Observation of Magnetic Order in the Heavy-Fermion Superconductor UBe_{13}

R. N. Kleiman[a] and D. J. Bishop
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

H. R. Ott
Eidgenössische Technische Hochschule Zürich-Honggerberg, Zürich, Switzerland

Z. Fisk and J. L. Smith
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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We have measured the magnetostriction, $L(H)$, of a single crystal of the heavy-fermion superconductor UBe_{13}, using an all-silicon, high-precision capacitance dilatometer. We find clear evidence for a transition to an antiferromagnetic state at $T_N \sim 8.8$ K, which is suppressed in a field by $dT_N/dH \sim 0.36$ K/T. At low temperatures we observe pronounced magnetostrictive oscillations, which we believe are de Haas-van Alphen oscillations due to an unusual aspect of the Fermi surface.

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In recent years the intimate relationship between magnetism and superconductivity in the heavy-fermion superconductors (HFSC) has become increasingly clear. In UPt_3, with $T_c \sim 0.55$ K, an antiferromagnetic transition has been observed[1] in neutron-scattering experiments with $T_N \sim 5.0$ K. In URu_2Si_2, with $T_c \sim 1.2$ K, there is a transition to an antiferromagnetic state at $T_N \sim 17.5$ K as seen in a number of measurements.[2] The coupling of the magnetic order parameter to the superconducting order parameter imposes symmetry constraints[3] on the nature of the superconducting state, thought to be paired with angular momentum $l \geq 1$ in all the HFSC.[4] Such studies have been performed on UPt_3 using neutron scattering,[5] and are in progress on URu_2Si_2 using neutron[6] and magnetic[7] x-ray scattering. Furthermore, it is believed that the pairing mechanism in the HFSC is due to mediation via antiferromagnetic spin fluctuations.[8] In this context we might expect that the presence of a magnetic transition with $T_N \sim 10T_c$ is a general feature of superconductivity in this class of materials, and an essential one to understand.

The heavy-fermion superconductor UBe_{13}, with $T_c \sim 0.8$ K, has the most pronounced low-temperature properties[9] of the HFSC, with $\gamma \sim 1100$ mJ/mol K^2; nevertheless, a magnetic transition above T_c has not been reported previously.[10] In this Letter we show clear evidence for an antiferromagnetic transition at $T_N \sim 8.8$ K, which is suppressed in a magnetic field by $dT_N/dH \sim 0.36$ K/T. At low temperatures we see unusual magnetostrictive oscillations, not previously observed in any system, which we believe are related to the de Haas-van Alphen effect and to the magnetism we observe. These may be related to the unusual features observed in both specific-heat[11,12] and resistivity[12] measurements at $T \sim 2$ K, neither of which have received a satisfactory explanation. Also of note is that pressure studies[13] show that the electronic Gruneisen constant γ_e changes sign at $T \sim 8$ K. UBe_{13} is to be distinguished from the other HFSC by its cubic crystal structure (UPt_3, hexagonal; URu_2Si_2, tetragonal). While we find again that $T_N \sim 10T_c$ in support of the generality of these as the relevant energy scales, the manifestation of the magnetic properties among the HFSC is quite different. This is in part due to the different crystal structures, and to their accompanying symmetry considerations.

Measurement of the magnetostriction of materials has long been a traditional tool[14] for the study of weakly magnetic systems, due to the high sensitivity of dilatometry techniques. This technique is especially justified for the heavy-fermion systems, since they not only have strongly enhanced electronic properties due to the mass renormalization, but also have strongly enhanced Gruneisen parameters,[15] typically $\gamma_e \equiv d\ln\gamma/d\ln V \sim 100$, where γ is the linear specific-heat coefficient. Thus, because of the strong volume dependence of the thermodynamic properties of the system, the thermal expansion and magnetostriction are more sensitive by γ_e and γ_m, respectively, than they would otherwise be, and hence more sensitive to magnetic order than the more direct thermodynamic measures (i.e., C_v, M). From a thermodynamic Maxwell relation[14]

$$V^{-1}\partial V/\partial H|_{T,P} = -\partial m/\partial P|_{T,H}, \quad (1)$$

where $m = M/V$, the magnetization per unit volume. Assuming that $\mathbf{M}$ points along $\mathbf{H}$, $m = \chi H$, and that the dilatations are isotropic, this is equivalent to

$$L^{-1}dL/dH = \tfrac{1}{3}\kappa\gamma_m m, \quad (2)$$

where κ is the compressibility, and γ_m is the magnetic Gruneisen parameter $\gamma_m \equiv d\ln\chi/d\ln V$. Since the compressibility is known, and assuming that γ_m is constant, Eq. (2) shows that magnetostriction measurements can be simply related to the bulk magnetization of the system.

Using a novel implementation of conventional capacitance dilatometry we have studied the thermal expansion $L(T)$ and magnetostriction $L(H)$ of UBe_{13}. For magnetostriction measurements it is conventional[14] to calibrate

a dilatometer using a slug of single-crystal silicon which is expected to have no magnetostriction component. Instead we have constructed the cell itself *only* from single-crystal silicon to achieve high stability and sensitivity for magnetostrictive measurements. Furthermore, the design is ideal for thermal-expansion measurements, since the length changes at low temperatures are approximately $\Delta L/L = 2.2\times10^{-13}T^4$ and are thus insignificant below 15 K. No materials other than the sample or the silicon were used, in order to eliminate the need for addendum corrections and to maintain high sensitivity. With this cell design we achieved a sensitivity of $\Delta L/L \sim 3\times10^{-9}$ for measurements either as a function of H or T.

The sample was grown from an Al melt using standard Czochralski techniques. The sample was approximately cubic, with dimensions $\sim5\times5\times5$ mm^3. The magnetic field was applied along the [100] direction and the dilatation measured along the perpendicular [100] direction. The capacitance ($C_0\sim5$ pF) was measured between a polished face of the sample (100) and a thin gold film evaporated onto a silicon electrode, using a standard three-terminal capacitance bridge.

The thermal expansion of UBe_{13} has been previously measured by Ott[16] at $H=0$ for $0.3<T<10$ K. Ott, Fisk, and Smith also reported[17] no significant magnetostriction up to 15 kG. Our detailed study of the thermal expansion in the superconducting state and up to 20 K is the subject of a later work,[18] but is generally consistent with their measurements. Our measurements up to 8 T show that the magnetostriction is not appreciable in magnitude, but a careful study of it shows significant structure which we now discuss.

In order to directly measure the quantity of interest, dL/dH [see Eq. (2)], we added a very-low-frequency ($f\sim10^{-3}$ Hz) field modulation of ~1 kG to the applied field, while recording the consequent modulations of the length of the sample. The frequency was kept low to eliminate sample heating and to maintain long integration times. The amplitude of the length modulations was measured at a number of temperatures, tracing out a curve, which is equivalent to $M(T)$. This is shown in Fig. 1 for $H_0=3$ and 7 T. The curves have the characteristic shape of an order parameter, being constant at low temperatures and vanishing at 7.65 and 6.33 K, respectively. We believe that the shape of the dL/dH curves is clear evidence for magnetic ordering and the suppression of the transition temperature in a field indicates that it is antiferromagnetic in nature. However, at a conventional antiferromagnetic transition the magnitude of the magnetic susceptibility decreases below T_N. The observed increase in $\gamma_m m$ may be due to a change in the magnetoelastic coupling (i.e., γ_m) associated with the magnetic ordering, or the result of a stronger coupling to one of the sublattice magnetizations, which would naturally increase below T_N.

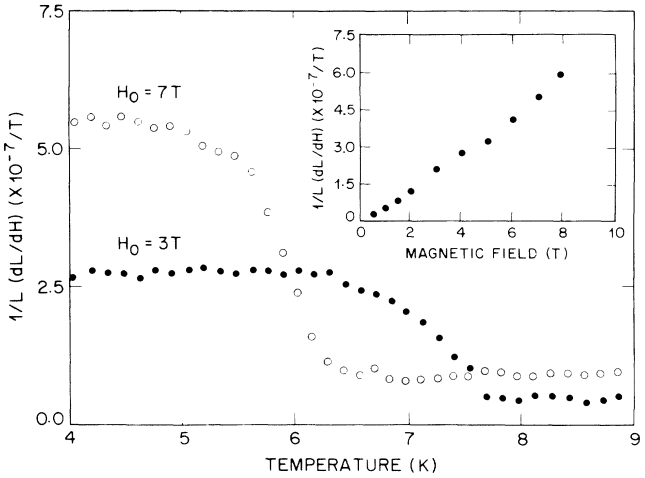

FIG. 1. Curves of $1/L(dL/dH)$ vs T taken using a field-modulation technique with $H=H_0+H_1\cos(\omega t)$. All data are taken with $H_1=1$ kG and $\omega/2\pi=10^{-3}$ Hz. The quantity $(1/L)dL/dH$ is proportional to the magnetization. Inset: Plot showing that the values of $(1/L)dL/dH$ in the saturated region ($T\sim3$ K) are proportional to the applied field as assumed in the text.

We have measured dL/dH vs T for a number of applied fields between 5 and 78.5 kG. The values of $(1/L)\times dL/dH$ in the saturated region ($T\sim3$ K) are plotted versus H in the inset of Fig. 1. The linear variation clearly supports the assumption made earlier of χ being independent of H. From the slope of this line, and a value for the compressibility[19] of $\kappa=8\times10^{-13}$ cm^3/erg, we find using Eq. (2) that $\gamma_m\chi\sim2.9\times10^{-3}$ emu/cm^3. This value is approximately 16 times higher than the measured[20] low-temperature value for the Pauli susceptibility χ_0, which confirms the fact that the observed effects are indeed due to a Gruneisen-enhanced susceptibility. Equivalently, we can express this as $(1/\chi_0)d\chi/dP\sim13$ Mbar^{-1}, which should be measurable with pressure studies provided that $d\chi/dP$ is constant up to the pressures typically used for measurement (~10 kbar).

The transition line in the H-T plane is plotted in Fig. 2. In the absence of a detailed model for $M(T)$, the transition temperatures T_N have been extracted assuming a linear variation of dL/dH near T_N. At low fields the error bars on T_N are high since the signal vanishes as $H\to0$. From Fig. 2 we find that T_N is suppressed by $dT_N/dH\sim0.36$ K/T. We conclude that for $H=0$, $T_N\approx8.8\pm0.2$ K.

While no hysteresis is observed in the curves of dL/dH as a function of temperature, there is significant hysteresis as a function of magnetic field for $T<T_N$. In Fig. 3 we show a number of plots of $L(H)$, taken at constant temperature, and relatively slow sweep rates $dH/dt\sim0.3$ mT/s. Figure 3, curve a is for $T>T_N$ ($T=10.0$ K) and shows no magnetic hysteresis. The curvature at low fields is most likely an artifact of the dilatometer.

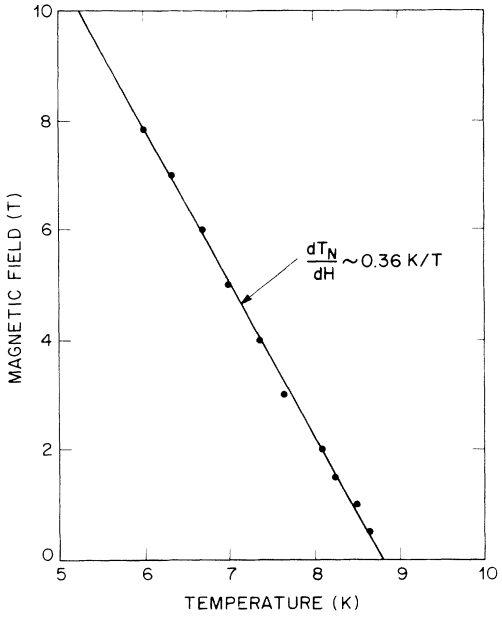

FIG. 2. The phase line separating the paramagnetic and antiferromagnetic region. At $H=0$, $T_N \sim 8.8$ K, and is suppressed in increasing fields by $dT_N/dH \sim 0.36$ K/T.

Figure 3, curve *b* is for $T < T_N$ ($T=4.0$ K) and shows significant magnetic hysteresis. Since $L(H) \sim MH$, the curves must meet at $H=0$. Figure 3, curve *c* is for $T=1.25$ K and only shows a trace for increasing field. It shows sharp jumps in $L(H)$ at specific fields, which appear to fall within an envelope roughly like that of Fig. 3, curve *b*. Upon repetition such a trace appears qualitatively similar, but the jumps do not occur at the same field each time. Figure 3, curve *d* is for $T=0.825$ K, and shows that the frequency of the jumps has increased significantly. They still remain in a well-defined envelope, now reduced in amplitude, but appear more convincingly to be oscillatory, perhaps periodic in $1/H$. With Fig. 3, curve *e* at $T=0.600$ K this trend continues. The anomaly at low fields is associated with H_{c2}(0.600 K) and is discussed elsewhere.[18]

The oscillatory magnetostriction at low temperatures, shown in Fig. 3, is clearly reminiscent of de Haas-van Alphen oscillations. It is not at all surprising to observe oscillatory magnetostriction in a clean metal at low temperatures due to the de Haas-van Alphen effect as was first noted by Chandresekhar,[21] and later measured[22] in bismuth. Magnetostriction measurements are particularly well suited due to their sensitivity, and the possible enhancements discussed earlier. However, there are a number of reasons why their appearance would be unlikely in this measurement; notably that the high effective masses implied by the specific heat, and the short mean free path implied by the resistivity would suggest that de Haas-van Alphen oscillations could only be observed at very low temperatures.[23]

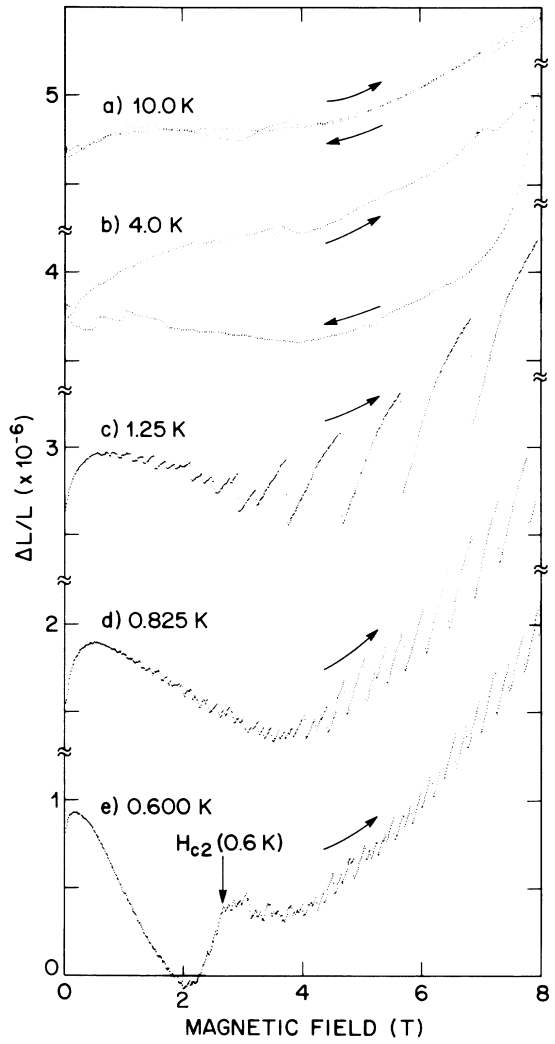

FIG. 3. The magnetostriction $L(H)$ taken at five different temperatures: curve *a*, $T > T_N$, no hysteresis is observed in the paramagnetic state; curve *b*, $T < T_N$, significant hysteresis is observed; curve *c*, $T=1.25$ K, magnetostrictive oscillations occur within an envelope similar to the above; curve *d*, $T=0.825$ K, magnetostrictive oscillations are much more rapid and lower in amplitude; curve *e*, $T=0.600$ K, the oscillations continue to decrease in amplitude and increase in rate. The anomaly below 2.6 T for curve *e* is due to H_{c2} for this temperature.

Furthermore, there are a number of ways in which our observations are not consistent with the conventional expectations[24] for the de Haas-van Alphen effect. First, the oscillatory frequency is given by $\omega_{1/B} \approx \phi_0 S$, where ϕ_0 is the flux quantum, hc/e, and S is an extremal area of the Fermi surface. The area S is given by the Fermi-surface topology and rarely deviates[25] by more than 10^{-4} over the range of temperatures where measurements are possible. In our measurements $\omega_{1/B}$ changes drastically with temperature, with $\omega_{1/B} \propto 1/T^3$ for $0.6 < T < 1.25$ K. Second, the oscillation amplitude is pre-

dicted to have a characteristic T and H dependence which we do not observe. It appears instead that the amplitudes are governed by the same domain effects responsible for hysteresis at higher temperatures. Finally, de Haas-van Alphen oscillations are usually sinusoidal and nonhysteretic. For this case, however, the oscillations appear more in the form of first-order jumps governed by nucleation processes.

While our data are clearly inconsistent with the conventional de Haas-van Alphen effect, we nevertheless believe that suitable modifications appropriate to UBe_{13} can help to explain our results. It has been shown that the amplitude of the magnetostrictive oscillations can be directly related[14] to the oscillatory magnetization by

$$\Delta L/L = \tfrac{1}{3}\kappa\gamma_s mB\,, \qquad (3)$$

where $\gamma_s = d\ln S/d\ln V$ is a Gruneisen parameter associated with the extremal orbit area S. Thus, there can be a tremendous enhancement over the usual oscillation amplitude if a part of the Fermi surface is particularly volume dependent. We can guess that $\gamma_s \sim \gamma_e \sim 100$, but it could easily be higher for one particular part of the Fermi surface.

The first-order nature of the jumps can likely be understood in terms of magnetic interaction.[26] The oscillations are governed not by the applied field, H, but by the internal field $B = H + 4\pi M$. Thus if M becomes comparable to the spacing between oscillations the system is in a metastable state during part of the oscillation cycle and undergoes a transition to a lower-energy state after a suitable nucleation process. Ordinarily such nucleation is not impeded and smooth variations are observed, but in our case it appears that the surface barriers are more significant than usual. In support of this argument we note that the jumps are only observed as a function of H and never as a function of T. It may be possible to understand the dependence of the oscillation amplitude on H and T in this context. It is not yet clear whether the observed hysteresis envelopes are a result of the same phenomena that lead to to the oscillations, or due to a separate magnetic effect which constrains the oscillation amplitude.

The strong temperature dependence to the oscillation frequency is difficult to understand. It may, in fact, be a change in the Fermi surface, related to the magnetic transition, such as the formation and opening up of a neck between parts of the Fermi surface. Alternately, in this temperature range the effective-mass renormalization is thought to be rapidly increasing, which may be reflected in the Fermi-surface topology. Finally, the change in oscillation rate may only be an apparent one, due to a change in the nucleation rate of magnetization jumps as a function of temperature.

We have shown that the capacitance dilatometry technique can be implemented to make very sensitive measurements of the magnetization of a system. It is particularly well suited to the study of the heavy-fermion systems, due to their greatly enhanced Gruneisen parameters. We have demonstrated that UBe_{13} orders into an antiferromagnetic state with $T_N \sim 8.8$ K, which is suppressed in a field with $dT_N/dH \sim 0.36$ K/T. Clearly, neutron-scattering experiments are warranted to elucidate the nature of the magnetic ordering. Below ~ 3 K we observe very unusual magnetostrictive oscillations, which we believe can be understood in the context of the de Haas-van Alphen effect with suitable modifications.

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