INFRARED ABSORPTION BY COUPLED COLLECTIVE CYCLOTRON EXCITATION–LONGITUDINAL-OPTIC PHONON MODES IN InSb

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The coupling of longitudinal-optic (LO) phonons and free-carrier collective excitations by "macroscopic" longitudinal electric fields in polar semiconductors has been treated theoretically by a number of investigators.[1-9] In the absence of a magnetic field, the coupled modes involve plasmons ($\omega=\omega_p$) as the free-carrier collective excitations, and LO phonons ($\omega=\omega_{\mathrm{LO}}$). The coupled modes are longitudinal in character and, as such, cannot interact with electromagnetic (EM) radiation in bulk samples or in thin films under normal incidence conditions. An interaction of the coupled modes with EM radiation can, however, be observed in thin-film, oblique-incidence transmission experiments[8] and also in Raman scattering experiments on semiconductors lacking a center of inversion.[5,10] In the presence of a magnetic field $\vec{H}_0$ there are two types of coupled modes, depending on the direction of the wave vector $\vec{q}$ of the modes relative to the direction of the magnetic field. One type involves plasmons and LO phonons with $\vec{q}$ parallel to $\vec{H}_0$; the other type involves collective cyclotron excitations ($\omega^2=\omega_c^2+\omega_p^2$) and LO phonons with $\vec{q}$ tilted away from $\vec{H}_0$.[11] Because of the requirement of momentum conservation, EM radiation will interact only with those modes having $\vec{q}$ parallel to the wave vector $\vec{k}$ of the radiation. Thus in thin-film transmission experiments carried out at normal incidence in the Faraday configuration ($\vec{k}\parallel\vec{H}_0$), one observes resonance absorption of EM radiation only by single-particle cyclotron excitations ($\omega=\omega_c$) and by transverse optical (TO) phonons phonons ($\omega=\omega_{\mathrm{TO}}$).[12] The coupled plasmon–LO-phonon modes with $\vec{q}\parallel\vec{H}_0$, which are purely longitudinal in character, do not interact with the EM radiation for $\vec{k}\parallel\vec{H}_0$. On the other hand, the coupled modes involving the collective cyclotron excitations and LO phonons are transverse as well as longitudinal in character. Thus they may be expected to interact with EM radiation when $\vec{k}\parallel\vec{q}$ is perpendicular to $\vec{H}_0$.

We report here the observation of resonance absorption by the coupled modes involving collective cyclotron excitations and LO phonons in thin-film transmission experiments on n-type InSb carried out at normal incidence in the Voigt configuration, i.e., $\vec{k}\perp\vec{H}_0$. The frequencies of the normal modes with $q\approx 0$ and $\vec{q}\perp\vec{H}_0$, which arise from the coupling of the collective cyclotron excitations with the LO phonons via the macroscopic longitudinal electric field, are given by[8]

$$2\omega_\pm^{\;2}=\omega_p^{\;2}+\omega_c^{\;2}+\omega_{\mathrm{LO}}^{\;2}\pm[(\omega_p^{\;2}+\omega_c^{\;2}+\omega_{\mathrm{LO}}^{\;2})^2-4(\omega_p^{\;2}\omega_{\mathrm{TO}}^{\;2}+\omega_c^{\;2}\omega_{\mathrm{LO}}^{\;2})]^{1/2}, \tag{1}$$

where $\omega_p^{\;2}=4\pi Ne^2/m^*\epsilon_\infty$, e and m^* are the charge and the effective mass of the carriers, respectively, N is the carrier density, and ϵ_∞ is the high-frequency dielectric constant. [When $H_0=0$, Eq. (1) reduces to the form which applies to the coupling of plasmons and LO phonons.]

The normal-incidence transmission of a thin film for which $2\pi d/\lambda_0 \ll 1$, where d is the thickness and λ_0 is the vacuum wavelength of the radiation, is given by[8,12]

$$T=1-\frac{2\pi d}{\lambda_0}\mathrm{Im}\chi_T^{\;r}(\omega), \tag{2}$$

where $\chi_T^{\;r}(\omega)=P_T(\omega)/E_T(\omega)$ is the transverse electric-susceptibility response constant and $P_T(\omega)$ and $E_T(\omega)$ are the macroscopic transverse polarization and transverse electric field in the medium.

For the Voigt configuration $\vec{E}\perp\vec{H}_0$, $\chi_T^{\gamma}(\omega)$ is given by

$$\chi_T^{\gamma}(\omega)=\frac{\epsilon_\infty-1}{4\pi}+\frac{\epsilon_\infty}{4\pi}\left[\frac{\omega_{LO}^2-\omega_{TO}^2}{\omega_{TO}^2-\omega^2-i\Gamma\omega}-\frac{\omega_p^2}{\omega^2+i\gamma\omega}+\frac{\omega_c^2\omega_p^2(\omega_{LO}^2-\omega^2-i\Gamma\omega)}{(\omega+i\gamma)^2D}\right], \tag{3}$$

where

$$D=[\omega_p^2-\omega^2-i\gamma\omega+\omega\omega_c^2/(\omega+i\gamma)](\omega_{LO}^2-\omega^2-i\Gamma\omega)-\omega_p^2(\omega_{LO}^2-\omega_T^2),$$

and Γ and γ are the damping constants for the phonons and electrons, respectively. Transmission minima occur at the frequencies where $\mathrm{Im}\chi_T^{\gamma}(\omega)$ exhibits maxima, i.e., at the frequencies ω_+ and ω_- where D exhibits minima, and also at ω_{TO}, the frequency of the $q\approx 0$ TO phonons.

Since the interaction of the coupled modes with EM radiation in the Voigt configuration takes place via the transverse electric moment of the collective cyclotron excitations, the absorption strengths of the two normal modes are proportional to the "collective cyclotron excitation" character of the modes. Analysis of $\chi_T^{\gamma}(\omega)$ on the assumption that Γ and γ do not vary with magnetic field indicates that the absorption strength of the high-frequency coupled mode ($\omega=\omega_+$) is zero at $H_0=0$, increases rapidly when the field is applied, and then levels off at high fields. The absorption strength of the low-frequency mode ($\omega=\omega_-$) is also zero at $H_0=0$, increases rapidly to a maximum as the field is increased, and then decreases slowly with further increase of field.

The width of the resonance absorption bands is determined by the magnitude of the imaginary part of D at $\omega=\omega_+$ and $\omega=\omega_-$, which is given by

$$\mathrm{Im}D=-\gamma[(\omega_1^2+\omega_\pm^2)/\omega_\pm](-\omega_\pm^2+\omega_{LO}^2)-\Gamma\omega_\pm(-\omega_\pm^2+\omega_p^2+\omega_1^2),$$

where $\omega_1^2=\omega_c^2/[1+\gamma^2/\omega^2]$. We note that when the low-frequency mode ω_- approaches ω_{LO}, its linewidth is determined primarily by Γ, the phonon damping constant. Also, when the high-frequency mode ω_+ approaches $(\omega_p^2+\omega_1^2)^{1/2}$, then $\omega_1\approx\omega_c$ and the width of the line is determined by γ, the electron damping constant.

The normal-incidence transmission measurements were carried out on samples of n-type InSb having carrier concentrations of 1.4×10^{17}, 5.5×10^{16}, and 1×10^{15} cm^{-3} at 4.2°K. The samples, glued to pure silicon backings, were ground and polished by conventional techniques to thickness in the range 5-15 μ. A Michelson interferometer adapted for light-pipe operation[13] was used with a superconducting magnet capable of producing 36 kOe. In this system, samples could be mounted in either the Faraday or Voigt configuration. The radiation was unpolarized. Transmission measurements were made in the spectral range 90-320 cm^{-1} using a gallium-doped germanium photoconductive detector. Most of the spectra were obtained with 2-cm^{-1} resolution, although some runs were made with resolutions of 1.0 and 0.5 cm^{-1}. In order to establish the sample carrier concentrations, zero-field room-temperature reflectivity was measured in bulk and thin samples in the spectral range 70-330 cm^{-1} to determine the reflection minima at $\omega\approx\omega_\pm$. Crystal orientation of the samples was not determined.

Typical results obtained for sample 1, which contained 1.4×10^{17} carriers cm^{-3}, are given in Fig. 1. The spectra are shown for applied

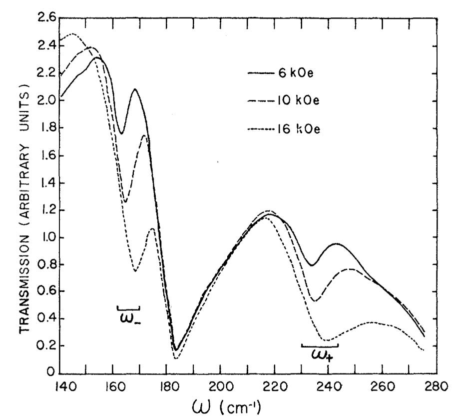

FIG. 1. Transmission spectra of InSb sample 1 with $N=1.4\times10^{17}$ carriers cm^{-3} for three values of magnetic field. The regions of coupled-mode absorption are indicated by brackets.

fields of 6, 10, and 16 kOe and are not corrected for the instrument spectral response. The latter is responsible for the gradually decreasing signal at higher energies. At 150 cm^{-1}, approximately 30% of the light was trasmitted through the sample. The main features of the spectra shown are the strong absorption line at 184 cm^{-1} corresponding to ω_{TO} and the lines corresponding to ω_+ and ω_-. The fact that a well-defined ω_{TO} absorption is observed indicates that the sample is sufficiently thin that there are no major complications from reflection effects. The ω_- line first appeared below ω_{LO} with low intensity in low magnetic fields. As the field was increased, the line became stronger and then disappeared into the absorption near ω_{TO}. The ω_+ line first appeared just above ω_{LO} with low intensity and then increased in intensity and frequency as the field was raised. This intensity variation in the range 0-36 kOe is in good qualitative agreement with the theoretical prediction. In addition, the ω_- absorption linewidth is much smaller then the ω_+ linewidth, in agreement with theory. The field dependence of the frequencies of the ω_+ and ω_- lines for the three samples is shown in Fig. 2. For the purest sample, an inflection was observed in the transmission curve for fields above 31 kOe in the range 192-198 cm^{-1}, where ω_- should occur, but no distinct minimum was observed. Measurements on sample 1 in the Faraday orientation did not show the coupled-mode absorption. While a

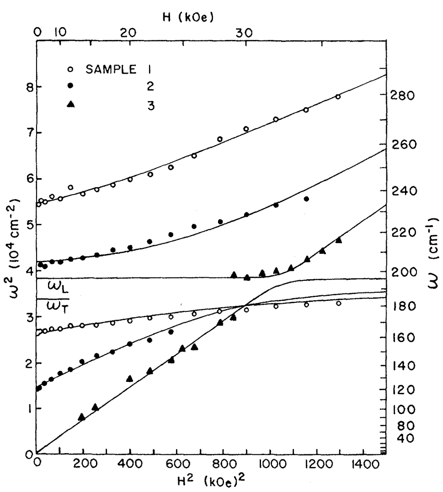

FIG. 2. Frequency positions of the coupled-mode absorption lines as a function of magnetic field. The solid lines were calculated with the use of Eq. (1) and the parameters of Table I.

Table I. Parameters obtained for samples 1, 2, and 3 by fitting the data as described in the text.

Sample	Thickness (μ)	ω_p (cm^{-1})	m^*/m	N (cm^{-3})
1	~5	205	0.019	1.4×10^{17}
2	~15	135	0.017	5.5×10^{16}
3	~10	18	0.016	1×10^{15}

general magnetic-field-dependent absorption was observed, no distinct resonance absorption by single-particle cyclotron excitations was seen. This is not understood at present since the criterion $\omega_c\tau > 1$ for distinct resonance was satisfied.

The theoretical curves shown in Fig. 2 were fitted to the experimental data by the use of Eq. (1). Throughout the calculation, the values $\omega_{TO} = 184$ cm^{-1}, $\omega_{LO} = 196$ cm^{-1}, and $\epsilon_\infty = 16$ were used. For each sample this procedure yielded the parameters m^*, ω_p, and therefore N. These parameters are given in Table I. They are in agreement with values of N and m^* obtained from zero-field reflection spectra and Hall-effect measurements and with the known variation of effective mass with carrier concentration.[14] Furthermore, the qualitative features of the transmission spectra obtained in the experiments, i.e., the dependence of the frequency, linewidth, and intensity of the resonances on magnetic field, are found to be in good agreement with theory.

We have not clearly observed the separation of the two modes in the region of bending in sample 3 with the lowest carrier concentration (1.5×10^{15} cm^{-3}), because of masking by the ω_{TO} absorption and the low strength of the lines when they approach the vicinity of ω_{TO} and ω_{LO}. Therefore, at the moment it is not possible to establish whether there is a polaron-induced anomaly[15,16] in the observed effect in sample 3 associated with the coupling of the single-particle excitations with the longitudinal coupled collective modes.[17]

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[1]B. B. Varga, Phys. Rev. 137, A1896 (1965).
[2]R. Tsu and D. L. White, Ann. Phys. (N.Y.) 32, 100 (1965).
[3]Y. C. Lee and N. Tzoar, Phys. Rev. 140, A396 (1965).
[4]K. S. Singwi and M. P. Tosi, Phys. Rev. 147, 658 (1966).
[5]A. Mooradian and G. B. Wright, Phys. Rev. Letters 16, 999 (1966).
[6]T. N. Casselman and H. N. Spector, Phys. Condensed Matter 4, 179 (1965).
[7]M. P. Greene, A. Houghton, and J. J. Quinn, Phys. Letters 20, 238 (1966).
[8]S. Iwasa, Y. Sawada, E. Burstein, and E. D. Palik, in Proceedings of the International Conference on Physics of Semiconductors, Kyoto, 1966 (unpublished).
[9]I. Yokota, in Proceedings of the International Conference on Physics of Semiconductors, Kyoto, 1966 (unpublished).
[10]E. Burstein, in J. Phys. (Paris) Suppl. 11, 688 (1965).
[11]The collective cyclotron excitations are collective modes consisting of single-particle cyclotron excitations ($\omega = \omega_c$) which are coupled by macroscopic electric fields having components in the plane of the cyclotron orbits. These have also been variously called magnetoplasma excitations and plasma-cyclotron excitations.
[12]E. Burstein, S. Iwasa, and Y. Sawada, "Proceedings of 1965 Enrico Fermi Summer School on Optical Properties of Solids" (unpublished).
[13]R. Kaplan, to be published.
[14]J. Kolodziejczak and S. Zukotynski, Phys. Status Solidi 16, K55 (1966).
[15]E. J. Johnson and D. M. Larsen, Phys. Rev. Letters 16, 655 (1966).
[16]E. J. Johnson, D. H. Dickey, and D. M. Larsen, Bull. Am. Phys. Soc. 11, 828 (1966).
[17]R. M. White and C. S. Koonce, Phys. Rev. Letters 17, 436 (1966).

SPIN-WAVE AND EXCITON DISPERSION RELATIONS OF COBALT FLUORIDE

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The dispersion relations of spin waves have been observed in antiferromagnetic cobalt fluoride at 4.5°K using inelastic neutron-scattering techniques. Measurements have been made not only of the excitations within the ground-state Kramers doublet of the paramagnetic phase, but also of the excitations to the first excited doublet of the Co^{++} ion. Insofar as we are aware, the latter are the first measurements of exciton dispersion relations in a single crystal obtained by neutron scattering techniques. The results are shown to be in at least qualitative agreement with a theory incorporating Heisenberg exchange forces between the spins on neighboring Co^{++} ions and with the crystal field parameters deduced by Gladney.[1]

The electronic-energy-level diagram of Co^{++} is shown in Fig. 1. Each Co^{++} ion is surrounded by an almost exactly cubic array of fluorine ions. The crystal field[2] from these splits the ^4F atomic state to give the ground state as an orbital triplet Γ_4. The rhombic and axial distortions of the field give rise to effects of similar size to those of the spin-orbit splitting, and Gladney[1] has deduced the relevant crystal-field and spin-orbit parameters from measurements of the g values[3] and infrared spectra.[4] The Γ_4 state is then split into six Kramers doublets, the lowest pair of which are separated by a frequency of only 4.5×10^{12} cps, while the next state is considerably higher in energy. The exchange field in the antiferromagnetic state then further splits the Kramers doublets, giving the lowest four states labeled A, B, C, D in Fig. 1.

The crystal of CoF_2 was grown at a rate of 2 mm/h by the Stockbarger process in a graphite crucible. The powder was first dehydrated by heating in a platinum crucible in an air furnace to 700°C, with an admixture of ammonium hydrogen fluoride to oppose hydrolysis. On first growth, the cone of the crystal was usually covered with a thin skin of cobalt, which could be peeled away before recrystallization. On regrowth, much less metal was formed, but it accumulated at the extreme tip of the