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RESONANT RAMAN SCATTERING BY TWO LO PHONONS IN InSb AT THE E_1 TRANSITION*

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Resonant Raman Scattering By Two LO Phonons in InSb at the E_1 Transition

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ABSTRACT

We have observed resonant Raman scattering of the 2 LO phonon mode in InSb in the region of the E_1 transition. The resonant enhancement peak was found to be up-shifted by 19 ± 4 meV from that of the one-phonon mode or by ~ 60 meV from the E_1 peak in the optical spectrum of InSb. Our experimental results are explained qualitatively by the double resonance theory.

Wir beobachteten eine resonante Verstärkung der Ramanstreuung durch die 2LO Phononmode in der Nähe des E_1 Übergangs. Diese Resonanz ist um 19 ± 4 meV gegenüber der Einphononmode verschoben, beziehungsweise um ~ 60 meV gegenüber dem E_1 Übergang im optischen Spektrum des InSb. Unsere

experimentellen Resultate können qualitative durch ein Doppelresonanzmodel erklärt werden.

Resonance effect in the Raman scattering of InSb near the E_1 transition (1.983 eV at 5°K) was first reported by Pinczuk and Burstein.[1] Subsequently, the dispersion of the Raman cross-section for the TO and LO phonons in the vicinity of the E_1 transition was measured by Yu and Shen.[2] These authors found that the resonant enhancement peak in the Raman cross-section of InSb occurred at an energy ~ 40 meV above the E_1 peak in the optical spectra.[3] This "blue shift" was also seen by Dreybrodt et al.[4] and was found to exist also in other semiconductors.[5-7] Here, we report the first observation and resonant Raman study of the 2 LO phonon in InSb near the E_1 transition. We found that compared with the 1 LO Raman mode, the resonant peak in the Raman cross-section of this 2 LO phonon is shifted by an additional 19 ± 4 meV towards higher energy.[1] This shift is explained qualitatively by the double resonance theory.[2]

Our measurements were performed with the back-scattering geometry on the [100], [110], and [111] faces of n-type InSb samples with a carrier concentration of 10^{14} - 10^{15} cm^{-3}. The experimental setup was the same as the one described in Ref. 2.

Figure 1 shows a typical Raman spectrum of InSb (T ~ 10°K) in the E_1 resonant region. Besides the TO (180 ± 2 cm^{-1}) and LO (192 ± 2 cm^{-1}) modes, we observed a strong well-defined peak at 389 ± 2 cm^{-1}, which is presumably the 2 LO (Γ) mode. This is the first time such a peak in InSb has ever been reported. As shown in Fig. 2, it has a resonant enhancement peak as narrow as the surface-field-induced (SFI) 1 LO Raman mode and narrower than the allowed 1 TO and 1 LO modes. It also appears to have the same polarization dependence as the SFI 1 LO mode, e.g., for all scattering configurations studied, the intensity for parallel incident and scattered polarizations is stronger than that for cross polarizations by more than a factor of 15. As suggested by Weinstein and Cardona in the case of GaP,[8] such a 2 LO (Γ) mode has a resonance behavior similar to the SFI 1 LO mode because of the Fröhlich interaction between electrons and LO phonons.[9]

Figure 2 shows the dependence of the 2 LO Raman cross-section on incident photon energy for back scattering from a [110] face. The corresponding curves for the TO and the SFI 1 LO modes are shown for comparison. The results for scattering from the [100] and [111] faces are similar. We notice in Fig. 2 that the 2 LO peak is comparable in width and in magnitude to the 1 LO peak, and that its peak position is shifted from the 1 LO and 1 TO peaks towards higher energy by 18 ± 4 meV. The magnitude of this shift varies between 16 and 21 meV for the different samples we have studied. The average shift is 19 ± 4 meV.

This "blue" shift of the 2-LO peak lends further support to the double resonance theory[2] proposed earlier which takes into account the finite energy and wave vector of the phonon. It cannot be explained by the quasi-static theory of Cardona and co-workers[4,10] which neglects the phonon energy. Recently, there is skepticism about the double resonance theory.[11] The objections are, however, based on a simple model which has not taken into account the complexity of the band structure[12] and the electron-hole correlation effects at the E_1

transition.[13] Physically, one would always expect to have such a double-resonance enhancement at least from the hot-luminescence point of view,[14,15] although the strength of this double resonance must be found from a more detailed theoretical study. Here, we shall discuss only qualitatively our experimental findings in terms of the double resonance theory.

In general, there are three types of processes which contribute towards the two phonon Raman scattering[16]: process (a) involving a repetition of the one-phonon Raman process; process (b) involving scattering of an electron (or hole) using the electron-two-phonon interaction Hamiltonian once and process (c) involving scattering of an electron (or hole) using the electron-one-phonon interaction Hamiltonian twice.

The Raman intensity of the 2 LO mode from process (a) is expected to be much weaker than that of the 1 LO mode, especially at their peak values. This is not consistent with our results and hence we can rule it out.

Process (b) has been shown recently to be important in scattering by 2 TO phonons.[8,17] Unfortunately, similar calculations on electron-2 LO (Γ) phonon matrix elements are not yet available. If we assume constant matrix elements, then, as in Reference 2, double resonance in this process should occur at $\omega_\ell = \omega_m$ with ω_m satisfying the equations (assuming a flat valence band)

$$\omega_g + (\vec{k}_\perp + \vec{q}_\perp)^2/2\mu_\perp = \omega_m$$
$$\omega_g + (\vec{k}_\perp - \vec{q}_\perp)^2/2\mu_\perp = \omega_S = \omega_m - 2\omega_o \qquad (1)$$

where ω_g is the E_1 energy gap, $\vec{q}_\perp$ is the projection of the incident photon wave vector, $\vec{q}_\ell$, onto the plane perpendicular to the [111] axis and $\mu_\perp$ is the transverse reduced mass of the valence and conduction band along [111]. We find from Eq. (1)

$$\omega_m = \omega_g + \left(2\omega_o + \frac{2q_\perp^{\;2}}{\mu_\perp}\right)^2 / (8q_\perp^{\;2}/\mu_\perp). \qquad (2)$$

Using the values of $q_\perp$, $\mu_\perp$, and ω_g in Reference 2, we obtain $\hbar\omega_m = 2.145$ eV. This energy is much higher than the position of the observed RRS peak for the 2 LO (Γ) mode (Fig. 2). We also realize that, since the strength of the E_1 transition is very low around 2.145 eV,[18] process (b) should not contribute significantly to the observed 2 LO mode.

The resonant Raman tensor for process (c) has an energy denominator ABC with

$$A = \omega_g + (\vec{k}_\perp + \vec{q}_\perp)^2/2\mu_\perp - \omega_\ell$$
$$B = \omega_g + (\vec{k}_\perp + \vec{q}_\perp - \vec{q}_\perp{}')^2/2\mu_\perp - \omega_\ell + \omega_o \qquad (3)$$
$$C = \omega + (\vec{k} - \vec{q})^2/2 - \omega + 2\omega_o$$

where again we assume a flat valence band. The wave vectors of the two LO phonons emitted are $\vec{q}'$ and $2\vec{q}_\ell - \vec{q}_\perp{}'$, and $\vec{q}'$ is the projection of $\vec{q}'$

onto the plane perpendicular to the [111] axis.

We note that the condition A = C = 0 is equivalent to Eq. (1). Hence, triple resonance with A = B = C = 0 and double resonance with A = C = 0 will occur at ω_ℓ = 2.145 eV where the E_1 transition has little oscillator strength to be significant. We are left with possible double resonances with (i) A = B = 0 and (ii) B = C = 0. Unlike the case of one-phonon scattering, these double resonance conditions can always be satisfied by an appropriate choice of $\vec{q}\,'$. As a result, the double resonance of case (i) will have an RRS peak at $\omega_g + \omega_o$ (= 2.031 eV) and that of case (ii) will have a peak at $\omega_g + 2\omega_o$ (= 2.055 eV). It is then possible that the observed 2 LO RRS peak at 2.047 ± 0.003 eV is the superposition of these two double resonant peaks.

Since the 2 LO(Γ) mode has one more electron-LO phonon matrix element than the SF-1 LO mode, one might expect its Raman cross-section to be weaker. But the smaller Raman matrix element is presumably compensated by a larger density of phonons in the 2 LO case.

In conclusion, we have observed a new Raman mode in InSb at 389 ± 2 cm^{-1} which is identified as due to scattering of two LO phonons. The Raman cross-section of this new mode shows a resonance enhancement peak which is up-shifted by ~ 60 meV from the E_1 peak in the optical spectrum of InSb. We have interpreted our results qualitatively in terms of the recently proposed double resonance theory.

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FIGURE CAPTIONS

Fig. 1. Raman spectrum of n-type InSb ($N = 1.2 \times 10^{15}$ cm^{-3}) at 10°K obtained by backscattering with parallel polarizations from a [110] surface. The exciting laser wavelength is 6085 Å (2.037 eV).

Fig. 2. Dispersion in the Raman cross-sections of the TO phonon (TO), the surface-field-induced LO phonon (SF-1 LO), and the 2 LO phonon (2 LO) modes of an n-type ($N = 1.2 \times 10^{15}$ cm^{-3}) InSb sample at ~ 10°K. The scattering configurations is the same as in Fig. 1. The intensity of the TO mode has been multiplied by a factor of 2 to bring it up to the same scale as the LO mode.

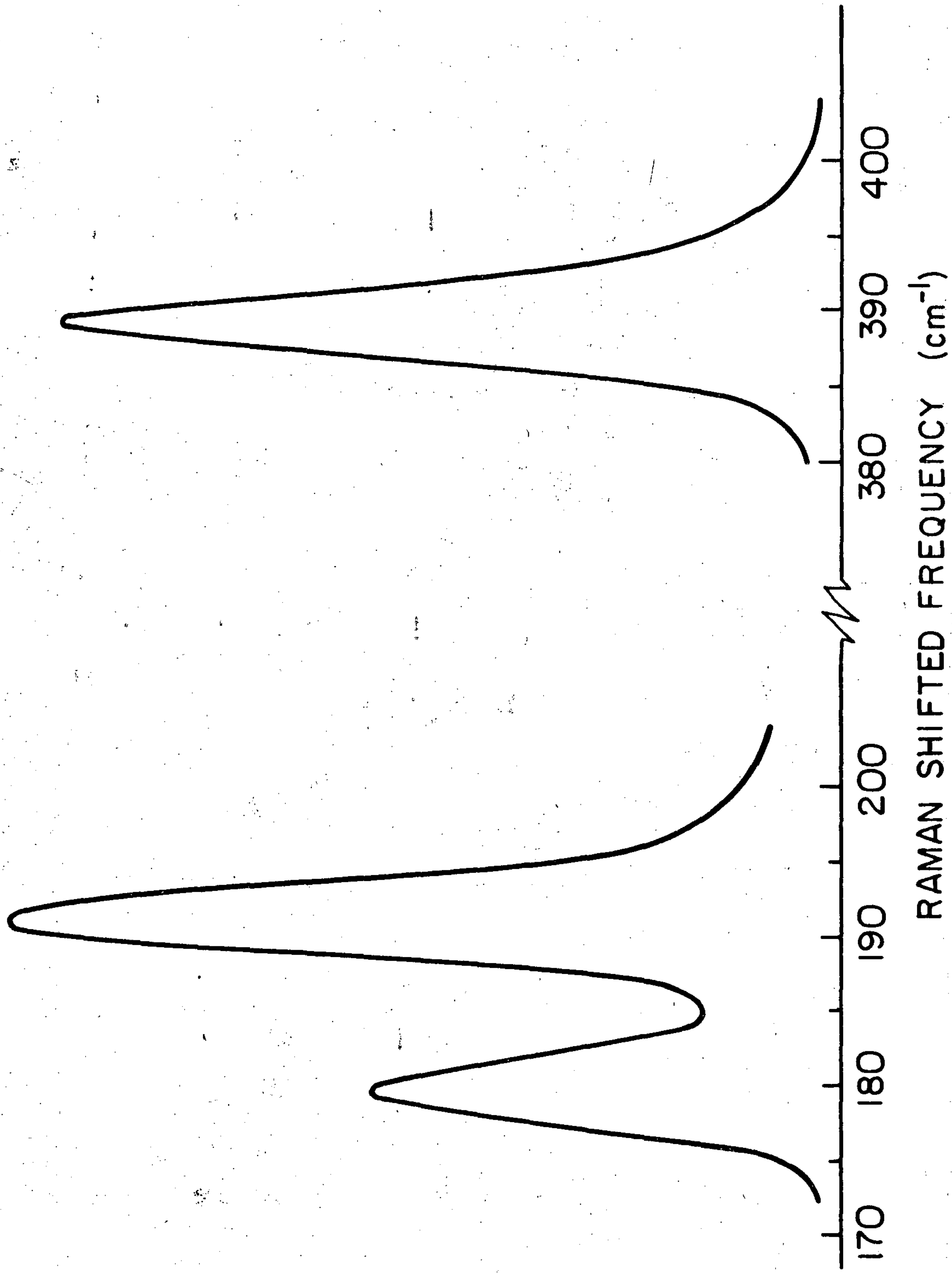

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Fig. 1

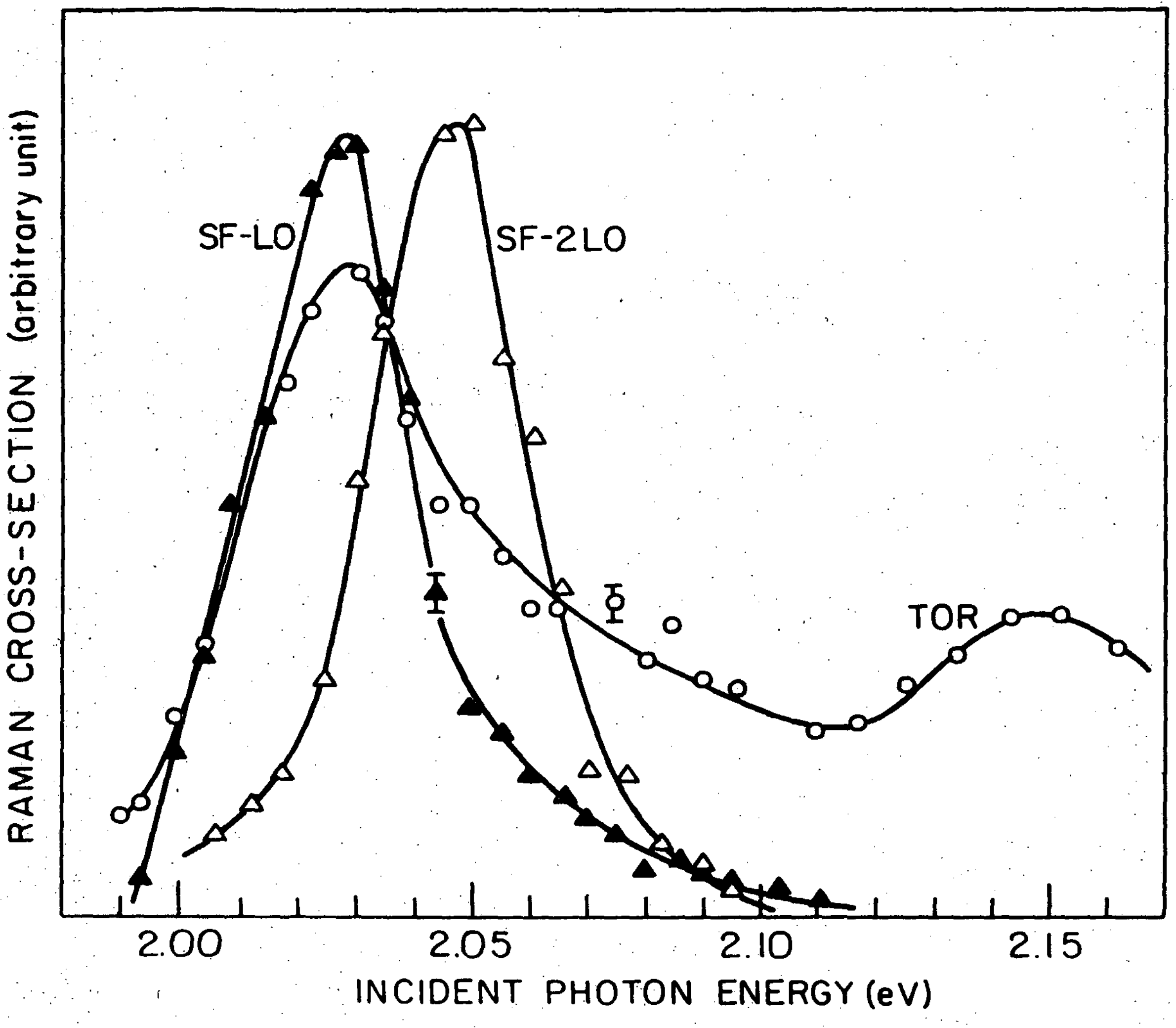

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Fig. 2

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