de Haas–van Alphen Study of Coherent Magnetic Breakdown in Magnesium

J. W. Eddy, Jr.,[a] and R. W. Stark
Department of Physics and Arizona Research Laboratories, The University of Arizona, Tucson, Arizona 85721
(Received 20 October 1981)

Studies of the de Haas–van Alphen spectra arising from the two-dimensional lattice of magnetic-breakdown–coupled orbits in magnesium contain spectral features which disagree qualitatively with the accepted theory of Falicov and Stachowiak. Their theory, which was thought to be equivalent to Pippard's theory for the magnetic band structure, is shown to be inequivalent.

PACS numbers: 71.25.Hc, 71.20.+c

Priestley's de Haas–van Alphen (dHvA) study of magnesium[1] resulted in the discovery of a high-frequency spectral component that was attributed to an equivalent semiclassical free-electron-like orbit. This "giant orbit," which circumnavigates the Brillouin zone when the magnetic field $\vec{H}$ is applied along the [0001] axis, led in turn to the discovery of magnetic breakdown[2] (MB). In a classic paper, Pippard[3] showed that this MB mechanism for quantum mechanically coherent interband coupling would lead to a two-dimensional lattice of coupled Landau orbitals. This coupled-orbit network would be characterized by a magnetic band structure (MBS) consisting of broad electron quantum-state energy bands instead of the usual highly degenerate Landau harmonic-oscillator energy-level structure.

Falicov and Stachowiak[4] (FS) then developed a path integral theory for the dHvA spectrum of the coupled-orbit network which contained a one-to-one correspondence between the individual spectral components and the specific semiclassical closed paths that could be traced on the network. This theory resurrected the historic and intuitive correlation between the dHvA spectrum and the semiclassical trajectories followed in $\vec{H}$ by electron wave packets at the Fermi energy (E_F). FS demonstrated the equivalence of their theory to Pippard's by numerically calculating, for comparison, a single Fourier component of the MBS density of states. As a result, it has been assumed for more than a decade that dHvA studies of coupled-orbit systems would yield no intrinsically new data about MBS.

On the experimental side, a number of experiments have yielded data which appear to agree qualitatively and semiquantitatively with the FS predictions.[5] One essential problem that arises in all such experiments results from a restriction imposed by the intrinsic electron quantum-state lifetime, τ. The realization of a MBS requires quantum coherence extending over many cells of the two-dimensional lattice of coupled orbits. This condition is usually expressed by $\omega_G\tau \gg 1$, where ω_G is the cyclotron frequency of the free-electron-like giant orbit. Although single crystals of extremely high purity have long been available, experimental samples have invariably contained enough dislocations to preclude this condition.[6] Electron quantum-state interference[7] studies carried on in this laboratory over the last several years have been partially focused on achieving magnesium single-crystal samples having much lower densities of dislocations than previously attained. As a result, our recent magnetoresistance studies have shown clear evidence of the fully coherent magnetic breakdown that characterizes this new MBS regime.[8] We used these same crystal-handling techniques to prepare a magnesium single crystal for the dHvA studies reported below.

Our dHvA studies were carried out at temperatures as low as 0.3 K, in magnetic fields up to 52 kG and oriented within 1° of the [0001] axis. Data were taken with the aid of a microcomputer-based data-acquisition system. The resultant data were processed by fast Fourier-transform analysis and represented graphically as plots of spectral intensity as a function of dHvA frequency expressed in atomic units (a.u.) of equivalent enclosed $\vec{k}$-space area.

Typical plots of spectral intensity are shown in Figs. 1(a) and 1(b). The location of the spectral peak corresponding to the semiclassical giant orbit is labeled by G; $2G$ locates the spectral component associated with a semiclassical path that makes two sequential traverses around the giant orbit. Figure 1(a) is the transform of dHvA data for 50.11 kG $\leq H \leq$ 50.25 kG; Fig. 1(b) is the transform of data for 50.25 kG $\leq H \leq$ 50.39 kG. These correspond to sequential data traces each of which contains approximately 28 dHvA oscillations for G and 56 oscillations for $2G$. The background noise content in these spectra is negligi-

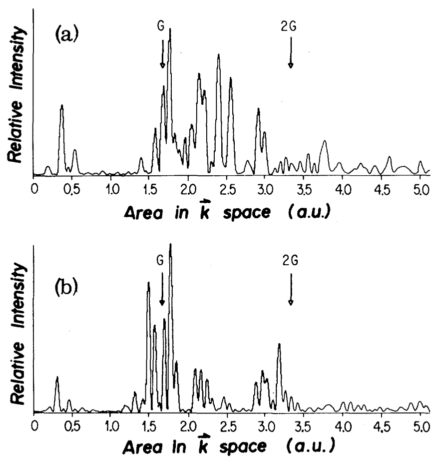

FIG. 1. (a) Fourier transform of dHvA data for 50.11 kG $\leq H \leq$ 50.25 kG. (b) Fourier transform of dHvA data for 50.25 kG $\leq H \leq$ 50.39 kG. The horizontal and vertical scales are the same for both transforms. Note the dramatic change in appearance resulting from $\frac{1}{2}$% change in H.

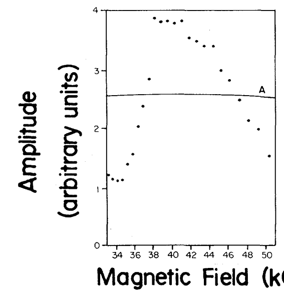

FIG. 2. Comparison of the FS theory with experimental data for the A spectral peak.

ble; additional retraces and transforms of new data exactly reproduced the transforms shown.

On the basis of all dHvA data taken to date, these two transforms would have been expected to be nearly identical. For this small a change in the magnetic field, the field-dependent amplitude changes for each spectral component should have been quite small. The dHvA effect is assumed to be a linear superposition of independent sinusoidal oscillations containing a one-to-one correspondence between each spectral peak and an equivalent semiclassical trajectory.[9] The spectral intensity shown in Fig. 1(a) is clearly different from that shown in Fig. 1(b). It is as if the dHvA spectrum for this regime of coupled orbits were *aperiodic*.

The data shown in Fig. 1 are representative of data taken for any field range in this experiment. The spectrum is not stable. Data samples containing more oscillations (resulting in greater spectral resolution) exhibited greater stability; that is, the spectrum appears more aperiodic when analyzed on an oscillation-by-oscillation basis than when analyzed by averaging over several hundred oscillations. Data samples containing enough oscillations to guarantee spectral resolution for all of the dHvA spectral peaks predicted by the FS theory show field-dependent changes in spectral intensity for these predicted peaks that disagree qualitatively with the intensity predictions of the FS theory.

An example of this is shown in Fig. 2. In the notation of FS, the A spectral peak corresponds to a convex lens-shaped semiclassical trajectory that requires four MB tunneling events each of which reduces the probability amplitude by the factor p, and two Bragg reflection events each of which reduces the probability amplitude by the factor q. These factors are related by $p^2+q^2=1$ with $p^2=\exp(-H_0/H)$; for this band gap in magnesium $H_0=5.8$ kG.[5a] Thus the probability amplitude for this orbit is reduced by a combined MB factor of p^4q^2. The data points shown in Fig. 2 are spectral amplitudes for the A peak taken from transforms of sequential data traces spanning the range 33.21 kG $\leq H \leq$ 50.42 kG. For comparison, the nearly straight line, drawn approximately horizontally across the graph, shows the relative amplitude variation for this field range as predicted by the FS theory. The form of this curve[4] is

$$A(H) \propto p^4 q^2 H^{1/2} \frac{X}{\sinh X}, \tag{1}$$

with

$$X = 2\pi^2 \frac{kT}{\hbar\omega_A}, \tag{2}$$

where ω_A is the H-dependent cyclotron frequency appropriate for the A orbit and $T=0.3$ K is the temperature at which the data were taken. The experimental data clearly do not agree with the predictions of the FS theory.

In addition, the spectral transforms, from which the data shown in Fig. 2 were taken, show spectral spikes at frequencies that disagree qualitatively with the predictions of the FS theory. These are partially identifiable as interference frequencies associated not with single closed semiclassical trajectories but with the interference of two closed semiclassical trajectories. For example, a strong peak is clearly evident at a frequency corresponding to 1.255 a.u., the cross-sectional area of the Brillouin zone. Although this has no semiclassical equivalent orbit, several combinations of two different semiclassical trajectories on the coupled-orbit network will have this as an interference difference frequency.[10]

In summary, for this very pure, carefully handled, magnesium single-crystal sample, the dHvA spectrum for the network of coupled orbits exhibits behavior that is characterized by (a) locally aperiodic structure, (b) amplitude variation with H for predicted spectral terms that disagrees qualitatively with the accepted FS theory, and (c) well-defined spectral spikes at unpredicted frequencies which seem to show close correlation with difference or interference frequencies between two predicted semiclassical orbitals.

This behavior was unexpected. Although we were confident that our crystal-handling techniques would produce a specimen having dislocation densities low enough to satisfy the criterion $\omega_G\tau \gg 1$ required to study the MBS regime, we nonetheless expected the dHvA effect for that regime to be in accord with the FS theory. The observed differences, together with the fact that dHvA data taken in the past on less perfect crystals seemed to agree with the FS predictions, suggests that this theory is a limiting-case theory applicable in a regime of quantum-state lifetimes too short to produce the long-range coherence required to achieve MBS.

Pippard's theory, on the other hand, should be applicable at the other lifetime extreme, $\tau \to \infty$. In order to recheck the equivalence of these two theories, we calculated the MBS density of states for the specific case appropriate for magnesium that is discussed in detail by Pippard.[3] For this case, the band structure is periodic with an energy period of $216\hbar\omega_G$. This calculation was carried out using techniques similar to those discussed in Appendix C of the FS paper.[4]

In their check for equivalence of the two theories, FS calculated the q dependence for only the single Fourier component of the MBS density of states corresponding to an energy period $\hbar\omega_G$. In contrast, we used the fast Fourier-transform algorithm, which had not been invented at the time of their earlier calculation, to obtain the complete energy spectral transform for the density of states. Portions of the spectral transforms calculated for $q = 0.75$ and 0.35 are shown labeled MBS in Fig. 3. For comparison, the spectral transforms directly predicted by FS are also shown for these two values of q.

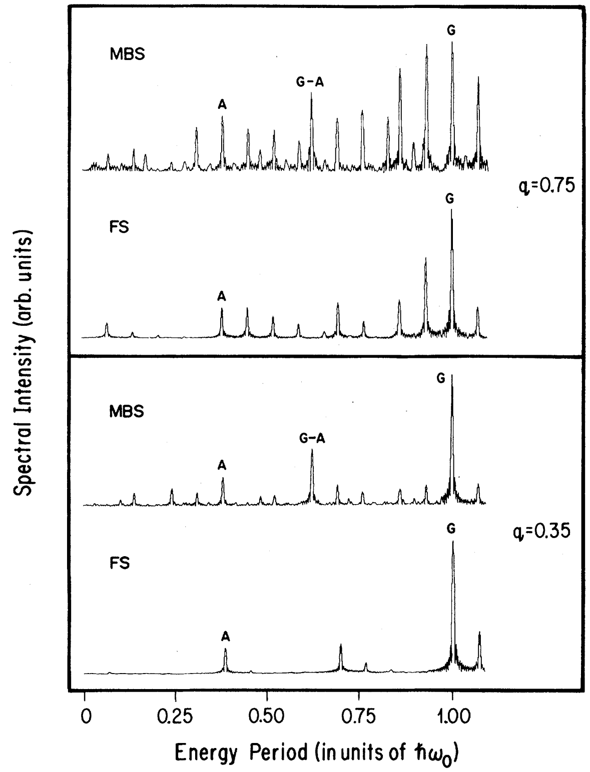

FIG. 3. Fourier transforms of the MBS for $q = 0.75$ and 0.35 compared with the Fourier transforms predicted by the FS theory. All of these transforms have been arbitrarily scaled to exhibit the same intensity for the G spectral spike. Note the presence of additional spectral spikes in the MBS transforms.

It is evident that these two theories are not equivalent. First, the relative intensities for those spectral components predicted by the FS theory do not agree with the equivalent MBS relative intensities. Second, the MBS transform contains additional spectral spikes not predicted by FS. All of these are clearly identifiable as interference difference periods; for example, the spike labeled G-A in Fig. 3 has an energy period equal to $\hbar(\omega_G - \omega_A)$.

Figure 4 shows the detailed q dependence for the spectral component having period $\hbar\omega_G$. The solid dots correspond to our calculated Fourier-transform values; the smooth curve, which has been arbitrarily normalized to obtain best agreement, shows the FS theory predictions. As expected, the two theories cannot be made to agree quantitatively as a function of q for all q. Our calculated values were not insensitive to calculational accuracy. If the MBS is represented numerically by a 9182-point histogram over the

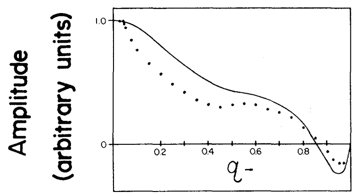

FIG. 4. The q dependence of the amplitude of the spectral spike labeled G in Fig. 3. The dots are MBS transform values; the smooth curve is the best fit prediction of the FS theory.

range $216\hbar\omega_G$, the calculated results agree well with those shown in Fig. 4 which were calculated using a 4096-point histogram. On the other hand, when only 2048 points were used, the calculated values seemed to agree much more closely with the FS theory. We conclude from this that in their earlier calculations, FS obtained fortuitous agreement as a result of an unconvergent calculation.

Pippard's MBS theory is strictly applicable only to the subset of H values that produce commensurable coupled orbit and ionic lattices. It is not clear exactly how one can extend his theory to arbitrary values of H in order to obtain new predictions for the dHvA spectrum to replace the FS predictions. However, it is evident that the spectral difference terms such as G-A that are inherent in the Fourier transform of the MBS density of states shown in Fig. 3 imply a significantly more complicated dHvA spectrum than previously believed. In particular, these interference difference terms in the energy spectrum imply interference difference terms in the H-dependent dHvA spectrum similar to those reported above. Also, Pippard's MBS solution is restricted to values of H that yield, for the lattice of coupled orbits, translation vectors that are integer multiples of the translation vectors for the atomic lattice. The MBS is expected to become more complex for other values of H and the dHvA effect may be even more complicated.

In conclusion, it is now evident that the FS theory provides an incomplete description for Pippard's MBS density of states. In contrast with our former expectations the dHvA effect will indeed be a most useful tool for studying this new regime of long-range quantum coherence.

This work was supported in part by National Science Foundation Grant No. DMR-7910533.

[(a)]Current address: Bell Telephone Laboratories, Murray Hill, N. J. 07974.

[1]M. G. Priestley, Proc. Roy. Soc. London, Ser. A 276, 258 (1963).

[2]M. H. Cohen and L. M. Falicov, Phys. Rev. Lett. 7, 231 (1961).

[3]A. B. Pippard, Philos. Trans. Roy. Soc. London, Ser. A 256, 317 (1964).

[4]L. M. Falicov and H. Stachowiak, Phys. Rev. 147, 505 (1966).

[5a]For a review, see R. W. Stark and L. M. Falicov, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1967), Vol. V, p. 235.

[5b]D. P. Karim, J. B. Ketterson, and G. W. Crabtree, J. Low Temp. Phys. 30, 389 (1978).

[6]A. B. Pippard, Proc. Roy. Soc. London, Ser. A 287, 165 (1965).

[7]R. W. Stark and R. Reifenberger, J. Low Temp. Phys. 26, 763 (1977).

[8]R. W. Stark and N. Sandesara, to be published.

[9]Exceptions to this arise principally because the electrons sense $\vec{B}=\vec{H}+4\pi\vec{M}$ instead of just $\vec{H}$; $\vec{M}$ is the oscillatory dHvA magnetization. Although some magnetic interaction effects were present in our experiment, these produced spectral perturbations that were small compared with those discussed above. A full account of the magnetic interaction effects will be published elsewhere.

[10]Such frequencies are expected to be present in the galvanomagnetic tensor. From that source, they could show up in this experiment as a result of eddy currents arising from the small signal $[\Delta h]\cos[2\pi ft]$ used to modulate H. We could find no evidence for magnetoresistive contributions for $f \lesssim 400$ Hz. All of the data discussed above were taken with $f = 22.5$ Hz.