Magnetic oscillations in the heavy-fermion superconductor UBe_{13}

G. M. Schmiedeshoff

Department of Physics, Occidental College, Los Angeles, California 90041

Z. Fisk and J. L. Smith

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 16 October 1992)

We present measurements of the magnetic torque acting upon a single crystal of UBe₁₃, measurements made with a capacitive magnetometer. We observe pronounced magnetic oscillations in the torque resulting from the de Haas-van Alphen effect. A conventional analysis reveals that these oscillations result from "light fermions" with an effective mass $m^* = 0.017m_e$ (m_e is the bare electron mass). The Fermi surface associated with these light quasiparticles is anisotropic. We present arguments that these oscillations represent an intrinsic phenomenon and compare our results with those of other groups.

The magnetic oscillations resulting from the de Haas-van Alphen (dHvA) effect have proven to be an effective probe of the Fermi surfaces and quasiparticles of heavy-fermion compounds.¹ Studies of the dHvA effect in UPt₃, for example, have revealed quasiparticles with a large effective mass² and coexisting quasiparticles with a wide range of effective masses have been identified in several other heavy-fermion compounds.³ In this paper we report measurements of dHvA oscillations in the heavy-fermion superconductor UBe₁₃.⁴

Magnetic oscillations have been observed in magnetostriction,⁵ ultrasound,⁶ and magnetic torque measurements⁷ on single-crystal samples of UBe₁₃. These oscillations have been attributed to the dHvA effect due to an unusual aspect of the Fermi surface.⁵ Despite being observed in three single-crystal samples, there is reason to believe that these oscillations are *not* intrinsic to UBe_{13} . Efforts to reproduce the effect in magnetostriction measurements on another sample have been unsuccessful;⁸ and our own technique, applied to a second single crystal, did not show the oscillations.⁷ However, based on information currently available, we shall presently argue that these oscillations are intrinsic to UBe₁₃. In the meantime, we will proceed under the assumption that these are dHvA oscillations, reflecting aspects of the Fermi surface of UBe₁₃.

The magnetization M can be derived from measurements of the magnetic torque τ acting upon the sample. These measurements are made with a capacitive, cantilever magnetometer described in detail elsewhere.⁹ Briefly, the sample is suspended by fine copper wires over a silvered glass plate, forming a capacitor in which the sample itself is one capacitor plate. This assembly is positioned at the magnetic center of a normal or superconducting magnet. Any nonspherical sample will experience a torque attempting to align the long axis of the sample with the external magnetic field H. Modeling the sample as an oblate spheroid, one can show that, to leading order in the magnetic susceptibility χ , these "shape effects" result in a torque $\tau \propto \chi^2 H^2$. If χ is not isotropic (e.g., if magnetic order exists) then an additional contribution to the torque can arise from a transverse (with respect to H) component of the magnetization M_{\perp} given by $\tau = M_{\perp}H$. If the change in capacitance of the magne- $[\Delta C/C \ll 1,$ tometer is small where $\Delta C \equiv C(T,H) - C(T,0)$ then $\Delta C \propto \tau$. Since γ is a naturally small parameter, even in heavy-fermion compounds, the effect of an M_{\perp} on ΔC can be much larger than that of the shape effects. If the magnetometer is positioned away from magnetic center, the sample will experience a force F, proportional to the field gradient, which also contributes to ΔC .⁹ This magnetometer has been used in dHvA studies of Pb (Ref. 10) and several organic superconductors.11

Our sample is a single crystal of UBe₁₃ with approximate dimensions $1 \times 1 \times 6$ mm³; the characteristics of this crystal have been described elsewhere.¹² Typical data showing dHvA oscillations at 4.2 K, for two orientations of the crystal with respect to *H*, are shown in Fig. 1. The oscillations are sinusoidal and periodic in 1/H, the hysteresis is much smaller than the oscillation amplitude. The oscillations exist predominantly in τ where they dominate the overall response of the magnetometer at this temperature. This dominance of the oscillations in τ over the background associated with the shape effects discussed above indicates that the oscillations are due to a transverse component of the magnetization M_1 .

We interpret these oscillations within the theory of the dHvA effect embodied in the Lifshitz-Kosevich (LK) formula.¹³ In this formalism, an extremal cross-sectional area of the Fermi surface is deduced from an oscillation frequency, the effective mass of the quasiparticles associated with this orbit is deduced from the temperature dependence of the oscillation amplitude at fixed field, and the mean-free path of the quasiparticles is deduced from the field dependence of the oscillation amplitude at fixed field triangle the field dependence of the oscillation amplitude at fixed fixed from the field dependence of the oscillation amplitude at fixed fixed temperature. In this theory, an oscillating M_{\perp} arises from an anisotropic Fermi surface which, therefore, requires the oscillation frequencies v to be anisotropic.¹³

Fast Fourier transforms (FFT) of the oscillatory component of the data shown in Fig. 1 are shown in Fig. 2 (the spectrum for the [100] data has been offset by 20 aF);

658



FIG. 1. Typical force (F) and torque (τ) response of a single crystal of UBe₁₃ at 4.2 K, for magnetic fields directed along (a) the [100] axis and (b) [110] axis.

the oscillation frequencies are clearly anisotropic. The high-frequency component is depressed to 295 T at $H \parallel [110]$ from 360 T at $H \parallel [100]$. The dominant low-frequency component increases to 45 T at $H \parallel [110]$ from 32 T at $H \parallel [100]$. Note that this increase is less than the overall width of the low-frequency peak(s).

Values of the effective mass m^* and mean-free path l_0 are deduced from measurements of $\Delta C(H)$ in fields to 23 T over a temperature range from 0.5 to 30.0 K. Our analysis focuses on the 360 T oscillation; the amplitude and frequency of the low-frequency component(s) are somewhat dependent on the method used to extract the oscillatory component of this data from the background. A frequency of 360 T yields an average wave vector $k_0 = 1.05 \times 10^7$ cm⁻¹ associated with this particular extremal Fermi surface cross section $[\pi k_0^2 = 0.09(2\pi/a)^2]$, where a = 10.025 Å is the lattice constant of UBe₁₃ (Ref. 14)].

Plots of the oscillation amplitude $[M_{osc}(H) = \tau_{osc}(H)/H]$ as a function of temperature at 20 T (solid circles) and 15 T (open circles) are shown in Fig. 3. The solid lines are fits of the data above 4.0 K to the LK formula. Note the peaks in $M_{osc}(T)$ near 3.0 K in both data sets. Although such a peak is consistent with earlier magnetostriction oscillations,⁵ it may be that this peak is associated with an anomalous behavior of the (dc) torque in UBe₁₃ discussed elsewhere.^{15,16} We, therefore, only fit the data at temperatures above 4.0 K to the LK formula; fits to both data sets yield an effective mass $m^*=0.017m_e$, where m_e is the bare electron mass. The dHvA oscillations are, therefore, associated with rather



FIG. 2. Fast Fourier transform (FFT) of the oscillatory component of the torque data of Fig. 1. The FFT spectrum from the [100] data has been shifted vertically by 20 aF.



FIG. 3. Amplitude of the magnetic oscillations as a function of temperature at fixed field. The solid and open circles denote data taken at 20.0 and 15.0 T, respectively. The solid lines are fits of the data at temperatures above 4.0 K to the LK formula (see text).

"light fermions" coexisting with the heavy fermions responsible for the enormous specific heat and magnetic susceptibility at low temperatures. The existence of light fermions has been suggested as the origin of the highly nonlinear Hall effect in UBe₁₃ at low temperatures.¹⁷

"Dingle plots" of the scaled oscillation amplitudes at 0.5 K (solid circles) and 7.5 K (open circles) are shown in Fig. 4 where $X = 2\pi^2 m^* ck_B T / e\hbar H$ and the symbols have their usual meanings. The solid lines represent a fit to the LK formula (for the data at temperatures above 4.0 K) which yield a mean-free path $l_0 = 320$ Å. Note that the data of 0.5 K are consistent with this value for the meanfree path at low fields, but fall below the fit at high fields. The oscillations in the high-field portion of the 0.5 K data may be due to the beating of another frequency component whose magnitude becomes appreciable at low temperatures and in high fields; if such an additional component exists, it might interfere destructively with the 360 T frequency component to create the peaks in the data of Fig. 3. The Dingle temperature¹³ associated with this data is 28 K, reflecting the small values of m^* and l_0 .

Next, we compare our results with those reported by Wolf, Blick, Bruls, Lüthi, Fisk, Smith, and Ott (WBBLFSO).⁶ The reader is cautioned that we are comparing data from different regimes of the H-T plane: the data of WBBLFSO result from measurements taken at temperatures between 45 mK and 3 K and in fields to 10 T. WBBLFSO find two high-frequency components of 305 and 384 T at 45 mK, as opposed to our single frequency at 360 T (which we find to be independent of temperature from 30 to 0.5 K). Perhaps this component



FIG. 4. Dingle plots of the amplitude of the magnetic oscillations as a function of the inverse magnetic field at fixed temperature. X is a quantity defined elsewhere (see text). The solid and open circles denote data at 0.5 and 7.5 K, respectively. The solid lines are fits of the data at temperatures above 4.0 K to the LK formula (see text).

splits with further lowering of the temperature. WBBLFSO find an effective mass $m^*=0.2m_e$, an order of magnitude larger than our result. High magnetic fields are known to depress the effective mass of the heavy fermions in UBe₁₃, although not by an order of magnitude.¹⁸ (On the other hand, such measurements are dominated by the heavy quasiparticles, not those responsible for these oscillations, and we find no field dependence between 15 and 20 T.) WBBLFSO find a mean-free path $l_0=2070$ Å, more than six times larger than ours. Mean-free paths larger than 2000 Å are characteristic of the very clean metallic crystals traditionally required to observe the dHvA effect;¹³ however, UBe₁₃ is a relatively dirty metal with a large resistivity above 2 K.¹⁹

We now argue that these oscillations represent a phenomenon intrinsic to UBe₁₃. Sample-to-sample variations can be significant in some heavy-fermion systems, as can aging effects which result from α decay of the U.²⁰ Since these oscillations have been reported to exist in three different crystals, one would like to attribute this effect to a particular impurity common to all three crystals. Aluminum is the most likely contaminant since the crystals are grown in an aluminum flux, and the existence of aluminum inclusions is well known in these samples. Indeed, the superconducting transition of aluminum is visible in the low-field superconducting quantum interference device (SQUID) data used to characterize our single-crystal sample and this aluminum signature is absent in similar data on a second single crystal which does not show dHvA oscillations. On the other hand, WBBLFSO report that their sample does not show such a signature. Also, the value of m^* we find is significantly smaller than any of the reported effective masses for aluminum that we are aware of^{21} (although this is not the case for the results of WBBLFSO at lower fields and/or temperatures). Further, for these oscillations to result from impurities, one would expect the impurities to exist as single crystals (yielding an impurity M_{\perp}) and/or to be of sufficient mass for the magnitude of their dHvA oscillations to be comparable to the (relatively) large magnetization of pure UBe_{13} ; we regard either situation as unlikely.

The last point in our case against (Al) impurities is that we have observed small magnetic oscillations in torque data on a *polycrystal* sample.⁷ This sample was fabricated by arc-melting appropriate quantities of the constituent elements on a water-cooled copper hearth in an argon atmosphere. The torque associated with an M_{\perp} need not average to zero in this polycrystal sample since its shape is not ellipsoidal and the internal field will vary from point to point. The dHvA effect might exist in polycrystals as a consequence of the small effective mass: $r_c = (m * v / eH)$, where r_c is the cyclotron radius, e the electronic charge, and v the velocity of the quasiparticles. As a lower limit on v, we take an estimate for the heavy fermions in UBe₁₃ at the Fermi surface (assumed spherical)²² of $v = 3.4 \times 10^5$ cm/s, yielding $r_c < 1$ Å $\ll l_0$. The frequency for the oscillations in the polycrystal at 4.2 K is about 14 T (Ref. 7), within the low-frequency peak of the single-crystal data shown in Fig. 2. Although we find these arguments compelling, they do not explain the ap<u>49</u>

In conclusion, we have observed dHvA oscillations in the magnetic torque acting upon a single crystal of UBe₁₃ and have argued that it is an intrinsic phenomenon. Applying the conventional theory of the dHvA effect to one of the two dominant frequencies in our data, we find an average wave vector $k_0 = 1.05 \times 10^7$ cm⁻¹ associated with this particular extremal Fermi surface cross section, an effective mass $m^*=0.017m_e$, and a mean-free path $l_0=320$ Å. The Fermi surface associated with these oscillations is anisotropic.

We would like to acknowledge the assistance of B. S. Held and Gao Hua. This work was supported by the National Science Foundation under DMR-9019661. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy. The high-field measurements were made at the Francis Bitter National Magnet Laboratory (supported by the NSF).

- ¹See, for example, Proceedings of the 6th International Conference on Crystal-Field Effects and Heavy Fermion Physics, J. Magn. Magn. Mater. 76&77 (1988).
- ²L. Taillefer and G. G, Lonzarich, Phys. Rev. Lett. **60**, 1570 (1988); L. Taillefer, R. Newbury, G. G. Lonzarich, Z. Fisk, and J. L. Smith, J. Magn. Magn. Mater. **63&64**, 372 (1987).
- ³See, for example, M. Springford and P. H. P. Reinders, J. Magn. Magn. Mater. **76&77**, 11 (1988).
- ⁴For a detailed review of the properties of UBe₁₃ see H. R. Ott, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (North-Holland, Amsterdam, 1987), Vol. XI, Chap. 5.
- ⁵R. N. Kleinman, D. J. Bishop, H. R. Ott, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. 64, 1975 (1990); R. N. Kleinman, D. J. Bishop, H. R. Ott, Z. Fisk, and J. L. Smith, Physica B 165&166, 321 (1990).
- ⁶B. Wolf, R. Blick, G. Bruls, B. Lüthi, Z. Fisk, J. L. Smith, and H. R. Ott, Z. Phys. B **85**, 159 (1991).
- ⁷G. M. Schmiedeshoff, Z. Fisk, and J. L. Smith, in *Physical Phenomena at High Magnetic Fields*, edited by E. Manousakis, P. Schlottmann, P. Kumar, K. Bedell, and F. M. Mueller (Addison-Wesley, Redwood City, CA, 1992), p. 209.
- ⁸A. de Visser, N. H. van Dijk, K. Bakker, J. J. M. Franse, A. Lacerda, J. Flouquet, Z. Fisk, and J. L. Smith, Phys. Rev. B 45, 2962 (1992).
- ⁹G. M. Schmiedeshoff, Philos. Mag. B 66, 711 (1992).
- ¹⁰J. S. Brooks, M. J. Naughton, Y. P. Ma, P. M. Chaikin, and R. V. Chamberlin, Rev. Sci. Instrum. 58, 117 (1986).

- ¹¹See, for example, A. G. Swanson, J. S. Brooks, H. Anzai, N. Konoshita, M. Tokomoto, and K. Murata, Solid State Commun. **73**, 353 (1990).
- ¹²G. M. Schmiedeshoff, Z. Fisk, and J. L. Smith, Phys. Rev. B 45, 10544 (1992).
- ¹³D. Shoenberg, Magnetic Oscillations in Metals (Cambridge University, Cambridge, England, 1984).
- ¹⁴J. L. Smith, Z. Fisk, J. O. Willis, B. Batlogg, and H. R. Ott, J. Appl. Phys. 55, 1996 (1984).
- ¹⁵G. M. Schmiedeshoff, R. T. Tisdale, J. A. Poulin, S. Hashmi, and J. L. Smith, Physica B 165&166, 325 (1990).
- ¹⁶G. M. Schmiedeshoff, Z. Fisk, and J. L. Smith, Phys. Rev. B (to be published).
- ¹⁷N. E. Alekseevskii, V. N. Narozhnyi, V. I. Nizhankosvkii, E. G. Nikolaev, and E. P. Khybov, Pis'ma Zh. Eksp. Teor. Fiz. 40, 421 (1984) [JETP Lett. 40, 1241 (1984)].
- ¹⁸M. J. Graf, N. A. Fortune, J. S. Brooks, J. L. Smith, and Z. Fisk, Phys. Rev. B 40, 9358 (1989).
- ¹⁹H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **50**, 1595 (1983).
- ²⁰J. L. Smith, Philos. Mag. B 65, 1367 (1992).
- ²¹See, for example, C. O. Larson and W. L. Gordon, Phys. Rev. 156, 703 (1967), and references therein.
- ²²M. B. Maple, J. W. Chen, S. E. Lambert, Z. Fisk, J. L. Smith, J. R. Ott, J. S. Brooks, and M. J. Naughton, Phys. Rev. Lett. 54, 477 (1985).