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N. Harrison
National High Magnetic Field Laboratory Los Almos

R. G. Goodrich
Louisiana State University

A. Teklu
Louisiana State University

L. Balicas
National High Magnetic Field Laboratory

J. S. Brooks
National High Magnetic Field Laboratory

See next page for additional authors

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The Kondo effect to heavy fermions studied using the de Haas–van Alphen effect

N. Harrison[a], R.G. Goodrich[b], A. Teklu[b], L. Balicas[c], J.S. Brooks[c], D. Young[c], Z. Fisk[c,*], J.C. Cooley[d], J.L. Smith[d]

[a]National High Magnetic Field Laboratory, LANL MSE-536, Los Alamos, NM 87545, USA
[b]Department of Physics and Astronomy, Louisiana State University, Baton Rouge, LA 70803, USA
[c]National High Magnetic Field Laboratory, Florida State University, 1800 E. Paul Dirac Drive, Tallahassee, FL 32310, USA
[d]Los Alamos National Laboratory, Los Alamos, NM 87545, USA

Abstract

In this paper, we briefly review some recent de Haas–van Alphen experiments on two contrasting types of heavy fermion/Kondo systems for varying concentrations x of the impurity ion, Ce or U. In both systems the Fermi surface is observed to vary continuously with x: in $Ce_xLa_{1-x}B_6$, the Ce ions behave as Kondo impurities while in $U_xTh_{1-x}Be_{13}$ they do not. In both systems, the f electrons are also found to contribute only partially to the Fermi surface volume, therefore raising questions concerning the applicability of Fermi liquid theory. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Heavy fermions; Kondo alloys; UBe_{13}; CeB_6; De Haas–van Aphen effect

1. Introduction

The de Haas–van Alphen (dHvA) effect can provide essential information on the electronic structure of heavy fermion systems. To begin with, it tells us when a material appears to behave like a Fermi liquid in the presence of a magnetic field, and can therefore potentially yield important Fermi liquid parameters, such as the electron g-factor or the quasiparticle effective mass enhancement (for reviews see Refs. [1,2]). When angle-resolved measurements are made, in some cases it becomes possible to discern whether a conventional band-structure-like picture appropriately describes the Fermi surface topology or whether a model based on the Anderson lattice Hamiltonian needs to be considered (see for examples Refs. [3–5]).

More generally, the actual electronic structure is expected to lie somewhere between these two extremes, although there exists no well-developed technique for performing electronic structure calculations within this intermediate regime. Interpretation of experimental results therefore relies upon assumptions that are often difficult to justify. One of the more common assumptions made, is that the effective mass measured by means of dHvA experiments is directly proportional the density of states obtained from specific heat measurements [1,2]. However, an exact agreement between these two quantities in heavy fermion systems is seldom

* Corresponding author. Fax: + 904-644-5038.
E-mail address: fisk@magnet.fsu.edu (Z. Fisk).

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obtained. Another matter that has been largely ignored, beyond the scope of dHvA measurements on pure heavy fermion systems is the question concerning the applicability of Luttinger's theorem and the extent to which the f electrons contribute to the Fermi surface volume [1,2].

In this paper, we review recent results on the $Ce_xLa_{1-x}B_6$ [6,7] and $U_xTh_{1-x}Be_{13}$ [8,9] intermetallic series that, while contrasting in many ways, have certain features in common. CeB_6 (i.e. $x = 1$) is a relatively weakly correlated antiferromagnetic 4f heavy fermion system that exhibits clear dHvA oscillations [10] and undergoes metamagnetic transition at $B \sim 2\,T$ [11]. UBe_{13}, on the other hand, is a very strongly correlated 5f electron superconductor [12] that does not appear to exhibit dHvA [8] nor does it undergo a metamagnetic transition at any field [13]. However, in both $Ce_xLa_{1-x}B_6$ and $U_xTh_{1-x}Be_{13}$, the cyclotron effective masses appear not to agree with specific heat measurements, and in both systems, the f electrons appear to contribute to the Fermi surface volume in a non-integral manner.

2. $Ce_xLa_{1-x}B_6$

Concentrating initially on $Ce_xLa_{1-x}B_6$ [7], here, all of the dHvA experiments are performed at magnetic fields greatly exceeding the metamagnetic transition field of CeB_6 [6,7,10,14–16]. We find that it is because the spin up and spin down spin components of the dHvA effect behave differently that the cyclotron effective mass measurements do not agree with the coefficient γ of the electronic specific heat [6]. This is evidenced by the observation that the dHvA signal at high magnetic fields originates from only one of these spin channels (presumably that with the lightest effective mass) [6]. The Fourier transform of the dHvA oscillations in CeB_6 in Fig. 1, for which the amplitude decays in an ideal exponential manner with increasing harmonic index, is a direct consequence of there being no phase interference when only a single spin component is present. One of the spin components must therefore be strongly damped. So, while specific heat measurements are always inclusive of both up and down spin states, this is certainly not the case with the dHvA effect [6].

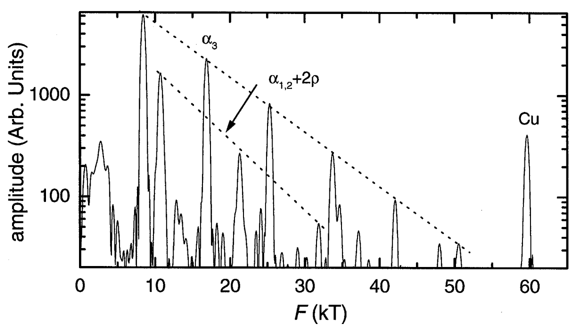

Fig. 1. A logarithmic plot of a Fourier transform of dHvA oscillations in CeB_6 over the field interval between 30 and 60 T at $\sim$ 500 mK. Two major frequencies are present.

It has, of course, already been anticipated that only one spin channel might dominate the dHvA signal [17]. It was only recently, however, that this was actually demonstrated by experiment [6]. According to the mean field theory of Wasserman et al. [17], the spin down component is thought to be the lightest, since, the hybridized f electron/conduction electron resonance feature in the density of states for this spin channel, is elevated to energies greatly exceeding the chemical potential by the Zeeman effect. Fits of this theory to the magnetic field dependence of the effective mass in CeB_6 had appeared to yield reasonable agreement [17]. By looking only at the field dependence of the effective mass in the pure heavy fermion compound CeB_6, however, there is no way of verifying which spin channel is dominant. Because one spin component is absent, it is also impossible to determine whether the different spins have different Fermi surface topologies, or, more importantly, different particle densities [6].

It is here that experiments at intermediate concentrations x, of Ce in $Ce_xLa_{1-x}B_6$, provide new vital information [7]. The effective masses and Fermi surface topology are found to vary continuously throughout the entire series [7], implying (i) that the Ce and La are thoroughly mixed and (ii) that a Fermi liquid description appears still to be appropriate in a randomly mixed alloy. The x-dependence of the effective mass of the principal α_3 dHvA frequency is shown in Fig. 2.

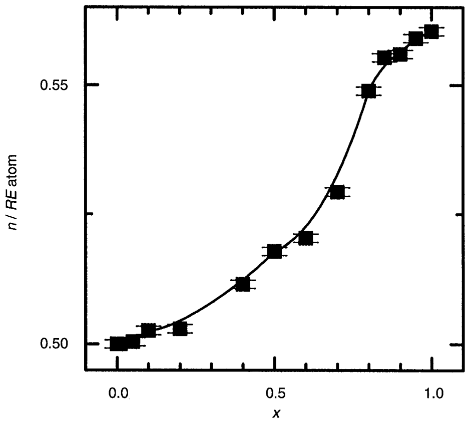

Fig. 2. The x-dependence of the effective mass in the $Ce_xLa_{1-x}B_6$ series at 40 T (a) as well as (b) the field dependence of the effective mass for various concentrations.

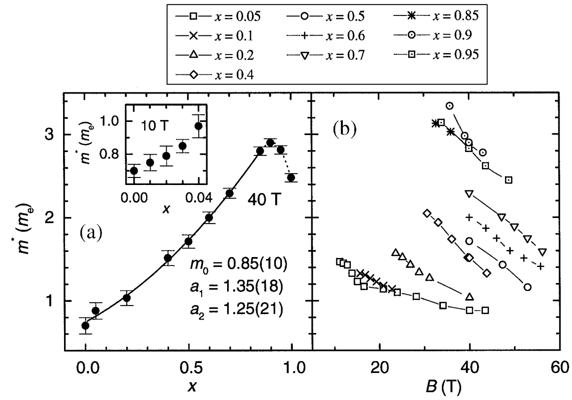

Fig. 3. The x-dependence of the electron density per spin in $Ce_xLa_{1-x}B_6$, determined by using the elliptical approximation for the Fermi surface.

Further information reveals itself at the low concentration end of the series, $x < 0.05$, since only here are both spin channels observed to be present. The two primary observations are (i) that it is the spin up channel that progressively dominates the dHvA signal as the concentration is increased [18], and (ii) that, in this particular system, the spin up and spin down Fermi surfaces appear to have identical cross-sectional areas [7]. The first of these observations (i) proves that the mean field theory of Wasserman et al. does not apply to this material [17,18]. These results are, however, consistent with the theory of Edwards and Green [19], which is applicable to heavy fermion systems in the limit where the Zeeman energy greatly exceeds the Kondo energy scale. The spin up component dominates the dHvA signal because, unlike the spin down component, it is relatively unaffected by spin fluctuation effects at high magnetic fields. For this reason, there is no observable increase in quasiparticle scattering across the alloy series for this spin component [7]. The second of these observations (ii) is difficult to explain in terms of the hybridized many body band picture, perhaps indicating the eventual limitation of this approach. This second observation does, however, enable us to precisely determine the total number of particles in the system, inclusive of both spins [7]. A plot of the particle number density for each spin is shown in Fig. 3. Rather than being integral, the Fermi surface volume accommodates ~ 1.1 electrons per unit cell per Ce ion, implying that the additional ~ 0.1 electrons (with reference to LaB_6) are 4f electrons. The only way that this observation can be interpreted is that only a fraction of the 4f electrons contribute to the Fermi surface volume, most likely to be that fraction that can be considered to be itinerant. The other ~ 0.9 4f electrons therefore appear to be localized. We will discuss the implications of this result in the concluding section of this paper.

There are no other heavy fermion systems for which it has been possible to perform a precise electron count. This has been due either to the fact that Fermi surface topology is too complicated, with possibly only a small fraction of it being observed, or, alternatively, to the fact that only one spin component of the dHvA signal can be observed. Again, there is no easy way to tell whether this is the case unless more than one harmonic is observed.

3. $U_xTh_{1-x}Be_{13}$

dHvA experiments have not been performed successfully on UBe_{13} [8], possibly because it has a non-Fermi liquid ground state [20–22]. A study of the dHvA effect within the $U_xTh_{1-x}Be_{13}$ series can still, however, provide useful information on how U interacts within the MBe_{13} lattice [9]. Th is

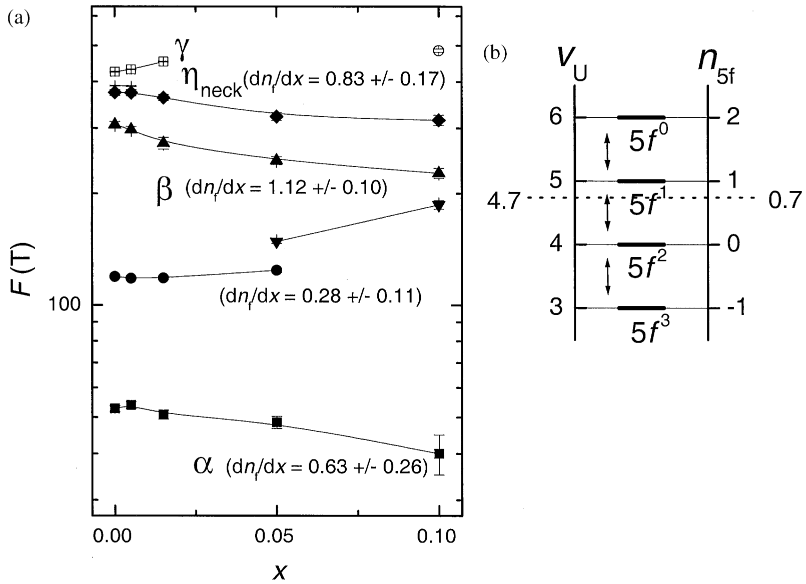

Fig. 4. (a) The x-dependence of several of the dHvA frequencies in $U_xTh_{1-x}Be_{13}$. (b) A schematic representation of the U valence (v_U), the extent to which the 5f electrons contribute to the Fermi surface volume (n_{5f}) and the electronic configuration.

the non-interacting actinide element that is closest to U in the periodic table, as is La considered to be the non-interacting lanthanide element closest to Ce.

Unlike the $Ce_xLa_{1-x}B_6$ series [7], however, in $U_xTh_{1-x}Be_{13}$, dHvA oscillations appear only to be observeable for $0 < x < 0.1$ [9]. The surprising result is that the dHvA oscillations die out with increasing x due to an increase in the quasiparticle scattering rate rather than the effective mass. The most likely physical reason for why U in $ThBe_{13}$ behaves differently from Ce in LaB_6, is that U is not a Kondo impurity at low concentrations, as was shown by Kim et al. [23]. Yet, the Fermi surface is still observed to transform gradually as a function of x, as depicted in Fig. 4. The gradual change in Fermi surface topology indicates that, like Ce, however, the valence of U is significantly different from that of Th. In fact, by monitoring the changes of the Fermi surface cross-sections for several orbits, we find that the valence of U in $U_xTh_{1-x}Be_{13}$ is ~ 4.7 compared to 4.0 for Th [9]. This would therefore appear to indicate that, as for 4f electrons in $Ce_xLa_{1-x}B_6$, only the itinerant fraction of 5f electrons in $U_xTh_{1-x}Be_{13}$ contribute to the Fermi surface volume.

The absence of any significant increase in the cyclotron effective mass with x appears to agree with the specific heat measurements, which indicate a weak but steady increase of γ with x [23]. Since γ is virtually independent of magnetic field in $U_xTh_{1-x}Be_{13}$, this can only imply that, unlike CeB_6, the various spin channels have virtually the same effective mass, with both of these contributing to the dHvA signal [6,7]. However, because no more than one harmonic of any of the dHvA frequencies in $U_xTh_{1-x}Be_{13}$ can be observed, there is no simple means to verify this conjecture directly.

The combined observation of only a marginal increase in γ with x, together with a considerable change in the Fermi surface topology, would appear to indicate that U enters the $ThBe_{13}$ lattice as a mixed valent impurity. This situation occurs whenever the highest occupied bare f electron level lies very close to the chemical potential, enabling it to mix directly with the conduction electron bands [24]. The question that therefore needs to be asked is whether the heavy fermion ground state in UBe_{13} is in any way related to the Kondo effect (as is the case in Ce-based heavy fermion systems such as CeB_6), and, if so, why does appreciable effective mass enhancement not take place until $x \to 1$ [23].

On the other hand, it was shown by Kim et al. [23] that the 5f electron Kondo effect in MBe_{13} systems occurs only when the lattice parameter is less than that of UBe_{13}. The likely explanation for this is that Kondo-like behaviour occurs only when the f electrons become more itinerant upon Kondo 'collapse', whereby the relative energy of the bare 5f electron levels sinks further below the chemical potential as the unit cell is reduced in size [9]. To fully test this hypothesis, it would be necessary to investigate whether the Kondo effect can be switched on in low concentration $U_xTh_{1-x}Be_{13}$ alloys under hysdrostatic pressure.

The configuration of the 5f electrons is another matter that is of relevance to the ground state of $U_xTh_{1-x}Be_{13}$ and UBe_{13}. While the J_z-spin state of the localized 5f electrons cannot be determined from dHvA measurements, the U valence of ~ 4.7 does imply that they exist of the $5f_2$ configuration prior to mixing, with the mixing then giving rise to $5f_2 \to 5f_1$ transitions. This could imply that the quadrupolar Kondo effect remains as one of the possible candidates for non-Fermi liquid-like behaviour in UBe_{13} [20–22].

4. Conclusions

While $Ce_xLa_{1-x}B_6$ and $U_xTh_{1-x}Be_{13}$ are systems that exhibit greatly contrasting physical properties, in both systems it is found that the f electrons contribute a non-integral amount to the electron density [7,9]. It is also found that the coefficient of the electronic contribution to the specific heat only matches the cyclotron effective mass obtained from dHvA measurements on consideration of the possibility of the spins having unequal masses [1,2,6,9,14–16].

The dHvA measurements on $Ce_xLa_{1-x}B_6$ indicate that the 4f electrons are predominantly localized in this system in strong magnetic fields. It is interesting to note that other heavy fermion systems, for example $CeRu_2Si_2$ [25], have been reported to transform via the metamagnetic transition from a system in which the 4f electrons are itinerant to one where they become localized at high magnetic fields. In $CeRu_2Si_2$, this was determined by comparisons of the measured Fermi surface with bandstructure calculations of $CeRu_2Si_2$ and $LaRu_2Si_2$, respectively. While it was not possible to perform a precise electron count in this case, the conjecture that the 4f electrons are predominantly localized at high magnetic fields is consistent with the observations made on $Ce_xLa_{1-x}B_6$ alloys.

In $U_xTh_{1-x}Be_{13}$, on the other hand, the question concerning the degree of itinerancy is more complicated, owing to the possible existence of several oxidation states [9]. The present dHvA measurements show that only ~ 1.3 5f electrons appear to remain localized [9].

According to some Fermi liquid descriptions, Luttinger's theorem implies that the degree to which the f electrons contribute to the Fermi surface volume is integral, irrespective of whether they exhibit localized or itinerant behaviour [26]. Thus, either the present experiments on $Ce_xLa_{1-x}B_6$ and $U_xTh_{1-x}Be_{13}$ imply that this conclusion is incorrect, or neither of these heavy fermion systems can be considered to be true Fermi liquids.

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