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To cite this version:

HAL Id: jpa-00228514
https://hal.archives-ouvertes.fr/jpa-00228514
Submitted on 1 Jan 1988

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MAGNETIC FIELD DEPENDENCE OF THE CYCLOTRON MASS IN THE KONDO LATTICE CeB₆

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Abstract. – The conduction-electron mass in the Kondo lattice system CeB₆ is found to be strongly field dependent. CeB₆ is an ordered heavy moment electron system with a zero field electronic specific heat coefficient of about 250 mJ/mole K². Using the de Haas-van Alphen effect at temperatures as low as 60 mK in steady magnetic fields as large as 22 T, we observe a cyclotron orbit of frequency 8680 T for fields along the [100] direction. The mass of this orbit was measured at eight fixed fields and found to decrease from 18 to 8 m_e as the field increases from 12 to 22 T. The field dependence of the cyclotron mass is consistent with recent specific heat results. The Fermi surface geometry is similar to that of LaB₆, except that the extremal cross-sectional areas observed are 10% larger than in LaB₆. The f-electrons are therefore largely local rather than itinerant in CeB₆, a picture confirmed by bandstructure calculations. The geometry of the Fermi surface does not depend on field. The observed field dependence of the cyclotron mass is consistent with the low energy scale of the system as measured, for example, by the Kondo temperature.

Heavy-electron systems form at low temperatures a highly correlated electronic state which exhibits unusual normal, antiferromagnetic, or superconducting behaviour [1]. The main characteristic, the anomalously large low-temperature specific heat (LTSH), indicates an exceptionally high electronic density of states at the Fermi energy. A high density of states results when the conduction-electron masses are very heavy. In CeCu₆ [2], CeB₆ [3] and UP₃ [4] recent de Haas-van Alphen (dHvA) measurements have confirmed the presence of conduction electrons with effective masses one to two orders of magnitude greater than observed in ordinary metals.

Here we discuss the results for the Kondo-lattice compound CeB₆ and show that the mass of the conduction electrons in this metal is strongly reduced in high magnetic fields. CeB₆ is one of the most typical dense Kondo lattices and many studies have been devoted to this system, see e.g. Kasuya et al. [5]. CeB₆ has a very low Kondo temperature, only 1-2 K [6] and orders magnetically at low temperatures. Even in the magnetically ordered phase the linear coefficient in the LTSH, γ, is very large and strongly magnetic field dependent [7, 8]. In zero field γ is about 250 mJ/mole K², which is to be compared to γ ≈ 2.6 mJ/mole K² [9] for the reference compound LaB₆, where the only difference is the absence of the f-electron. Two phase transitions are observed at T_Q = 3.2 K and T_N = 2.4 K. The transition temperatures are strongly affected by the application of an external magnetic field and the magnetic phase diagram has been studied by many groups, e.g. Effantin et al. [10]. Below T_N a complex antiferromagnetic order sets in, which is destroyed by fields larger than 1 to 2 T depending on the field direction. The phase between T_Q and T_N shows antiferro-quadrupolar ordering: the quadrupolar moment on the Ce ions alternates from site to site in all three perpendicular directions. In addition to a large uniform magnetic moment, a staggered magnetic moment is induced by an external field due to the difference in susceptibility on the +Q and −Q sites [10, 11]. Crystal field levels have been extensively studied in CeB₆, with recent measurements of Zirngiebl et al. [12] showing that the four fold degenerate Γ₇ level is the ground state and the Γ₇ state is 545 K higher in energy.

The dHvA effect is the oscillatory variation of the magnetization of a metal with the magnetic field H. The usual formalism for the interpretation of the dHvA signal is based on a formula similar to the one originally derived by Lifshitz and Kosevich [13]. The component of the oscillatory magnetization parallel to the

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field arising from one extremal cross-section may be expressed as:

\[ M = \sum_{r=1}^{\infty} M_r \sin \left( 2\pi r F / H + \beta_r \right), \quad (1) \]

where \( \beta_r \) is a constant for a given orbit. The magnetization varies periodically in \( H^{-1} \) with the dHvA frequency \( F = h A / 2\pi e \), where \( A \) is the extremal cross-sectional area of the Fermi surface normal to \( H \). The amplitude factor \( M_r \) depends on the cyclotron effective mass \( m^* \) (given in units of the free electron mass \( m_e \)), the Dingle temperature \( T_D \) (both quantities are renormalized by the many-body interactions) and on field \( H \) and temperature \( T \) in the following way:

\[ M_r = D_r T (rH)^{-1/2} \exp \left( -\alpha m^* T / H \right) / \sinh \left( \alpha m^* T / H \right), \quad (2) \]

where the precise form of \( D_r \) may be found in the review by Shoenberg [13] and \( \alpha = 14.69 \ T/K \) is a constant. Consequently, \( m^* \) and \( T_D \) can be determined from the temperature and field dependence of \( M_r \). The first harmonic \( (r=1) \) is generally dominant and for metals with a single or with separable oscillations the mass is calculated by an iterative procedure which properly accounts for the sinh term in equation (2). The first iteration fit \( \ln \left( M_1 / T \right) \) to \( T \) using a linear-regression algorithm. The slope \( \alpha m^* / H \) of this fit gives the first estimate of the mass, which is then used in the second iteration, where \( \ln \left( [M_1 \exp (-2 \alpha m^* T / H)] / T \right) \) is fitted to \( T \) by linear regression. This process is continued until the slope of the line changes by less than some test amount.

The cyclotron effective mass is an orbital average of the inverse Fermi velocity \( v^{-1} \) around an extremal orbit on the Fermi surface

\[ m^* = \frac{\hbar}{2\pi} \int \frac{dk}{v_{\parallel}}, \quad (3) \]

with \( v_{\parallel} \) the component perpendicular to the applied magnetic field. Integration of \( v^{-1} \) over the entire Fermi surface gives the electronic density of states at the Fermi level

\[ N_F = \frac{1}{4\pi} \int_{PS} \frac{ds}{\hbar v}, \quad (4) \]

which is proportional to \( \gamma \), the contribution of the itinerant electrons to the LTSH.

The dHvA effect was measured by a low-frequency large-amplitude field-modulation technique. Samples of typical dimensions \( 0.8 \times 0.8 \times 3 \ mm^3 \) and with its long axis parallel to [001] were selected out of a batch of Al flux-grown single crystals and mounted inside a tight-fitting set of compensated pickup coils. For the angle-resolved study a sample with its long axis parallel to [110] was additionally cut by spark erosion. The angular dependence of the dHvA frequencies were studied at 0.3 K in a \(^3\)He cryostat, where the assembly of pickup coils and sample can be rotated in one plane over \( \pm 90^\circ \). The plane of rotation was determined by Laue X-ray diffraction. The field-dependent mass measurements were carried out in a tailor-made dilution refrigerator designed to operate in the 25 T polyhelix magnet of the Grenoble High Field Facility. The dilution refrigerator has an all-plastic mixing chamber to eliminate eddy-current heating. It permits a warming from 60 mK to room temperature, changing of samples, and cooling to 60 mK in about 5 h. For more experimental details see reference [3].

Fig. 1. - De Haas-van Alphen frequencies as a function of field orientation, for magnetic fields in the (100) and (110) planes. The labels on the curves refer to the position of the centers of the relevant orbit in the Brillouin zone. In the inset the Fermi surface, Brillouin-zone together with high symmetry points are shown.
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They are qualitatively similar to those of the isostructural compound LaB₆ [15], which has no f-electron. Therefore the Fermi surface of CeB₆ like LaB₆ consists of spheres centred at the X-points of the Brillouin zone and connected by necks in the [110] directions. We were not able to detect directly the small necks with our field-modulation technique but recently they were observed in ultrasonic measurements [16]. Further evidence for the necks is given by the sudden disappearance of the X-centred and the formation of the R- and T-centred orbit for field angles close to [111]. The Fermi surface topology of CeB₆ does not seem to be affected much by the extra f-electron, except that the areas of the X-centred extremal orbits are 10 % larger than in LaB₆. The f-electron is therefore localized and sits below the Fermi energy. Strong evidence for this interpretation is also found from the band structure calculations by Norman and Min [17]. They performed band structure calculations within the local density approximation to the density functional theory using several approximations. First they treated the f-electrons as band states and next as atomic states, where the f-electrons are not frozen, yet are not allowed to hybridize with the conduction states. Only for the second approach a satisfactory agreement with the experimental dHvA frequency branches is found. CeB₆ is therefore a heavy-electron system with local f-electrons, different to UP₃ [4] where the f-electrons form a band which intersects the Fermi energy.

The cyclotron effective mass of orbits on the main part (belly) of the Fermi surface has been measured for a number of field orientations. The measurements were done for magnetic fields pointing 0, 15, 30 and 45 degrees from the [001] direction in the (100) plane, and for fields 0, 30, and 90 degrees from the [001] direction in the (110) plane. The masses were determined by fitting the temperature dependence of the amplitudes in the Fourier spectrum to equation (2). The Fourier spectrum was taken over a fixed field range of 22 T to 18.6 T and the temperature was varied between 0.35 K and 0.9 K. The effective masses were found to be within the experimental accuracy proportional to the orbit size, indicating an isotropic Fermi velocity over the belly part of the Fermi surface.

The cyclotron effective mass measurements as a function of field were performed for the field parallel to [001]. In this field direction and with the present technique one strong frequency \( F = 8 \, 680 \, \text{T} \) is observed. This frequency is constant with field and temperature to a precision of 0.5 % showing that the number of quasiparticles is field and temperature independent. The electron mass was determined at eight different fixed field values between 12.9 and 21.4 T and the dHvA amplitudes were measured in the temperature range 60 mK to 0.8 K [3]. The resulting masses as a function of field are plotted in figure 2 together with the mass measured in pulsed fields at 30 T by van Deursen et al. [14]. A large suppression of the cyclotron effective mass with the field is observed. Even in high fields the masses are very high compared to \( m^* = 0.61 \, m_e \) for the equivalent orbit in LaB₆ [15].

In order to compare the observed cyclotron effective mass to the electronic specific heat \( \gamma \) one has to make according to equations (3) and (4) an integration over the entire Fermi surface. We cannot apply this scheme rigidly yet since we have only measured the mass over the belly part of the Fermi surface. However the necks are relatively small so that, although they may have a very different mass enhancement, the weight of these orbits is small, when calculating the total electronic specific heat. Thus, we can find \( \gamma \) from \( m^* \) if we ignore the small differences between the Fermi surfaces of CeB₆ and LaB₆ and assume a nearly uniform enhancement of the cyclotron masses over the Fermi surface. In figure 2 the high-field LTSH data of Müller et al. [8] and the cyclotron masses are adjusted in this way. In view of the approximations made in converting \( m^* \) to a value for \( \gamma \) the agreement is good. The field dependence is reproduced in both properties. The difference in magnitude of about 30 % may not be significant. We will investigate this point further by extending our search for the neck frequencies and measurement of its mass. If the discrepancy is genuine it will be much more obvious at lower fields, closer to the transition at 2 T. This is under investigations. It should be emphasized that where the dHvA effect
probes only the itinerant electrons and is insensitive to local effects, the LTSH may also show contributions from the entropy of the local f moments. Therefore deviations between the LTSH and dHvA masses may show up at lower fields where the phase transition is observed in the specific heat $\gamma$ [7].

The common feature between Kondo-lattice, heavy-electron, and intermediate-valence compounds is the hybridization of the normally local f-electrons with the conduction band of s, p, and d-electrons. If the hybridization is strong the f-electrons form a narrow band which is situated at the Fermi energy. This situation is encountered in the intermediate-valence compound CeSn$_3$ [18]. Band-structure calculations predict Fermi-surface geometries close to those observed in dHvA experiments if the f-electrons are treated as itinerant, but in strong disagreement with observation if the f-electron is treated as part of the ion core. On the other hand, if the hybridization is very weak the f-electrons will be local as, e.g., in CeSb, which behaves as an ordinary local-moment rare-earth system [19]. Heavy-electron materials are found for hybridization strength in between these two extremes. In CeCu$_6$ and UPt$_3$ only a very small local-moment order is observed at low temperature, suggesting the formation of a hybridized f-conduction band which quenches the local moment. Indeed, band-structure calculations bear out this expectation, predicting Fermi-surface geometries remarkably close to those observed in UPt$_3$ [4] if the f-electron is treated itinerant.

CeB$_6$ on the other hand, has a local f-electron which sits below the Fermi energy, as discussed above. Nevertheless, there is significant hybridization as shown by the Kondo-type behaviour of the resistivity and the large electron mass. Thus CeB$_6$ represents a different limit of heavy-electron behaviour from UPt$_3$. In CeB$_6$ the hybridization is not strong enough to destroy the local moment but it is strong enough to allow many-body interactions which raise the electron mass by nearly two orders of magnitude. We find that these interactions are strongly suppressed by a magnetic field, whereas the number of conduction electrons and the occupation of states in the wave-vector space remain unchanged. We note that in CeB$_6$ the characteristic temperatures are all small: $T_{kondo} = 1 - 2$ K, $T_N = 2.4$ K, and comparable to the Zeeman energy $\mu_B H$. We further show that the field dependence of the electron masses is consistent with recent high-field LTSH measurements. This result stands in contrast to CeCu$_6$ where the LTSH is suppressed by a factor 2 to 3 in high fields but in dHvA experiments a search for a field dependence in the electron mass did not show any corresponding effect [2].

Acknowledgments

We would like to thank J. Flouquet and P. Wyder for their stimulating support and M. R. Norman for helpful discussions. The assistance of M. Caussignac in development and maintenance of the dilution refrigerator is gratefully acknowledged. The work was partially supported by the US-DOE-BES-Materials Sciences under contract No. W-31-109-ENG-38.


