

Electrons and holes in InSb under crossed magnetic and stress fields. IV. Stimulated Raman scattering in the valence bands

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Anticrossing of magnetic levels in InSb leads to high combined densities of states in the valence band at nonzero values of the longitudinal momentum k_B . Using pump laser radiation with photon energies near the band gap we observed stimulated Raman scattering due to transitions between repelling energy levels under uniaxial deformation. The stress was applied perpendicular to the magnetic field and perpendicular to the wave vector of the incident light ($T \parallel [100]$, $\mathbf{B} \parallel [001]$, $\mathbf{k} \parallel [010]$). The energy levels involved in the scattering process could be identified as states with $k_B \neq 0$ in the heavy-hole bands 3V ($0, -\frac{1}{2}$) and 6V ($1, \frac{1}{2}$). The range of frequency of the Raman radiation and of the magnetic field, where the observation is possible, depends very sensitively on stress. Detailed calculations of the valence-band structure show that the deformation shifts the anticrossing point in k_B space and varies the energy difference of the levels 3V and 6V.

I. INTRODUCTION

Raman scattering experiments supply a powerful method for investigation of band structure of semiconductors. From such measurements, for example, the effective g value of electrons could be derived very exactly.¹ In the present paper we report on stimulated Raman scattering from photoexcited holes in InSb. The anticrossing of energy levels in a magnetic field leads to a high joint density of states in the valence band at $k_B \neq 0$. Raman scattering by holes in these levels was first observed by Ebert *et al.* in InSb.²

The behavior of the levels in the k_B region of interest can be studied in more detail if the sample is subjected to different stresses. These cause strong interactions of the valence-band levels, especially near anticrossing points in k_B space. The Landau levels depend both on the direction of the magnetic field and of the deformation. In our experiment the direction of stress $T \parallel [100]$ was perpendicular to the magnetic field. In this configuration the fourfold symmetry of the crystal is reduced to a twofold symmetry.

II. THEORY

To explain our experimental results we need an exact model of the magnetic field and stress-dependent Landau states in the valence band (VB) of InSb. A very detailed

calculation of the Landau states was done by Trebin *et al.* in paper I of this series.³ Figure 1 shows the valence-band energy levels (on an inverted scale) near the band edge at a magnetic field of 6.0 T in the orientation $\mathbf{B} \parallel [001]$ for zero stress with k_B values up to 13×10^5 cm⁻¹. The band parameters are the same as are used in papers I–III.³ The solid (dashed) lines belong to states of the group theoretical representations $P = 1$ ($P = 0$). Energy levels with different quantum numbers P are allowed to cross while those with the same P repel each other. The valence-band levels 3V and 6V of the representation $P = 1$ display very clearly this anticrossing behavior. In the k_B region, where the levels approach, the dispersion curves of the two energy levels become parallel, which yields a large joint density of states ρ_B . This fact is formulated in Eq. (1) for two energy levels with energies $E_1(k_B)$ and $E_2(k_B)$ at $\hbar\omega = E_1(k_B) - E_2(k_B)$ without considering any quenching;

$$\rho_B(\hbar\omega) = \frac{eB}{(2\pi)^2 \hbar^2} \left| \frac{1}{\left[\frac{\partial E_1}{\partial k_B} \right] - \left[\frac{\partial E_2}{\partial k_B} \right]} \right|_{E_1(k_B) - E_2(k_B) = \hbar\omega} \quad (1)$$

A maximum of ρ_B arises whenever E_1 and E_2 become parallel. $E_1(k_B)$ and $E_2(k_B)$ are the energies of the levels 3V and 6V. These two levels are of particular interest to

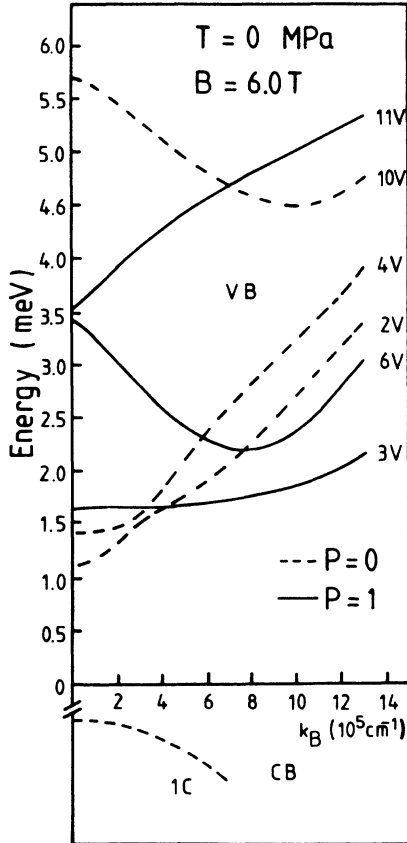


FIG. 1. k_B dependence of the valence-band levels near the band edge in InSb at a magnetic field of 6.0 T and zero stress. The energy scale is inverted. All energies are referred to the lowest conduction-band level 1C at $k_B=0$. Levels drawn with dashed (solid) lines belong to representation $P=0$ ($P=1$).

us, because of all maxima of ρ_B where Raman scattering is possible due to the polarization selection rules, their anticrossing point is situated closest to the quasi-Fermi-level (QFL) in a sample which is not too much excited.

III. EXPERIMENTAL DETAILS

The samples were cut from single crystals of p -type InSb with carrier concentrations of $\rho=1.0 \times 10^{14} \text{ cm}^{-3}$ and were oriented by means of the Laue method. We used a Q -switched CO laser oscillating on one of the lines near the band gap of InSb. The sample was mounted in a stress apparatus fixed in a superconducting magnet at a temperature of 1.5 K. The scattered radiation was observed in the configuration where the uniaxial stress is perpendicular to the magnetic field, i.e., $\mathbf{B} \parallel [001]$, $T \parallel [100]$, $\mathbf{k} \parallel [010]$. The transmitted and scattered beam was detected with a liquid-nitrogen-cooled InSb detector in conjunction with a grating spectrometer.

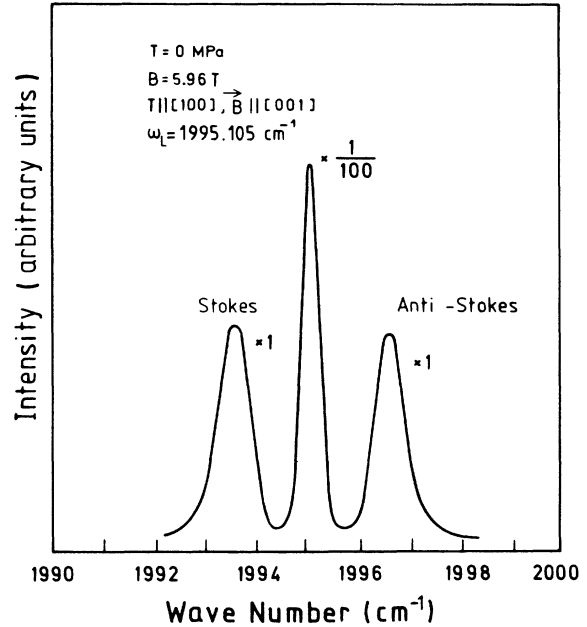


FIG. 2. Spectrum of the stimulated Raman scattering from photoexcited holes in InSb taken at a magnetic field of 5.96 T and zero stress. The intensity of the transmitted laser beam is higher by a factor of 100 than the scattered radiation.

IV. RESULTS AND DISCUSSION

In the spectrum of Fig. 2, taken at zero stress, a Stokes, an anti-Stokes, and the transmitted laser line are seen. The Raman shift is about 1.5 cm^{-1} . The photon energy of the pump radiation ($\omega_L = 1995.105 \text{ cm}^{-1}$) differs only

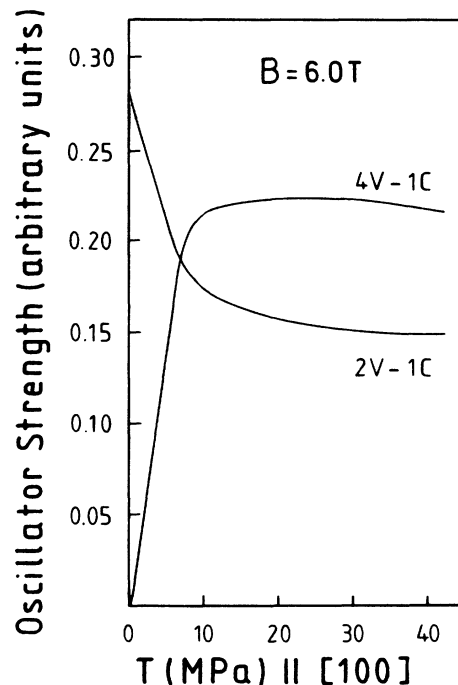


FIG. 3. Stress dependence of the oscillator strength for the transitions from 2V and 4V to the conduction band 1C at a magnetic field of 6.0 T.

slightly from the gap energy at 6 T. In the discussion of our experimental data interband absorption plays an important role since the light is scattered by photoexcited holes. The scattering process is only possible if the initial state is occupied by electrons and if there are holes in the final state. In other words, the QFL must be situated between the two levels. Its particular position determines the k_B values of the electrons involved in the scattering process. In order to determine this position, one has to discuss the interband absorption first.

A. Interband absorption

The quasi-Fermi-level depends on the magnetic field and the uniaxial deformation, provided intensity and energy of the pump radiation are kept constant. Since the magnetic field widens the fundamental gap the absorption coefficient diminishes with growing field and the quasi-Fermi-level moves up to the valence-band maximum. With photon energies near the band-gap energy and in a magnetic field of 6 T, interband absorption is mainly caused by transitions $2V \rightarrow 1C$, since these have a high oscillator strength.

Transitions between the subsequent level in the valence band (4V) and the lowest conduction-band level (1C) are forbidden for zero stress. This is shown in Fig. 3, where the oscillator strengths of these transitions are drawn versus stress. In Fig. 4 the corresponding transition energies are presented. Other transitions than those men-

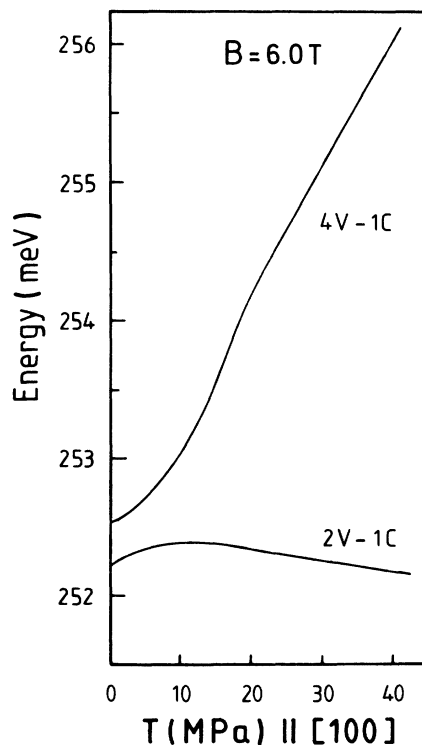


FIG. 4. Stress dependence of the transition energies $E(1C) - E(4V)$ and $E(1C) - E(2V)$ for 6.0 T.

tioned have a low oscillator strength or are not possible due to energy reasons.

The valence-band levels 2V and 4V belong to the same group-theoretical representation. They are strongly interacting under uniaxial deformation. This is manifested in the oscillator strength, too, where some contributions of the 2V level are transferred to the lower valence-band state 4V. The absorption of the laser beam decreases already under very small stresses, because the oscillator strength for $2V \rightarrow 1C$ declines and the now favored transition $4V \rightarrow 1C$ widens in energy (see Figs. 3 and 4). Thus, when applying uniaxial stress the absorption of photons with energy near the band gap is diminished. In order to maintain the density of free holes and to keep the position of the quasi-Fermi-level the magnetic field has to be decreased. The enhancement of the transmission under uniaxial stress is demonstrated impressively in our experiments. Figure 5 shows two spectra taken with the same magnetic field ($B = 5.82$ T) but different values of stress ($T = 0$ and $T = 21$ MPa). Under zero stress the pump laser radiation is absorbed strongly, and hence intense stimulated recombination radiation is generated (a). A small uniaxial deformation reduces the absorption and makes the luminescence vanish. At a stress of $T = 21$ MPa the stimulated Stokes and anti-Stokes Raman lines can be observed (b). By then the intensity of the transmitted laser beam is increased by a factor of 20. When stress is applied, not enough electron-hole pairs are

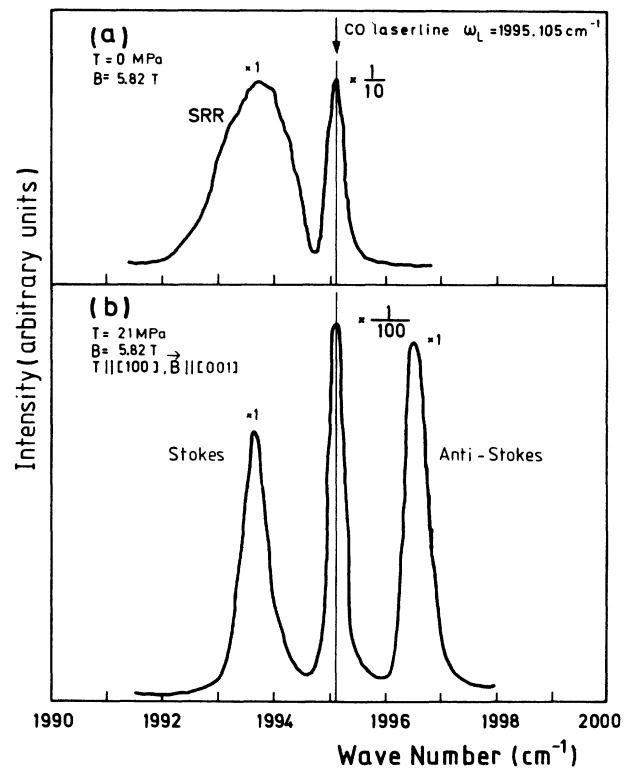


FIG. 5. Spectra of the stimulated recombination radiation and of the stimulated Raman scattering by free holes taken at the same magnetic field of 5.82 T. (a) Stress $T = 0$; (b) $T = 21$ MPa.

created for exciting stimulated recombination radiation and abundant pump intensity is left in the sample to excite stimulated Raman scattering. By further increasing the stress the intensity of the Stokes and anti-Stokes radiation rapidly diminishes until the scattered light becomes unobservable.

B. Stimulated Raman scattering

Similarly as for Raman scattering, for stimulated Raman scattering one also has to take into consideration the energy difference between the levels 3V and 6V and the position of the quasi-Fermi-level. The QFL determines the range of longitudinal momentum k_B where scattering is possible. It depends on energy and intensity of the pump radiation as well as on magnetic field and stress, as discussed in the preceding section. Calculations indicate that there is no significant shift of the level 3V relative to 6V if in zero stress the magnetic field rises from 5.5 to 6.5 T.² The magnetic field only changes the quasi-Fermi-level. For different magnetic fields B_i the positions of the quasi-Fermi-level are plotted schematically in Fig. 6. When applying a field B_1 , the QFL lies at point F_1 in the 6V valence band. At first sight Raman scattering can take place both for $k_B > k_1$ and for $k_B < k_1'$. However, for $k_B < k_1'$, ρ_B is too small to reach the threshold for stimulated Raman emission. As explained before, in increasing magnetic fields the quasi-Fermi-level moves towards the top of the valence band (downwards in the inverted scale in Fig. 6) and therefore the point k_1 (k_1') in k_B space shifts to lower (higher) values.

Optimum conditions for Raman scattering are expected if the QFL is situated between F_2 and F_3 (fields between B_2 and B_3). Then, in the range of the k_B space where the joint density of states is largest (near k_2, k_3) the 6V states are occupied by holes and the 3V states are

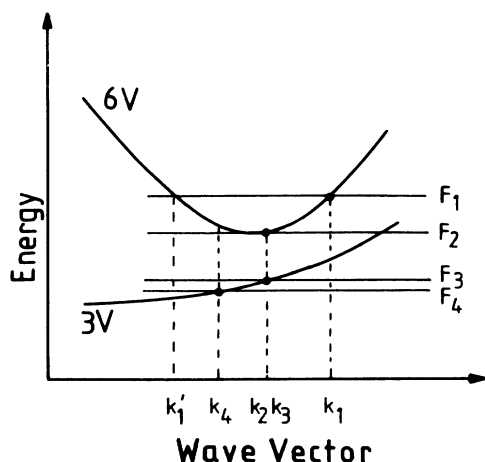


FIG. 6. Schematic representation of the quasi-Fermi-level near the valence-band levels 3V and 6V for different magnetic field [$F_i = F(B_i)$, $B_1 < B_2 < B_3 < B_4$].

occupied by electrons. For magnetic fields exceeding B_3 (e.g., B_4) scattering is only possible for $k_B < k_4$ with a very reduced density ρ_B .

Between B_2 and B_3 the Raman shift should be almost independent of the magnetic field. For $B > B_3$, however, transitions occur at momenta $k_B < k_3$ and the 6V-3V excitation energy increases with B due to the decrease of k_B . In experiments for all fields $B < B_3$ the absorption of the pump radiation turns out too high and Raman scattering cannot be observed. Only in the magnetic field interval between B_3 and B_4 Raman scattering by free holes is stimulated. If we further raise the magnetic field, the slopes of the 3V and 6V bands deviate more and more from each other in the k_B region where transitions are possible (see Fig. 6). The joint density of states becomes too small and stimulated Raman scattering ceases to occur.

In Fig. 7 the measured Raman shift is plotted versus magnetic field for different deformations. The frequency of the pump radiation used is 1995.105 cm^{-1} . Obviously, Raman scattering by holes of nonzero longitudinal momentum k_B can only be detected in a small magnetic field range around 6 T and is strongly dependent on uniaxial stress. With increasing stress the magnetic field range in which Raman scattering is observed shifts to smaller values because the pump radiation is absorbed less.

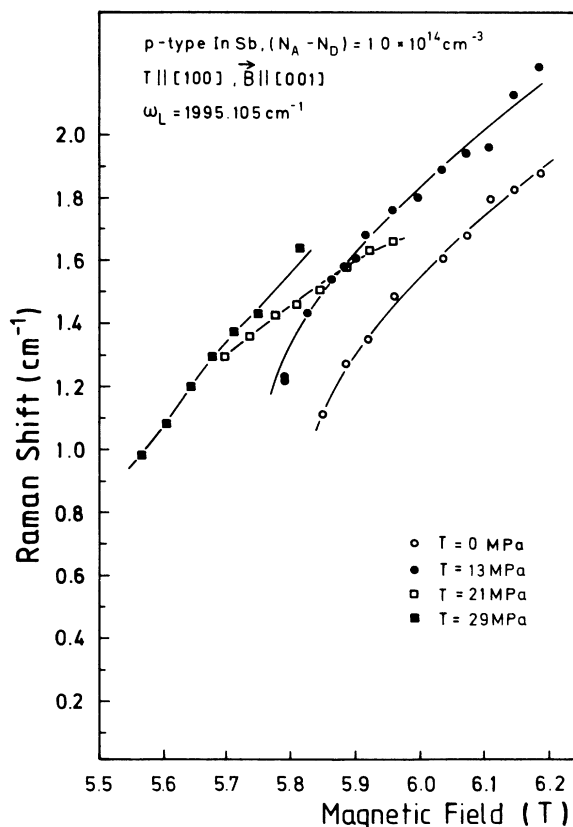


FIG. 7. Magnetic field dependence of the frequency shift in stimulated Raman scattering by holes in InSb at different uniaxial stresses.

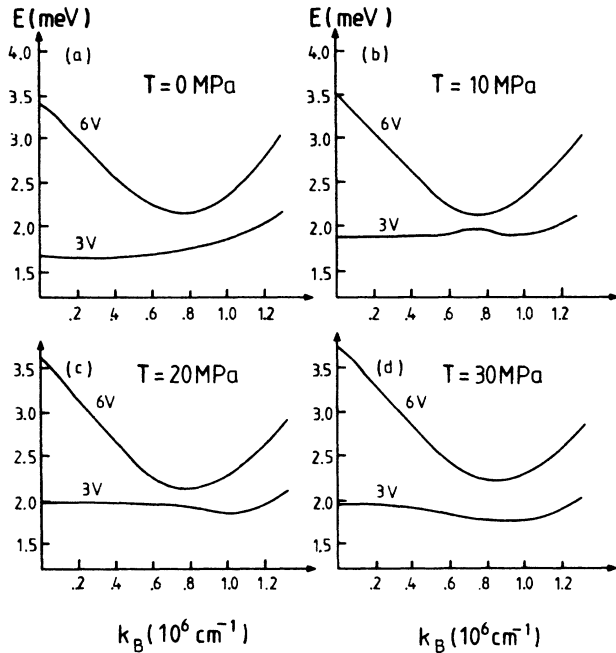


FIG. 8. The valence-band levels 3V and 6V at a magnetic field of 6.0 T under different uniaxial stresses.

Figure 8 presents calculated energy bands 6V and 3V for 6 T and several stress values. When the stress is increased the points of closest approach of the levels are displaced slightly. We must discuss the position of the quasi-Fermi-level only for that magnetic field range in which Raman scattering is observable. With increasing stress the 6V level preserves its shape in the anticrossing region, whereas the 3V level is repelled by the 6V level and by deeper valence-band levels for large k_B values.

This leads to greatly differing slopes of the 3V and 6V bands. So, for magnetic fields between B_3 and B_4 we expect for $T \approx 0$ and $T \geq 20$ MPa the Raman shift to rise notably with increasing field. From Fig. 8 we conclude that at a stress of about 10 MPa the 3V level is nearly independent of k_B . Therefore, the slope of the tuning curve should be rather small. For $T > 30$ MPa the absorption of the pump radiation is too small for observing the scattering process. The theoretical calculations explain the characteristic features of our experimental results quite well. In particular, we understand why the Raman frequency changes much less with the magnetic field at a certain stress than at other stresses in our measurements. This critical stress is found at 21 MPa whereas theory predicts such a behavior at 10 MPa. But we have to bear in mind that the exact amount of the Raman shift and the value of this stress depend very sensitively on the band parameters used (paper I of this series³). Thus, in our calculations, which are not fitted to the present experimental results, quantitative agreement within a fraction of a wave number cannot be expected.

V. SUMMARY

Anticrossing effects of neighboring bands 3V and 6V cause noncentral extrema of the joint density of states. Ebert *et al.* have shown that the noncentral maxima are significant for Raman scattering experiments.² The behavior of the levels in the k_B region of interest can be studied more intensively when applying uniaxial stress, because it influences the interactions of the bands. We have measured the Raman scattering near an anticrossing point of the levels 3V and 6V under uniaxial stress and have interpreted the characteristic features of our observations using detailed theoretical calculations of the band structure.

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²R. Ebert, H. Pascher, and H. G. Häfele, *Phys. Rev. B* **23**, 6570 (1981).

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