de Haas–van Alphen effect, magnetic transitions, and specific heat in the heavy-fermion system UCd_{11}

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(Received 21 December 1998)

We have used high pulsed magnetic fields to 50 T to observe de Haas–van Alphen oscillations in the heavy fermion antiferromagnet UCd_{11}, which has a stongly enhanced value of the electronic specific-heat coefficient ($\gamma = 803$ mJ/mol K^2). The low-temperature magnetization shows the existence of two magnetic phase transitions. The presence of quantum oscillations above the first transition indicates that UCd_{11} has a coherent Fermi liquid, although there is little or no change in the Fermi-surface topology on passing through the second transition. [S0163-1829(99)00421-X]

UCd_{11} crystallizes in the cubic $BaHg_{11}$-type structure with a lattice constant of 9.29 Å thus having one of the largest U-U separations $d_{\text{U-U}} = 6.56$ Å of any uranium compound.[1] This value of $d_{\text{U-U}}$ is much larger than the Hill limit ($d_{\text{U-U}} \approx 3.5$ Å),[2] and using the Hill criterion, one would expect magnetic ordering in UCd_{11}. Indeed, it is found that at $T_N \approx 5$ K, UCd_{11} orders antiferromagnetically.[1] Neutron-scattering experiments suggest that the magnetic structure is rather complicated.[3] The electronic specific heat coefficient ($\gamma = 803$ mJ/mol K^2) for UCd_{11} is the highest for any full moment magnetically ordered uranium heavy fermion compound.[4]

Previous measurements of the specific-heat in an applied magnetic field,[4] together with measurements of the magnetization and resistivity under pressure,[5] have shed some light on the unusual ground-state properties of UCd_{11}. For $T > 80$ K, the magnetic susceptibility exhibits Curie-Weiss behavior with $\mu_{\text{eff}} = 3.45\mu_B/\text{U}$ and $\Theta_p = -20$ K, while at low temperatures it becomes constant.[1] This behavior is typical of heavy fermion systems.[6] The application of a magnetic field causes a transition to be observed in specific-heat measurements for $B > 10$ T. When external pressure is applied to UCd_{11}, two further transitions are inferred from anomalies in the resistivity.[5] While the exact nature of these transitions is unclear, it has been proposed that at least one of them corresponds simply to a spin reorientation of the complicated magnetic structure.[5] These results show that UCd_{11} indeed has $5f$ electrons which hybridize with the conduction electrons, and the ordered $5f$ moment is sensitive to applied magnetic fields and pressure.

In this paper, we report measurements of the magnetization and de Haas–van Alphen (dHvA) effect in UCd_{11} in pulsed fields extending to 50 T, with the magnetic field applied along the $\langle 100 \rangle$ axis of the cubic crystal. At the lowest temperature measured (0.49 K), magnetic transitions are found to occur at applied magnetic fields of $B_{M1} = 6$ T and $B_{M2} = 16$ T. For $B_{M1} < B < B_{M2}$, six distinct dHvA frequencies are observed. For $B > B_{M2}$, we detected eleven frequencies, including all six that are seen for $B_{M1} < B < B_{M2}$. The overlap in the observed frequencies between the two field intervals suggests that the magnetic transition at 16 T does not significantly affect the Fermi-surface topology of UCd_{11}. The effective masses of the dHvA frequencies for $B > B_{M2}$ range from $m^* = 2$ to 11 m_e (where m_e is the free-electron mass). While these values are clearly larger than the free-electron value, they are significantly lower than what should be expected for the rather high value of γ.

Single crystals of UCd_{11} were grown using a standard flux technique described elsewhere.[7] The resulting well separated single crystals were typically cubes of a few mm on a side. The samples were found to crystallize in the $BaHg_{11}$-type structure with cubic lattice parameters of 9.29 Å, as evidenced by x-ray-diffraction studies. Using a standard four-probe measurement, the residual resistivity ratio between 4 and 300 K was found to be 43, which is much higher than the previously reported value of 2.6.[1] A clear kink was also observed in the resistivity at $T_N = 5$ K. The specific heat was measured on a small (~ 4 mg) sample employing a thermal relaxation method, with magnetic fields between 0 and 10 T provided by a superconducting magnet. The magnetization and dHvA effect were measured using counterwound highly compensated pickup coils in pulsed magnetic fields up to 50 T at the National High Magnetic Field Laboratory, Los Alamos. Throughout the pulsed field experiments, the sample was immersed in a ^{3}He environment in which the temperature could be varied between 0.49 and 2.1 K.

In Fig. 1, we show the measured inductive signal (which is directly proportional to dM/dH) versus magnetic field at 0.49 K. The two magnetic transitions at $B_{M1} = 6$ T and $B_{M2} = 16$ T are clearly visible, with the corresponding increases of the magnetic moment amounting to $0.06\mu_B/U$ and $0.05\mu_B/U$, respectively. The exact nature of the transitions is unclear, but it is unlikely that either is truly a metamagnetic transition where one expects a drastic change in the character of the f electron which manifests itself as an increase in the magnetization on the order of $1\mu_B/U$. However, we know that the magnetization increases at both transitions so it is likely there is some spin reorientation. The observation of two transitions in applied field is similar to the high-pressure results,[5] though without more detailed studies of the magnetic structure we cannot say they are analogous. Further investigations, such as high-field neutron

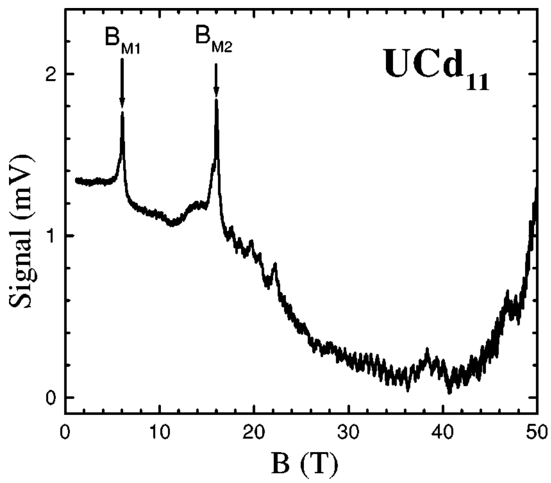

FIG. 1. The measured signal (induced voltage) versus applied pulsed magnetic field for UCd_{11} at 0.49 K. Two magnetic transitions are clearly observed at $B_{M1}=6$ T and $B_{M2}=16$ T.

diffraction or dHvA measurements as a function of pressure, are needed to determine the field dependence of the magnetic ground state.

The zero field data from specific-heat measurements are shown in Fig. 2. The line represents a fit to the data using

$$C/T=\gamma+\beta T^2, \tag{1}$$

where γ is the electronic specific-heat coefficient and β is the lattice Debye term. The data were fit to Eq. (1) above T_N over the range $70<T^2<200$ K^2. From the zero-field data shown in Fig. 2, we find $\gamma=803\pm3$ mJ/mol K^2 and $\beta=5.13\pm0.02$ mJ/mol K^4; the latter corresponds to a Debye temperature of $\Theta_D=166$ K. The entropy associated with the magnetic transition is $0.34R\ln 2$. These values agree reasonably well with previously published data,[1] but the value of

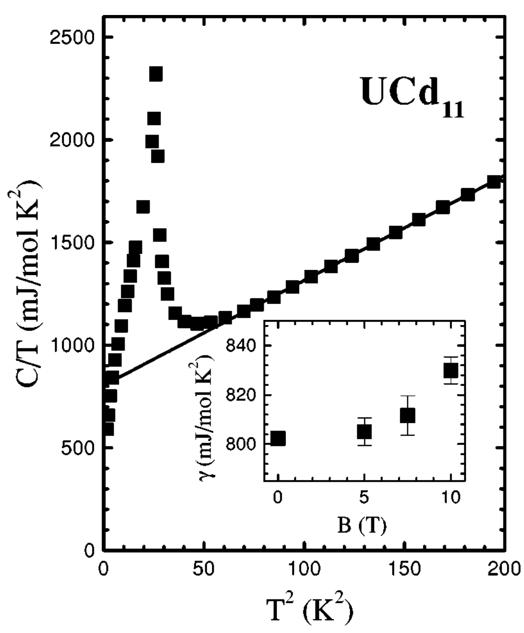

FIG. 2. The zero-field specific heat C divided by temperature T versus T^2 for UCd_{11}. The line is a fit as described in the text. The inset shows the field dependence of the linear specific-heat coefficient γ.

Θ_D is somewhat smaller than the reported value of 200 K of Andraka *et al.*[4] This discrepancy may be due to a difference in sample quality or the temperature range used to fit the specific-heat data. Application of magnetic fields to heavy fermion systems can lead to dramatic effects in the measured specific heat. In general, the value of $\gamma(B)$ tends to decrease with applied field.[8–10] There are, however, systems which do not behave according to the standard theory. For example, in systems exhibiting a metamagnetic transition field B_M, it has been shown experimentally in $CeCu_2Si_2$ (Ref. 11) and UPt_3 (Ref. 12) and theoretically[13] that $\gamma(B)$ can increase for $B<B_M$ but then decrease for $B>B_M$. The measured values of $\gamma(B)$ for UCd_{11} changes with magnetic field as shown in the inset of Fig. 2, while the measured value of $\beta(B)$ (not shown) does not change. The rapid increase of $\gamma(B)$ on the approach to 10 T in Fig. 2 would appear to be consistent with a metamagnetic transition in fairly close proximity to that field; i.e., in good agreement with the transition observed in Fig. 1 at $B_{M2}=16$ T, but in disagreement with a previous report which did not observe a change in $\gamma(B)$ in fields to 16 T.[4] However, the relatively small change in the magnetization at B_{M2} makes a metamagnetic transition unlikely. If we look at temperatures below T_N, we can determine $\gamma_o\equiv C/T$ $(T\to 0)$ from our data and that of Andraka *et al.*,[4] and the value of γ_o decreases as B increases and appears to be reaching a nearly constant value near at high fields. From our results, we would estimate this value to be of the order of 300 mJ/mol K^2, while Andraka *et al.*,[4] who measured to higher fields and lower temperatures, find a value of $\sim$200 mJ/mol K^2. It would be useful to continue the measurement of the specific heat to higher fields to determine how γ and γ_o behave over a broader field range.

The dHvA effect is the most definitive probe of the Fermi-surface properties of metals; the frequency of the oscillations F are directly proportional to extremal cross-section areas of the Fermi surface, while the amplitude of the signal yields important information concerning the electronic interactions. Much theoretical and experimental work has been done on the Fermi surfaces of heavy fermion systems.[14–18] The main finding of the studies is that a Fermi-liquid description fits but is characterized by a heavily renormalized effective mass m^*. Since the dHvA amplitudes decrease drastically as m^*/B increases, it is very difficult to observe dHvA oscillations in heavy fermion systems where large values of γ imply large m^*. The use of very high pulsed fields to 50 T can greatly reduce the value of m^*/B and make the observation of dHvA oscillations in heavy fermion systems possible where traditional low-field techniques cannot. According to the mean-field Anderson lattice model, the dHvA oscillations are thought to be dominated by a single spin sheet of the Fermi surface.[14,19]

The dHvA measurements were performed on a cylindrical sample with a diameter of $\sim$0.8 mm that was cut from a single crystal with the cylindrical axis along the crystalline (100) direction. The measured signal for UCd_{11} versus the inverse applied field at 0.49 K is displayed in Fig. 3(a). There is clearly an oscillatory dHvA signature in the data. The fast Fourier transform (FFT) of the measured data in the ranges $B_{M1}<B<B_{M2}$ and $B>B_{M2}$ for falling fields is shown in Fig. 3(b). Numerous peaks appear in the FFT data in Fig. 3(b), and the frequencies of the eleven observed peaks

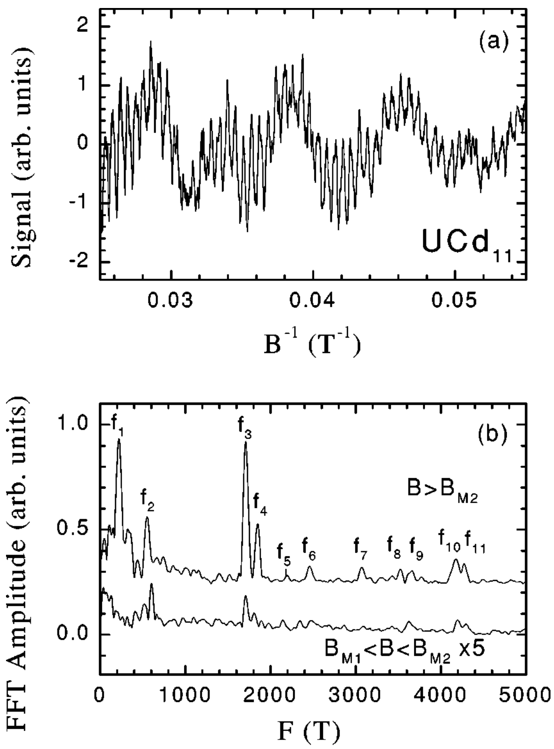

FIG. 3. The measured signal versus inverse applied field for fields above B_{M2} is shown in (a). The fast Fourier transform (FFT) amplitudes of the measured signal for fields above and below B_{M2} (note that for $B_{M1}<B<B_{M2}$ the amplitude has been multiplied by a factor of 5) is shown in (b). The peaks that appear above B_{M2} are labeled f_1-f_{11} and have their physical properties summarized in Table I.

along with the corresponding mass of the orbit for $B>B_{M2}$ (labeled f_1-f_{11}), as determined by fitting to the Lifshitz-Kosevich theory, are listed in Table I. The masses listed in Table I are averages of the values obtained for rising and falling field data over the range $24<B<46$ T. Six of the frequencies (f_2, f_3, f_4, f_9, f_{10}, and f_{11}) observed for $B>B_{M2}$ are definitely seen in the range $B_{M1}<B<B_{M2}$. The two frequencies with the largest amplitude where the orbit mass could be determined in both field ranges (f_2 and f_3), do not exhibit a change in m^* or T_D within the experimental uncertainty at B_{M2}. Four of the other frequencies (f_5, f_6, f_7, and f_8) are impossible for us to say for sure if we can or cannot detect them, and if it is assumed that m^* and T_D are the same for fields above and below B_{M2} (as seen for f_2 and f_3), the estimated amplitude of these frequencies in the range $B_{M1}<B<B_{M2}$ would be within our measured noise. Using these same assumptions, the 11th frequency (f_1) should be well above the noise level for $B_{M1}<B<B_{M2}$, and it would appear that this small piece of Fermi surface disappears as the field is increased above B_{M2}. From these results, while there may be some change of the Fermi surface at B_{M2}, there is not a major Fermi-surface reconstruction as has been observed in other U systems such as UPt_3 and UPd_2Al_3 that display a metamagnetic transition.[20,21] Since the masses listed in Table I appear to be rather low compared to the value of γ and γ_o, it is possible that there may exist Fermi-surface sheets with heavier masses that we are unable to detect. It is also conceivable that we are only seeing half of the Fermi surface, since only the spin channel with the lightest effective masses contributes appreciably to the dHvA effect. The specific heat, on the other hand, represents the entire Fermi-surface integrated effective mass of all spin channels. A more complete knowledge of the Fermi-surface topology is required for us to make any definite conclusions. Angle-dependent magnetization data together with band-structure calculations would also be helpful in this regard.

TABLE I. Measured de Haas–van Alphen frequencies F, effective masses m^*, and Dingle temperatures T_D above the magnetic transition at B_{M2} in UCd_{11}. The frequencies f_1-f_{11} correspond to the labeled peaks in Fig. 3.

	F (T)	m^* (m_e)	T_D (K)
f_1	221	4.9 ± 0.5	1.1 ± 0.2
f_2	551	2.3 ± 0.3	2.5 ± 0.4
f_3	1714	1.7 ± 0.3	2.2 ± 0.2
f_4	1856	5.0 ± 0.8	1.8 ± 0.2
f_5	2204	7.5 ± 3.3	0.6 ± 0.1
f_6	2460	10.7 ± 2.5	0.5 ± 0.1
f_7	3069	8.5 ± 2.1	1.3 ± 0.1
f_8	3544	8.6 ± 1.4	1.1 ± 0.1
f_9	3685	5.5 ± 1.2	1.6 ± 0.2
f_{10}	4183	5.6 ± 0.4	1.2 ± 0.1
f_{11}	4280	7.6 ± 1.8	0.8 ± 0.1

We also examined the dHvA data for any field or temperature dependences of the frequencies or the masses. In weak ferromagnets[22,23] and some heavy fermion systems, notably UPt_3,[20,24] it is known that there can be a large field dependence of the dHvA frequencies due to spin-split bands. We did not observe such an effect in UCd_{11} for $B>B_{M2}$. In fact, the discernible change of the dHvA frequency at B_{M2} for the three frequencies with the largest dHvA amplitude for $B_{M1}<B<B_{M2}$ is less than 30 T. We also did not observe any temperature dependence of the dHvA frequencies. Within the experimental uncertainty of our measurements, we could not discern any field dependence of m^* for $B>B_{M2}$, and as discussed earlier, there was no change in m^* at B_{M2} for the two frequencies where m^* could be determined above and below B_{M2}. These results are consistent with the specific-heat measurements which show that γ_o appears to be reaching a nearly constant value at high fields.

In summary, we have observed dHvA oscillations in the heavy fermion antiferromagnet UCd_{11}. The value of the electronic specific-heat coefficient in applied field increases with increasing field up to 10 T. The magnetic ground state is sensitive to applied magnetic fields as two magnetic transitions are clearly observed at $T=0.49$ K for fields $B_{M1}=6$ T and $B_{M2}=16$ T. Due to the similarity of the Fermi surface above and below B_{M2}, we conclude that the phase transition that occurs at B_{M2} is probably just a spin reorientation rather than a metamagnetic transition. The 11 de Haas–van Alphen frequencies that are observed for $B>B_{M2}$ have field independent effective masses in the range 2–11 m_e. While these values are clearly larger than the free-electron value, they are significantly lower than what should be expected for the rather high electronic contribution to the specific heat. Further angle-resolved dHvA measurements and band-structure calculations are needed to fully determine the Fermi surface of UCd_{11}, and to enable a more careful comparison of the specific heat to the dHvA effective masses.

We would like to thank J. L. Smith for cutting the sample and M. E. Torelli for a low-temperature magnetic susceptibility measurement to confirm the magnitude of the magnetic transition at $B_{M1}=6$ T. Work at LANL is performed under the auspices of the U.S. Department of Energy, and the NHMHL is supported by the NSF and the State of Florida.

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