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OBSERVATION OF EXCITON FINE STRUCTURE IN THE INTERBAND MAGNETOABSORPTION OF InSb AND GERMANIUM

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Careful examination of individual absorption peaks in the low-temperature magnetoabsorption spectra of InSb and germanium reveal fine structure attributable to excitons. This confirms theoretical predictions concerning excitons in a high magnetic field and provides a means of experimentally determining the role of excitons in the analysis of the magnetoabsorption spectra for band parameters and for polaron effects.

We have observed and identified exciton fine structure in InSb and germanium with applied magnetic field. These results provide a means of experimentally evaluating the contribution of exciton effects to the position of the absorption lines. Now one can obtain more precise band parameters from the interband magnetoabsorption spectra of these materials than those obtained previously by neglecting exciton energy shifts^{1,2} or by using theoretical estimates.³ Excitons can be expected to add complications to the analysis of the polaron effects previously observed in the interband magnetoabsorption of InSb,^{4,5} and a good knowledge of the role of excitons should contribute to the understanding of the polaron observations. The exciton problem in the interband magnetoabsorption of these materials is essentially that of a hydrogen atom in a high magnetic field, which is a problem of long-standing theoretical interest.⁶⁻¹¹

Absorption peaks associated with exciton formation have not yet been observed in InSb for $H = 0$. A likely explanation for this situation is that the linewidths of the hypothetical absorption lines are too large relative to the spacings of the exciton energy levels and in-

dividual peaks cannot be resolved from the continuum absorption. One might say that the broadened discrete exciton states form a quasicontinuum. Elliott¹² has shown theoretically that the infrared absorption involving the exciton quasicontinuum is continuous, with the absorption involving the true continuum. If one compares the quadratic Zeeman effect on the exciton ground state with the behavior of the $n = 0$ level, it becomes obvious that the exciton ionization energy should increase with magnetic field, and several workers have extended this consideration to high magnetic fields where the cyclotron energy is comparable with and much greater than the zero-field exciton binding energy.⁶⁻¹¹ One might expect that at sufficiently high magnetic fields one would begin to resolve discrete exciton lines from the continuous absorption. Indeed, experimentally it is found that the application of a small magnetic field (~ 2 kG) results in the appearance of peaks in the absorption. The difficulty, however, is that in the presence of a magnetic field, peaks occur in the density of states in the continuum and corresponding peaks might be expected in the absorption. There is then a question of experimentally distinguishing be-

tween peaks in the absorption due to discrete exciton states and peaks in the absorption due to Landau-level formation. Zwerdling, Kleiner, and Theriault³ found in analyzing their magnetoabsorption data for InSb that, in order to fit with band theory the energy of each peak observed, a certain exciton binding energy had to be assumed in each case. Pidgeon and Brown, however, found that they could neglect exciton effects in fitting their data on InSb with an improved band theory.² In analyzing our data showing polaron effects in the interband magnetoabsorption of InSb, we encountered certain difficulties which appear to be due to excitons.⁵

A single exciton line is observed for $H = 0$ in the case of germanium.¹³ Early work on the magnetoabsorption spectrum of germanium^{1,14} was complicated by effects due to sample strain. Later work by Edwards and Lazareva¹⁵ on unstrained samples showed a lowest absorption peak which displayed a behavior with magnetic field suggestive of a quadratic Zeeman effect and which extrapolated to the exciton peak at $H = 0$. Higher energy peaks, observed with magnetic field, extrapolated to an energy value below the $H = 0$ exciton peak. These results were taken to indicate that the absorption lines observed were all associated with discrete exciton states.

To clarify the situation in regard to InSb and germanium, we have closely examined individual absorption peaks in the magnetoabsorption spectrum of each of these for fine structure which might demonstrate the presence of exciton effects and provide a means of experimentally determining the size of these effects and their behavior with magnetic field. The InSb studied was n type with $n \approx 10^{14} \text{ cm}^{-3}$. The germanium was n type with $n \approx 4 \times 10^{12} \text{ cm}^{-3}$. In Fig. 1 we show the observed transmission with $E \parallel H$ for photon energies in the neighborhood of the transitions involving the two lowest energy conduction-band Landau levels. In each of these the conduction-band Landau quantum number is 0. They differ in the sense of the conduction-band electron spin and the valence-band level involved. With the anomalously large conduction-band g factor in InSb, these two transitions are well separated in energy, whereas in germanium they are essentially degenerate for the magnetic fields used. The data were taken with $H = 39 \text{ kG}$ and $T \approx 20^\circ\text{K}$. In each case symmetric strong absorption peaks con-

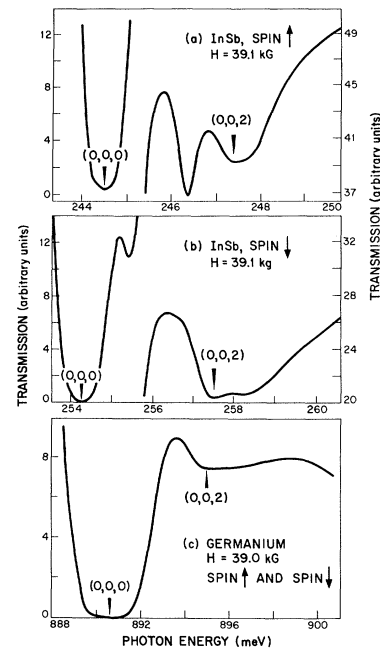


FIG. 1. Observed exciton fine structure in the magnetoabsorption spectra of (a), (b) InSb and (c) germanium. The traces on the right-hand side of (a) and (b) go with the transmission scale on the right-hand side.

stitute the main absorption. To higher energies fine structure is indeed observed. For InSb, but not for germanium, a second symmetrical peak is observed at intermediate energies. At higher energies an absorption edge with absorption tailing off slowly to higher energies is observed in each case. Somewhat similar structure has also been observed in the electromagnetoreflection spectrum of InSb by Pidgeon, Groves, and Feinleib.¹⁶

This type of line shape is to be compared with the line shape predicted by Elliott and Loudon⁷ shown in Fig. 2. If one neglects the Coulomb interaction between electron and hole, the transition probability is independent of energy and the absorption shows a sharp rise at $E_g + \frac{1}{2}\hbar\omega_c$ and follows the density of states in a Landau level. The effect of the Coulomb interaction on the transition probability is such as to remove the peak in absorption expected at $E_g + \frac{1}{2}\hbar\omega_c$, giving an absorption that varies slowly beyond the threshold for Landau-level absorption. Discrete exciton lines occur at lower energy. With spectral linewidths the order of the zero-field exciton binding energy, only a single exciton peak is resolved. The highly excited exciton states give an ab-

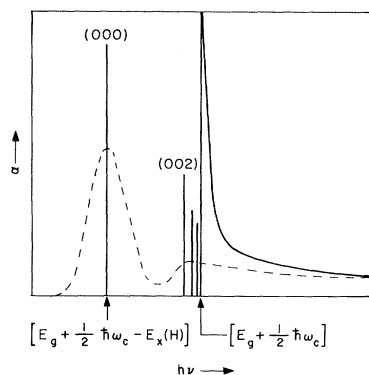


FIG. 2. The theoretically predicted exciton fine structure for the case of optical linewidths approximately equal to R , and $\gamma=2$ (dashed curve). Here $E_x(H)$ is the exciton ionization energy. The solid curve shows the Landau-level absorption in the absence of broadening and Coulomb interaction. The solid lines show the discrete exciton absorption in the absence of broadening. Only the first few of these are shown, and the theoretical strengths of these lines are orders of magnitude greater than shown. The lines in the diagram are merely used to illustrate the decrease in intensity and the crowding together of the discrete lines as they approach the series limit at $E_g + \frac{1}{2}\hbar\omega_c$. (After Elliott and Loudon.⁷)

sorption edge with absorption at higher energies continuous in magnitude and slope with the absorption involving the Landau-level transitions. The theoretically predicted spectrum is shown dashed in Fig. 2.

In discussing an exciton in a magnetic field it is convenient to define a reduced magnetic field:

$$\gamma = \frac{1}{2}\hbar\omega_c/R, \quad (1)$$

where $\hbar\omega_c$ is the cyclotron energy,

$$\hbar\omega_c = 2(m/\mu)\beta H, \quad (2)$$

and R is the zero-field exciton ionization energy,

$$R = (\mu/m)(R_0/\kappa^2). \quad (3)$$

Here μ is a suitable reduced electron-hole effective mass; κ is the static dielectric constant. The quantities m , β , and R_0 are, respectively, the free-electron mass, the Bohr magneton, and the Rydberg constant. A magnetic field is considered high for a particular material if $\gamma \gg 1$. The theoretical spectrum shown in Fig. 2 corresponds to $\gamma=2$. For InSb $\gamma=2$ implies $H \approx 4$ kG and for germanium, $H \approx 25$ kG.

The line shape observed experimentally in InSb and in germanium agrees with that predicted theoretically except that in InSb an additional symmetric absorption peak is observed. This latter peak is anomalous and appears to be associated with transitions involving additional close-lying valence-band levels as predicted theoretically by Bell and Rogers.¹⁷ We will discuss these peaks more extensively in a future publication. We identify the strong symmetric peak with the transition involving the exciton ground state, designated by Elliott and Loudon⁷ (000); and the edge, with a transition involving the (002) exciton excited state. The latter is the lowest even excited state. The absorption to the high-energy side of the edge involves additional exciton levels and the true continuum.

We now consider the qualitative behavior of these features with magnetic field. At high magnetic field (i.e., large γ , which is achieved in the present experiments only for InSb) the separation between the exciton peak and the edge changes slowly with H in qualitative agreement with the theory, which will be discussed briefly below. At lower magnetic fields only the main absorption peak is resolved. The behavior there is suggestive of a quadratic Zeeman effect which is consistent with the identification of the main peak with the exciton ground state. The position of the edge absorption extrapolates at $H=0$ to a value 0.4 meV above the extrapolation of the main absorption peak. This value should be slightly less than the zero-field exciton binding energy.

The experimental data display the qualitative features of the exciton fine structure predicted by Elliott and Loudon.⁷ It is not so easy to obtain good quantitative comparison between experiment and theory. Unfortunately, the Elliott and Loudon theory does not take into account several complications known to be of consequence in InSb and in germanium. The most difficult of these is the complicated nature of the valence band. In the absence of a suitable theory, one might attempt to fit the Elliott and Loudon theory to experiment using some empirically determined average hole mass which is independent of magnetic field. In the first attempt to fit the data we shall assume this average hole mass to be equal to the heavy-hole mass.

Another complication is due to the fact that the exciton states involved are s -like states

where there is a large probability of small separations between electron and hole. The resulting central-cell corrections should give larger binding energies than those predicted by effective mass theory.¹⁸

For $\gamma \ll 1$ the shift of the exciton ground state is given by¹⁹

$$E_{(000)}(H) - E_{(000)}(0) = \frac{1}{2}R\gamma^2. \quad (4)$$

The numerical results of Elliott and Loudon,⁷ which apply for $\gamma \gg 1$, can be given by¹⁹

$$E_{(000)}(H) - E_{(000)}(0) = R(\gamma - 1.6\gamma^{1/3}). \quad (5)$$

So that for very high magnetic fields the exciton ground state moves nearly parallel with the Landau level as the magnetic field changes. In comparison, the separation of the excited state from the Landau level varies much more slowly with magnetic field and is equal to R in the limit as γ goes to infinity. The shift with magnetic field for $5 < \gamma < 200$ can be approximated by

$$E_{(002)}(H) - E_{(002)}(0) = R(\gamma - 0.342\gamma^{1/9.4}). \quad (6)$$

In Fig. 3 we compare the experimental energy separation between the exciton peak and the edge with the calculation of the separation between the (000) exciton state and the (002) exciton state. The theoretical values fall about 15% below the experimental values for the spin-up transition and one is tempted to vary parameters to obtain a better fit. An infinite hole mass would raise the theoretical energy by only 5%. The other parameters are firmly established so that the ~15% discrepancy must be due to the simplicity of the theory. One might attribute the extra binding to central-cell corrections to the binding energy. However, the 12% difference between the data for the two sets of transitions indicates that complications due to the complex valence band may be significant. One way to obtain better agreement is to identify the absorption edge directly with the Landau level. However, as has already been stated, this would not be consistent with the theory of Elliott and Loudon,⁷ and Hasegawa and Howard.⁹

Similar quantitative comparison of experiment and theory for germanium is more uncertain since at 37.5 kG γ is only 3 and the Elliott and Loudon theory is not good as γ approaches 1. The magnitude of the theoretical and experimental energy separations agree within 10%, but the experimental separations de-

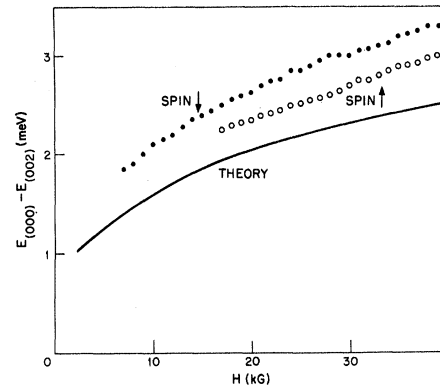


FIG. 3. Comparison of the experimental energy separation between the exciton peak and the absorption edge for InSb with the theoretical calculation of the separation between the exciton states $E_{(000)}$ and $E_{(002)}$. The values $\mu = 0.0136m$, $\kappa = 17.8$ were used in the calculation.

crease more rapidly with decrease in field than would be expected theoretically. A good understanding of the situation must await further experimental and theoretical work.

Finally, we have looked for exciton fine structure in magnetoabsorption peaks corresponding to higher Landau quantum numbers, where the Coulomb binding should decrease. Structure in transitions involving the $n = 1$ conduction-band Landau levels has been seen both for InSb and for germanium, but the analysis of these results is still tentative.

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AC MEASUREMENT OF THE HEAT CAPACITY OF NICKEL NEAR ITS CRITICAL POINT*

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Recent revival of interest in critical-point phenomena^{1,2} creates a need for data giving the temperature dependence of the heat capacity, $C(T)$, at temperatures extremely close to the critical value, T_c . This Letter reports preliminary measurements of the specific heat of Ni near its Curie point ($\sim 625^\circ\text{K}$). The data were obtained using a simple but exceptionally sensitive ac calorimetric technique which permits measurement of the temperature variation of $C(T)$ over intervals less than 0.01°K (i.e., $\epsilon = (T - T_c)/T_c \approx 10^{-5}$).

The method of measurement is shown schematically in Fig. 1(a). The sample was a 0.025-mm thick nickel foil weighing about 0.4 mg, to which was spot-welded a Chromel-Alumel junction of mass much less than that of the foil. The "cold junction" of the thermocouple was also situated in the oven. The foil was periodically heated by chopped light (26 Hz) from a tungsten lamp with intensity constant to 0.5%. The temperature variation of the sample relative to the "cold junction" is converted by means of the Peltier effect into an ac voltage. The voltage is amplified and measured by means of lock-in detection referred to the phase of the chopped light.

A second thermocouple placed very close to the sample monitors the average temperature, T , of the oven. The output of the lock-in amplifier is proportional to $\Delta T = \Delta Q/C(T)$, where ΔT is the rms temperature rise, ΔQ is the energy absorbed by the Ni foil per cycle, and $C(T)$ is the heat capacity of the foil. An x-y recorder is used to plot ΔT vs T as the specimen temperature drifts slowly past

the critical point. A typical chart record is shown in Fig. 1(b) on which the sharp minimum of ΔT corresponds to the cusplike singularity in $C(T)$ at the critical point. The rms temper-

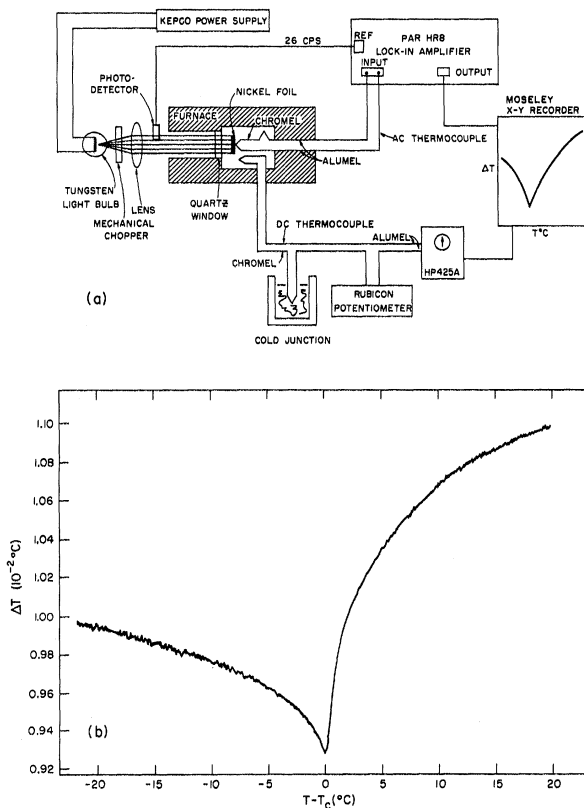


FIG. 1. (a) A schematic diagram of the experimental apparatus. (b) X-Y recorder trace of the experimental data. ΔT vs T over a range of 20°C on either side of the critical point.