Development of the High-Field Heavy-Fermion Ground State in $Ce_xLa_{1-x}B_6$ Intermetallics

R. G. Goodrich,[1] N. Harrison,[2] A. Teklu,[1] D. Young,[3] and Z. Fisk[3]
[1]*Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803*
[2]*National High Magnetic Field Laboratory, Los Alamos National Laboratory, MS-E536, Los Alamos, New Mexico 87545*
[3]*National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32310*
(Received 15 July 1996)

de Haas–van Alphen measurements in $Ce_xLa_{1-x}B_6$ intermetallics reveal the existence of long-lived quasiparticles for all $0 < x < 1$. This is accompanied by the enhancement of the field-dependent effective mass, together with changes to the topology of the Fermi surface developing very early in the series, and with the effective mass eventually exhibiting a maximum near $x \sim 0.9$. One of the spin contributions to the signal is also observed to disappear at very low x, indicating a spin polarity-dependent scattering mechanism. [S0031-9007(99)09074-2]

PACS numbers: 71.18.+y, 71.27.+a

The de Haas–van Alphen (dHvA) effect has played a key role in the understanding of dense Ce- and U-based Kondo metals. It has shown quite definitively the existence of long-lived quasiparticles with heavily renormalized effective masses, and has shown that the topology of the Fermi surface (FS) is often modified by the presence of partially delocalized f electrons [1,2]. In Ce compounds, for example, the effect of the $4f$ electrons has been inferred by comparing the FS with that of the equivalent La compound in which there are no $4f$ electrons [1]. While compounds at the two opposite extremes containing either Ce or La have been the subject of many dHvA studies, the transitional regime, consisting of intermediate concentrations x of Ce substituted in place of La, has not been investigated with dHvA. The dilute limit is understood to conform to the single impurity model [3], but with the interactions becoming more complicated as their concentration is increased. This has to do largely with the effects of magnetic correlations between impurities [4,5], which are known to exist in a number of heavy-fermion (HF) alloys [6,7]. CeB_6 is one example of a compound in which heavy quasiparticles can coexist with antiferromagnetic ordering [8].

Given that our familiar understanding of the dHvA effect in metals predisposes a crystalline lattice that is nearly free from imperfections and impurities (especially magnetic impurities) [9], one expects alloys consisting of arbitrary impurity concentrations x, in which the magnetic Ce ions are disordered, to be inaccessible to studies by means of the dHvA effect. A study of the effects of Fe impurities in Au, the only well-documented study of the dHvA effect in a dilute Kondo alloy, has shown that the quasiparticles are rapidly dephased on incorporating only very low concentrations [10]. If the same arguments were to apply to alloys with varying concentrations of Ce, we would expect a dHvA investigation to be limited only to the situations where $x \ll 1$ or $x \equiv 1$.

In this paper, we report the results of performing dHvA experiments on alloys of $Ce_xLa_{1-x}B_6$ for $0 \leq x \leq 1$. The surprising result is that in $Ce_xLa_{1-x}B_6$ at high magnetic fields, the Fermi liquid (FL) is remarkably coherent throughout the entire series. As x is increased, both the FS topology and the quasiparticle effective masses are observed to transform gradually from that of pure LaB_6 [11] to that of pure CeB_6 [8]. In addition, the dHvA signal is found to originate from only a single spin FS sheet at very small values of x. The nonmonotonic variation of the quasiparticle effective masses with increasing x, exhibiting a maximum at $x \sim 0.9$, represents a marked deviation from any of the current theoretical models.

$Ce_xLa_{1-x}B_6$ intermetallic samples with fourteen different compositions x were grown in Al flux. The dHvA susceptibility was measured using a compensated arrangement of detection coils, with pulsed magnetic fields of up to 60 T provided by the National High Magnetic Field Laboratory, Los Alamos. Temperatures between ~400 mK and 4 K were obtained by pumping on ^3He and ^4He reservoirs.

The simple FS topologies of trivalent rare earth (RE) hexaboride compounds afford a clear interpretation of the experimental results. The FSs of both LaB_6 and CeB_6 consist of prolate electron ellipsoids situated at the X points of the cubic Brillouin zone (BZ) which overlap along the ΓR symmetry axes [8,11]. By directing the magnetic field B along the $\langle 100 \rangle$ axis, the minimum cross section area of this ellipse α_3 can be observed directly, while the maximum area $\alpha_{1,2}$ can be observed through magnetic breakdown (MB) [12–14]. Figure 1 shows an example of a Fourier transform (FT) of a dHvA signal measured in a sample of $Ce_xLa_{1-x}B_6$ for which $x = 0.4$; the actual dHvA oscillations are shown in the inset. The very fact that dHvA oscillations can be observed at such high concentrations x clearly implies the existence of a coherent FL with long-lived quasiparticles or, equivalently, long mean-free paths; we will address the mean-free paths of the quasiparticles later in this paper. Since the signal in Fig. 1 originates from a sample containing 60% La and 40% Ce, the α_3 frequency ($F_{\alpha 3}$), which dominates the spectrum, lies between that of LaB_6

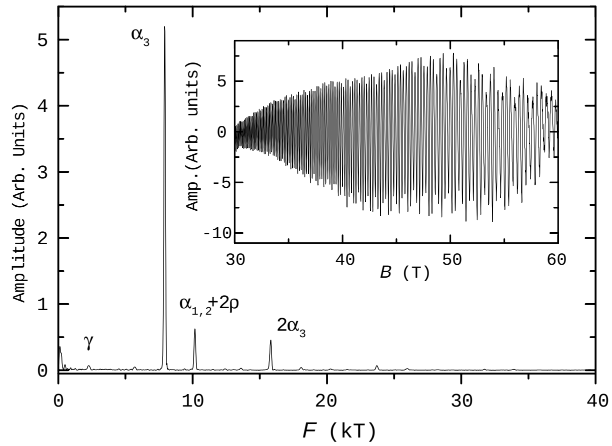

FIG. 1. A FT of dHvA oscillations measured in $Ce_{0.4}La_{0.6}B_6$ for the interval in the magnetic field between 30 and 60 T at ~450 mK, with the principal frequencies indicated. The raw oscillations are shown in the inset.

and CeB_6. This frequency is easily identifiable as α_3 since it is seen to vary in a continuous manner as the composition x is changed [see Fig. 2(a)]. A similar behavior is observed for the $\alpha_{1,2}$ MB frequency ($F_{\alpha 1,2}$). Given that both the α_3 and the $\alpha_{1,2}$ frequencies in Fig. 2(a) increase steadily on increasing x, the volume of the FS [at least for one spin (see below)] must also increase. Angle-dependent dHvA experiments have shown that the FS topologies of these compounds can be modeled by an ellipsoid of revolution [8], making an estimate of the number of carriers possible. Figure 2(b) shows a plot of the estimated conduction electron density (of the observed spin) relative to that of pure LaB_6, for which $n = 0.5$ per spin per RE atom [11]. This shows rather conclusively that the FS transforms gradually from that of pure LaB_6 to that of pure CeB_6. Our previous quantum interference measurements have shown that the areas of the BZ in LaB_6 and CeB_6 are the same to within 0.1% [12,13]. Hence, the observed ~12% increase in FS volume in Fig. 2(b) cannot be attributed to a change in the lattice parameters.

Evidence for the gradual transformation of the ground state from a simple low-effective-mass metal at one extreme to an enhanced-mass HF compound at the other emerges as a result of measuring the x dependence of the quasiparticle effective mass. Figure 3(a) shows the results of fitting the Lifshitz-Kosevich theory [9,15] to the temperature dependence of the amplitude of the α_3 oscillations for different concentrations x at ~40 T (and at 10 T for smaller x in the inset). Not only is the effective mass $m^*_{\alpha 3}$ (of the α_3 frequency) seen to increase with x, even at very low concentrations of Ce, but is also field dependent at intermediate concentrations, as evident in Fig. 3(b). The enhanced effective mass together with its partial suppression at high magnetic fields is a property characteristic of the HF ground state of pure CeB_6 [8,13,16–18]. Figures 3(a) and 3(b) show that

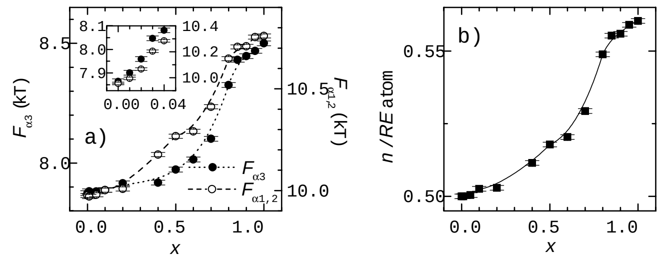

FIG. 2. (a) Plots of the concentration dependence of the α_3 and $\alpha_{1,2}$ dHvA frequencies at ~40 T, and at 10 T using the more sensitive field modulation technique in the inset. Error bars are shown. (b) The concentration dependence of the FS volume of the dominant spin sheet.

mass enhancement due to interactions with delocalized f electrons starts to develop at $x \lesssim 0.05$. More unusually, the x dependence of $m^*_{\alpha 3}$ becomes nonmonotonic at the highest concentrations, reaching a maximum at $x \sim 0.9$ before falling again for $x > 0.9$ (i.e., as the concentration corresponding to pure CeB_6 is approached). We note that the x dependence of the measured FS areas also exhibits a change in slope at $x \sim 0.85$, thus suggesting that the mechanism responsible for both the enhanced masses and the increased FS volume changes at this concentration.

The enhancement of the effective masses in ordered HF systems is usually understood in terms of the Anderson lattice model [7,19]. This model has also been shown to approximately account for the field dependence of the effective mass [2,20,21]. Owing to the absence of an ordered lattice of Ce ions, there are conceptual difficulties in applying this model to $Ce_xLa_{1-x}B_6$ intermetallics for which $x < 1$. Since magnetic correlations are absent at very low concentrations ($x \ll 1$), the single impurity model provides an alternative point of departure from which to investigate the effects of interactions. According to this model, thermodynamic functions of state, such as the Sommerfeld coefficient γ and the Pauli susceptibility χ, increase linearly with x [3,7]. Since the enhancement of γ in low concentration Kondo alloys is

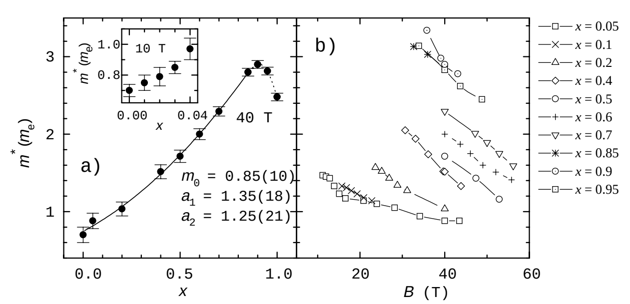

FIG. 3. (a) A plot of the quasiparticle effective mass of the α_3 frequency versus concentration at ~40 T, and at 10 T using field modulation in the inset. The parameters of the quadratic fit (solid line) for $x < 0.85$ are shown, and the dotted line is a spline fit. (b). The field dependence of the effective mass of the α_3 frequency for different concentrations. Spline fits join the points, and error bars are omitted for clarity.

proportional to the enhancement of the quasiparticle effective mass, $m^*_{\alpha,3}$ should therefore also vary linearly with x. Instead, we find in Fig. 3(a) that the x dependence of $m^*_{\alpha,3}$, for $x < 0.9$, is better described by a quadratic function of the form $m^* = m_0(1 + a_1 x + a_2 x^2)$ (where m_0 is the effective mass for pure LaB_6). It was suggested recently by Gor'kov and Kim that an additional x^2 term in the dependence of γ and χ on x could arise if two-impurity corrections are considered [22]. While the antiferromagnetic order is destroyed at high magnetic fields where the measurements are performed, antiferromagnetic exchange interactions (one of the essential ingredients for a n-impurity Kondo model) should still exist. However, given that the Ce moment ground state is a Γ_8 quartet, which may split into two doublets [3], it is unclear to what extent such a model can apply in $Ce_xLa_{1-x}B_6$ intermetallics. Two-impurity corrections alone evidently cannot explain our high-field results at the highest concentrations. The existence of antiferroquadrupolar order at high concentrations could lead to yet further complications, although the nature of this phase is still far from understood.

Another property of CeB_6 that was recently revealed as a result of high magnetic field studies is that the dHvA signal originates from only a single spin FS sheet [13]. Given that the Anderson lattice model anticipates that many-body bands of more than one spin channel cross the chemical potential μ at high magnetic fields [2,20,21], this model is apparently inconsistent with the experimental observations [13]. The contribution of only one spin channel to the dHvA signal in CeB_6 was inferred from the study of the harmonics [13]. In a simple metal with a Zeeman split band, the two spin states have the same frequency but different phases and their dHvA contributions interfere, with the degree of phase cancellation varying from one harmonic to the next. While there are additional factors, such as the thermal and Dingle amplitude reduction factors [9], which lead to an exponential damping of the harmonics, spin damping causes these to decay in an erratic manner. Pure LaB_6 fits this description quite well, as shown in Fig. 4(a). The small amplitude of the odd α_3 harmonics compared to the even harmonics results from the fact that the phase difference between the two spin channels is close to an odd integer multiple of π. In CeB_6, on the other hand, the nearly ideal exponential decay of the harmonics with increasing harmonic index indicates that there is only a single spin [13].

When we plot the logarithm $\ln(A_p/p^{1/2})$ versus x for $Ce_xLa_{1-x}B_6$ intermetallics in Fig. 4(a) (where A_p is the amplitude of the pth harmonic), we find that some aspects of the behavior of LaB_6 are reproduced in the sample with $x = 0.01$, and somewhat less at $x = 0.02$. The change in the spectral weight of certain harmonics relative to those of pure LaB_6 suggests that, even at this low concentration, the Ce moments are already starting to modify the ground

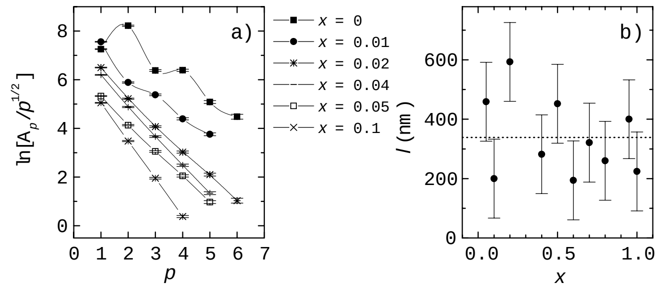

FIG. 4. (a) Plots of $\ln(A_p/p^{1/2})$ versus p at selected values of x, with spline fits shown for clarity. The linear slopes for $x \geq 0.05$ indicate that the oscillations originate from a spin polarized FS sheet. (b) Estimates of the mean-free path for alloys of different concentrations. The dotted line is the mean and the error bars represent the standard deviation.

state. As soon as $x \gtrsim 0.05$, the harmonics begin to fall in a nearly ideal exponential manner in the same way as in CeB_6. This would appear to suggest that the contribution of one of the spin states begins to disappear as soon as the high field HF state develops. Thus, the question of the missing spin quasiparticles applies not only to CeB_6 [13] but also to alloys with surprisingly low concentrations of Ce. In CeB_6 it was found that, at the high magnetic fields where dHvA oscillations are observed [13], the magnetic field was sufficiently strong (i.e., $|mg\mu_B B| \gg k_B T_A$, where g is the electron g factor and T_A is the Kondo temperature) so that, in addition to the spin-down component of the Kondo resonance being Zeeman shifted to energies far above μ, the spin-up component (which is above μ at $B = 0$) is shifted to energies below μ. This scenario was recently discussed by Edwards and Green [21]. While it is unclear to what extent the mean-field Anderson lattice model might apply at such high magnetic fields [2], it is apparent from Eq. (1) of Ref. [13] that the effective masses for both spin channels (i.e., $m = \pm\frac{1}{2}$) should fall with increasing field when $|mg\mu_B B| > k_B T_A$. However, because the effective masses determined for the low concentration alloys (i.e., with x as low as 0.05) are much lighter than those in pure CeB_6, it is unlikely that the absence of one of the spin components to the dHvA signal can be attributed to very heavy quasiparticles of that spin. According to Edwards and Green [21], the vanishing of the dHvA signal of the spin-down channel is attributed to spin fluctuation effects, which predominantly scatter and enhance the mass of this spin channel when $|mg\mu_B B| > k_B T_A$. The results presented here on $Ce_xLa_{1-x}B_6$ alloys would therefore appear to suggest that a related spin polarity-dependent scattering mechanism operates for all concentrations $0 < x < 1$. The scattering of only one spin from fluctuating Ce moments would remove this contribution from the dHvA signal, but not the specific heat.

It is interesting to note that, as the amplitude of the damped spin channel falls on increasing x to 0.05, there is no detectable change in its frequency from that of the dominant unscattered spin channel. Thus, at least for the lower concentrations ($x < 0.05$), where both spins are observed, there is no discernible difference in the FS topologies of the two spin sheets. While it is unclear to what extent the two spin sheets continue to have the same topology for $x > 0.05$, the evidence at hand strongly suggests that n [in Fig. 2(b)] increases with x for both spin sheets. One can therefore conclude that, at least at the lower concentrations, the total FS volume (comprising both spins) increases with x, with a noninteger number of f electrons per Ce ion contributing to this volume. The noninteger contribution of f electrons to the FS volume is anticipated by the renormalized-band picture [21]. Given that the FS volume is observed to be field independent in CeB_6, we should expect this increase in FS volume with x to apply at all fields $|mg\mu_B B| > k_B T_A$.

While the persistence of dHvA oscillations to high concentrations of Ce in LaB_6 (or, alternatively, La in CeB_6) implies that the FL is coherent, it is instructive to see to what extent the impurities affect the quasiparticle scattering rate τ^{-1}. The contribution of only one spin channel for samples with $x > 0.05$ makes the determination of τ rather straightforward. This can be obtained directly at one specific field by equating the slope of $\ln(A_p/p^{1/2})$ versus p in Fig. 4(a) to $\sim 2\pi^2 kT/\hbar\omega_c + \pi/\omega_c\tau$ [9] (where T is the temperature and $\omega_c = eB/m^*$ is the cyclotron frequency). To make the estimate of the sample quality independent of the degree to which the m^* is renormalized by the interactions, it is more meaningful to extract the mean-free path l. This is related to the mass-dependent τ via the relation $l/l_c = \omega_c\tau$ which is mass independent, where $l_c = (2\hbar F/eB^2)^{1/2}$ is the cyclotron length for a particular orbit. Upon performing this analysis on samples with different concentrations, rather than having any kind of dependence on x, in Fig. 4(b), l is found to vary randomly between ~200 and 600 nm and is of about the same order as that found for pure CeB_6. We can therefore conclude that the arbitrary substitution of La with Ce (or vice versa) contributes little or nothing to the quasiparticle scattering rate of the dominant spin sheet at high magnetic fields. The x independence of l has two implications. Firstly, the substitution of La with Ce does not lead to any significant potential scattering due to charge differences or lattice distortions. Secondly, the scattering from spin fluctuations does not occur for the dominant spin, while these scatterings completely remove the other spin component from the signal at high concentrations. It is therefore likely that the pronounced negative magnetoresistance observed in $Ce_xLa_{1-x}B_6$ alloys at high fields [23] is due entirely to the suppression of spin fluctuation scattering of only one spin sheet.

In summary, these first ever dHvA measurements on a complete alloy series $Ce_xLa_{1-x}B_6$ reveal the existence of long-lived quasiparticles for all x, but only for one of the spin channels. The second spin channel is rapidly attenuated at very low x. The persistence of one of the spin channels throughout the alloy series enables a detailed investigation of the development of the FS of the HF ground state in the high magnetic field regime. Both the dHvA frequencies and, in particular, the quasiparticle effective masses vary in a nonmonotonic fashion with x; the maximum in the effective mass at $x \sim 0.9$ is not anticipated by any existing theoretical model.

Work conducted at the National High Magnetic Field Laboratory was supported by the National Science Foundation (NSF), the State of Florida, and the Department of Energy. Additional support from the NSF (DMR95-01419) is acknowledged by one of us (R. G. G.). We would like to express our gratitude to Pedro Schlottmann for useful comments.

[1] For reviews, see G. G. Lonzarich, J. Magn. Magn. Mater. **76–77**, 1 (1988); M. Springford, Physica (Amsterdam) **171B**, 151 (1991).
[2] A. Wasserman, M. Springford, and A. C. Hewson, J. Phys. Condens. Matter **1**, 2669 (1989).
[3] See, e.g., P. Schlottmann, Phys. Rep. **181**, 1 (1989).
[4] C. Jayaprakash *et al.*, Phys. Rev. Lett. **47**, 737 (1981).
[5] B. A. Jones *et al.*, Phys. Rev. Lett. **61**, 125 (1988); B. A. Jones and C. M. Varma, Phys. Rev. Lett. **58**, 843 (1987).
[6] G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).
[7] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, England, 1993).
[8] Y. Onuki *et al.*, J. Phys. Soc. Jpn. **58**, 3698 (1989).
[9] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, England, 1984).
[10] D. H. Lowndes *et al.*, Proc. R. Soc. London **331**, 497 (1973).
[11] A. J. Arko *et al.*, Phys. Rev. B **13**, 5240 (1976).
[12] N. Harrison *et al.*, Phys. Rev. Lett. **80**, 4498 (1998).
[13] N. Harrison *et al.*, Phys. Rev. Lett. **81**, 870 (1998).
[14] The actual MB frequency that is observed is $\alpha_3 + 2\rho$. However, since the size of the ρ ellipsoid is much less than that of the α ellipsoid, we introduce only a small error by ignoring the ρ contribution.
[15] I. M. Lifshitz and A. M. Kosevich, Sov. Phys. JETP **2**, 636 (1956).
[16] W. Joss *et al.*, Phys. Rev. Lett. **59**, 1609 (1987).
[17] E. G. Haanappel *et al.*, Physica (Amsterdam) **177B**, 181 (1992).
[18] N. Harrison *et al.*, J. Phys. Condens. Matter **5**, 7435 (1993).
[19] J. W. Rasul, Phys. Rev. B **39**, 663 (1989).
[20] Y. Ono, J. Phys. Soc. Jpn. **65**, 19 (1996).
[21] D. M. Edwards and A. C. M. Green, Z. Phys. B **103**, 243 (1997).
[22] L. P. Gor'kov and Ju H. Kim, Phys. Rev. B **51**, 3970 (1995).
[23] N. Sato *et al.*, J. Phys. Soc. Jpn. **54**, 1923 (1985).