

QUANTUM EFFECTS IN CYCLOTRON RESONANCE IN *p*-TYPE TELLURIUM

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Quantum effects of cyclotron resonance of holes have been observed in tellurium by using high-resolution HCN and D₂O submillimeter laser spectrometers. The multiplicity of resonance absorption lines in both parallel and perpendicular directions with respect to the principal axis suggests complex energy surfaces with highly warped bands. In addition, cyclotron resonance of electrons thermally excited across the gap was obtained.

We have observed cyclotron resonance of holes and electrons in tellurium using steady magnetic-field intensities up to 165 kOe and an HCN laser source operating at a wavelength of 337 μm . A large number of absorption lines have been resolved as shown in Fig. 1. Since the quantum conditions are satisfied in the present experiments, namely, the laser photon energy $\hbar\omega$ is larger than the Fermi energy and also $\hbar\omega > kT$, we shall interpret the multiplicity of these absorption lines as the first observation of quantum effects in tellurium. In previous cyclotron resonance experiments¹⁻³ at millimeter wavelengths, the dual conditions $\omega\tau \gg 1$ and $\hbar\omega > kT$ were only marginally satisfied and quantum effects were not resolved. The cyclotron-resonance nature of the present absorption lines was confirmed by observing the main features at a higher frequency using a continuous-wave D₂O laser.

There have been indications from Shubnikov-de Haas experiments⁴ in heavily doped tellurium that the shape of the Fermi surface is complicated. The quantum effects in submillimeter cyclotron-resonance experiments due to warping are

consistent with these studies. The first indication of structure in cyclotron-resonance observations was seen at a wavelength of 700 μm by Picard and Carter² in the form of a second harmonic but they observed no structure when the magnetic field was oriented parallel to the principal axis. Nevertheless, in view of the various experimental data accumulated over the past few years, a consensus has been developing that the surfaces of constant energy appear to consist of four ellipsoids of revolution at low energy close to the point *H* at the edge of the Brillouin zone, which then merge into two dumbbells as the energy is increased to a few millielectron volts. The axis of revolution of the dumbbells coincides with the hexagonal edges of the zone. Although we have not yet completed the detailed theory for these quantum effects in cyclotron resonance, we believe that the structure shown in the spectra of Fig. 1 indicates that this particular model of the Fermi surface is inadequate.

Our experiments were performed with a continuous-wave HCN molecular gas-laser spectrometer described previously.⁵ The magnetic field was furnished by a water-cooled copper so-

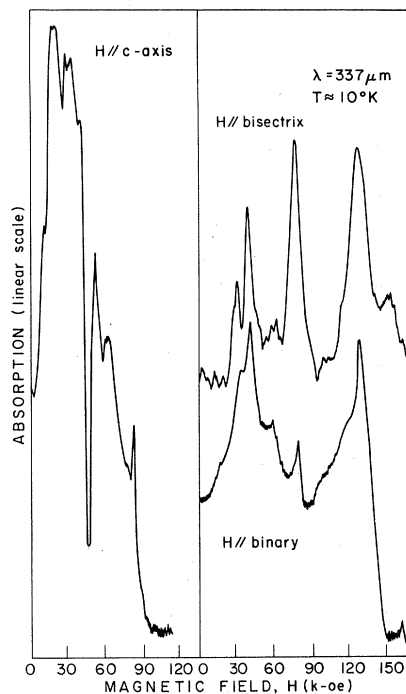


FIG. 1. Low-temperature cyclotron-resonance absorption spectra in tellurium for $\hbar\omega > kT$. Both the direction of propagation and static magnetic field were parallel (left side) or perpendicular (right side) to the c axis of tellurium. All curves are tracings of pen recordings.

lenoid. The field was applied parallel to the direction of propagation of the unpolarized monochromatic laser radiation. Alternate single-crystal specimens were used having the c axis, bisectrix axis, and binary axis parallel to the magnetic field. The stabilized laser output was of the order of microwatts and had a noise level of less than $\frac{1}{2}\%$. The signal was detected with a Golay cell. All crystals were spark-cut cylinders prepared to fill the $\frac{3}{8}$ -in-diam light pipe of the laser spectrometer. The end faces had a mirrorlike finish prepared by polishing and etching. Some specimens were annealed in the presence of tellurium powder after this treatment. The crystals were of high purity having hole concentrations between $(1 \text{ and } 5) \times 10^{14} \text{ cm}^{-3}$ at room temperature. Thus the submillimeter frequency was much larger than the plasma frequency.

Figure 1 shows representative spectra recorded at about 10°K for three different orientations. The spectrum at the left side of the figure shows a multitude of absorption lines characteristic of the spectra observed whenever the magnetic field was applied along the c direction. The c -ax-

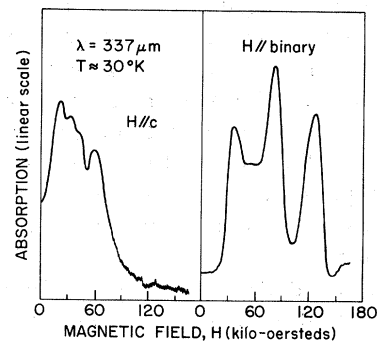


FIG. 2. Higher temperature spectra for $\hbar\omega \approx kT$. Each broad strong envelope covers several unresolved quantum transitions and, in the perpendicular direction, the relative intensities have changed with temperature.

is specimens were essentially transparent at field intensities above 90 kOe. When the field was applied perpendicular to the c axis, however, absorption lines were observed at field intensities as high as 130 kOe. The main features of the spectra for the binary and bisectrix directions appear at the same fields. Some differences in the spectra may exist but they have not yet been fully investigated.

Figure 2 shows that as the temperature is increased to about 30°K where $\hbar\omega \approx kT$, the fine structure merges into broad strong envelopes. This is characteristic of the quantum effects previously observed in p -InSb and p -Ge as a function of temperature.⁶ Then only the envelopes of several unresolved quantum transitions remain. In this case of tellurium, the centers of the envelopes are observed at magnetic field intensities of 29 and 63 kOe for the c direction corresponding to effective masses of $0.09m_0$ and $0.2m_0$. For the binary direction, the three envelopes are observed at magnetic fields of 36, 82, and 127 kOe corresponding to effective masses of $0.11m_0$, $0.26m_0$, and $0.40m_0$. Previous observers¹⁻³ have quoted cyclotron masses of about $0.11m_0$ (c direction) and $0.17m_0$ (perpendicular direction). It should be noted that we also observe a mass of about $0.4m_0$ in the perpendicular direction which has never been seen before in a low-temperature experiment. It is interesting to note that high-frequency conductivity experiments⁷ at high temperatures have indicated the presence of a mass of the order of $0.4m_0$.

In order to confirm that the absorption lines represent cyclotron resonance rather than impurity transitions or other phenomena, we have performed one experiment on the c -face specimen

using the higher frequency ($\lambda = 172 \mu\text{m}$) from a continuous-wave D_2O laser. The weaker D_2O laser radiation handicapped our efforts to reveal all of the structure but the prominent features are clearly evident and correspond to those observed with HCN scaled to this new frequency. Again the appearance of the spectrum changes with temperature, which is characteristic of quantum effects.

The pronounced structure which we see in Fig. 1 with $\vec{H} \parallel \vec{c}$ implies that the constant-energy surfaces are exhibiting warping in the c plane because, for rotational symmetry, only a single line would be expected in the classical limit for an ellipsoid model. Pseudopotential calculations by Picard and Hulin⁸ suggested that trigonal warping is definitely present. If trigonal warping is strong, anisotropy should also appear for $\vec{H} \perp \vec{c}$ as a difference in the spectral structure for the binary and bisectrix direction. However, the main absorption lines appear at the same field intensities, but distinct differences in the intensities of the two spectra do exist. Hence the nature of the trigonal warping has to be re-examined in light of these results.

Our high-temperature studies of cyclotron resonance were carried out using the HCN laser. The photon energy $\hbar\omega$ for the HCN laser is not large enough to see the structure of quantum effects at temperatures higher than about 30°K but $\omega\tau$ is still large enough to observe the envelope of unresolved absorptions up to about 200°K . At a temperature of 150°K , for example, the peak of the envelope is at a magnetic field intensity of about 30 kOe with $\vec{H} \parallel \vec{c}$ which corresponds to a cyclotron mass of slightly less than $0.1m_0$. At this same temperature, 159°K , the cyclotron mass for $\vec{H} \perp \vec{c}$ is $0.28m_0$. We find little evidence at this time for a considerable change of the mass with temperature. As the temperature is increased above 200°K , the cyclotron-resonance absorption of electrons is seen (Fig. 3) because of the thermal excitation of electrons across the forbidden gap. This electron resonance is not visible for $\vec{H} \parallel \vec{c}$ but is well separated from the hole peak for $\vec{H} \perp \vec{c}$ as shown in Fig. 3. It appears at 43 kOe corresponding to a mass of $0.135m_0$. The value of $\omega\tau$ was large enough to follow the electron resonance up to a temperature of about 250°K . This effective cyclotron mass of $0.135m_0$ is considerably higher than the value of 0.047 which can be derived from magneto-optical data.⁹ The reason for the absence of the electron resonance for the direction $\vec{H} \parallel \vec{c}$ is

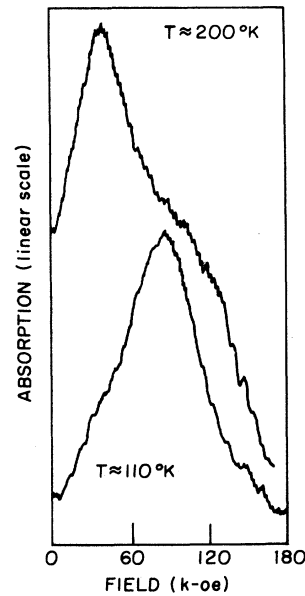


FIG. 3. Cyclotron resonance of electrons (upper spectrum) and holes (lower spectrum) in $p\text{-Te}$ with $\vec{H} \parallel$ bisectrix direction and $\lambda = 337 \mu\text{m}$. The electrons are thermally excited across the forbidden gap as the specimen is allowed to warm up.

not clear at present. Either the effective mass is so high that the resonance would occur beyond 165 kOe, or the electron resonance is superimposed on the hole absorption at about 30 kOe. If the latter occurs, it should be possible to sort out the effect by using circularly polarized radiation.

One of our c -face specimens of slightly higher carrier concentration was examined in an attempt to discover the possible existence of very weak absorption lines at high fields which could not be seen for the c direction in Fig. 1. This sample did not transmit at field intensities below 60 kOe because of strong cyclotron-resonance absorption but then showed oscillatory transmission in the transparent region at field intensities above 60 kOe. The oscillations were clearly resolved and grew larger and wider spaced at higher fields but did not display a simple $1/H$ dependence. The analysis of this interference phenomenon will be discussed separately.¹⁰ In the present data, the two phenomena of cyclotron absorption and oscillatory interference are clearly separated because multiple internal reflections cannot survive in the presence of significant cyclotron-resonance absorption.

In conclusion; it appears that a more refined theoretical model of the valence bands near the band edge will have to be formulated. The sim-

ple model that has served to explain previous experimental results is inadequate to deal with the latest findings. The $\mathbf{k} \cdot \mathbf{p}$ perturbation method is probably appropriate because a relatively small portion of the zone needs to be considered. This theoretical approach is now being pursued for the purpose of calculating the Landau levels and the selection rules. At this time it is possible to say with reasonable confidence that transitions from impurity levels¹¹ are not present in these data because not only do we find the main features to scale with frequency but also the necessary carrier freeze-out phenomenon has never been observed in tellurium either at low temperature or high magnetic field.⁷ Further experiments are to be attempted using linear and circular polarized radiation to reduce complications that may be introduced by birefringence.

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¹J. H. Mendum and R. N. Dexter, *Bull. Am. Phys. Soc.*, **9**, 632 (1964).

²J. C. Picard and D. L. Carter, *J. Phys. Soc. Japan Suppl.*, **21**, 202 (1966).

³P. L. Radoff and R. N. Dexter, *Bull. Am. Phys. Soc.*, **14**, 330 (1969).

⁴E. Braun and G. Landwehr, *J. Phys. Soc. Japan Suppl.*, **21**, 380 (1966); C. Guthmann and J. M. Thuillier, *Solid State Commun.*, **6**, 835 (1968); M. S. Bresler, I. I. Farbstein, D. V. Mashovets, Yu. V. Kosichkin, and V. G. Veselago, *Phys. Letters* **29A**, 23 (1969).

⁵K. J. Button, H. A. Gebbie, and B. Lax, *IEEE J. Quantum Electron.*, **2**, 202 (1966); C. C. Bradley, K. J. Button, B. Lax, and L. G. Rubin, *IEEE J. Quantum Electron.*, **4**, 733 (1968).

⁶K. J. Button, B. Lax, and C. C. Bradley, *Phys. Rev. Letters* **21**, 350 (1968); K. J. Button, A. Brecher, B. Lax, and C. C. Bradley, to be published.

⁷P. Grosse, *Z. Physik* **193**, 318 (1966).

⁸M. Picard and M. Hulin, *Phys. Status Solidi* **23**, 563 (1967).

⁹P. Grosse and K. Winzer, *Phys. Status Solidi* **26**, 139 (1968).

¹⁰K. J. Button, G. Landwehr, C. C. Bradley, B. Lax, and D. Cohn, to be published.

¹¹Y. Couder, *Phys. Rev. Letters* **22**, 890 (1969).

NONANALYTIC BEHAVIOR ABOVE THE CRITICAL POINT IN A RANDOM ISING FERROMAGNET*

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It is shown that in a class of randomly diluted Ising ferromagnets the magnetization fails to be an analytic function of the field H at $H=0$ for a range of temperatures above that at which spontaneous magnetization first appears.

It is commonly assumed¹ that the critical point of a simple ferromagnet represents a complete termination of the first-order phase transition indicated by the presence of spontaneous magnetization, and above the critical temperature T_c where this spontaneous magnetization disappears, the free energy is an analytic function of temperature T and magnetic field H . In other words, for $T > T_c$ there is no "higher-order" transition representing a continuation of the first-order transition for $T < T_c$. We shall show that in the case of certain random Ising ferromagnets this assumption is demonstrably false, and the tem-

perature T_c^* where nonanalyticity begins as T decreases definitely exceeds T_c .

The random ferromagnets of interest are obtained by starting with an ordinary or regular Ising model with spins located on the vertices of a regular two- or three-dimensional lattice, with nearest-neighbor exchange interactions. In the corresponding random ferromagnet² only a fraction p of the lattice sites are occupied with Ising spins, the rest remaining vacant, and exchange interactions exist only between spins on neighboring pairs of occupied sites.³ The probability p of occupancy of a given site is independent of H , T ,