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Citation for published version (APA):
Joss, W., Ruitenbeek, van, J. M., Crabtree, G. W., Tholence, J. L., Deursen, van, A. P. J., & Fisk, Z. (1987). Observation of the magnetic field dependence of the cyclotron mass in the Kondo lattice cerium boride (CeB6). *Physical Review Letters*, *59*(14), 1609-1612. https://doi.org/10.1103/PhysRevLett.59.1609

DOI:
10.1103/PhysRevLett.59.1609

Document status and date:
Published: 01/01/1987

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Observation of the Magnetic Field Dependence of the Cyclotron Mass in the Kondo Lattice CeB_6

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(Received 26 May 1987)

Low-temperature, high-field de Haas–van Alphen measurements are presented which show that the conduction-electron mass in the Kondo lattice CeB_6 decreases strongly with field. This field dependence is consistent with recent specific-heat results. The geometry of the Fermi surface does not depend on field. Thus we observe a reduction in the many-body enhancement of the electronic density of states at the Fermi energy which is described by a change of the itinerant-electron mass alone; the number of particles and the occupation of states in **k** space remain unchanged. We argue that CeB_6 represents a different limit of heavy-fermion behavior as compared to UPt_3.

PACS numbers: 71.25.Jd, 71.28.+d, 75.20.Hr

The unusual behavior of heavy-fermion metals is a major unsolved puzzle in metals physics.[1] These materials are well known for their large, nonlinear electronic specific heats and their large temperature-independent magnetic susceptibilities at low temperatures. In addition some of the compounds show anomalous superconducting and magnetically ordered states. The main characteristic, the 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 electrons are very heavy or, in other words, very slow. Recently de Haas–van Alphen (dHvA) experiments have directly measured the electron mass at the Fermi surface in UPt_3[2] and $CeCu_6$,[3] and have shown dramatically that this picture is correct, i.e., that the conduction electrons are very heavy and that the masses can, according to estimates, potentially explain the large values of the LTSH. A similar conclusion was reached earlier for the mixed-valent material $CeSn_3$.[4] The experiments further demonstrated that the conduction-electron scattering time is exceptionally large but the mean free path is like that of ordinary metals (> 1000 Å) as a result of the reduced velocity.

Here we present dHvA experiments in the Kondo-lattice compound CeB_6 and show that the mass (or velocity) of the conduction electrons in this metal is strongly reduced (increased) in high magnetic fields. No such field dependence has yet been observed in UPt_3, $CeCu_6$, and $CeSn_3$. We find that the Fermi surface is not affected by the field. Thus we show that there is a reduction of the density of electronic states in high fields which is solely due to a reduction of the electron mass; the number of conduction electrons and the occupation of the states in wave-vector space remain unchanged. These results represent important information for a description of these systems in terms of coherent Kondo-lattice models. We further show that the obtained electron masses are consistent with recent measurements of the LTSH.

CeB_6 is well studied for its pronounced Kondo-lattice properties.[5] It is unlike $CeSn_3$, $CeCu_6$, and UPt_3 in that it orders magnetically at low temperatures. Even in the magnetically ordered phase the linear coefficient in the LTSH, γ, is very large,[6] about 250 $mJ/mole \cdot K^2$, which is to be compared to $\gamma \simeq 2.6$ $mJ/mole \cdot K^2$ for the reference compound[7] LaB_6 (where the only difference is the absence of the f electron). In CeB_6, like in $CeCu_6$, γ is strongly field dependent.[6] Surprisingly, however, in the latter compound no field dependence of the electron mass has been observed in the dHvA experiments.[3]

CeB_6 was first studied with the dHvA effect by van Deursen *et al.*[8] The main features of its Fermi-surface geometry are qualitatively similar to those of LaB_6,[8,9] lending support to the view that the f electron is largely local in the Ce compound. Preliminary band-structure calculations[10] confirm this picture showing that a local rather than an itinerant f electron is required to reproduce the observed Fermi-surface geometry. The electron mass given in Ref. 8 for CeB_6 is $m^* = (6 \pm 2)m_e$, where

m_e is the free-electron mass. This is an order of magnitude larger than $m^* = 0.61 m_e$ for the corresponding orbit in LaB_6 but still not sufficient to explain the 2 orders of magnitude difference in the LTSH between the two compounds. Here it will be shown that this discrepancy is naturally explained by the field dependence of the electron mass.

The dHvA effect measures the oscillatory component of the susceptibility[11] and gives a direct measure of the Fermi-surface geometry and the cyclotron effective mass m^*. This mass gives a cyclotron-orbit average of the inverse Fermi velocity v_F. Integration of $1/v_F$ over the Fermi surface gives the contribution of the *itinerant* electrons to the specific heat. The large electron mass for heavy-fermion metals ordinarily prevents detailed investigation of their Fermi-surface properties because strong thermal damping of the dHvA oscillations by a factor which is exponential in the cyclotron effective mass attenuates the signal. In order to see the high-mass oscillations, the combination of a large magnetic field and low temperatures is required. Measurements of the field dependence of the mass require in addition a magnetic field which is variable over a large range and a detection system capable of measuring the signals over a finite temperature range. In order to obtain these conditions, our experiments were carried out in a special 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. A low-frequency, large-amplitude modulation technique and phase-sensitive detection at the second harmonic of the modulation frequency were used to measure the signals. The sample, of typical dimensions $0.8 \times 0.8 \times 3$ mm^3 and with its long axis parallel to [100], was selected out of a batch of Al-flux-grown single crystals and mounted parallel to the field inside a tight-fitting set of compensated pickup coils. The dHvA frequencies were calibrated by comparison to those of a well-known reference sample in a second set of pickup coils mounted above the first set in the mixing chamber. In this experiment the 2.57×10^4-T frequency of a Pt sample along [111] was used as reference.[12]

The temperature inside the mixing chamber was set by the application of a constant heating power and was measured with a Speer resistor which was calibrated outside the field. Corrections for the magnetoresistance are small for the lowest temperatures, only 2 mK at 60 mK and 22 T. At higher temperatures magnetoresistance corrections become more important but can be made to an accuracy of about 5 mK at 0.5 K and 10 mK at 0.7 K. The errors in temperature at high T do not seriously affect the mass measurement since the accuracy at higher temperature is limited by the signal-to-noise ratio of the dHvA oscillations.

The sample dimensions were chosen small enough to avoid eddy-current heating in the sample due to the applied modulation field. For the dHvA frequency of 8680 T in CeB_6 and a field of 22 T the optimal modulation amplitude is 0.028 T. The modulation frequency was 19 Hz. In order to verify that no important sample heating occurred the dHvA oscillation amplitude was measured as a function of the modulation amplitude at a temperature of 86 mK. The correct second-Bessel-function dependence was reproduced and no deviations were found even for 0.05-T modulation, which corresponds to about 4 times the maximum heating power in the sample during measurement.

All experiments were performed for the field parallel to [100]. In this field orientation there is only one frequency of $f = 8680$ T corresponding to an orbit area of 36% of the cross section of the Brillouin zone. To determine this frequency corrections for the internal magnetization of the sample were made and these amount at maximum to 2%. This frequency showed no indication of any beat structure and was constant with changes in field and temperature to within an accuracy of 0.5%. Thus, despite the large uniform magnetic moment of about $1 \mu_B$/Ce which is induced in high fields, there is no indication of spin splitting of the Fermi surface. In view of the highly nonlinear magnetization, the absence of any exchange splitting other than Zeeman splitting[13] is a surprising result.

The Fermi surface of[9] LaB_6 can be visualized as spheres centered on the X points of the Brillouin zone and connected by necks in the [110] directions. Although the Fermi surfaces of LaB_6 and CeB_6 are quite similar, there are a number of differences. In particular all the dHvA frequencies related to the necks between the spheres are not observed in the case of CeB_6. Also the volume of the spheres in CeB_6 seems to be larger by

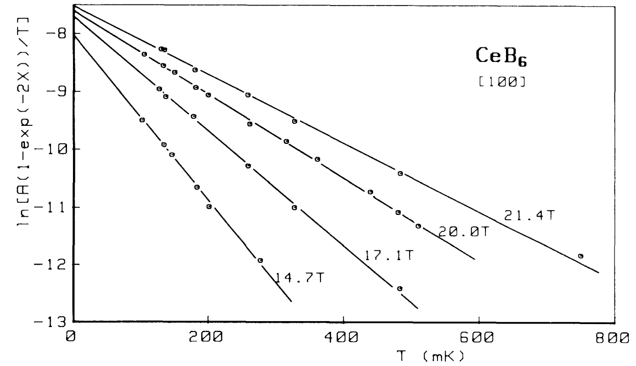

FIG. 1. Temperature dependence of the dHvA amplitudes for representative field values. The slope $X = 14.7 m^* T/B$ of the fitted lines determines the cyclotron effective mass. The value of X for each field is found by iteration. The resulting effective masses are shown as a function of field in Fig. 2.

about 10%. It is most likely that these two points can be explained by one's taking into account the nonspherical charge distribution of the localized *f* electron in close analogy to CeSb.[14]

The effective mass for the electron orbit was determined in the usual way[11] from the temperature dependence of the dHvA amplitude shown in Fig. 1. This procedure was repeated for different values of the magnetic field and the resulting effective mass as a function of the field is plotted in Fig. 2. At lower fields the accuracy the accuracy of the mass measurement is limited by the reduced signal-to-noise ratio. In extracting the effective mass of the electrons from the experimental data we have assumed that the Lifshitz-Kosevich theory remains valid even for a material with strong electron-electron correlations. There have been a few attempts to improve the Lifshitz-Kosevich theory[16] but so far no important corrections to the theory have been proposed. The fact that we find a good fit of the theory to the temperature and field dependence of the amplitude, as is the case for $CeCu_6$[3] and UPt_3,[2] gives us confidence in the validity of the procedure.

In order to compare the observed effective mass to the electronic specific heat one has to make an integration of m^* over the Fermi surface. Since the full angular dependence of m^* has not been measured we cannot follow this procedure. However, we can find γ from m^* if we ignore the small differences between the Fermi surfaces of CeB_6 and LaB_6 and assume a nearly uniform enhancement of the cyclotron masses over the Fermi surface. Justification for this assumption is found in the dHvA data on UPt_3[2] and $CeCu_6$,[3] where a fairly uniform enhancement over the band mass is observed. In the inset of Fig. 2 the γ values thus calculated from the measured cyclotron effective mass are compared to the specific-heat results of Ref. 6. It is found that the large low-temperature specific heat is in semiquantitative agreement with the cyclotron effective mass of the itinerant electrons. Thus, the apparent discrepancy between the LTSH and the high-field dHvA measurements[10] is largely resolved. A full quantitative comparison requires specific-heat data on the same single crystal (because of an apparent sample dependence of γ) in the same fields as used here. Such experiments are under way. It should be emphasized that where the dHvA effect measures only the conduction electrons, the LTSH may also show contributions from the entropy of the local *f* moments. Therefore deviations between the LTSH and the dHvA masses may still show up, especially at lower fields where the phase transition is observed in the specific heat γ (inset of Fig. 2).

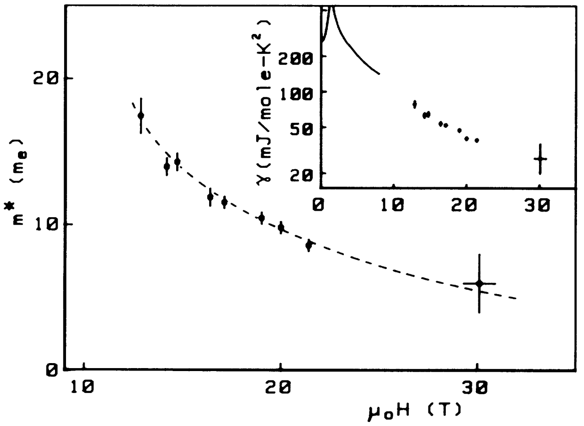

FIG. 2. Field dependence of the effective mass in CeB_6 for the 8680-T orbit with the field along [100]. The results are obtained as described in the caption of Fig. 1. The value of m^* from Ref. 8, measured in pulsed fields above 30 T, is also included. Inset: The field dependence of the electronic specific heat γ, on semilogarithmic scale. The curve at low fields represents the specific heat as measured in Ref. 6. The divergence at 2 T is related to the phase transition going from antiferromagnetic to quadrupolar order (Ref. 15). The points are calculated from the measured cyclotron masses on the assumption that the mass enhancement going from LaB_6 to CeB_6 is roughly isotropic over the Fermi surface and scaling to the mass $m^* = 0.61$ for LaB_6.

The common feature between Kondo-lattice, heavy-fermion, 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 $CeSn_3$.[4] Band-structure calculations predict Fermi-surface geometries close to those observed in dHvA experiments if the *f* electron is 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.[14] Heavy-fermion materials are found for hybridization strength in between these two extremes. In $CeCu_6$ and UPt_3 no 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[2] UPt_3 if the *f* electron is treated as 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 behavior of the resistivity and the large electron mass. Thus CeB_6 represents a different limit of heavy-fermion behavior from UPt_3. In CeB_6 hybridization is not strong enough to destroy the local moment but it is strong enough to allow many-body effects which raise the electron mass by nearly 2 orders of magnitude.

This enormous mass enhancement would be expected in a Kondo description of the low-temperature properties in CeB_6. In an impurity Kondo system at low temperatures a so-called Abrikosov-Suhl resonance develops in the density of electronic states near the Fermi energy (see Lee *et al.*[1]). Also for the Kondo lattice such a resonance is expected though it may have a more complicated structure due to coherence between the lattice sites. Here the situation is complicated by the onset of magnetic transitions below 3.2 K. Apparently the local-moment order does not destroy the resonance. In fact, a full description of this resonance may have to take in account the interplay between the Kondo-type interactions and local-moment intersite (Ruderman-Kittel-Kasuya-Yosida) interactions. However, the effect of magnetic ordering on the electron mass is probably limited to low fields close to the transition at 1.5 K: There the magnetic susceptibility is large, flattening to an almost constant value[5] at higher fields. The strong suppression of the electron mass as observed here in higher fields is exactly what one would expect in a Kondo system when the Kondo temperature is smaller than the Zeeman energy $\mu_{\mathrm{B}}H$. The effect is so clearly observed here because the Kondo temperature of CeB_6 is very low, only a few degrees kelvin.[17]

We would like to thank J. Flouquet and P. Wyder for their stimulating support and P. Stamp for helpful discussions. The assistance of M. Caussignac in the development and maintenance of the dilution refrigerator is gratefully acknowledged. This work was partially supported by the U. S. Department of Energy, Basic Energy Sciences—Materials Sciences under Contract No. Δ-31-109-ENG-38.

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