York, 1968), p. 669.

[2]D. Magde and H. Mahr, Phys. Rev. B 2, 4098 (1970).

[3]The excitonic molecule may not be a stable entity in all materials. See R. R. Sharma, Phys. Rev. 170, 770 (1968); R. K. Wehner, Solid State Commun. 7, 457 (1969).

[4]A. Mysyrowicz, J. B. Grun, R. Levy, A. Bivas, and S. Nikitine, Phys. Lett. 26A, 615 (1968).

[5]J. B. Grun, S. Nikitine, A. Bivas, and R. Levy, J. Lumin. 1,2, 241 (1970); H. Souma, T. Goto, T. Ohta, and M. Ueta, J. Phys. Soc. Jap. 29, 697 (1970).

[6]S. Nikitine, J. Ringeissen, M. Certier, C. Wecker, S. Lewonczuk, J. C. Merle, and C. Jung, in *Proceedings of the Tenth International Conference on the Physics of Semiconductors, Cambridge, Massachusetts, 1970*, edited by S. P. Keller, J. C. Hansel, and F. Stern, CONF-700801 (U. S. AEC Division of Technical Information, Springfield, Va., 1970), p. 196.

[7]S. Nikitine and J. Ringeissen, Proc. Phys. Soc., London 26, 171 (1965).

[8]K. L. Shaklee and R. F. Leheny, to be published.

[9]A. F. Armington and J. J. O'Connor, J. Cryst. Growth 3, 367 (1968).

[10]M. Certier, C. Wecker, and S. Nikitine, J. Phys. Chem. Solids 30, 2135 (1969).

[11]Such saturation effects have been observed in many materials and are a consequence of the recombination dynamics.

[12]J. R. Haynes, Phys. Rev. Lett. 17, 860 (1966).

[13]R. S. Knox, S. Nikitine, and A. Mysyrowicz, Opt. Commun. 1, 19 (1969).

[14]Knox *et al.* (Ref 13) assumed negligible radiationless decay of excitonic molecules and still calculated results in agreement with experiments in CuCl (Ref. 5).

Observation of Nonextremal Fermi-Surface Orbits in Bulk Bismuth*

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Using giant quantum oscillations in the attenuation of ultrasound, we have observed nonextremal areas of the electron Fermi surface of bismuth. The giant oscillations occur simultaneously with oscillations due to extremal areas, and both the periods and the general shape of these oscillations can be explained by the theory of Gurevich, Skobov, and Firsov.

We have observed *nonextremal* Fermi-surface orbits by means of giant quantum oscillations in the attenuation of ultrasound in bismuth. The possibility of seeing such orbits in metals and semimetals in a magnetic field was first predicted by Gurevich, Skobov, and Firsov[1] in 1961. Considering the problem of the absorption of a single acoustic phonon by an electron which remains on the same Landau level, they found several interesting features of the acoustic attenuation. One of the properties of these "giant quantum oscillations" is that when the phonon wave vector $\vec{q}$ is nearly perpendicular to the magnetic field $\vec{H}$, the orbits of the electrons responsible for acoustic attenuation are displaced from the extremal Fermi-surface orbit by an amount which depends on the angle between $\vec{q}$ and $\vec{H}$. It should thus be possible to study *any* cross section of the Fermi surface by varying the relative orientation of $\vec{q}$ and $\vec{H}$, a property unique to this type of acoustic attenuation. Giant quantum oscillations have been seen in several materials, and many features of the theory have been confirmed,[2] but there have been no unambiguous measurements of nonextremal Fermi-surface areas.[3,4] When $\vec{q}$ is nearly normal to $\vec{H}$, the attenuation versus magnetic-field curves $\alpha(H)$ become complex, but the periods found in previous experiments correspond to extremal areas.[4,5] We report here the first observation of giant quantum oscillations due clearly to nonextremal orbits. They occur simultaneously with oscillations due to extremal orbit electrons in the vicinity of $\vec{q}$ normal to $\vec{H}$. The resulting Fermi-surface areas agree with the theory of Gurevich, Skobov, and Firsov,[1] assuming an ellipsoidal-parabolic model for the electrons.

The sample was a 1-cm cube spark cut from a single-crystal boule grown by the Czochralski method from 99.9999%-pure bismuth. Resistivity ratio measurements made on small ($15\times1\times1$ mm^3) bars cut from the same boule yielded values of $\rho(300°K)/\rho(4.2°K)\approx 230$. A conventional pulse-transmission method was used for the ultrasonic measurements, with gating circuits monitoring the amplitude of the first transmitted pulse. Longitudinal sound waves at 185 MHz were generated by resonant quartz transducers glued to the sample. All measurements were made at a temperature of 1.5°K. The orientation of the sample in the Dewar could be varied by a gear-driven sample holder and rotatable magnet, with an accuracy of about 0.1°. The relative orientation of $\vec{q}$ and $\vec{H}$ was determined by using the tilt effect.[6]

Because of the simultaneous occurrence of oscillations from all four sheets of the Fermi surface, an orientation was chosen which would simplify the data as much as possible. The magnetic field was kept near the binary direction in the binary-trigonal plane, and the sound was propagated along the trigonal axis. In this orientation two of the electron ellipsoids are degenerate and have small cross-sectional areas (about 14 kG), and the third electron and the hole ellipsoids have very large areas (around 200 kG). [Area in $\text{cm}^{-2} = 9.53\times10^{10}\times$(area in kG).] Quantum oscillations due to nonextremal orbits on the degenerate electron sheets should become distinguishable from the extremal ones when H is within about $\pm8°$ of the binary axis, their frequency tending to zero near $\pm1.5°$ (i.e., the plane of the orbits does not intersect the Fermi surface for angles smaller than $\pm1.5°$).

Some of the pulse-height versus magnetic-field traces taken in the range $0° < 90° - \theta < 9°$, where θ is the angle between the magnetic field and the sound wave vector, are shown in Fig. 1. The vertical scale is the same for all traces. The long-period spikes (periodic in $1/H$) are due to the two (nearly) degenerate electron sheets, while the short-period oscillations, visible above 5 kG, are due to the last electron and the hole sheets. We shall not consider the latter here. Since the difference in the areas of the two electron sheets is only observable for $(90°-\theta) > 5°$, we shall speak of them as degenerate in the region of these measurements. The spin splitting can be observed on the two highest-field peaks in this orientation. As θ approaches 90°, the large ("giant") attenuation spikes broaden, and their period becomes longer (i.e., the electron orbits are moving farther away from the belly of the Fermi surface). Smaller amplitude (de Haas–van Alphen–type) oscillations become visible at the magnetic field locations previously occupied by the giant peaks and eventually dominate. At $90°-\theta = 0.2°$, even these have disappeared.

All of the peaks in the experimental data can be accounted for assuming the presence of only two periods. (Recall that we are ignoring the high-frequency oscillations.) These periods are plotted as solid dots in Fig. 2. The solid line is a theoretical curve for nonextremal areas (with no adjustable parameters), assuming an ellipsoidal-parabolic model for the electrons,[7] and the dashed line corresponds to extremal cross sections.

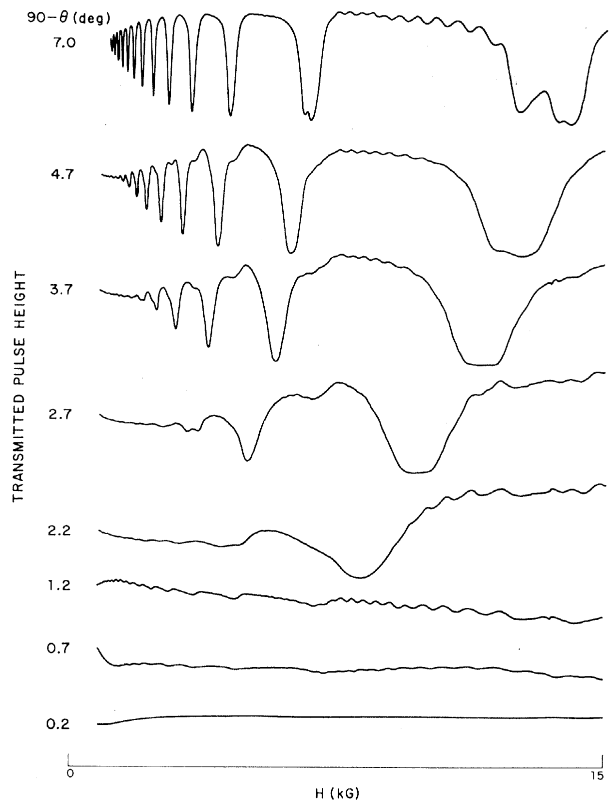

FIG. 1. Experimental transmitted pulse height versus magnetic field. θ is the angle between $\vec{H}$ and $\vec{q}$.

To make sure that the oscillations seen are due to nonextremal orbits, sound was propagated along the binary axis, with H still near the binary. In this configuration only extremal orbits contribute to the attenuation, and there should be little angular dependence of the amplitude in the region of our measurements. Such was the case, with all traces similar to the first one in Fig. 1. The periods measured are plotted as open circles in Fig. 2. The small differences in the extremal areas measured in the two configurations, which are outside experimental error, have not yet been explained.

The fact that long-period oscillations occur only when $\vec{q}$ is nearly normal to $\vec{H}$ confirms our interpretation that they are due to nonextremal cross sections. Nothing else shows such an angular dependence. The areas of the nonextremal orbits in this orientation are virtually independent of small deviations of $\vec{q}$ from the trigonal axis (the deviation here is less than 1°). They depend only on the shape of the Fermi surface and the relative orientation of $\vec{q}$ and $\vec{H}$, which is known to $\pm0.1°$ from the tilt effect. The details of the attenuation theory have no effect on the measured areas either, only on the line shapes and the relative strength of the two oscillations. Thus the areas measured by means of giant quantum oscillations provide unambiguous information about nonextremal properties of the Fermi surface.

To explain the detailed form of the experimental data, we consider the theory of Gurevich *et al.*[1] for $\Delta n = 0$ (n is the Landau-level quantum number), including electron relaxation. Taking H in the z direction, the acoustic attenuation coefficient is then given by

$$\alpha = \mathrm{const}\left(\frac{H}{T}\right)\sum_n \int_{-\infty}^{\infty} dk_z \frac{1}{\tau}\left[\frac{1}{\tau^2} + \left(\frac{\hbar k_z q\cos\theta}{m_z} + \frac{\hbar q^2\cos^2\theta}{2m_z} - \omega\right)^2\right]^{-1} \times \cosh^{-2}\left(\frac{\hbar\omega_c(n+\frac{1}{2}) \pm \frac{1}{2}g\mu_0 H + \hbar^2 k_z^2/2m_z - E_F}{2kT}\right), \tag{1}$$

where T is the temperature, τ is the electron relaxation time (assumed constant and isotropic), $\hbar k_z$ is the z component of electron momentum, m_z is the corresponding effective mass, ω is the sound frequency, and E_F is the Fermi energy. The constant contains various crystal properties, plus the electron-phonon–coupling matrix element.[1]

Nonextremal orbits enter through momentum conservation in Eq. (1). The collision-broadening Lorentzian in the integrand is centered at

$$k_{z0} = m_z\omega/\hbar q\cos\theta - \tfrac{1}{2}q\cos\theta, \tag{2}$$

and for large $\omega\tau$ it becomes a δ function. The electrons contributing to the attenuation are then only those whose orbits lie k_{z0} away from the extremal orbit. To see the magnitude of this orbit displacement, we can rewrite Eq. (2) in the form

$$\langle v_{z0}\rangle = v_s/\cos\theta - \hbar q\cos\theta/2m_z, \tag{3}$$

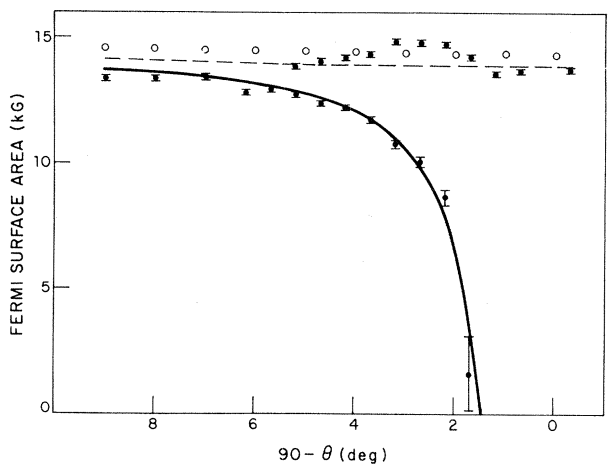

FIG. 2. Fermi-surface areas (in kG). Closed circles are for $\vec{q}$ along the trigonal axis, $\vec{H}$ in binary-trigonal plane; θ is the angle between $\vec{H}$ and $\vec{q}$. Open circles are for $\vec{q}$ along the binary axis, $\vec{H}$ in binary-trigonal plane; $(90°-\theta)$ is the angle between $\vec{H}$ and $\vec{q}$. Solid line is predicted (Ref. 1) nonextremal area, and dashed line is predicted extremal area.

where $\langle v_{z0}\rangle$ is the z component of the electron velocity averaged over the orbit and v_s is the velocity of sound. The last term in Eq. (3) is generally small for any value of θ, and goes to zero when $\theta = 90°$. Since the Fermi velocity is much larger than the sound velocity, $\langle v_{z0}\rangle$ (and hence k_{z0}) is only significantly different from zero when θ is very close to 90°. For intermediate values of $\omega\tau$, the Lorentzian, while still centered at k_{z0}, is broad enough to overlap $k_z = 0$. Because of the singularity in the density of states at $k_z = 0$, peaks in the attenuation then occur both when a Landau level passes k_{z0} (at the Fermi level) and when it passes $k_z = 0$. For $\omega\tau \ll 1$, the Lorentzian is so broad that only the peaks at $k_z = 0$ remain, giving de Haas–van Alphen–type oscillations.

In order to compare the theory with the data, the attenuation was computed numerically and $e^{-\alpha L}$ was plotted, where L is the sample length. Only two adjustable parameters enter in the expression; τ and the matrix element in the constant in Eq. (1). The latter is only necessary because of the nonlinear dependence of pulse height on attenuation. A value of $\omega\tau = 7$ was used, although values between 5 and 10 gave reasonably good agreement with experiment. The spin splitting was taken equal to the Landau-level spacing for convenience, an assumption which is only approximately correct.[8] It has an effect on the phase of the oscillations, but not on their period. Figure 3 gives the resulting pulse height versus H for the same angles as the data in Fig. 1. The motion of the peaks with angle is the same as that described above for the experimental results. In fact, curve-by-curve comparison shows a close resemblance between the two.

The electron relaxation time predicted from the above analysis is about 4-9 nsec (at 1.5°K), consistent with some of the best values reported in bismuth.[9] The resistivity ratio is also char-

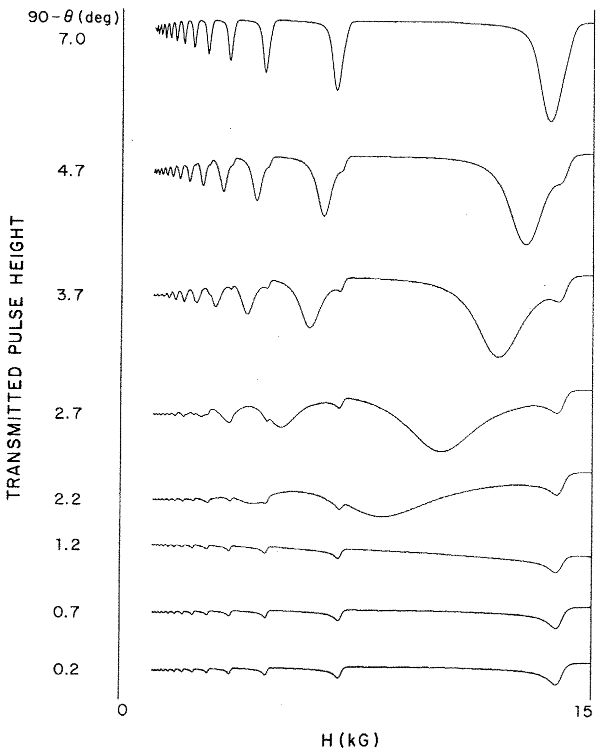

FIG. 3. Theoretical pulse height versus magnetic field for $\omega\tau = 7$.

acteristic of good bismuth.[10] The measured value of 230 is a lower limit on the bulk value since it was measured on small bars where surface scattering can increase the low-temperature resistivity.[11] The value in the sample may be several times as large. The sample seems to be of sufficient quality that further work with giant quantum oscillations may be able to shed some light on the reported anomaly in the hole Fermi surface.[4,12] This work is currently under way.

The only feature of the data that is in significant disagreement with theory is the complete disappearance of even the de Haas-van Alphen-type oscillation when $\vec{q}$ is normal to $\vec{H}$. The presence of these oscillations is dependent upon the assumptions made in deriving the theory, some of which are open to question.[13] Perhaps measurements of this type will prove to be a sensitive test of the theory.

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[1]V. L. Gurevich, V. G. Skobov, and Y. A. Firsov, Zh. Eksp. Teor. Fiz. 40, 786 (1961) [Sov. Phys. JETP 13, 552 (1961)].

[2]For a review of work through 1968, see Y. Shapira, in *Physical Acoustics*, edited by W. P. Mason (Academic, New York, 1968), Vol. V.

[3]J. W. Dooley and N. Tepley, Phys. Rev. 187, 781 (1969), reported some evidence of nonextremal areas at 9.3 GHz in bismuth, but the areas did not agree with theory and could not be explained. Also, the small deviations shown in Fig. 1 of their paper could be due to the effective masses that they used, which are slightly different from more recent measurements (see T. Sakai, Y. Matsumoto, and S. Mase, J. Phys. Soc. Jap. 27, 862 (1969).

[4]M. Giura, R. Marcon, T. Papa, and F. Wanderlingh, Phys. Rev. 179, 645 (1969), observe a sharp tilt effect in bismuth and the disappearance of oscillations for $\vec{q}$ normal to $\vec{H}$, but their data do not indicate the existence of nonextremal areas.

[5]For example, see A. P. Korolyuk, Zh. Eksp. Teor. Fiz. 51, 697 (1966) [Sov. Phys. JETP 24, 461 (1967)].

[6]H. N. Spector, Phys. Rev. 120, 1261 (1960).

[7]Sakai, Matsumoto, and Mase, Ref. 3.

[8]G. E. Smith, G. A. Baraff, and J. M. Rowell, Phys. Rev. 135, A1118 (1964).

[9]R. A. Murphy, private communication; R. Hartman, Phys. Rev. 181, 1070 (1969).

[10]A. M. Forrest and A. C. Hollis Hallett, J. Phys. D: Brit. J. Appl. Phys. 2, 753 (1969).

[11]For example, see A. N. Friedman and S. H. Koenig, IBM J. Res. Develop. 4, 158 (1960).

[12]M. Giura and R. Marcon, Phys. Rev. B 1, 1528 (1970).

[13]F. A. Blum, private communication; J. J. Quinn, Solid State Commun. 3, 25 (1965); J. Mertsching, Solid State Commun. 3, 267 (1965).