Unusual Evolution of the Conduction-Electron State in $Ce_xLa_{1-x}B_6$ from Non-Fermi Liquid to Fermi Liquid

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We report unusual evolution of the conduction-electron state in the localized f electron system $Ce_xLa_{1-x}B_6$ from normal electron state to heavy Fermi liquid (FL) state through local FL and non-FL states with increasing Ce concentration and/or with increasing magnetic field. The effective mass of quasiparticle or the coefficient A of T^2 term of resistivity is found to increase divergently near the boundary between FL state and non-FL state. The features of the non-FL state are also different from those of the typical non-FL systems previously observed or theoretically predicted.

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The heavy fermion compound CeB_6 and its alloy compounds $Ce_xLa_{1-x}B_6$ have been most intensively studied because of various interesting magnetic and electronic properties. $Ce_xLa_{1-x}B_6$ crystallizes in the CaB_6 structure. The ground crystal electric field state of the $4f$ electron is the Γ_8 quartet which carries the magnetic dipolar, electric quadrupolar, and magnetic octupolar moments. Because of the plural degrees of freedom, it exhibits an intriguing phase diagram as a function of magnetic field and Ce concentration. Figure 1 shows the magnetic phase diagram when magnetic fields are applied parallel to the [001] axis [1]. For CeB_6 the ground state is the antiferromagnetic (AFM) state (phase III) where the quadrupolar order coexists with the magnetic order. With increasing magnetic field it changes to the antiferroquadrupolar (AFQ) state (phase II). No transition to the paramagnetic (P) state (phase I) has been observed up to 60 T. With decreasing Ce concentration to $x \sim 0.75$ phase III changes to phase IV whose order parameter is controversial but is suspected to be octupole [2]. With further decrease of Ce concentration the system becomes a paramagnetic state around $x = 0.6$. With increasing magnetic field the phase IV and phase I above $x \sim 0.4$ change to the AFM state through weak first order transition. It is found that the AFQ phase (phase II) changes to phase I at 29 T for $x = 0.5$ [3], implying that the transition field decreases quickly with decreasing x. When $x < 0.3$ no clear first order transition is observed under magnetic fields. The dotted lines in Fig. 1 are hypothetical boundaries estimated from these observations.

The resistivity of CeB_6 shows the typical Kondo behavior of the $-\log T$ temperature dependence down to the AFQ transition at 3.2 K and shows a heavy Fermi liquid (FL) behavior in the ground state in spite of the magnetic order. That is, at lowest temperatures below 0.5 K, the specific heat is proportional to T with electronic specific heat coefficient of 0.25 J/K^2 mol [4] and the resistivity increases proportionally to T^2 [5]. Various anomalous properties of the conduction electrons have been reported from the de Haas-van Alphen (dHvA) effect studies. It is found that the effective mass (EM) of CeB_6 is very strongly suppressed by magnetic field [6]. The observed dHvA oscillations are likely to arise from the conduction electrons of only one spin direction [7]. The evolution from the normal electron state in LaB_6 to the heavy FL state in CeB_6 has been studied in $Ce_xLa_{1-x}B_6$ at very high magnetic fields [8]. The behavior of the evolution as well as the anomalous properties of the conduction electrons at high magnetic fields have not been understood [7,8]. We have studied the conduction state at zero and low magnetic fields in this unique system whose $4f$ electron has the plural degrees of freedom and have found unusual evolution of it with magnetic field and Ce concentration.

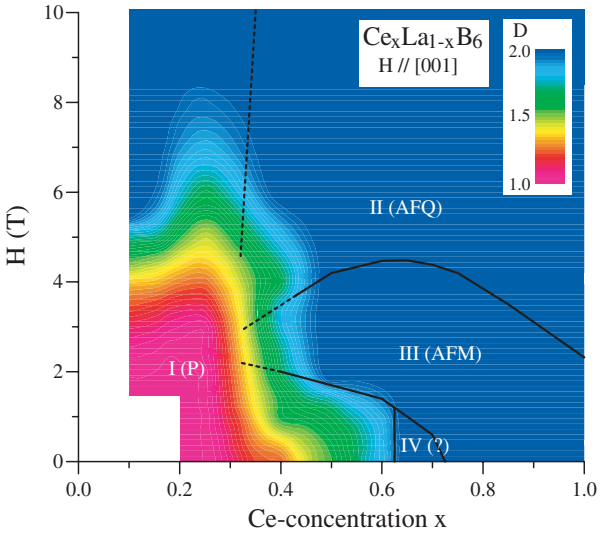

FIG. 1 (color). Magnetic phase diagram of $Ce_xLa_{1-x}B_6$ for $H \parallel [001]$ as a function of Ce concentration (x) and field (H) at low temperatures. The color denotes the value of exponent D in the temperature dependence of resistivity $\rho(T) = \rho(0) + AT^D$.

We have measured resistivity, the dHvA effect, and ac magnetic susceptibility with fields applied parallel to the [001] axis. All the measurements were performed in a dilution refrigerator in magnetic fields up to 17 T. The samples are single crystals grown by a floating zone method. For the measurements of the resistivity, Au thin films were deposited as electrodes. The samples and small pieces of sintered silver were both glued on the Cu plate and immersed into the mixture. The resistivity was measured as a function of applied current to confirm that the self-heating does not affect the measurements. The current was applied parallel to the [110] axis. The dHvA effect measurements were performed by the conventional field modulation method [9]. The modulation field strength was kept low enough so that the heating does not affect the EM measurements.

Figure 2(a) shows the resistivity ρ of $Ce_xLa_{1-x}B_6$ at zero field as a function of T^2. The origin of each data is shifted by an appropriate amount to compare with each other. For $x = 1.0$ and 0.75, the temperature dependence of the resistivity at lowest temperatures is given by a formula $\rho(T) = \rho(0) + AT^2$, which indicates the FL behavior. For $x = 0.7$ the plot is straight only at lowest temperatures. At higher temperatures between 0.22 K and 0.89 K, another straight line with a different slope can be fitted to the data points. For $x = 0.65$, a straight line can be fitted to the data points in a relatively wide temperature range. The temperature dependence changes qualitatively around $x = 0.65$. Those for $x = 0.6$–0.25 can be described well with formulas $\rho(T) = \rho(0) + AT^D$ with the exponents D less than 2.0 as shown in Fig. 2(b).

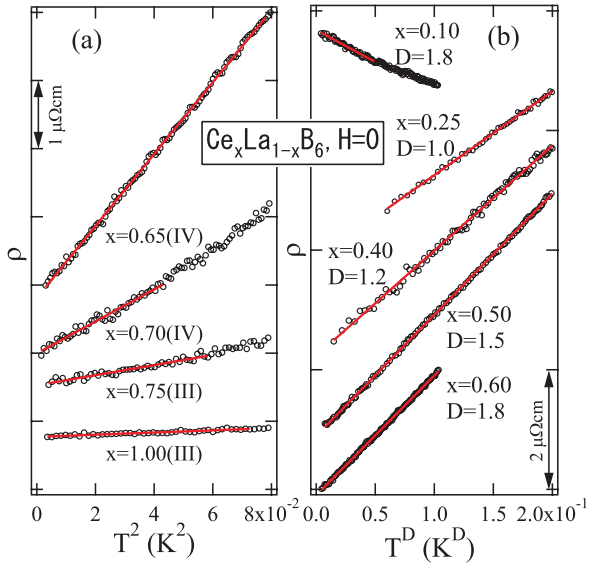

FIG. 2 (color). (a) Temperature dependence of the electrical resistivity of $Ce_xLa_{1-x}B_6$ at zero field plotted as a function of T^2 in the ordered phases III and IV and (b) as a function of T^D in the paramagnetic phase (phase I). The origin of each data set is shifted vertically by an appropriate amount and the solid lines are guides to the eye which are obtained by least squares fits to the data points.

It was reported that the specific heats of high Ce concentration samples ($x \geq 0.9$) are proportional to T at low temperatures, but for $x = 0.75$ it becomes difficult to fit the temperature dependence to a linear T relation in a sufficiently wide temperature range [4]. The specific heats of the samples with $x = 0.65$, 0.6, and 0.5 no longer show the FL behavior but are described well with the formula $C = \alpha T^\delta$ with the exponent δ being less than 2 [10]. The present observations together with these previous results indicate that FL state is formed down to $x = 0.75$ at lowest temperatures, but becomes more unstable with decreasing Ce concentration. It is not clear whether the T^2 dependence of resistivity observed for $x = 0.7$ and 0.65 points to the FL or is a consequence of competing effects.

In the dilute samples the sign of the coefficient A becomes minus. For $x = 0.1$, the resistivity at low temperatures is given by $\rho(T) = \rho(0) + AT^{1.8}$, while in a very dilute sample ($x = 0.0061$, 0.012) $\rho(T) = \rho(0) + AT^{2.0}$ [11,12] which is expected for the local FL [13]. Above 1.5 T, the temperature dependence is given by $\rho(T) = \rho(0) + AT^D$ with a plus A. At sufficiently high fields the value of D becomes 2.0. The T^2 dependence is consistent with the observation of the dHvA oscillations at high magnetic fields.

In Fig. 1 we show the contour plot of the exponent D derived from the resistivity measurements at various magnetic fields for various $Ce_xLa_{1-x}B_6$ samples. It is noted that $D = 2.0$ at sufficiently high magnetic fields and in the ordered phases II, III, and IV except for the region near the boundaries between phase I. In phase I, D starts to decrease from 2.0 with decreasing field and/or decreasing Ce concentration, and approaches 1.0. Then, as noted from Fig. 2(b), the value of A changes from plus to minus and finally the local FL behavior appears at low concentration and low field region. The detailed evolution of the values of D and A in the low concentration and low field region has not been clarified yet. The figure indicates that starting from the normal state in LaB_6 the conduction-electron state evolves first into the local FL state and then to the non-FL (NFL) state and finally to the FL state with increasing Ce concentration and magnetic field. This is compared with the observation in a typical nonmagnetic heavy fermion system $Ce_xLa_{1-x}Cu_6$ where the resistivity follows the relation $\rho(T) = \rho(0) + AT^{2.0}$ for all concentration samples and A changes from minus to plus with increasing Ce concentration, i.e., from the local FL to heavy FL without passing through the NFL state [14]. The f electrons are thought to behave as itinerant electrons in $CeCu_6$. On the other hand, the present studies demonstrate a different way of evolution to heavy FL in a localized f electron system with plural degrees of freedom.

Figure 3 plots the value of A as a function of magnetic field when $D = 2.0$. The values increase rapidly with

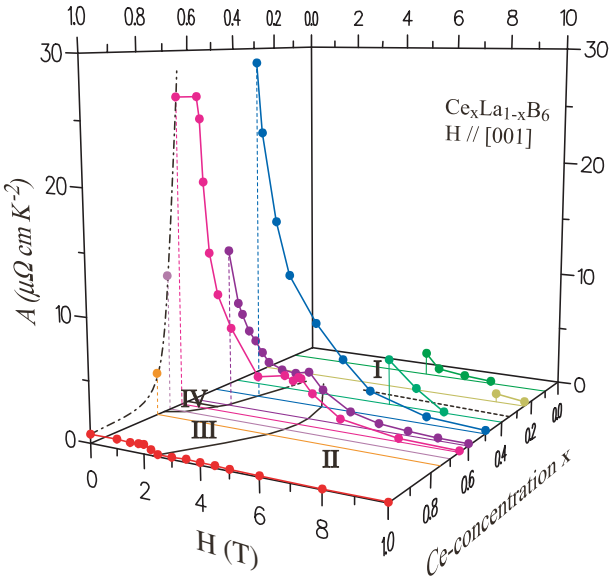

FIG. 3 (color). Field dependence of the coefficient A for various $Ce_xLa_{1-x}B_6$ samples.

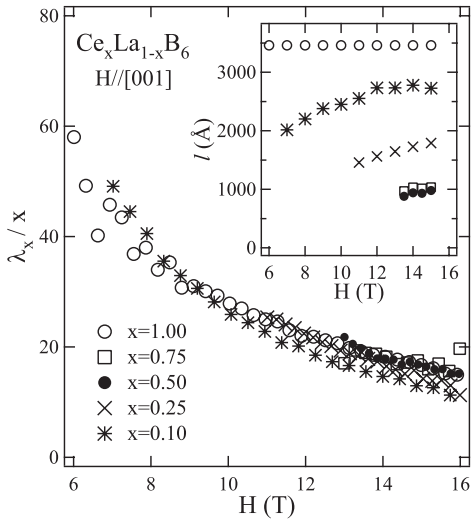

FIG. 4. Field dependence of mass enhancement factor per Ce λ_x/x and mean free path l (inset) for various Ce concentration samples.

decreasing field, implying that the EM increases rapidly with decreasing field. The very rapid increase with decreasing field has been observed also for the electronic specific heat coefficient of CeB_6 [15] and that of $Ce_{0.5}La_{0.5}B_6$ [16]. When the ground state is either phase III ($x = 1.0$) or IV ($x = 0.65$), the increasing rate of the value decreases with decreasing field and the value becomes finite and constant. On the other hand, when the ground state is phase I, the value increases divergently at low fields and the transport property starts to deviate from that of the FL. It is also noted that when we trace the value of A at zero or a constant field, the value of A appears to increase very rapidly or divergently with decreasing x near the boundaries between phase I and ordered phases II, III, and IV.

To investigate the FL states at high fields and how they evolve to the NFL state at lower fields, we have measured the EM and the mean free path of the conduction electron via the dHvA effect. The EM's are found to increase divergently with decreasing field. In Fig. 4 we plot the mass enhancement factor λ_x of the α_3 oscillation which arises from the main ellipsoidal Fermi surface. The factor λ_x is given by the formula $m_x^* = (1 + \lambda_x)m_{\mathrm{La}}^*$, where m_x^* and m_{La}^* are the EM's of $Ce_xLa_{1-x}B_6$ and LaB_6, respectively. That is, (λ_x/x) of Fig. 4 gives the mass enhancement factor per Ce. It is noted that the data points of various samples lie approximately on a single curve, indicating that the mass enhancement is nearly proportional to the number of Ce ions. The effective masses appear to increase divergently towards finite fields of 2–4 T, which is consistent with the behavior of A. The good scaling relation of the mass enhancement with the number of Ce ions indicates that the rapid increase of the EM may arise from a single site effect like the Kondo effect and the fields could be compared with the magnetic Kondo field of 1.1 T which is derived from the magnetoresistance measurements in low concentration samples [12].

The mean free path of the conduction electron in CeB_6 was determined by the conventional Dingle plot which plots the appropriately reduced amplitude of the oscillation as a function of inverse field [9]. The plot ordinarily becomes a straight line and the slope gives the value of m^*T_D. Here, m^* is EM and T_D is the Dingle temperature which is inversely proportional to the lifetime of the conduction electron or is a measure of damping of the oscillation due to scattering. The mean free path l is obtained from the following equation:

$$m^*T_D = \frac{\hbar^{3/2}}{\pi k_B}\left(\frac{eF}{2}\right)^{1/2}\frac{1}{l}. \qquad (1)$$

Here F is the dHvA frequency. The Dingle plot of CeB_6 gives a straight line, indicating that m^*T_D is independent of magnetic field even when the EM of CeB_6 changes with field. However, in other $Ce_xLa_{1-x}B_6$ compounds, it is found that the Dingle plot is not straight implying that m^*T_D depends on magnetic field. In these cases the Dingle plot analysis cannot give the Dingle temperature. We have made the absolute measurements of the dHvA oscillation amplitudes and compared them with that of CeB_6 to determine the magnitude of the damping or the Dingle temperature.

In order to derive the Dingle temperature by this procedure, we need to know the values of the spin splitting factor and the curvature factor. The former factor arises from the dephasing effect of the up and down spin electron oscillation [9] and it is very difficult to determine the value of the factor experimentally. However, owing to the peculiar fact that the observed oscillation arises dominantly from the only one spin state for all the $Ce_xLa_{1-x}B_6$ samples, this

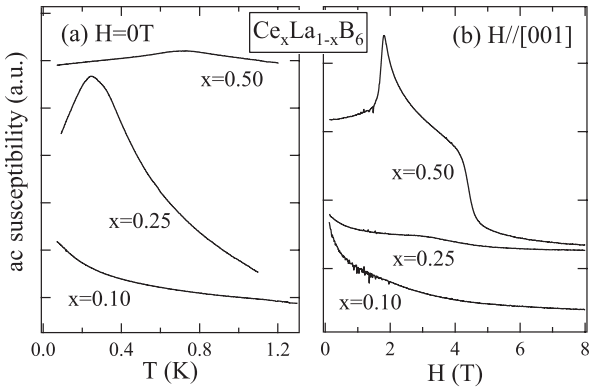

FIG. 5. Magnetic susceptibility of $Ce_xLa_{1-x}B_6$ ($x = 0.1$, 0.25, and 0.5) samples (a) as a function of temperature at zero magnetic field and (b) as a function of magnetic field at 50 mK for $x = 0.1$ and at 100 mK for $x = 0.25$ and 0.5. The origin of each curve is shifted by an appropriate amount.

factor can be assumed to be the same except for LaB_6. The latter factor depends on the curvature of the Fermi surface. We have studied the angular dependence of the frequency of the α oscillation around [001] and have found that this factor can be assumed to be nearly the same for all the $Ce_xLa_{1-x}B_6$ [17].

The inset of Fig. 4 shows the l's thus determined. l decreases with decreasing field except for CeB_6 and appears to decrease to zero at a finite field. The increasing l with increasing magnetic field is consistent with our observation that the magnetoresistance at low temperatures is negative except for CeB_6. The successful observation of the dHvA oscillation indicates that a coherent state is formed over a distance of several hundreds of atoms, although the Ce and La atoms are distributed randomly. This fact leads to the idea that the field dependent l thus obtained may not be interpreted as a measure of scattering amplitude by impurities but may give a measure of the distance over which the heavy quasiparticle is coherent. In this situation we speculate that the destruction of coherence would lead to the NFL transport property.

The transition from FL to NFL seems to be accompanied by the rapid increase of EM or A. This feature may be compared with the observations near the magnetic quantum critical point (QCP). However, in the present system the same feature is also observed in phase I under magnetic fields where no phase transition takes place. It is also noted that the NFL behavior is observed not only near the boundary of magnetic order-disorder transition but also over a broad range of Ce concentration and magnetic field. These observations are obviously different from those near the QCP.

As described above the specific heat observed in phase I ($x = 0.5$, 0.6) does not show a divergent behavior with decreasing temperature like that observed in the typical NFL system [18,19]. The behavior of the susceptibility also differs from that of the typical NFL system where the susceptibility shows a divergent behavior with decreasing temperature. Figures 5(a) and 5(b) show the susceptibilities of $Ce_xLa_{1-x}B_6$ samples with $x = 0.5$, 0.25, and 0.1 as functions of temperature and magnetic field, respectively. For $x = 0.5$ and 0.25 the susceptibility exhibits a broad hump with decreasing temperature. Under magnetic fields, the susceptibility for $x = 0.5$ exhibits two features corresponding to the two first order magnetic transitions, while for $x = 0.25$ a weak hump is observed. For $x = 0.1$, the susceptibility increases with decreasing temperature signaling a NFL behavior at zero magnetic field. However, under magnetic fields it exhibits a very broad and faint shoulder around 1.5 T. These observations indicate that a short range antiferromagnetic correlation survives in the low concentration $Ce_xLa_{1-x}B_6$ samples, in spite of the NFL transport properties. The NFL behavior presently observed is also different from those expected for Griffiths phase, distribution of Kondo temperature, or multichannel Kondo effect [18,19].

In summary, we have demonstrated the unusual evolution from normal electron state to the heavy FL state through the local FL and NFL states in the localized f electron system with plural degrees of freedom. The NFL behavior is also different from those of the typical NFL systems which are previously observed or theoretically predicted.

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[1] S. Nakamura *et al.*, J. Phys. Soc. Jpn., Suppl. **71**, 112 (2002).
[2] H. Kusunose and Y. Kuramoto, J. Phys. Soc. Jpn. **74**, 3139 (2005).
[3] M. Akatsu *et al.*, Phys. Rev. Lett. **93**, 156409 (2004).
[4] T. Furuno *et al.*, J. Phys. Soc. Jpn. **54**, 1899 (1985).
[5] N. Sato *et al.*, J. Phys. Soc. Jpn. **54**, 1923 (1985).
[6] W. Joss *et al.*, Phys. Rev. Lett. **59**, 1609 (1987).
[7] N. Harrison *et al.*, Phys. Rev. Lett. **81**, 870 (1998).
[8] R. G. Goodrich *et al.*, Phys. Rev. Lett. **82**, 3669 (1999).
[9] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, England, 1984).
[10] S. Nakamura *et al.*, Phys. Rev. B **61**, 15 203 (2000).
[11] K. Winzer, Solid State Commun. **16**, 521 (1975).
[12] K. Samwer and K. Winzer, Z. Phys. B **25**, 269 (1976).
[13] K. Yamada, Prog. Theor. Phys. **54**, 316 (1975).
[14] A. Sumiyama *et al.*, J. Phys. Soc. Jpn. **55**, 1294 (1986).
[15] T. Müller *et al.*, J. Magn. Magn. Mater. **76–77**, 35 (1988).
[16] S. Nakamura *et al.*, Phys. Rev. B **68**, 100402 (2003).
[17] M. Endo *et al.*, J. Phys. Soc. Jpn. **75**, 114704 (2006).
[18] G. R. Stewart, Rev. Mod. Phys. **73**, 797 (2001).
[19] E. Miranda and V. Dobrosavljević, Rep. Prog. Phys. **68**, 2337 (2005).