INFRARED MAGNETOREFLECTION IN BISMUTH. I. HIGH FIELDS

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The pulsed-field infrared experiments on bismuth reported by Keyes et al.[1] at wavelengths between 10 and 22 μ and at room temperature, were originally interpreted as transitions corresponding to cyclotron resonance. Well-defined dispersion curves were obtained by reflection techniques. The value of the magnetic field at the center of each line and the measured wavelength were utilized to obtain an apparent effective mass from the well-known cyclotron equation $m^* = eH/\omega_c c$. Three distinct sets of mass values obtained from the strongest absorption data[1] yielded apparent effective masses for electrons which showed a large variation with magnetic field, with mass values above and below those deduced from the de Haas-van Alphen effect[2] and microwave cyclotron resonance.[3] This suggested that the large change in the effective masses was due to a rapid change in the curvature of the energy bands. To explain this a theoretical expression was developed for the energy-momentum relation, assuming only two sets of interacting energy bands which are located near the edge of the zone. The derivation uses the $\vec{k}\cdot\vec{p}$ perturbation method, following the work of Kane[4] on InSb and that of Cohen and Blount[5] on bismuth. We showed that the nonparabolic energy dependence of the bands could be represented by the expression[6]

$$\epsilon\left(1+\frac{\epsilon}{\epsilon_g}\right)=\vec{p}\cdot\frac{\alpha}{2}\cdot\vec{p}, \tag{1}$$

where ϵ_g is the energy gap and α is the inverse mass tensor at the bottom of the conduction band and (with a minus) at the top of the valence band. When a magnetic field is applied, the Hamiltonian including spin becomes

$$\mathcal{H}_{\text{eff}}=\left[\vec{\Pi}\cdot\frac{\alpha}{2}\cdot\vec{\Pi}+\frac{\beta}{2}\vec{\sigma}\cdot g\cdot\vec{H}\right]\frac{1}{1+\epsilon/\epsilon_g}, \tag{2}$$

where $\vec{\Pi}=\vec{p}+e\vec{A}/c$, $\vec{A}$ is the magnetic vector potential, β is the Bohr magneton, and g is the effective spectroscopic splitting tensor[5] at the bottom of the bands. The solution of Eq. (2) with the magnetic field taken in the z direction yields an equation for the eigenvalues for the valence and conduction bands,

$$\epsilon_n\left(1+\frac{\epsilon_n}{\epsilon_g}\right)=(n+\tfrac{1}{2})\hbar\omega_c+\frac{\hbar^2k_z^2}{2m_z}\pm\tfrac{1}{2}\beta g_{\text{eff}}H, \tag{3}$$

where

$$\epsilon_n=-\tfrac{1}{2}\epsilon_g\pm\left\{(\tfrac{1}{2}\epsilon_g)^2+\left[(n+\tfrac{1}{2})\hbar\omega_c+\frac{\hbar^2k_z^2}{2m_z}\pm\tfrac{1}{2}\beta g_{\text{eff}}H\right]\epsilon_g\right\}^{1/2}, \tag{4}$$

$\omega_c=eH/m^*c$, cyclotron mass $m^*=[\alpha_1\alpha_2\alpha_3 m_z]^{-1/2}$, $m_z=\alpha_1^{-1}\lambda_1^2+\alpha_2^{-1}\lambda_2^2+\alpha_3^{-1}\lambda_3^2$, and $g_{\text{eff}}=[g_1^2\lambda_1^2+g_2^2\lambda_2^2+g_3^2\lambda_3^2]^{1/2}$; the α_i are the components of the inverse mass tensor α and the λ_i are the directional cosines of the magnetic field relative to the principal ellipsoidal axes. The plus sign in front of the radical in Eq. (4) gives the energy of the conduction band and the minus sign, that of the valence band.

In attempting to fit the experimental curves for bismuth with Eq. (4), assuming cyclotron resonance, it was necessary to choose very small values for the energy gap ϵ_g (~0.01 ev) and also for the effective masses; at room temperature the data yielded masses at the Fermi level approximately one quarter of those found at low temperatures.[2,3] In addition, two other dilemmas were present. The first involved the existence of well-defined "resonant" reflection traces, such as shown in Fig. 1. This was in contradiction

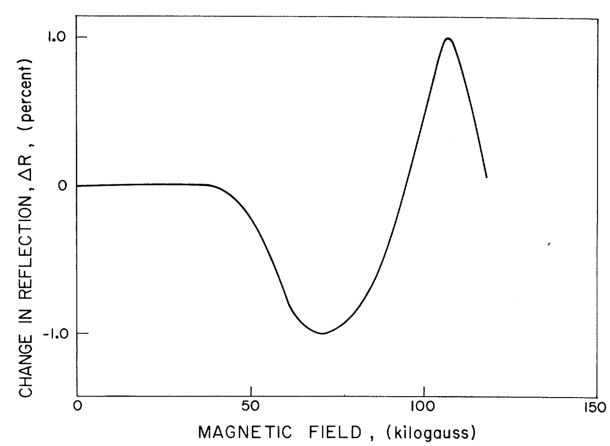

FIG. 1. Trace of the "resonance" dispersion curve for bismuth obtained by reflection using pulsed magnetic fields at infrared wavelength, $\lambda=13.0$ microns.

with the expected magnetoplasma effect for bismuth at room temperature, where the plasma wavelength occurs at $\sim 10\ \mu$ for a carrier concentration of $n \approx 10^{18}/\mathrm{cm}^3$ as derived from galvanomagnetic data.[7] From theoretical considerations, a resonant reflection trace should not have been observed over at least part of the wavelength range (i.e., near $20\ \mu$). The second puzzle involved the appearance of two resonances with the magnetic field in the binary direction $[11\bar{2}0]$, where only one was expected. The intensity of the extra line appeared greater than that of the line which agreed best with known values of masses.

These difficulties all seem to be resolved if it is assumed that the observed transitions do not correspond to intraband cyclotron resonance but are interband transitions between magnetic levels of the valence and conduction bands near the edge of the Brillouin zone. Such direct transitions are known to have high absorption coefficients from similar studies in semiconductors.[8] The estimated penetration depth for this process in bismuth is of the order $\sim 0.1\ \mu$, whereas that for free carrier penetration due to the presence of a plasma (for $\omega\tau \gg 1$) would be one order of magnitude larger, i.e., $\sim 1\ \mu$.

The explanation of the first dilemma can be considered in a more general context as suggested by Dresselhaus.[9] The argument is similar to that of Anderson[10] for the direct observation of cyclotron resonance of minority carriers in the presence of majority carriers even at a frequency well below the plasma frequency. If the complex conductivity of the medium consists of a large part $\sigma(\omega, H)$ which varies slowly with magnetic field and a small part $\Delta\sigma(\omega, H)$ which varies "resonantly" with magnetic field, then the "resonance" line will be observable, modified in shape only slightly by the background absorption. Thus a trace as in Fig. 1 of reflection versus magnetic field will represent the "resonance" due to $\Delta\sigma(\omega, H)$. In the particular case of bismuth the dominant contributions to $\sigma(\omega, H)$ are the nonresonant background interband transitions and $\Delta\sigma(\omega, H)$ is due to the small "resonant" interband transitions between Landau levels. The analysis for such a situation in the absence of a plasma has been treated for semiconductors.[11] The important conclusion for other metals is that the presence of a large plasma contribution $\sigma(\omega, H)$ due to the conduction electrons should not interfere with the observation of "resonant" interband transitions between Landau levels, although the frequency at which these observations are made is below the plasma frequency. However, the intensity of the magnetoreflection lines in a more highly conducting metal would probably be smaller than that in bismuth.

In order to test the hypothesis that interband transitions have been observed, the pulse data reported were replotted in terms of photon energy as a function of magnetic field. The three sets of lines converged to a value at zero field of 0.040 $\pm$ 0.005 ev, corresponding to the energy gap near the edge of the zone at room temperature. Had these been cyclotron resonance transitions the lines would have converged to the origin. Furthermore, upon re-examining the original photographic traces, we found additional weak lines corresponding to what we believe to be transitions between higher quantum states. With this in mind, it is now possible to interpret the data from a model of the energy level diagram shown in Fig. 2, including spin splitting.[5] However, since the scatter of the present data was large, only a semiquantitative interpretation was possible. It appears that some of the points in the $[11\bar{2}0]$ direction can be explained as transitions between the large mass levels corresponding to the resonance mass value $m^* \sim 0.2m_0$. These appear as reasonably well-defined lines.

We again attempted to interpret the data, this time assuming interband transitions and using the theory for nonparabolic bands since on theoretical grounds Eq. (4) should apply; however, the pulsed data and those in the following Letter indicate that the deviation from parabolicity is not as large as that predicted by this equation. It is possible that an analysis similar to the pres-

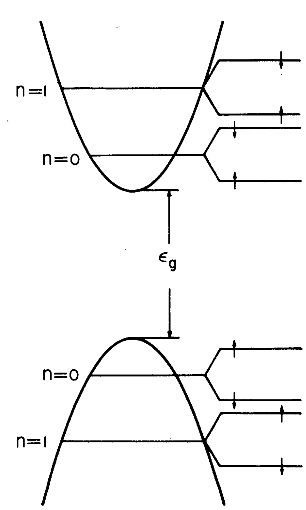

FIG. 2. Model for bismuth of the energy levels near the edge of the Brillouin zone in a magnetic field. For the low-mass directions, the spin splitting of the Landau levels is determined from $g_{\mathrm{eff}} \approx 2m_0/m^*$. The arrows indicate $\vec{j}$ states rather than pure spin states.

ent one, but modified to take into account the effects of other bands, would provide a better model for the quantitative treatment of the data. The effective Hamiltonian given in Eq. (2) can be readily extended to the case of a band interacting with a number of other bands, but the location and character of other bands near the zone edge in bismuth are not at present known.

This experiment indicates the power of the method of interband magnetoreflection measurements in exploring the energy band structure of metals. Masses and g values for both conduction and valence bands can be studied. In addition, bands can be studied not only at the Fermi surface (as with de Haas-van Alphen effect and cyclotron resonance) but at energies above the Fermi energy; and information about lower lying bands can be obtained. Furthermore, the temperature dependence of the band parameters can be studied up to room temperature and possibly higher. It is interesting that the detection of transitions due to a mass as large as $\sim 0.2m_0$ is possible at room temperature and with transient pulse techniques. This lends encouragement to experiments in other metals with high carrier masses at low temperatures, with steady magnetic fields. By using magnetic fields of the order of 100 kilogauss and higher, this method could be applicable to the investigation of a number of metals.

The pulse experiments logically suggested the possibility that this phenomenon could be observed at lower fields and at low temperatures with photon energies well above those used in the pulse experiments, thereby permitting the observation of interband transitions to higher quantum numbers. Such experiments were indeed successfully realized and are discussed in the following Letter.

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[1]R. J. Keyes, S. Zwerdling, S. Foner, H. H. Kolm, and B. Lax, Phys. Rev. 104, 1804 (1956). In this experiment damped oscillatory magnetic fields up to 150 kgauss, with a half period of approximately 150 microseconds, were used.

[2]B. Lax, K. J. Button, H. J. Zeiger, and L. M. Roth, Phys. Rev. 102, 715 (1956).

[3]J. K. Galt, W. A. Yager, F. R. Merritt, B. B. Cetlin, and A. D. Brailsford, Phys. Rev. 114, 1396 (1959).

[4]E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957).

[5]M. H. Cohen and E. I. Blount, Phil. Mag. 5, 115 (1960).

[6]B. Lax, Bull. Am. Phys. Soc. 5, 167 (1960); B. Lax and J. G. Mavroides, Advances in Solid-State Physics, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1960), Vol. 9. Recently P. A. Wolff has also obtained the above result (private communication).

[7]B. Abeles and S. Meiboom, Phys. Rev. 101, 544 (1956).

[8]W. C. Dash and R. Newman, Phys. Rev. 99, 1151 (1955).

[9]G. Dresselhaus (private communication).

[10]P. W. Anderson, Phys. Rev. 100, 749 (1955).

[11]B. Lax, Phys. Rev. Letters 4, 511 (1960).

INTERBAND MAGNETOREFLECTION IN BISMUTH. II. LOW FIELDS

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Magnetoreflection measurements in bismuth have been carried out at liquid air temperatures and below with static magnetic fields up to 38 kilogauss. The measurements were made using a magnetospectrometer similar to that of Zwerdling *et al.*,[1] except that infrared reflection techniques were used. Best results were obtained by sweeping the magnetic field at a fixed wavelength. This was done at wavelengths between 6 and 14 μ, using an NaCl prism.

Single-crystal specimens were grown from Bi of "99.999" % purity. Surfaces were obtained by cleaving at nitrogen temperature along a trigonal plane. A diamond saw was used to cut two samples 2 mm × 3 mm × 25 mm from the crystal. The cuts were made so that the magnetic field parallel to the exposed trigonal surface was in one case along the bisectrix axis $[10\bar{1}0]$, and in the other case along the binary axis $[11\bar{2}0]$. The surface was then polished with Buehler No. 3 grit to obtain a good reflecting surface. The surface strains were removed by electropolishing.[2] Back-reflection Laue patterns gave the crystal orientation and revealed minor strains. The samples