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Title
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Permalink
https://escholarship.org/uc/item/50x9353s

Journal
Journal of Applied Physics, 63(8)

ISSN
0021-8979

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Publication Date
1988-04-15

DOI
10.1063/1.340595

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Peer reviewed

Magnetic field dependence of the cyclotron effective mass in the Kondo lattice CeB$_6$

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We report the first observation of a field-dependent mass in a hybridizing f-electron system. CeB$_6$ is an ordered moment heavy fermion system with an electronic specific heat coefficient γ of order 225–300 mJ/mole K^2. 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 $18m_e$ to $8m_e$ as the field increases from 12 to 22 T. The observed Fermi surface is very similar to that of LaB$_6$, indicating that the f-electrons are largely local rather than itinerant in CeB$_6$, a picture confirmed by bandstructure calculations. 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. Our results are compared with Fermi surface observations in other heavy fermion systems.

The heavy fermion materials form a major challenge to our understanding of the physics of metals.[1] They are unusual in almost every physical property but the most characteristic one is the high value of the low-temperature specific heat (LTSH). Many of these systems show properties resembling the impurity Kondo effect or intermediate valence behavior and the physics of heavy fermions is closely related to the physics of both the Kondo effect and valence fluctuations. Recently de Haas–van Alphen (dHvA) experiments have provided conclusive microscopic information in a number of these compounds: the very heavy fermion system CeCu$_6$ (Ref. 2) ($\gamma \simeq 1500$ mJ/mole K^2), the heavy fermion superconductor UPt$_3$ (Ref. 3) ($\gamma = 420$ mJ/mole K^2), the intermediate valence compound CeSn$_3$ (Ref. 4) ($\gamma = 17$ mJ/mole K^2) and the Kondo lattice CeB$_6$ (Ref. 5) ($\gamma \simeq 260$ mJ/mole K^2). The information obtained in these experiments can be summarized as follows.

(1) At low temperatures the conduction electrons form coherent states and the mean free path is several hundreds of nanometers long.

(2) For UPt$_3$ and CeSn$_3$ the dHvA frequencies are consistent with bandstructure calculations treating the f electrons as itinerant. The f electrons form a band at the Fermi energy. (CeB$_6$ will be discussed below.)

(3) The electron mass is very high and one or two orders of magnitude larger than found by conventional bandstructure calculations. Estimates show that the masses found in dHvA experiments explain the large values of the LTSH (with the possible exception of CeCu$_6$).

Here we discuss the results for CeB$_6$ and we show in addition that the electron mass is strongly suppressed in high magnetic fields (see also Ref. 5). This is the first observation of a field-dependent electron mass in this type of compound. It shows that the many body interactions which make the electrons heavy (or in other words slow) are strongly reduced in high fields and the electrons become light again (speed up). At the same time it is observed that the size of the Fermi surface, as measured by the dHvA frequency, is field independent. Thus the *number* of particles is conserved.

CeB$_6$ is one of the most typical Kondo lattice systems and has therefore received a lot of attention, see, e.g., Kasuya *et al.*[6] The Kondo temperature is very low, only 1–2 K.[7] It is interesting to compare the experimental Fermi surface information to that of LaB$_6$ and to bandstructure calculations by Norman and Min.[8] The results indicate that the f electron of CeB$_6$ is localized and can be treated as part of the ion core. This is in sharp contrast to the situation for UPt$_3$ and CeSn$_3$. We conclude that CeB$_6$ belongs to a different class of heavy fermion compounds: the hybridization is apparently not strong enough to bring the f electrons to the Fermi level. Yet an electron mass enhancement of roughly a factor 100 is observed.

The dHvA effect measures the oscillatory magnetization in high magnetic fields which arises due to the quantiza-

tion of the electron motion on Landau orbits.[9] The frequency observed when changing the applied field is directly proportional to the cross-sectional area of the Fermi surface. The field and temperature dependence of the oscillation amplitude allow a determination of the mean free path with respect to the effective mass for this orbit. The dHvA effect is usually only observed in high magnetic fields and at low temperatures. These restrictions are the more severe as the mean free path is shorter and the effective mass is larger. In order to obtain these conditions our experiments were carried out in a special dilution refrigerator designed to operate in the 25T polyhelix magnet of the Grenoble high magnetic field facility (SNCI/MPI). 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 angular dependence of the dHvA frequencies[10] closely resembles that for the isostructural compound LaB$_6$.[11] The latter is an ordinary metal with a LTSH coefficient[12] $\gamma = 2.6$ mJ/mole K^2. Its Fermi surface consists of spheres centered at the X points of the Brillouin zone which are connected by small necks and contain one electron per cell per spin.[11] In addition, CeB$_6$ has one f electron. However, the Fermi surface does not seem to be affected very strongly by this extra electron and we conclude that the f electron is localized and sits below the Fermi energy. Strong evidence for this interpretation is also found from the band-structure calculations by Norman and Min.[8] They performed calculations using several approximations first treating the f electron on equal footing with the other valence electrons and next treating it as part of the ion core allowing no hybridization with the conduction electrons. Only for the second approach a satisfactory agreement with the experimental Fermi surface information is found, although an important discrepancy of 10% in size remains to be explained. This conclusion is opposite to the results for UPt$_3$ (Ref. 3) and CeSn$_3$ (Ref. 4) where the f electrons form a band which clearly intersects the Fermi level.

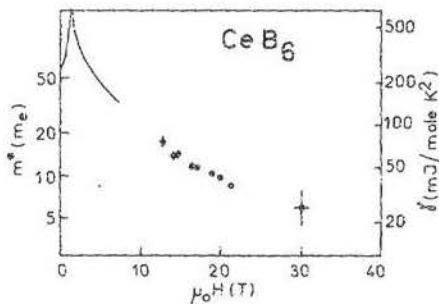It is of importance to note that we do not find a double frequency, not even a beating, as would be expected for a spin splitting of the Fermi surface. In the strong fields used here the induced magnetic moment is close to $1\mu_B$/Ce. As a consequence the exchange interaction between the conduction electrons and the local magnetic moments of the f electrons must be very small.

The effective mass study was carried out for the field along [100]. In this field direction and with the present technique one strong frequency is observed of $F = 8680$ T. This frequency is constant with field and temperature to a precision of 0.5% showing that the number of particles is field and temperature independent. The electron mass was determined at different field values by fitting the temperature dependence to the usual Liftshitz–Kosevich theory for the dHvA effect.[9] The resulting masses are plotted as a function of field in Fig. 1. The mass measured in fields above 30 T by van Deursen et al.[10] is also included in the figure. A substantial suppression of the electron mass with field is observed. Also, even in high fields, the masses are very high compared to $m^* = 0.61 m_e$ for the equivalent orbit in LaB$_6$. In order to allow a comparison to the specific heat the data in Fig. 1 are presented on a semi logarithmic scale. The LTSH of CeB$_6$ has been measured to very low temperatures and in fields up to 8 T by Marcenat and by Bredl.[13] There appears to be some sample dependence of the absolute values: the linear term γ in zero field ranges between 225 and 300 mJ/mole K^2. However, the general behavior is the same and it was found that γ initially increases with field toward the transition from the low-field antiferromagnetic phase to the high-field phase II. Then a strong decrease of γ with field is observed. The curve in Fig. 1 represents this field dependence of γ schematically.

The cyclotron mass is an integral of the inverse Fermi velocity $1/v_F$ over the cyclotron orbit. The LTSH coefficient γ, if conventional theory applies, is proportional to the density of electronic states at the Fermi level which in turn is an integral of $1/v_F$ over the entire Fermi surface. The zero field specific heat for CeB$_6$ $\gamma_{Ce} = 260$ mJ/mole K^2 is enhanced over the value of LaB$_6$ $\gamma_{La} = 2.6$ mJ/mole K^2. This enhancement corresponds to an enhancement of the electron mass and a reduction of the Fermi velocity. If we assume that the enhancement is nearly isotropic then we can relate the measured γ value for CeB$_6$ to the cyclotron mass for the present orbit via $m^*_{Ce} = (\gamma_{Ce}/\gamma_{La})m^*_{La}$. The mass for the corresponding orbit in LaB$_6$ is $m^*_{La} = 0.61 m_e$.[11] The relation above is used to adjust the scales in Fig. 1. It shows that the zero field effective mass for this orbit should be roughly $60 m_e$. We find from Fig. 1 that there is a fairly good agree-

FIG. 1. Field dependence of the effective mass in CeB$_6$ for the 8680 T orbit with the field along [100]. The data are presented on semi-logarithmic scale in order to allow comparison to the electronic specific heat γ. For the positioning of the scale of γ with respect to that for m^* see text. The curve at low fields represents the specific heat as measured in Ref. 13. The divergence at 2 T is related to the phase transition going from antiferromagnetic to quadrupolar order.
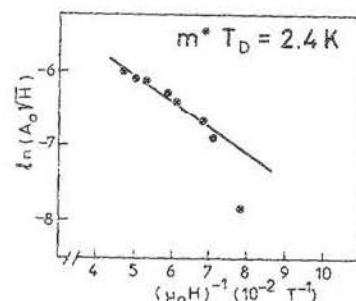

FIG. 2. Field dependence of the zero-temperature dHvA amplitude. After correction for the temperature dependent terms the field dependence of the dHvA amplitude is $(1/B)\exp(-am^*T_D/B)$ where $\alpha = 14.69$ T/K is a constant. The product m^*T_D, which in turn gives the mean free path, is found from the slope of this plot.

ment, though not yet quantitative, between γ and m^* and that these are both enhanced by the same amount.

The mean free path of the electrons on this orbit was determined by analysing the field dependence of the amplitude of the dHvA oscillations. First the temperature dependent factor in the amplitude was eliminated by linear extrapolation to $T = 0$. The field dependence of the resulting zero temperature amplitudes is given in Fig. 2. The straight line gives[14] $m^* T_D = 2.4$ K where T_D is the so-called Dingle temperature which is inversely proportional to the scattering time τ. Now, since m^* is field dependent, so are T_D and τ. A possibly more fundamental property is the mean free path l which is inversely proportional to the product $m^* T_D$. We find no evidence that this product is field dependent although this might be hard to distinguish in a limited field interval. (For a discussion of the 12.7 T point in Fig. 2 see below.) From the quoted value for $m^* T_D$ we calculate a mean free path $l = 0.30$ μm. The circumference of the real space orbit at 10 T is $2\pi r = 2.13$ μm. Thus, we find that the electrons form coherent states which extend over a very large number of unit cells.

Finally, a remarkable effect was observed on thermal cycling of the samples. In three different samples from two different batches the signal after the first cool down was roughly of the same amplitude. Thermal cycling reduced the signal amplitude drastically. After 2 or 3 cycles the signal in all three samples was below noise level. To our knowledge there is no crystalline phase transition below room temperature which would explain this phenomenon. The samples were carefully mounted in cotton wool and no glue or grease was used, in order to avoid stress due to differential thermal contraction on cooling. All data in Figs. 1 and 2 were taken without heating above 1 K, except for the lowest field point. This was taken after one room temperature cycle and $m^* T_D$ for this point appears significantly higher. The field dependence of the cyclotron mass was reproduced in one other sample and found to be consistent with the results presented here.

The most salient feature of the results presented here is the direct observation of a strong suppression of the heavy mass in CeB_6. In order to describe this effect one could start from an impurity Kondo model or, alternatively, from a spin fluctuation model. However, the situation here is complicated by the fact that Kondo effect and magnetic order play an important role and the characteristic temperatures are all small: $T_K = 1-2$ K, $T_N = 2.4$ K. It is due to the smallness of these energy scales that the effect is so clearly observed. In UPt_3 the characteristic temperatures are an order of magnitude higher and up to 15 T no field effects are observed.[3] For $CeCu_6$ the problem is even more interesting: the electronic specific heat is suppressed by a factor 2 or 3 in high fields but in dHvA experiments a search for a field dependence in the electron mass did not show any corresponding effect.

Further, it is observed that the f electron is local and has only minor effects on the Fermi surface. These small effects, however, deserve our full attention and should be studied in more detail. More experiments are under way.

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