Unambiguous determination of the g factor for holes in bismuth at high B/T

S. G. Bompadre,[1] C. Biagini,[1,2] D. Maslov,[1] and A. F. Hebard[1]
[1]*Department of Physics, University of Florida, Gainesville, Florida 32611-8440*
[2]*Istituto Nazionale di Fisica della Materia, L.go E. Fermi 2, 50125, Firenze, Italy*
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Magnetotransport has been investigated in high-purity bismuth crystals in static magnetic fields as high as 20 T and temperatures as low as 25 mK. This high B/T ratio permits observation of pronounced Shubnikov–de Haas oscillations over a wide field range and up to fields where most of the carriers are in the lowest Landau level. For transport currents in the bisectrix or binary directions, and field in the perpendicular trigonal direction, we have observed doublet splittings centered on each Shubnikov–de Haas oscillation. These splittings exhibit a quadratic dependence on field and disappear before the last oscillation. Our observations allow us to conclude unambiguously that when the Landau-level index is as high as 2, the carriers are fully polarized and the g factor for holes with the field in the trigonal direction is 35.3(4).

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In this paper we report on magnetotransport measurements of crystalline bismuth at high magnetic fields and low temperatures. These measurements are motivated in part by the unusual electronic properties of the semimetal bismuth that reflect its unique location in an intermediate position between good metals and semiconductors. The rhombohedral unit cell of bismuth can be obtained by slight deformation of a double cubic cell and contains two atoms with five electrons each. These ten electrons fill the first five Brillouin zones and then spill into the sixth zone leaving behind hole pockets in the fifth zone. The Fermi surface is well described by three ellipsoids containing electrons lying almost in the bisectrix-binary plane and one ellipsoid containing holes and oriented along the trigonal axis. The small cross section of this highly anisotropic Fermi surface leads to a small carrier density that is the same for both electrons and holes ($n_{\mathrm{hole}} = n_{\mathrm{electron}} \approx 3\times 10^{17}$ cm^{-3}). Accordingly, bismuth is a perfectly compensated semimetal. This unusual electronic structure together with the ready availability of high-quality crystals with long mean-free-paths, has generated a large field of research[1,2] on magnetic-field-induced oscillations in the magnetization (de Haas-van Alphen effect) and conductivity (Shubnikov–de Haas effect).

For the highest magnetic fields, the carriers can be placed into the lowest Landau level and the quantum limit realized. The direction of the field is important. For example, if the field is along the bisectrix direction (aligned closely with the electron ellipsoids) then the quantum limit can be realized at a few Tesla for the light electrons, while the heavier holes remain in the quasiclassical regime in fields up to 50 T. Guided by these considerations and cognizant of the very limited experimental evidence suggesting the existence of high-magnetic-field-induced correlated states,[3–8] we have initiated magnetotransport studies of bismuth at high B/T, with the aim of finding magnetic-field-induced instabilities that might drive bismuth into a charge-density-wave insulating state. Although our initial results do not show any evidence of such instabilities, they have clarified the meaning of doublet structures that coincide with the Shubnikov–de Haas oscillations and that have a heretofore-unobserved spacing that is quadratic in a field. The observations and analysis reported here reveal an unambiguous determination of the g factor or equivalently the ratio of the Zeeman energy to the cyclotron energy for holes confined to Landau orbits by a field directed along the trigonal axis.

The bismuth crystals used in this paper were obtained from a variety of sources and had purities of 99.9995% or better. The crystals were cleaved at liquid-nitrogen temperatures to expose the trigonal plane and then aligned along the bisectrix or binary directions according to the striation marks created during the low-temperature cleave. These alignments were confirmed using Laue diffraction. Typical crystals were cut with a string saw to 5 mm length and had cross sectional areas on the order of 4 mm^2. After cutting, the crystals were etched in a nitric acetic acid solution and then annealed to temperatures as high as 250 °C for periods as long as a few days. The small area contacts made to the samples using gold wires and low melting temperature solder (Woods metal) had contact resistances on the order of 0.1 Ω. The current direction was chosen to be along the binary (bisectrix) direction and the samples were mounted on a rotating stage so that the applied magnetic field could be rotated in the plane determined by the trigonal and binary (bisectrix) directions. All measurements were taken using low-frequency (<20 Hz) ac techniques.

Shown in Fig. 1 is the dependence of the longitudinal resistance on the reciprocal magnetic field for a binary sample with the magnetic field aligned perpendicular to the trigonal plane. The temperature is at 25 mK and the magnetic field was swept up to 20 T. This high B/T allows more than 30 Shubnikov–de Haas oscillations to be clearly discerned. For clarity, double logarithmic axes were used to accommodate the greater than two-decade range in field and greater than three-decade range in resistance. The choice of logarithmic axes however obscures the presence of almost uniform spacing $\Delta(1/B)$ of the Shubnikov–de Haas oscillations.

Assigning consecutive integers n to the resistance minima of Fig. 1 and then plotting the values of $1/B$ at the respective minima versus n, addresses this aspect of the data analysis. The plot of Fig. 2 shows the results of such a plot for a set of integers that are consistent with a smooth extrapolation of the data to $n=0$. The set of integers assigned in Fig. 2 and

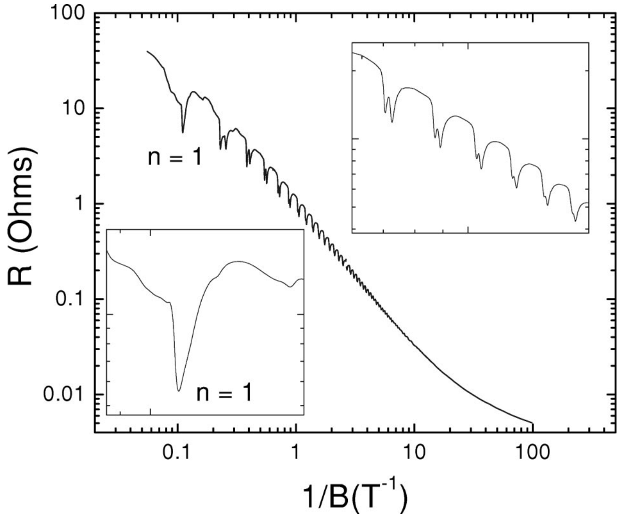

FIG. 1. Resistance vs reciprocal field on logarithmic axes. The upper right inset is an expanded view of the $n=5$ through $n=10$ oscillations, and the lower left inset is an expanded view of the $n=1$ oscillation showing the sharpness of the resistance change as the Fermi energy passes through the fully polarized $\nu=1$ Landau level. The tic marks on the unlabeled inset axes correspond to the tic marks of the labeled axes.

labeled for $n=1$ in Fig. 1, uniquely identifies the order of each Shubnikov–de Haas oscillation. As will be shown below, when spin splitting is taken into account, the order of oscillation n need not be the same as the integer ν identifying the order of the Landau level.

At high fields the local slope in Fig. 2 gives a period for the Shubnikov–de Haas oscillations $\Delta(1/B)=0.146\ \mathrm{T}^{-1}$, in good agreement with experimental values obtained by others for holes[9,10] with the field applied in the trigonal direction. The positive curvature reflects the fact that the valence and conduction bands are affected by the field, giving rise to a change in the carrier density and thus in the Fermi energy. For the field along the trigonal axis, both the carrier density

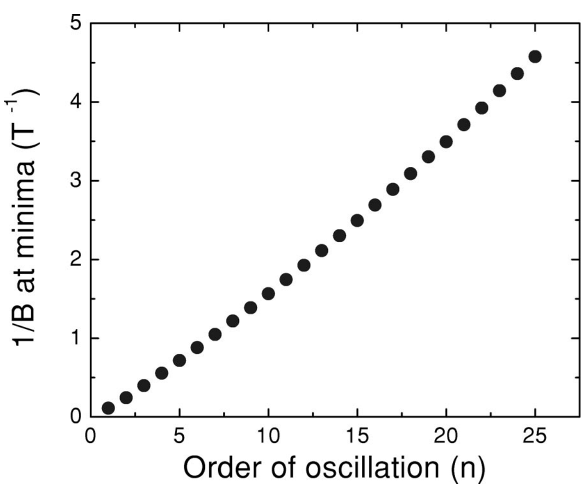

FIG. 2. Plot of the reciprocal fields as a function of integer n for the first 25 resistance minima of Fig. 1. The integers n identify the order of the Shubnikov oscillation with $n=1$ labeled in Fig. 1.

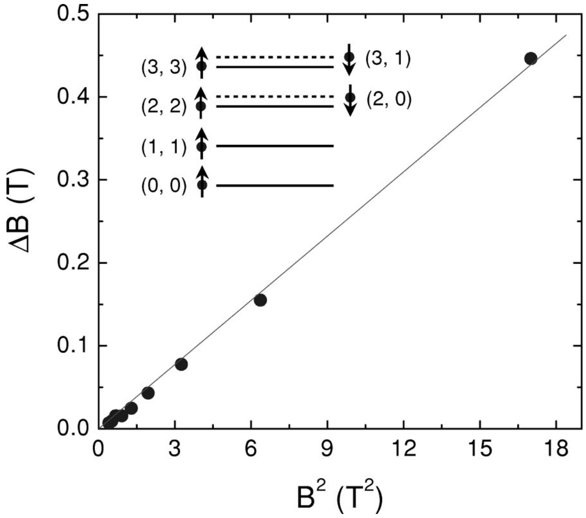

FIG. 3. Plot of the doublet spacings against the square of the field. The inset is a schematic of the energy levels showing the spin direction and the identifications (n,ν) of the order of oscillation n and Landau level ν.

and the Fermi energy increase with the field.[9] This field-induced increase in the Fermi energy and the concomitant decrease in the $1/B$ spacings of the Shubnikov–de Haas oscillations, qualitatively explains the positive curvature. We note that this effect is actually quite small when compared to what is expected when the field is applied in the basal plane, where the Fermi energy has a stronger dependence on B.

A salient feature of the data presented in Fig. 1 is the appearance of doublets coinciding with the valleys of the Shubnikov–de Haas oscillations. These doublets persist to low fields as shown in the inset and as many as 10 can be clearly identified. In addition, the spacing in the field between the subpeaks of a doublet increases with the field. This observation is made more quantitative in the plot of Fig. 3 where the doublet spacings are plotted against the square of the field.

For the oscillatory contribution to the holes' conductivity for the field along the trigonal axis, one has[11]

$$\tilde{\sigma}_{11}=\tilde{\sigma}_{22}\propto\frac{1}{B^{3/2}}\sum_{p=1}^{\infty}\frac{R_D(p)R_T(p)}{\sqrt{p}}\times\left\{\cos\left[2\pi p\left(\frac{l_B^2\epsilon_F}{\hbar^2\alpha_1}-\frac{1}{2}-\frac{g}{2m_0\alpha_1}\right)-\frac{\pi}{4}\right]+\cos\left[2\pi p\left(\frac{l_B^2\epsilon_F}{\hbar^2\alpha_1}-\frac{1}{2}+\frac{g}{2m_0\alpha_1}\right)-\frac{\pi}{4}\right]\right\}, \quad (1)$$

where

$$R_T(p)=\frac{2\pi^2 p(Tl_B^2/\hbar^2\alpha_1)}{\sinh[2\pi^2 p(Tl_B^2/\hbar^2\alpha_1)]} \quad (2)$$

is the temperature smearing factor, and $R_D(p)$ is the disorder smearing factor. The parameters α_i are the components of the inverse mass tensor with respect to the crystallographic axis (1 corresponds to the binary axis, 2 to the bisectrix one, and 3 to the trigonal axes: for the holes, $\alpha_1=\alpha_2$), m_0 is the bare electron mass, $\epsilon_F\simeq 0.012$ eV the hole Fermi energy at zero field, g the hole gyromagnetic ratio, and $l_B=\sqrt{\hbar/eB}$ the magnetic length. The disorder smearing factor has the form

$$R_D(T)=\exp\left[-\frac{2\pi^2 p T_D l_B^2}{\hbar^2\alpha_1}\right],$$

where $T_D=\hbar/2\pi\tau$ is the Dingle "temperature" and τ is the hole mean-free time. Using Eq. (1), we analyzed the smearing of the Shubnikov–de Haas oscillations at the relatively high temperature of 5 K, where the doublets are washed out and the first harmonic ($p=1$) dominates. At this temperature, all of the broadening is found to be thermal, implying that T_D is well below 1 K. This estimate is consistent with our experimentally determined estimate of τ ($\tau\simeq 2\times 10^{-10}$ s) and the corresponding lower bound on T_D ($T_D \geqslant 0.006$ K) calculated using the Drude formula and the measured resistivity at low temperature and zero field.

Equation (1) gives us the observed period with the magnetic field.[1] The signature of the crossing of a Landau level by the Fermi energy in a perfect crystal at zero temperature is a *singularity* in the oscillations and a sharp peak in a real crystal at a high B/T ratio. In the experiment, we measured the resistivity but not the conductivity. However, because bismuth is a perfectly compensated semimetal, the Hall components of the conductivity tensor are small compared to the diagonal ones. Therefore, the relation between the conductivity and resistivity in a strong magnetic field is very simple, namely, $\rho_{11}\approx 1/\sigma_{11}$. Hence the oscillatory corrections are related by $\tilde{\rho}_{11}/\rho_{11}=-\tilde{\sigma}_{11}/\sigma_{11}$. This means that the maxima in $\tilde{\sigma}_{11}$, corresponding to the Fermi energy crossing the center of the Landau level, appear as *minima* in $\tilde{\rho}_{11}$, as is observed experimentally (Fig. 1).

In Eq. (1), we emphasize the fact that the oscillatory behavior is given by the superposition of two different oscillations, one for holes with spin parallel to the field ("spin-up" holes) and the other for holes with spin antiparallel to the field ("spin-down" holes). As has already been pointed out, Eq. (1) shows sharp features when the argument of one of the cosines is not p dependent; in fact, in such a case $\nu \propto \Sigma_{p=1}^{\infty} 1/\sqrt{p}\rightarrow\infty$. This happens when the factor multiplying $2\pi p$ is an integer number. This singularity corresponds to the one-dimensional singularity in the density-of-states for the energy approaching the bottom of the Landau band. Using this observation, one readily finds the spacing between resistance minima for spin-down and spin-up holes, taking the difference between the position of two singularities in Eq. (1) with opposite spin: this is given simply by

$$\epsilon_F\Delta\left(\frac{1}{\hbar\omega_c}\right)=\frac{\epsilon_Z}{\hbar\omega_c}+l, \tag{3}$$

where ϵ_Z is the Zeeman energy and l is an arbitrary integer number. The spacing in the inverse magnetic field is then given by

$$\Delta\left(\frac{1}{B}\right)=\frac{e\hbar}{\epsilon_F}\left(\frac{g_0}{m_0}+l\alpha_1\right). \tag{4}$$

g_0 is the part of the gyromagnetic ratio that contributes only to the *fractional* part of the ratio between the Zeeman energy and the cyclotron energy. The value of g_0 can then be obtained from the fit to the quadratic dependence in Fig. 3: $g_0=2.68(4)$. The error here is determined by the scatter of the data.

At this point, we need to obtain the exact value of l, i.e., the integer part of $\epsilon_Z/\hbar\omega_c$, which will be related to a second contribution to the gyromagnetic ratio g_1: usually, this is done from theoretical considerations on the band structure, but in our case it is straightforward to obtain this value from the observation that *the last minimum* in Fig. 1 is not split. It is helpful to interpret Eq. (1) as the sum of two oscillations with the same period, only *slightly displaced in phase*, due to the contribution of the spin, in such a way that the first peak of the doublet (in an increasing field) corresponds to spin down holes and the second to spin up holes *with different Landau indices*. The last split minimum at $n=2$, corresponds to the crossing of the Fermi surface of the levels ($\nu=0,-$) and ($\nu=2,+$). The spin-up holes are still oscillating: this is the meaning of the unsplit minimum that signs the crossing through the Fermi surface of the level ($\nu=1,+$); therefore we can conclude that the integer part of the ratio of the Zeeman energy to the cyclotron energy is equal to 2, which gives us $g_1=2m_0\alpha_1=32.6(3)$. The precision in this evaluation is primarily due to uncertainties in the effective-mass determination.[1] Because the g factor is anisotropic, there is also an error due to possible misalignment of the magnetic field with the trigonal axis. The accuracy of our alignment is on the order of one degree, which we calculate from the anisotropic mass tensor of the hole ellipsoid to give a relative error in g on the order of 1.6×10^{-3}. This error is significantly smaller than the error determined by scatter.

Summing up the two contributions, we obtain

$$g=g_1+g_0=35.3(4), \tag{5}$$

which coincides with a ratio between the Zeeman energy to the cyclotron energy equal to $\epsilon_Z/\hbar\omega_c=2.16(2)$. The two values of this ratio reported in the literature, i.e., $\epsilon_Z/\hbar\omega_c=2.16$ by Edel'man[2] and $\epsilon_Z/\hbar\omega_c=1.94$ by Smith *et al.*,[9] straddle an integer value of two. Because we do not observe a spin-split minimum at $n=1$ (see Fig. 1 inset), we can state unambiguously that the ratio is greater than two. Accordingly, our finding $\epsilon_Z/\hbar\omega_c=2.16(2)$ precludes values less than two and is in close agreement with Edel'man.[2]

Note that an alternate interpretation, in which spin-split peaks are associated with the same orbital Landau level, would require one to assume that the value of the g factor around the last minimum is much larger than that for higher minima, i.e., that the g factor depends strongly on the field. Such a scenario is very unrealistic, if not impossible.

In agreement with previous investigations,[10] no sign of an electronic contribution to the Shubnikov–de Haas effect has been identified. The electrons are expected to have a period three times shorter than the holes, the difference being due to the nonparabolicity of their energy spectrum. At the same time, the temperature and the Dingle temperature smearing factor should have a bigger numerical factor, in such a way that the electronic oscillation should be depressed by a factor of 3. But the only signature of the presence of the electrons is related to some modulation of the hole oscillation that can be quantitatively interpreted *assuming* a Dingle temperature for electrons almost ten times bigger than for the holes. On the other hand, no theoretical arguments can be put forward to explain the difference in the Dingle temperatures, viz. in the carrier mean free time.

In conclusion, we have shown in this work that the study of magneto-oscillatory phenomena at high B/T can reveal detailed information about electronic structure that is not evident at higher temperatures or lower fields. By high B/T we mean fields high enough and temperatures low enough such that $\hbar\omega_c \gg k_B T$. In addition, the system must be clean enough, as are our bismuth crystals, to assure that the additional condition $\hbar\omega_c \gg k_B T_D$ or, equivalently $\omega_c \gg 1/2\pi\tau$, holds. When both of these conditions are satisfied, higher harmonics in Eq. (1) become manifest and quantitative information on the g factor and spin polarization can be obtained. With increasing field, the spin polarized Landau levels that cross the Fermi energy give rise to increasingly sharp structures that are limited only by the disorder broadened width of each Landau level. At $n=1$, near a field of 9 T, the sharpness is most pronounced (lower left insert of Fig. 1) with a factor of 2 change in resistance for a less than 3% change in field. We have shown that at high B/T, the appearance of these sharp features is a direct consequence of the high harmonic content of the Shubnikov–de Haas oscillations, which together with the occurrence of well-defined spin-split doublets, reveals important details in the electronic structure.

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