Cyclotron Resonance in Aluminum*

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Preliminary results of cyclotron resonance experiments at 35 kMc/sec in aluminum having a relatively long relaxation time at 4.2°K ($\omega\tau\sim20$) are presented. Discrepancies and errors in earlier cyclotron resonance experiments are resolved. A tentative orbit identification scheme is proposed which indicates (a) that the second-zone Fermi surface is consistent with the single orthogonalized plane wave (OPW) model, and (b) that the third-zone Fermi surface is connected and that the connecting region is thicker than in the single OPW model. The effective masses obtained are compared with the values predicted by the single OPW model and with other experimental values, where pertinent. Departures of the effective mass value of orbits in the second zone from values given by the single OPW model are noted and may be attributable to electron-electron or electron-phonon effects.

INTRODUCTION

PRELIMINARY reports of cyclotron resonance experiments in aluminum have been made by Langenberg and Moore[1] and by Fawcett.[2,3] We present at this time the results of more extensive, but still incomplete experiments which resolve discrepancies in the earlier work. We assume reasonable familarity with the Azbel'-Kaner effect,[4] and shall use the concepts and nomenclature developed by Kip *et al.*[5]

Recent theoretical and experimental progress in Fermi surface physics[6] make it possible to compare experimental results with fairly accurate theoretical calculations made within the single-particle scheme. For data interpretation we shall rely heavily upon the Fermi surface model due to Harrison.[7] This model is based upon the "nearly free electron" or "single orthogonalized plane wave" (OPW) approximation corrected by the addition of up to four OPW's near points of high symmetry and normalized to earlier energy-band calculations by Heine.[8] The single OPW model is shown in Fig. 1. The low-field de Haas–van Alphen data of Gunnersen[9] has been fitted to this model by Harrison without unreasonable modification. The region connecting the surfaces in the third zone depends very sensitively on the value of the Fermi energy and upon the lattice potential; the "arms" are pinched off for the Fermi energy which gives the best fit to the de Haas–van Alphen data. Previous experimental evidence is inconclusive regarding the connectivity of the third-zone surface.

EXPERIMENTAL

The present experiments were performed in almost exactly the same manner as those in copper described by Kip *et al.*[5] except at a frequency of 35 kMc/sec. The aluminum sample had a residual resistivity ratio of 10 000. A chemically cut and electropolished (110) surface was lightly clamped to a choke joint to form the bottom wall of a TE_{111} cylindrical cavity. A pin placed near the cylindrical wall of the cavity was used to remove the degeneracy of the microwave modes in the cavity and to allow the use of either of two alternate modes, giving rf currents in either of two perpendicular directions. All data were taken at 4.2°K.

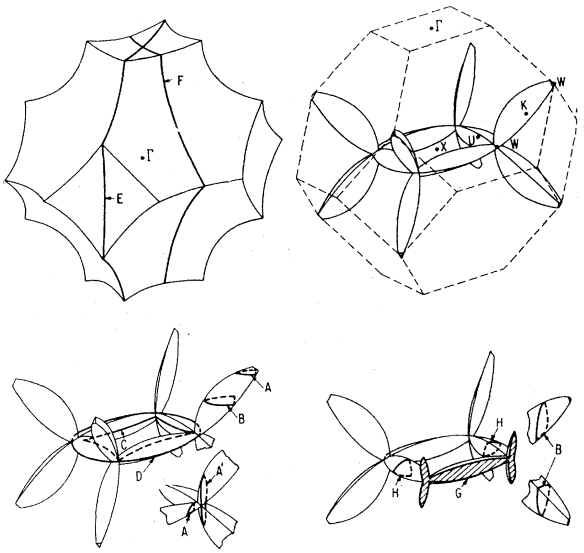

FIG. 1. The single OPW model of the Fermi surface of aluminum. The second-zone region of holes is shown in the upper left panel with the center of the zone at Γ. Orbits for H parallel to a ⟨100⟩ axis are labeled E and to a ⟨110⟩ axis are labeled F. The third-zone region of electrons is shown in the upper right panel, with the center of the zone at X. Proposed orbits in the third zone are shown for H parallel to a ⟨100⟩ axis (lower left) and to a ⟨110⟩ axis (lower right).

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[1] D. N. Langenberg and T. W. Moore, Phys. Rev. Letters **3**, 137 (1959).
[2] E. Fawcett, Phys. Rev. Letters **3**, 139 (1959).
[3] E. Fawcett, in *Proceedings of the International Conference on the Fermi Surface in Metals at Cooperstown, New York, on August 22–24, 1960* (John Wiley & Sons, Inc., New York, 1960).
[4] M. Ya Azbel' and E. A. Kaner, J. Phys. Chem. Solids **6**, 113 (1958).
[5] A. F. Kip, D. N. Langenberg, and T. W. Moore, Phys. Rev. **124**, 359 (1961).
[6] An excellent review of recent progress in Fermi surface physics may be found in reference 3.
[7] W. A. Harrison, Phys. Rev. **116**, 555 (1959); **118**, 1182 (1960); **118**, 1190 (1960).
[8] V. Heine, Proc. Roy. Soc. (London) **A240**, 3611 (1957).
[9] E. M. Gunnersen, Phil. Trans. Roy. Soc. (London) **A249**, 299 (1957).

EXPERIMENTAL RESULTS

Data were taken for all orientations of the magnetic field parallel to a (110) surface. We adopt the notation that θ is the angle between the magnetic field direction and a $\langle 100 \rangle$ axis in the (110) plane. The rf current was either $7\frac{1}{2}$ degrees from a $\langle 110 \rangle$ or $\langle 100 \rangle$ axis, as indicated, so that the rf polarization angle, that is the angle between the rf current and the magnetic field, varied as the magnetic field direction was rotated in the sample surface.

For purposes of presentation and discussion it is convenient to divide the data into two classes, one a low-field region having low cyclotron masses ($m^*/m_e \approx 0.2$) and the other a high-field region having high masses ($m^*/m_e \approx 1$). Figure 2 shows typical recorder tracings in each region.

The anisotropy of the masses observed in the low-field region is shown in Fig. 3. The orbits involved here are believed to arise from electrons in the third zone. The single-valued branch A was observed only for the rf current $7\frac{1}{2}$ degrees from the $\langle 110 \rangle$ axis, while the double-valued branch B was observed with both polarization modes. The data were symmetrical about the cubic axes within experimental error and scatter, which is about 1%. No splitting of branch B was observed near the $\langle 100 \rangle$ direction. This is surprising, since this branch appears to intersect this twofold symmetry axis at an acute angle, and a second mass is therefore expected. However, the data in this region are complicated by the presence of the A resonance and high-order harmonics of the high masses, so that a splitting could have been obscured. (See the low-field trace in Fig. 2.)

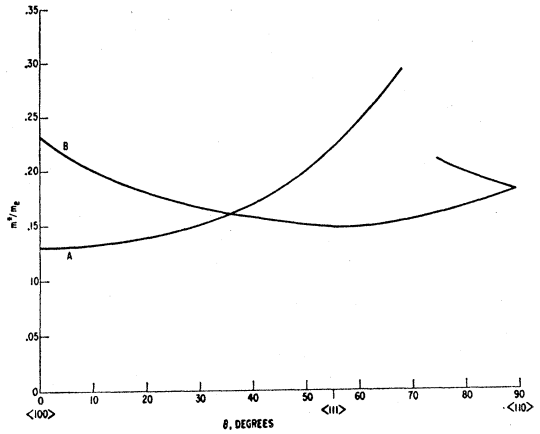

FIG. 3. Observed effective mass anisotropy in the low-field region.

The mass anisotropy in the high-field region is shown in Fig. 4. The effective cyclotron mass of central, stationary ($k_H=0$) orbits in the second zone according to the single OPW model is shown by the dashed light line. Branches E and F are thought to arise from the central (hole) orbits in the second zone. Branches C, D, and G, which are believed to arise from third zone electrons, were observed only with the rf current $7\frac{1}{2}°$ from a $\langle 110 \rangle$ axis, branch F only with the current $7\frac{1}{2}°$

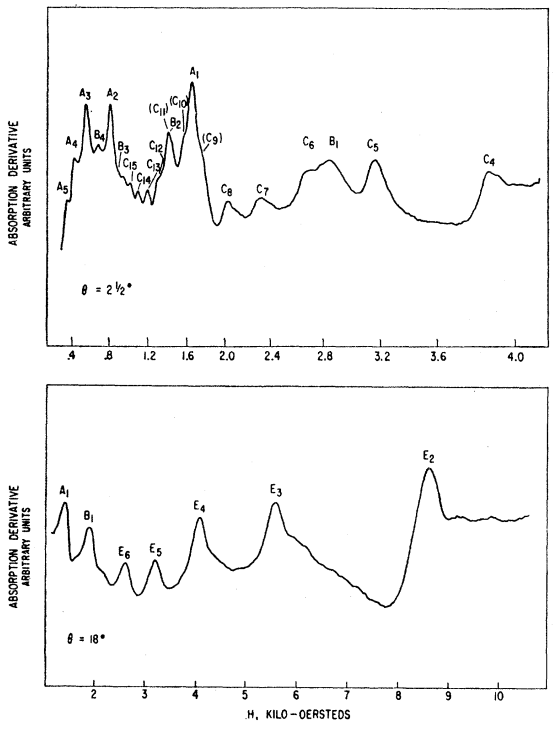

FIG. 2. Typical recorder tracings in the low-field region (upper panel) and high-field region (lower panel). The magnetic field was essentially perpendicular to the rf currents in both traces. The harmonic order numbers of various resonance series as described in the text are indicated.

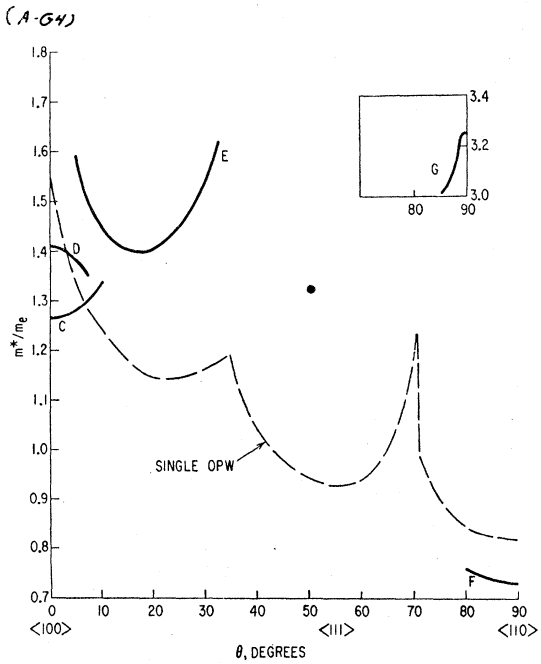

FIG. 4. Observed effective mass anisotropy in the high-field region. The light dashed line gives the mass values of central, stationary ($k_H=0$) orbits in the second zone according to the single OPW model.

from a ⟨100⟩ axis, and branch E was observed with both modes of rf polarization.

Data taken in the region between $\theta=33°$ and $\theta=80°$ indicated at least weak resonances in one or the other modes of rf polarization; effective mass values between 1.3 and 1.8 were found. The mass values and signal strengths were very anisotropic and the data rather fragmentary, so no attempt has been made to indicate the mass anisotropy in this preliminary report. Strong resonances having an effective mass of 1.32 were observed at $\theta=51°$ with the rf current $7\frac{1}{2}°$ from a ⟨100⟩ axis. In this region the rf current direction in both modes is far from the magnetic field direction, and further experiments with better polarization control are needed.

No large "phase shifts" such as those observed by Kip *et al.*[5] in copper have been observed in aluminum.

The relatively high relaxation time ($\omega\tau\approx 20$) and excellent quality of the present data make it possible to resolve discrepancies in earlier cyclotron resonance results in aluminum reported by Langenberg and Moore[1] and by Fawcett.[3] Langenberg and Moore reported an isotropic effective mass of $1.5\pm 10\%$ in a (110) plane, while Fawcett observed an isotropic mass of 0.87 ± 0.05 in both a (110) and (100) plane. The relaxation time of the samples used in both of these earlier investigations was rather low; the $\omega\tau$ of the sample used by Langenberg and Moore was probably less than their estimated value of 10. The present data for the magnetic field along the ⟨110⟩ axis show the fourth through fourteenth harmonics of a mass of 3.24 with the rf current nearly parallel to the field direction. With the rf current nearly perpendicular to the field direction, the fundamental and three harmonics of a mass of 0.73 are observed. We believe the second harmonic of the high mass together with the low-mass resonance peaks were misinterpreted by Langenberg and Moore as a resonance series having effective mass 1.5. We do not find the high degree of isotropy indicated by either Langenberg and Moore or Fawcett.

INTERPRETATION

Fairly satisfactory orbit identification can be made on the basis of the single OPW model although the modifications to the third-zone structure required to fit the present cyclotron resonance data are perhaps inconsistent with Gunnersen's low-field de Haas–van Alphen data as reinterpreted by Harrison. We present a tentative identification scheme below, and have indicated the proposed orbits on Fig. 1.

We interpret branches E and F and the point at $\theta=51°$ to be due to stationary ($k_H=0$) orbits on the large second-zone surface of holes. The absence of a resonance signal in the second zone with H along the ⟨100⟩ axis is expected, and need not imply contact with the Brillouin zone boundary at the corners W; the central orbit for this orientation coincides with the contact line between hexagonal faces of the surface and the range of k_H for which the drift velocity along the magnetic field direction is zero is vanishingly small. Priestley[10] also failed to observe an orbit in this orientation in high-field de Haas–van Alphen experiments. The anisotropy is consistent with this interpretation. Notice that the experimentally determined mass values depart significantly from predicted values. This departure from the calculated single OPW values is probably not due to the introduction of the lattice potential, since in the second zone most of the Fermi surface is far from zone boundaries. We believe it much more likely that the departure results primarily from electron-electron or electron-phonon effects. This is probably the first case for which cyclotron resonance data have been obtained in a situation where convergent, self-consistent single-particle band calculations are available. Such comparisons between calculations and experiment may prove to be an important method of investigating the importance of electron-electron and electron-phonon effects. The change in the ratio of experimental masses to predicted masses from approximately 1.2 near the ⟨100⟩ to approximately 0.9 near the ⟨110⟩ direction is interesting but puzzling.

The resonances C and D are thought to be from orbits around the inner and outer surfaces of four contacting arms in the third zone. This is the strongest experimental evidence to date for this contact. The existence of these orbits out to $\theta=10°$ indicates that the contact areas are considerably thicker than the single OPW prediction of $\theta=3°$, at least in the ⟨100⟩ direction normal to an arm axis. This conclusion is difficult to reconcile with Harrison's identification of the lowest frequency de Haas–van Alphen signals as arising from orbits with minimal cross-sectional area at the contacts. The area indicated by these de Haas–van Alphen oscillations coincides with the single OPW prediction; however, this agreement must be regarded as fortuitous since this region of the Fermi surface (near the symmetry point W) can be significantly influenced by the introduction of the lattice potential and is very sensitive to the exact value of the Fermi energy, which is also affected by the lattice potential.

Branch B orbits encircle the middle part of eight separate arms, the plane of an orbit passing through the symmetry point K; these orbits are stationary. With the magnetic field aligned along either the ⟨100⟩ or ⟨110⟩ axes, all eight orbits are equivalent; for intermediate field orientations in a (110) plane these orbits split into two nonequivalent sets of four orbits each. Hence the mass splitting observed at the ⟨110⟩ axis. No splitting was observed at the ⟨100⟩ axis, as discussed above. However, such a splitting must occur if $dm^*/d\theta\neq 0$ at the ⟨100⟩ axis. The mass minimum at the ⟨111⟩ axis, where the angle between the magnetic field and four of the arm axes [⟨110⟩ axes] is a minimum, is consistent with this interpretation.

[10] M. G. Priestley, *Low-Temperature Physics*, edited by G. M. Graham and A. C. Hollis Hallett (University of Toronto Press, Toronto, 1961).

It is tempting to assign the A branch to orbits having minimal cross sections in the contact regions of the eight arms mentioned above; there would then be sixteen such orbits, one at both ends of each arm. With the magnetic field along a ⟨100⟩ axis all sixteen orbits are equivalent. As the magnetic field is rotated toward the ⟨110⟩ axis in the (110) plane, this resonance should split in the same manner as the B branch, and signals should persist as long as there is a minimal ($v_D=0$) cross section in the contact region. The single mass in the region near the ⟨100⟩ axis of branch A is not consistent with this orbit identification; however, a small splitting could be unresolved or overlooked in the complicated region around the ⟨100⟩ direction. Another possible identification for these resonances is an orbit around the contact region of two arms, labeled A' in Fig. 1. This assignment has reflection symmetry about a ⟨100⟩ axis but it is not clear that such a minimum-cross-section orbit exists over the observed range of θ.

The G branch could arise from "butterfly" orbits around the long dimension of one arm and the contact region and two neighboring arms at each end of the orbit. The thickening of the contact regions inferred from the C and D orbits would allow this orbit to traverse a large section of the four adjacent arms so that the high mass might not be unreasonable. Alternatively, this resonance might arise from extended orbits made possible by "magnetic breakdown"[11] between the second and third zone surfaces in regions where the band gaps are very small. Recent theoretical work by Blount[12] has indicated that the criteria for magnetic breakdown is not $\hbar\omega_c \gtrsim E_g$ but rather $\hbar\omega_c \gtrsim E_g^2/E_F$, where E_g is the gap energy. Harrison[7] has found that there are degeneracies (lines of contact) near the corners W which would be lifted by spin-orbit coupling; the resulting energy gaps would be of the order of 10^{-3} ev and would tend to disappear in fields as small as 10 gauss.

One disturbing feature of our whole identification scheme in the third zone is the absence of any signal from stationary orbits around the middle of the arms with the magnetic field in a ⟨110⟩ direction (labeled H in Fig. 1). These orbits would be expected to give strong signals over a large range of θ about a ⟨110⟩ axis. It might be possible that this class of orbits has a large range of k_H over which the drift velocity along the magnetic field is small and that mass spread destroys any resonance.

We do not attach great importance to comparisons of the experimental mass values assigned to the third zone with the single OPW model masses, since the effect of the lattice potential and the modifications to the model necessary to fit the cyclotron resonance data make such a comparison seem unrealistic.

[11] M. H. Cohen and L. M. Falicov, Bull. Am. Phys. Soc. **6**, 145 (1961).
[12] E. I. Blount (private communication from M. H. Cohen).

TABLE I. Identification of orbits.

θ	m^*/m_e	Orbit identification	m^*/m_e Other sources	m^*/m_e Single OPW
0° ⟨100⟩	1.27	inner ring, third zone		0.40
	1.41	outer ring, third zone		1.00
	0.23	maximum cross section, third zone	0.15[a]	
	0.13	minimum cross section, third zone	0.10[b]	
	···	central orbit, second zone		1.54
5°	1.59	central orbit, second zone		1.33
51°	1.32	central orbit, second zone		0.94
54.7° ⟨111⟩	···	central orbit, second zone	1.3[c]	0.93
90° ⟨110⟩	0.73	central orbit, second zone		0.82
	0.18	maximum cross section, third zone	0.15[d]	
	3.17	"butterfly," third zone		

[a] Low-field de Haas–van Alphen data (reference 9); high-frequency oscillation.
[b] Low-field de Haas–van Alphen data (reference 9); low-frequency oscillation, 0.8° from ⟨100⟩ in (100) plane.
[c] High-field de Haas–van Alphen data (reference 10).
[d] Low-field de Haas–van Alphen data (reference 9); high-frequency oscillation, 2.8° from ⟨110⟩ in (100) plane.

CONCLUSION

We summarize in Table I the mass values and orbit identification for the magnetic field parallel to the principal crystal axes and for a few other orientations, together with the mass values from the single OPW model and other experimental sources, where pertinent.

Our proposed orbit identification scheme cannot at this point be regarded as the only scheme which will fit a modified single OPW model, nor can it be taken as conclusive evidence that this model correctly represents even the general topology of the Fermi surfaces of aluminum. It is, however, strong corroboration for the model and indicates that the third-zone arms are connected, with the contact regions being considerably thicker than in the unmodified single OPW surface.

A complete report of this work will be presented at a later date after experiments with more flexible rf polarization and perhaps with different surfaces have been performed.

ACKNOWLEDGMENTS

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