de Haas–van Alphen Effect in the Heavy-Electron Compound $CeCu_6$

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We report measurements of the de Haas–van Alphen effect in the heavy-electron metal $CeCu_6$ in the coherent state. Cyclotron masses of up to $40m_0$ are observed and we deduce a many-body enhancement 20 times larger than in $LaCu_6$. We conclude from the absence of any light electrons that the many-body renormalization leading to the coherent state influences all electrons, and not just those with primarily f character.

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At low temperature $CeCu_6$ has one of the largest known electronic specific heat coefficients[1–3] but unlike many other heavy-electron compounds it has neither a magnetic nor a superconducting transition down to 20 mK.[1–4] It therefore provides a very suitable material for the study of the low-temperature coherent state. At higher temperatures it behaves as if it was a system of isolated "Kondo" impurities but below about 10 K the resistance drops from a peak value of about 80 $\mu\Omega$ cm to very low (sample dependent) values,[1–4] the magnetoresistivity becomes small and positive rather than large and negative,[5,6] and the Hall effect changes sign.[7] These are all indications of the formation of a "Kondo lattice" in which the scattering from the Ce ions becomes coherent in a manner which is not yet understood. The electronic specific heat coefficient and the susceptibility are both very large but have a Wilson ratio of order 1.[8] Many of these low-temperature properties can be understood in terms of many-body Abrikosov-Suhl or "Kondo" resonance at the Fermi level although there is some evidence that an additional double-peaked structure appears as coherence develops.[9]

We report here the first measuremtns of the de Haas–van Alphen (dHvA) effect in a true heavy-electron metal. The measurements were made down to 20 mK where the electrons are certainly in the coherent state as indeed the observation of dHvA oscillations shows. In addition to determining Fermi surface parameters the measurements also yield scattering data and cyclotron effective masses.

The cyclotron masses obtained from the temperature dependence of dHvA oscillations are known to be renormalized by the electron-phonon interaction,[10,11] and electron-electron effects are expected to appear similary.[12,13] The cyclotron masses m^* are the orbital integral of $1/v_F^*$, where v_F^* is the renormalized velocity, so that, with a suitable parametrization scheme, cyclotron masses can be deconvoluted to obtain velocities. Suitably integrated over the whole Fermi surface, these should give γ as determined from the electronic specific heat which is enhanced by many-body effects in the same way. Indeed, this process has been carried through in detail for a number of cases.[14–16] The very large values of γ in heavy-electron metals therefore imply correspondingly large cyclotron masses and weak dHvA signals which will only be detectable at very low temperatures. If, however, as has sometimes been suggested, both light and heavy electrons coexist in certain heavy-electron metals,[17] the light electrons would signal their presence unambiguously by a relatively much stronger dHvA effect.

Field-dependent studies of dHvA-effect amplitudes give information on the electron scattering rate. They yield the ratio of cyclotron mass to electron lifetime, m^*/τ^*, in which both parameters are renormalized by the many-body interactions.

$CeCu_6$ is orthorhombic at room temperature but undergoes a phase transition to a monoclinic structure around 200 K.[18] As a precaution, the sample was cooled slowly through this point to minimize sample deterioration. The crystal distortion, however, is small ($\gamma = 90°$ becomes $\beta = 91.36°$) so that we can usefully retain the orthorhombic notation for the low-temperature phase. The present measurements were made on a c-axis crystal whose resistivity at 20 mK was close to 1 $\mu\Omega$ cm. The c axis was selected as it seemed likely to yield the most abundant dHvA signals, based on the work of Ōnuki *et al.*[19] *on the reference* materials $LaCu_6$ and $PrCu_6$. Additionally we expect the modifications to the Fermi surface induced by the phase transition to give rise to beats which will have least significance in the c direction.

Experiments were performed in a dilution refrigerator in fields up to 14.5 T with use of the field modulation technique at 6 Hz to avoid sample heating. Temperatures were in the range 20–250 mK as determined by a Ge thermometer in a zero-field region in the mixing chamber.[20]

The measured dHvA frequencies and cyclotron effective masses are given in Table I. A feature was observed in fields ≤ 2.5 T which might be explained as an extremely low dHvA frequency of ~ 8 T with a cyclotron mass of 2.5, but this assignment is not certain. The large orthorhombic unit cell which contains four formula units of $CeCu_6$ leads to a correspondingly small Brillouin zone. The highest frequency observed (1300 T) is 13% of the cross section of the Brillouin zone in this direction and corresponds therefore to a Fermi surface feature of significant size. The measured dHvA frequencies were independent of temperature to 1%, the frequency $F=122$ T being studied over the widest temperature range of 20–250 mK. Cyclotron effective masses were determined from the temperature dependence of dHvA amplitudes at constant field, as shown in Fig. 1.

In the absence of a band-structure calculation for $CeCu_6$ the measured cyclotron masses can best be discussed by comparison with those found[19] in $LaCu_6$ and $PrCu_6$. As there are no f electrons in $LaCu_6$ and those in $PrCu_6$ are localized, they form convenient reference compounds, with cyclotron masses typical of a d-band transition metal. Plotted versus dHvA frequency on log-log scales the data for these compounds and for $CeCu_6$ fall on two curves with similar slopes but with the masses in $CeCu_6$ larger by a factor of 30. This ratio may be compared with the corresponding ratio of the electronic specific-heat coefficients. For $LaCu_6$,[21] $\gamma = 8$ mJ/mole$\cdot$K^2, so that using a value for $CeCu_6$ (extrapolated to $T=0$) of 1.5 J/mole$\cdot$K^2 gives the significantly larger ratio value of 190.

The discrepancy between these two ratios may have its origin in the effect of a magnetic field. The electronic specific heat of $CeCu_6$ is strongly field dependent.[22] With the field parallel to the c axis, as used in these experiments, the maximum value of C/T is reduced by a factor of about 2 in a field of 5.5 T, and at 7.5 T[23] is about 0.5 J/mole$\cdot$K^2, that is, only a factor of 60 larger than in $LaCu_6$. At the lowest temperature at which the specific heat was studied, 0.15 K, C/T had not saturated, and a direct comparison with the cyclotron masses which are measured at lower temperature is therefore not possible. The cyclotron mass ratio of 30, measured in fields greater than 10 T, corresponds to C/T for $CeCu_6$ of about 0.25 J/mole$\cdot$K^2 and is broadly consistent with the field dependences quoted above. It implies, however, that measured cyclotron masses should be field dependent. While they may be tending to saturate for fields greater than 10 T, effects mirroring the changes in electronic specific heat should be observed at lower fields. This point was examined for the one dHvA frequency, $F=122$ T, that could be observed at lower fields. Measurements were made at several magnetic fields in the range 6–13 T but, to within the experimental uncertainty, the cyclotron mass was found to be constant at $m^* = (6.0 \pm 0.2) m_0$.

Two factors contribute to the cyclotron masses in $CeCu_6$ being ~ 30 times greater than in $LaCu_6$ and $PrCu_6$. Firstly, hybridization of the narrow f band

TABLE I. Measured dHvA frequencies F, cyclotron masses m^*/m_0, and Fermi velocities v_k^*, in a c-axis crystal of $CeCu_6$. Cyclotron masses are determined from the temperature dependence of dHvA amplitudes as illustrated in Fig. 1. The v_k^* are derived as explained in the text. The highest frequency observed with $m^*/m_0 \sim 80$ is thought to be a harmonic of the fundamental $F=1034$ T. Harmonics of other frequencies are also observed, notably of $F=40$ T and $F=122$ T, but are not listed.

F (T)	m^*/m_0	$10^{-3} v_k^*$ (m$\cdot$s^{-1})
200 ± 100	80 ± 10	...
1300 ± 20	24 ± 2	9.6
1034 ± 5	40 ± 2	5.1
740 ± 10	23 ± 1	7.5
638 ± 10	14 ± 1	11.5
210 ± 10	...	...
122 ± 2	6.0 ± 0.2	11.6
40 ± 4	11 ± 3	3.7

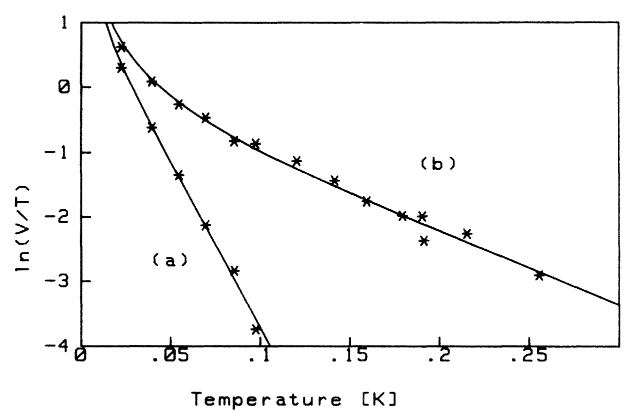

FIG. 1. Temperature dependence of dHvA amplitudes in a c-axis crystal of $CeCu_6$ for two dHvA frequencies: curve a, $F=1034$ T at $B=11.51$ T; and curve b, $F=122$ T at $B=7.25$ T. The variation of $\ln(V/T)$ vs T is shown, where V is proportional to the dHvA amplitude. The solid lines are fits to the data with the theoretical expression $y = \ln(P/\sinh QT)$ by the method of least squares. P and Q are constants and the cyclotron mass is obtained from $Q = 14.69 m^*/m_0 B$. The data for curve a yield $m^* = (40 \pm 2) m_0$ and for curve b $m^* = (6 \pm 0.2) m_0$. The results for all orbits are summarized in Table I.

with other bands increases the overall "band structure" mass. Additionally we expect a large many-body enhancement which is presumably related to the "Kondo" resonance at the Fermi level. A useful comparison may be made with $CeSn_3$ which, although not a heavy-electron compound, also has anomalously large masses and in which a direct comparison is possible with both band-structure calculations[24] and with the reference compound $LaSn_3$. The dHvA cyclotron masses in $CeSn_3$ are about 5 times larger than those in $LaSn_3$[25] and the γ values for the two compounds are in the same ratio. Comparison with band-structure calculations[26] suggests that f hybridization accounts for an increase in the masses in $CeSn_3$ by a factor of ~ 1.6, the difference being attributable to the difference in the enhancement term $(1+\lambda_{ep}+\lambda_{ee})$ in the two cases. Using, as a guide, the same factor of 1.6 for $CeCu_6$, we deduce that the many-body enhancement in $CeCu_6$ in fields over 10 T is greater than in $LaCu_6$ by a factor of ~ 20.

From our measured cyclotron masses in Table I we may estimate the renormalized quasiparticle velocities v_k^* characteristic of particular orbits on the Fermi surface. If we assume that they are circular with radius k_r, the velocity, assumed isotropic, is given by

$$v_k^* = \hbar k_r / m^*,$$

where

$$\pi k_r^2 = 2\pi eF/\hbar.$$

The values deduced in this way are given in Table I. They vary by a factor of about 3 from sheet to sheet which is typical of what is expected from band-structure effects. An estimate for the average velocity and the total Fermi surface area S_F can be deduced from the value of γ (which is given by $S_F\langle 1/v_k^*\rangle$) and the ratio of Dingle (scattering) temperature to the impurity resistivity which is proportional to $S_F\langle v_k^*\rangle$. With use of the measured values of 0.05 K for the Dingle temperature and 1 $\mu\Omega$ cm for the impurity resistivity the corresponding values of $\langle v_k^*\rangle$ are 3600 m/s if γ is taken as 1.5 J/mole$\cdot$K^2, or 8800 m/s if γ is 0.25 J/mole$\cdot$K^2. While it is not clear exactly what value of γ the cyclotron masses should be compared with, it is nevertheless apparent that the velocities deduced from the dHvA cyclotron masses (Table I) are consistent with the values deduced from the large electronic specific-heat coefficient. We note also that whereas γ is dominated by regions of lowest v_k^* (highest m^*), the dHvA effect is most sensitive to regions of highest v_k^* (lowest m^*) which, expressed in terms of a *single* sheet of Fermi surface would have a Fermi radius of about 10^{10} m^{-1}, a dHvA frequency of about 4×10^4 T, and an effective mass of $380 m_0$ and would contain 10 electrons/unit cell, i.e., 2.5 electrons per formula unit. However, $CeCu_6$ is a compensated metal and the Fermi surface must consist of a number of smaller sheets, with both hole and electron character, having the same total surface area. The scattering time deduced from the Dingle temperature is $\sim 10^{-11}$ s which is several hundred times larger than the corresponding quantity for simple metals with a resistitivy of 1 $\mu\Omega$ cm, but the free path, $v_k^*\tau^*$, is about 10^{-7} m which is quite typical for resistivities of this magnitude. The parameters are therefore consistent with a many-body enhancement which reduces all the velocities over the Fermi surface and increases the scattering times by the same amount so as to leave the mean free path constant, and which accounts qualitatively for the large electronic specific heat.

In summary, these first dHvA measurements in a heavy-electron metal have yielded results which influence our perception of the heavy-electron state. The observation of quantum oscillations implies a sharply defined Fermi surface even in the strongly interacting Fermi liquid that comprises the coherent state. At 10 T the orbits extend ~ 1 μm and so retain coherence over at least ~ 1000 lattice constants, and the dimensions of at least one small sheet of the Fermi surface change by less than 1% between 20 and 250 mK. We conclude from the measured cyclotron masses that no "light" electrons exist in $CeCu_6$ in the coherent state but that *all* are renormalized by an amount which, at the high magnetic fields used in these experiments is ~ 20 times greater than in $LaCu_6$. Finally, on one small sheet of the Fermi surface ($F=122$ T), we find no evidence of any field dependence of the cyclotron mass to reflect the reported field dependence of the electronic specific heat.

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