INFLUENCE OF THE ELECTRON INTERACTIONS ON ORBITAL QUANTIZATION*

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(Received 3 January 1966)

Since the observation by Shoenberg[1] of an unexpectedly large harmonic content in the de Haas–van Alphen (dHvA) effect in the noble metals and the explanation of the phenomenon by Shoenberg and Pippard[2] in terms of magnetic interaction among the electrons, there has been considerable interest in the nature of the dHvA effect when the magnitude of the oscillatory magnetization becomes comparable to the field spacing of the oscillations. Pippard has shown by a thermodynamic argument that if the field acting on an electron is taken to be B within the metal rather than the applied field H, then the expression for the oscillatory magnetization is obtained by replacing H by B in the relation derived neglecting interactions. For a metal having a Fermi surface with a single extremal cross-sectional area, the magnetization can be written as

$$I = I_0 \sin(2\pi f/B), \qquad (1)$$

where f is the dHvA frequency. We have neglected here the influence of the nonsinusoidal nature of the dHvA effect in the absence of magnetic interaction, which results when the thermal energy is comparable to or less than the spacing of the Landau levels. Such complications do not appreciably affect the discussion to follow.

Using $B = H + (4\pi - N)I$ for the field within a sample of regular shape having a demagnetizing coefficient N in the direction of the applied field, Eq. (1) can be written, for $(4\pi - N)I/H \ll 1$, as

$$y = a \sin(x - y), \qquad (2)$$

where $y = 2\pi f(4\pi - N)I/H^2$, $a = 2\pi f(4\pi - N)I_0/H^2$, and $x = 2\pi f/H$. Since Eq. (2) for y becomes multivalued for $a > 1$, Pippard has predicted on this model that the magnetization would make a step change once during every cycle in the region $x = 2n\pi$ (n is an integer) and thus would appear as a saw-tooth function in the dHvA effect. The magnitude of the step in I is expected to vary from zero at $a = 1$ to $(\Delta H)/4\pi$ at $a = \infty$ where $\Delta H = H^2/f$ is the field separation of the oscillations.

Irrespective of the validity of using B for H as the field acting on the electrons, a simple thermodynamic argument requires that the variation in the magnetization be no larger than $H^2/4\pi f$. If the magnitude of the change in I were to become larger than this value, then for some field region in every oscillation $\partial B/\partial H < 0$, that is, more diamagnetic than a perfect diamagnet.

The basic premise in the treatment of Pippard is the assumption that the effect of electron-electron interactions can be described macroscopically by an effective field, the electron experiencing the field B within the metal rather than H. The interesting effects arise from the interactions among electrons in the same highly degenerate Landau level as it passes through an extremal area of the Fermi surface. Electrons on other parts of the surface interact with an electron having an extremal orbit but presumably the net result of such interactions is a temperature-independent, nonoscillatory Landau diamagnetism, which is of no consequence to this discussion. Also, the large steady internal field in a ferromagnet such as iron[3,4] resulting from localized, unpaired intrinsic spins is a different case than we are considering here of the interaction among electrons in the same orbital state.

Influenced by the observations of Plummer and Gordon[5] and Halloran and Hsu,[6] we have made a study of dHvA magnetization oscillations and magnetothermal oscillations[7,8] in carefully shaped samples of beryllium[9] at very low temperatures to determine if the simple substitution of B for H is sufficient to describe the effect of the electron interactions. Measurements on three different samples will be reported, a cylinder having length-to-diameter ratio of 1.6:1, an ellipsoid of revolution with axial ratio 1.6:1, and a circular disk having a thickness-to-diameter ratio of 1:40.

Beryllium is particularly suited for an investigation of magnetic interactions. Its Fermi surface[10,11] consists in part of electron "cigars" that are slightly pinched in the middle, resulting in two extremal cross sections, "waists" and "hips," for the magnetic field parallel to the [0001] axis of the hexagonal crystal. Since the two extremal areas differ by approximately 3%, the dHvA pattern in the absence of appreciable interaction appears at low fields as a single frequency, amplitude modulated at the difference frequency (f_1-f_2). This is illustrated by measurements of $(\partial I/\partial H)_T$ of the ellipsoid in Fig. 1(a). The curvature of the cigar surface with respect to k_z is very small at the extremals. Thus, the number of electrons having extremal orbits and the interaction among them becomes large at low temperature. The modulation at f_1-f_2 is no longer sinusoidal; the maximum amplitude of $\partial I/\partial H$ saturates [see Fig. 1(b)], in that it is independent of the phase of the two beating oscillations, except very close to the region of a node.

The magnitude of the measured value of $\partial I/\partial H$ is not in agreement with the simple prediction that in the absence of eddy currents I makes a discontinuous step change and $\partial I/\partial H$ becomes infinite. The data illustrated in Fig. 1 were taken under the conditions of sufficiently slow field sweep, and sufficiently low-frequency modulation of small amplitude, that changing the magnitude of any experimental parameter did not affect the measurements. The question then arises as to the reason for the broadening of the step change in I. Electron scattering in the usual sense is not the cause as that has already been included in the calculation of I_0 in the absence of interactions, if important. Two possible explanations come to mind: (1) field inhomogeneity in the metal resulting from imperfect sample geometry, or (2) an intrinsic broadening by the electron interaction. The following discussion is directed to showing that possibility (1) is untenable and that (2) is likely to exist.

A simple extension of Pippard's analysis

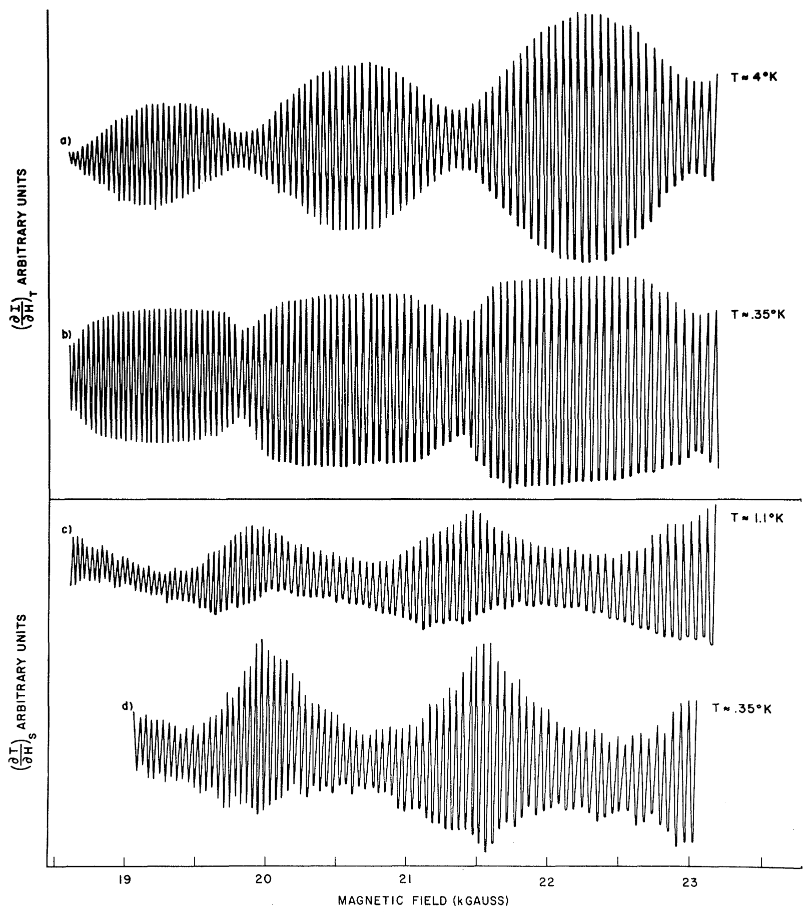

FIG. 1. (a) and (b). Measurement of $(\partial I/\partial H)_T$ vs H for ellipsoid at two temperatures. (c) and (d). Measurement of $(\partial T/\partial H)_S$ for thin disk at two temperatures. These two different measurements are included on the same graph to show the relation of the beat envelopes.

of the consequences of replacing H by B to include the effects of two extremal orbits of almost equal cross section can be obtained by starting from the equation

$$I = I_0 \sin\frac{2\pi f_1}{B} + I_{02} \sin\frac{2\pi f_2}{B}. \quad (3)$$

This relation is more conveniently expressed as

$$y = a\sin(x - y + \delta), \quad (4)$$

where $x = 2\pi(f_1+f_2)/2H$, $y = 2\pi(f_1+f_2)(4\pi-N)I/2H^2$,

$$\delta = \arctan\left[\frac{I_{01}-I_{02}}{I_{01}+I_{02}}\tan\frac{2\pi(f_1-f_2)}{2H}\right],$$

and

$$a \simeq \frac{\pi(f_1+f_2)(4\pi-N)}{H^2} \times \left[I_0^{\,2} + I_{02}^{\,2} + 2I_{01}I_{02}\cos\frac{2\pi(f_1-f_2)}{H}\right]^{1/2}. \quad (5)$$

The condition $(4\pi-N)I \ll H$ has been used.

The coefficient a of Eq. (4) as given by Eq. (5) can be varied in either of two ways: (1) by changing the temperature and hence the magnitude of the magnetization amplitudes I_{01} and I_{02} in the absence of interactions, or (2) by varying the applied field from one region of the beat envelope to another. For the ellipsoid at $H = 20$ kG and $\cos[2\pi(f_1-f_2)/H] = 1$, the value of a was estimated at high temperature from measurements and calculated at low temperatures to range from $a = 0.06$ at $T = 4°$K to $a = 3$ at $T = 2°$K, $a = 6$ at $T = 1.2°$K, and $a = 7$ at $T = 0.35°$K, in reasonable agreement with the value quoted by Plummer and Gordon at 2°K.

Returning to Fig. 1(b) it can be seen that the magnitude $\partial I/\partial H$ at $T = 0.35°$K is independent of the value of the coefficient a as it varies from $a = 7$ at $\cos[2\pi(f_1-f_2)/H] = 1$ to $a = 3$ at $\cos[2\pi(f_1-f_2)/H] = -1/2$. However, the magnitude of $\partial I/\partial H$ measured at $T = 1.2°$K, while still showing strong saturation in the sense discussed above, is almost 40% less than at 0.35°K. Thus, the magnitude of $\partial I/\partial H$ or I does not depend only upon a as the analysis assuming an effective field would lead one to believe. A broadening of the step change in the I-vs-H curve caused by field inhomogeneity within the sample should be only dependent upon the sample shape and the magnitude of a, not on the parameters giving rise to a. It appears that there exists, therefore, a temperature-dependent mechanism within the electron-electron interaction which causes a broadening of the I-vs-H curve.

To observe the dependence of the field B within the ellipsoid of beryllium as a function of applied field, a thin cut was made in the sample and a magnetoresistance probe of bismuth inserted. The field experienced by the probe, which is very nearly B within the material for this configuration, is plotted against H at $T = 0.35°$K in Fig. 2. This plot very clearly shows the deviation of B from the behavior predicted on the basis that the electron interactions are completely describable in terms of an effective field.

The temperature dependence of the magnetization was further studied by performing magnetothermal measurements using a field-derivative technique.[8] The quantity measured, the field dependence of the temperature at constant entropy, is related to the magnetization by the expression

$$(\partial T/\partial H)_S = -(T/C_H)(\partial I/\partial T)_H, \quad (6)$$

where C is the specific heat. Measurements of $(\partial T/\partial H)_S$ on the cylindrical sample are plotted in Fig. 3 at several different temperatures for H close to the hexagonal axis. At $T = 2.9°$K, where there is little magnetic interaction, the

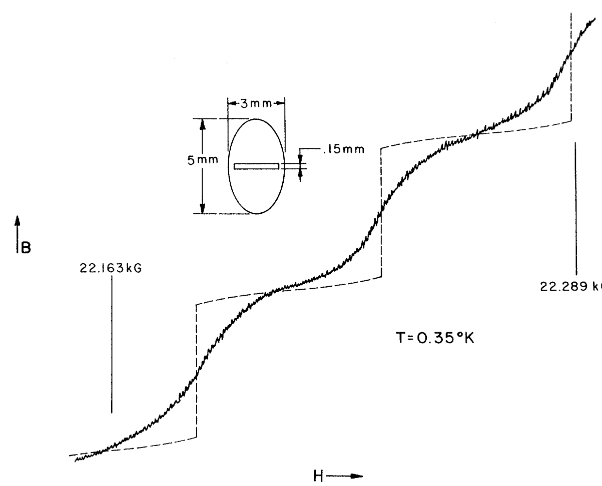

FIG. 2. Measurement of B vs H for ellipsoid at 0.35°K. Dashed curve indicates expected behavior for $a = 7$ of Eq. (4) in text.

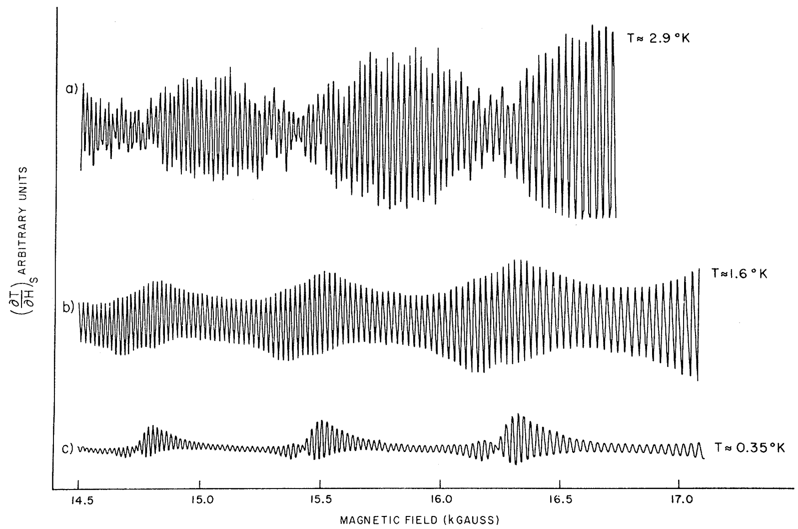

FIG. 3. Plot of $(\partial T/\partial H)_S$ vs H for cylinder at three different temperatures.

envelope is sinusoidal, but at low temperatures it becomes the very odd-looking pattern first observed by Halloran and Hsu.[6] This pattern derives its shape from the fact that I tends to saturate in what would normally be the antinodal region of the beat envelope, i.e., for $\cos[2\pi(f_1-f_2)/H]\simeq 1$; and, consequently, $\partial I/\partial T$ is less in that region than for other values of $\cos[2\pi(f_1-f_2)/H]$. As discussed above, I is still temperature dependent at the lowest temperatures obtained in these experiments, but the dependence is not that which is predicted either in the absence of interactions or by the simple substitution of B for H.

Additional magnetothermal measurements were performed on a very thin disk, 0.0048 in. thick and 0.20 in. in diameter. The hexagonal axis and the magnetic field were both less than 1° from the normal. For this configuration the field B within the metal is approximately equal to the applied external field. Assuming the disk to be an oblate ellipsoid and thus having a demagnetizing coefficient of $N=0.96(4\pi)$, the predicted value of the coefficient a in the antinodal region of the beat envelope at 20 kG and 0.35°K is $a=0.3$. The measured magnetothermal oscillations are shown in Figs. 1(c) and 1(d). The degree of saturation of I as exhibited by the decrease in $\partial I/\partial T$ in the antinodal region at $T=0.35$°K is comparable to that observed in the cylinder at $T=1.2$°K where $a\simeq 6$. The deviation of the disk geometry from that of a regular ellipsoid or the quality of the surface finish are not considered to be likely causes of this discrepancy.

In conclusion, these measurements indicate that the interaction among electrons on a highly degenerate Landau level cannot be described completely in terms of the field $(4\pi-N)I$ within the metal. This would require that the interaction shift all the levels exactly the same amount. The measurements of the shape and the temperature dependence of the B-vs-H curve suggest that there must be a broadening, not due to a spatial field inhomogeneity or normal scattering process, but due to the nature of the interaction itself. In addition, the measurements on a very thin disk indicate that the field experienced by an electron averaged over an orbit is not simply the macroscopic field within the metal. It appears that a microscopic calculation treating problems such as electron-correlation effects must be considered.

A more lengthy paper will treat these measurements in detail, as well as the observation of spin splitting of the hip orbits at high fields, the influence of magnetic interaction on this splitting, the observation of magnetic breakdown between the electron cigar waists and the hole coronet, and the measurement of high-frequency dHvA oscillations associated with the coronet.

We are grateful to J. Quinn for several helpful discussions.

†This work was supported in part by the National Science Foundation and the Advanced Research Projects Agency.

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OBSERVATION OF PHONON ASSISTED INDIRECT TRANSITIONS BY STRESS MODULATED OPTICAL TRANSMISSION

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(Received 10 January 1966)

Recently, the periodic stress modulation of the reflectivity of semiconductors[1,2] and metals[1,3,4] has been shown to provide useful information about the band structure of solids. In principle, if these data are taken over a sufficiently wide energy range, they will yield, after a Kramers-Kronig analysis,[3,4] all of the information that may be obtained concerning the changes in optical constants due to strain. In practice, however, this reflectivity technique is sufficiently sensitive to detect only the changes in the strong direct optical transitions. The relatively weak phonon-assisted transitions at the indirect edge of Ge, for example, are not observed.[1] This Letter reports the results of an experiment where the stress modulation of the optical properties is measured by means of the transmitted beam. The results at room temperature clearly show the phonon and exciton structure associated with the indirect edge of Ge. The allowed transitions are clearly distinguished from the forbidden ones, and the indirect band gap, the dilatation and pure shear deformation potentials, and the phonon energies at L can all be directly observed.[5] This experimental method should be generally useful for studying the band structure and phonon dispersion curves of less well-known indirect semiconductors.

Figure 1 shows a schematic diagram of the apparatus used for the experiment. A polarized monochromatic beam, modulated at f_1, is sent through the sample onto a detector. The sample, which is the central leg of a three-post yoke, undergoes a periodic strain at the frequency f_2. The sample is driven by two matched lead-zirconate-titanate-type transducers which comprise the other two legs of the yoke. The signal received at the detector is analyzed for both frequencies f_2 and f_1 by means of two phase-sensitive lock-in amplifiers. A voltage proportional to the amplitude of the signal at f_1, $A(f_1)$, is fed back to a servo controlling the monochromator which maintains a constant transmitted intensity. To first order the ratio of the am-

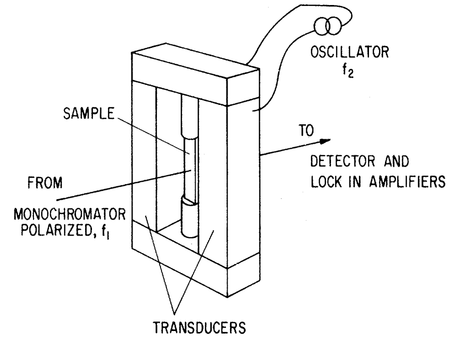

FIG. 1. Schematic view of experimental apparatus.