonon wave vector for small scattering angles.

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