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Field-induced transition from non-Fermi-liquid state to heavy Fermion state in $Ce_{0.5}La_{0.5}B_6$

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Measurements of the electrical resistivity, specific heat, and dHvA oscillations have been made on a cubic Kondo system $Ce_{0.5}La_{0.5}B_6$ with a Γ_8 ground state at very low temperatures. In the paramagnetic phase I which appears in lower fields, the electrical resistivity deviates from the formula $\rho(T)=\rho(0)+AT^2$ below ~300 mK, implying an appearance of non-Fermi-liquid (NFL) state. In the field induced antiferromagnetic phase III, the large γ coefficient in the specific heat and the large coefficient A in the electrical resistivity have been found, indicating a heavy fermion (HF) state. The coefficient A diverges when the field approaches the phase III–phase I transition point. On the other hand, by increasing the Ce concentration phase IV with a long range order appears and a HF state is found to be formed there. We argue that the origin of the NFL behavior is related with the quadrupolar moment.

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In the last decade, non-Fermi-liquid (NFL) systems have attracted much attention. To date a lot of NFL systems have been found.[1] Several mechanisms have been proposed as the origin of NFL: the multichannel Kondo effect,[2,3] long-range fluctuation of spins in the neighborhood of quantum phase transition,[4] a distribution of the Kondo temperature driven from disorder (Griffiths phase),[5,6] etc. These theories have succeeded in explaining a part of the properties of NFL systems. However, the physics of NFL is now in progress and far from settled. Finding new NFL systems is desirable.

$Ce_xLa_{1-x}B_6$ is known as a typical Kondo system. The boron octahedra form a simple cubic network and a Ce or a La ion occupies each body center of the network. The ground crystal electric field state is a Γ_8 quartet and a Γ_7 excited sits at 540 K.[7] The Γ_8 state carries two kinds of independent degrees of freedom. One is the degrees of freedom for magnetic dipolar moment and the other is that for electric quadrupolar moment. When Ce-concentration x shows high values ($x \gtrsim 0.5$), attractive magnetic phases appear. We show the magnetic phase diagrams of $Ce_xLa_{1-x}B_6$ ($x=0.5$ and 0.65, $H\|[001]$) in Fig. 1 for convenience of later discussion. The solid lines in this figure are taken from previous experiments[8,9] and the solid circles are present results obtained from the magnetoresistivity (not shown). Phase I is a paramagnetic phase where $-\log T$ temperature dependence is shown in the electrical resistivity above ~2 K.[10] Several microscopic measurements suggest a mysterious splitting of the Γ_8 state in this phase.[7,11] Phase II which extends to a higher field side in the H-T plane is an antiferroquadrupolar (AFQ) phase. The neutron scattering[12] and NMR measurements[13] performed on CeB_6 indicate the absence of magnetic dipolar order in this phase. Phase III is an AFQ phase where an antiferromagnetic (AFM) order coexists. The magnitude of the spontaneous magnetic moment in this phase is only ~0.3 μ_B, which is much smaller than that expected for the Γ_8 state (1.57 μ_B).[12] In phase IV which appears in the low temperature (low-T) and low field (low-H) region, no Bragg peak has been found in the neutron scattering using a powder sample.[14] On the other hand, a clear λ-shape anomaly in the specific heat[15] and a pronounced discontinuous anomaly in the elastic constant C_{44} (Refs. 8 and 16) have been observed at the phase I–phase IV transition point. Recent muon spin relaxation measurements indicate the absence of static magnetic internal field in this phase.[17] Nakamura *et al.* have suggested that two characteristic temperatures can be identified in the concentrated $Ce_xLa_{1-x}B_6$;[15] one is the temperature for cancellation of dipolar moment (~10 K) and the other is that for quadrupolar cancellation (~1 K). They claimed phase IV is an electric quadrupolar ordered phase where dipolar moments are quenched by the Kondo effect. Very recently, Akatsu *et al.* have found a spontaneous shear lattice distortion in this phase through the thermal expansion measurements, which implies the generation of spontaneous quadrupolar moments.[18] They suggested the ferroquadrupolar order in phase IV.

With reducing the Ce-concentration x, the ground state of $Ce_xLa_{1-x}B_6$ in the low-T, low-H region changes from phase IV to paramagnetic phase I at $x_c \sim 0.6$.[19] In phase I of $Ce_{0.5}La_{0.5}B_6$, no discontinuity is found both in the specific

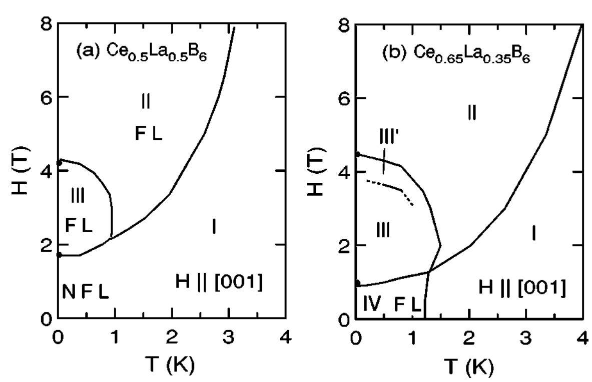

FIG. 1. The magnetic phase diagrams of $Ce_xLa_{1-x}B_6$ (a) $x=0.5$ (b) $x=0.65$ for $H\|$ [001]. The solid lines are taken from Refs. 9 and 8, respectively. Solid circles are the present results.

heat and C_{44} down to very low temperatures,[15,20] indicating the absence of long range-order. The ground state of $Ce_{0.5}La_{0.5}B_6$ in the low-T, low-H region shows various mysterious features. The magnetic susceptibility χ_m of this system shows only weak temperature dependence below 1 K,[21] indicating the quenching of the dipolar moments. The quadrupolar susceptibility χ_q at 0 T increases with decreasing temperature, shows a broad peak at around 240 mK, and decreases gradually at least down to 15 mK.[15] This provides the view that quadrupolar degrees of freedom remain down to ~240 mK but the quadrupolar moments are screened by conduction electrons at very low temperatures. The ground state of $Ce_{0.5}La_{0.5}B_6$ in the low-T, low-H region is not a spin glass, because only a small hysteresis is observed in the low-T magnetization.[21] The specific heat of $Ce_{0.5}La_{0.5}B_6$ at 0 T shows $T^{1.5}$ behavior below ~700 mK down to at least 170 mK, suggesting a deviation from FL picture.[15] When fields are applied along the [001] axis, this system undergoes successive transitions from phase I to phase III at 1.7 T and further to phase II at 4.3 T.[9] A small hysteresis loop in the magnetization at the phase I–phase III transition point is an indication of weak first order transition.[21]

$Ce_xLa_{1-x}B_6$ is a nice system for studying NFL physics, because one can change the competition between the Kondo effect and the intersite interactions by changing the magnetic field or Ce concentration. Interestingly, the ground state of the $4f$ electron carries quadrupolar moments. In the present paper, we show low-T properties of $Ce_{0.5}La_{0.5}B_6$ and the electrical resistivity of the reference system $Ce_{0.65}La_{0.35}B_6$. The transition from a NFL to a FL occurs in $Ce_{0.5}La_{0.5}B_6$ by tuning the magnetic field in association with magnetic phase transition.

The single crystals of $Ce_xLa_{1-x}B_6$ are grown by the floating zone method. The samples for the resistivity measurements were cut from ingots by the spark wire cutter and were polished to rectangular shapes. Dimensions of samples of $Ce_{0.5}La_{0.5}B_6$ and $Ce_{0.65}La_{0.35}B_6$ were $0.265\times0.65\times8$ mm^3 and $0.215\times0.74\times8$ mm^3, respectively. Both of the samples are longest along the $[1\bar{1}0]$ axis and the current is directed along this axis. We measured the resistivity using the dc four-terminal method in a top loading dilution refrigerator. The samples were glued on copper plates for a good thermal contact to the ^{3}He-^{4}He mixture. The sample of $Ce_{0.5}La_{0.5}B_6$ for the specific heat measurements is 162.7 mg in weight. The specific heat was measured by the semiadiabatic method. The de Haas-van Alphen (dHvA) oscillations were detected by the conventional field modulation method, using a superconducting magnet.

The electrical resistivity of $Ce_{0.5}La_{0.5}B_6$ in phase III is shown in Fig. 2 (a). The resistivity well follows the formula $\rho(T)=\rho(0)+AT^2$. The coefficient A is found to be 28.8 $\mu\Omega\,\mathrm{cm\,K}^{-2}$ at 1.8 T. This value is 35 times larger than that of CeB_6 at 0 T (Ref. 10) and is almost the same as that of $CeAl_3$.[22] The large A strongly suggests that heavy quasiparticles (QP's) are realized in phase III. Similar T^2 dependence in the resistivity is observed in phase II (not shown). We plot A and its inverse A^{-1} in Fig. 3 as a function of magnetic field, respectively. The coefficient increases with decreasing field and diverges as $A=21.2/(H-H_0)$ in phase III. The pronounced change in A indicates that the correlation between the conduction electrons depends on the field strongly. The positive value of H_0 (=1.1 T) implies that the FL composed of heavy QP's become unstable at lower fields.

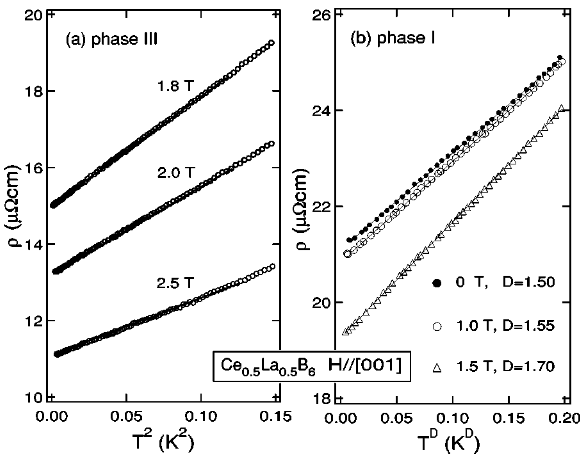

FIG. 2. The electrical resistivity of $Ce_{0.5}La_{0.5}B_6$ (a) in phase III and (b) in phase I shown as a function of (a) T^2 and (b) T^D. The field and current are directed along the [001] and $[1\bar{1}0]$ axes, respectively. Solid lines are fits.

With further increasing field in phase II, the dHvA oscillations start to be detected from about 13 T.[23] Figure 4 shows the α_3 oscillation and its Fourier spectrum under fields parallel to the [001] axis. The frequencies of the signal obtained in the present study is consistent with the previous dHvA experiments by Goodrich *et al.*[24] They observed the signal under high magnetic fields ~40 T produced by a pulsed magnet. The effective mass $6.7m_0$ obtained in the present study in the range $14<H<16$ T is much larger than $\sim2m_0$ obtained by Goodrich *et al.* at around 40 T. The large effective mass $6.7m_0$ indicates the formation of a coherent heavy fermion state in spite of the disorder (random distribution) of Ce ions.

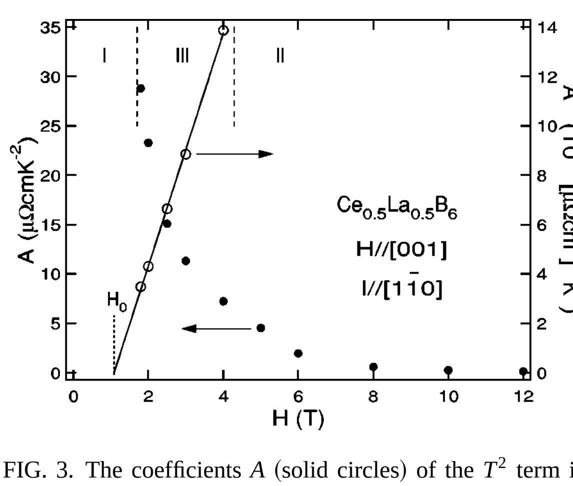

FIG. 3. The coefficients A (solid circles) of the T^2 term in the resistivity of $Ce_{0.5}La_{0.5}B_6$ shown as a function of magnetic field. The field and current are directed along the [001] and $[1\bar{1}0]$ axes, respectively. This figure includes A^{-1} (open circles) in phase III. The solid line is a fit to A^{-1}.

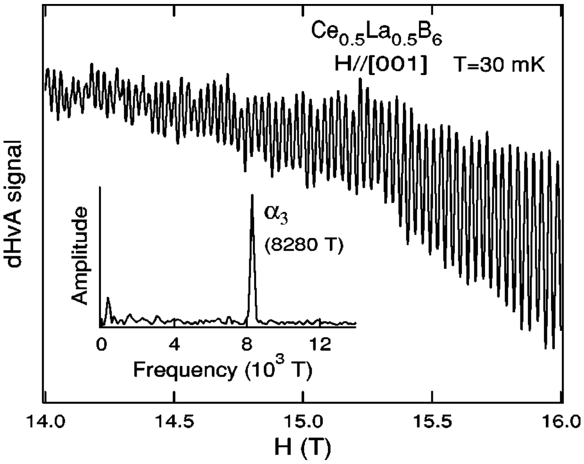

FIG. 4. The dHvA signal of $Ce_{0.5}La_{0.5}B_6$. Fields are applied along the [001] axis. Fourior spectrum is presented in the inset.

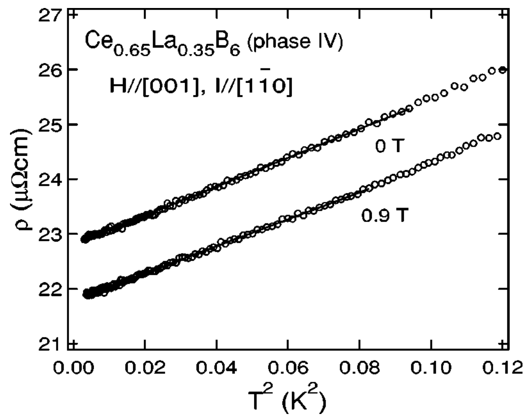

FIG. 6. The electrical resistivity of $Ce_{0.65}La_{0.35}B_6$ in phase IV shown as a function of T^2. The fields and current are directed along the [001] and $[1\bar{1}0]$ axes, respectively. The solid lines are fits.

We show the specific heat of $Ce_{0.5}La_{0.5}B_6$ under the fields applied along the [001] axis in Fig. 5. This system is in phase III at low temperatures in this field range. The arrows indicate the phase I–phase III or phase II–phase III transition points. The specific heat follows a formula $C/T=\gamma+\beta T^D$ in phase III. Similar to the case of CeB_6,[25] the βT^D term can be explained by the contribution of AFM magnon. The γ coefficient for $Ce_{0.5}La_{0.5}B_6$ in phase III is found to be 1.53, 1.45, 0.40, and 0.71 J(Ce mol)$^{-1}$K^{-2} for $H=1.85$, 2.0, 2.5 and 3.0 T, respectively. The ratio A/γ^2 obtained are 1.1×10^{-5} and 2.2×10^{-5} $\mu\Omega$ cm(Ce mol K/mJ)^2 for 2.0 and 3.0 T, respectively. These values roughly agree with the ratio widely observed in heavy fermion systems.[26]

In Fig. 2(b) we show the electrical resistivity of $Ce_{0.5}La_{0.5}B_6$ in phase I down to ~60 mK as a function of T^D. It is noted that the resistivity follows the formula $\rho(T)=\rho(0)+AT^D$ below ~300 mK, where D is not equal to 2.[27] The resistivity for 0 T shows a $T^{1.5}$ behavior indicating a NFL. The exponent D increases slightly in the neighborhood of the phase–I-phase III transition point.

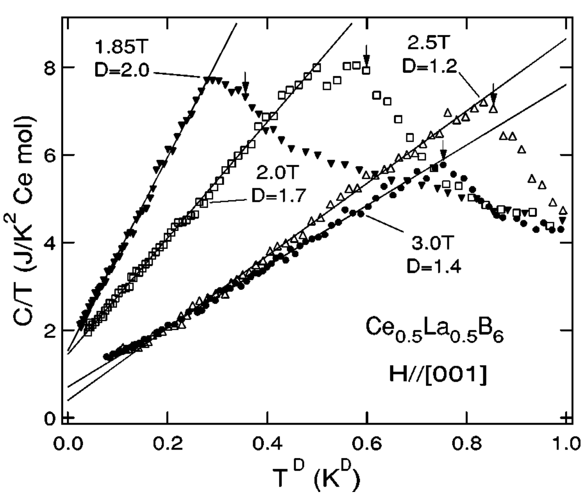

FIG. 5. The specific heat over temperature of $Ce_{0.5}La_{0.5}B_6$ per Ce mole as a function of T^D. The arrows indicate the phase I–phase III or phase II–phase III transition points and this system is in phase III at low temperatures. Solid lines are fits by the formula $C/T=\gamma+\beta T^D$.

For comparison, let us see the electrical resistivity of $Ce_{0.65}La_{0.35}B_6$ in phase IV, which locates at the higher concentrated side of x_c. In this system the quadrupolar degeneracy is lifted in association with the generation of spontaneous lattice distortion. Figure 6 shows the electrical resistivity of this system as a function of T^2. The field and current are directed along the [001] and $[1\bar{1}0]$ axes, respectively. The resistivity shows the FL behavior $\rho(T)=\rho(0)+AT^2$ from ~300 mK down to at least ~60 mK regardless of the disorder of Ce ions. The coefficient A is found to be 26.2 and 24.5 $\mu\Omega$ cm K^{-2} for 0 and 0.9 T, respectively, implying strong correlation between the conduction electrons.

In the case of $YbRh_2Si_2$, a divergence in A is observed in the vicinity of the field-induced quantum critical point (QCP) between AFM phase and paramagnetic phase.[28] One may expect that the divergence in A in phase III of $Ce_{0.5}La_{0.5}B_6$ could also be explained as a quantum critical phenomenon. However, present results are different from the case of $YbRh_2Si_2$ in the sense: (1) the AFM phase is induced by the magnetic field, (2) the characters of the phase I–phase III transition is a weak first order, (3) no divergence in A is found in the lower field side of the transition point (in the region of phase I). In phase I of $Ce_{0.5}La_{0.5}B_6$, the NFL behaviors in the resistivity are not only shown in the vicinity of the phase transition point, but in the wide field range. This feature of the resistivity in phase I is difficult to explain in terms of a quantum critical phenomenon.

Let us compare the low-T properties of $Ce_{0.5}La_{0.5}B_6$ in phase I with theoretical predictions. According to the self-consistent renormalization theory for the itinerant electron systems,[4] the resistivity shows $T^{1.5}$ behavior and the specific heat follows the formula $C/T=\gamma_0-T^{0.5}$ in the neighborhood of a QCP. As we mentioned before, however, actual C/T of $Ce_{0.5}La_{0.5}B_6$ observed at 0 T is nearly in proportion to $T^{0.5}$ at very low temperatures. In Griffiths phases, C/T and χ_m expected are both in proportion to $T^{-1+\lambda}$ ($\lambda<1$),[6] which is far from the observations[15,21] for $Ce_{0.5}La_{0.5}B_6$ at very low temperatures. According to the theory of the impurity quadrupole Kondo effect, negative slope in the resistivity and $-\log T$ temperature dependence of the specific heat are expected.[3] But none of them are observed in phase I. The behavior of the NFL in $Ce_{0.5}La_{0.5}B_6$ does not agree with any theoretical prediction. The disorder of Ce ions is not the sufficient condition for the appearance of NFL in $Ce_xLa_{1-x}B_6$.

We have reported that the NFL state appears in the region $H<1.7$ T, $x=0.5$. On the other hand, in the surrounding region in the H-x plane, that is, in phase IV of $Ce_{0.65}La_{0.35}B_6$ and in phase III of $Ce_{0.5}La_{0.5}B_6$, very strongly correlated states appear with long-range orders. Associated with the phase III–phase I transition, both the spontaneous quadruolar moments and the magnetic moments disappear simultaneously, while as described above we believe that the spontaneous quadrupolar moments disappear with the transition from phase IV to phase I. From these points we speculate that the NFL state in phase I of $Ce_{0.5}La_{0.5}B_6$ is related to the disappearance or quenching of the quadrupolar moments of the localized $4f$ electrons.

In conclusion, we have reported that the heavy fermion (HF) state and the NFL state appear in $Ce_{0.5}La_{0.5}B_6$ depending on the magnetic field applied along the [001] axis. In the field-induced phase III, the HF state coexists with the antiferromagnetic and antiferroquadrupolar orders. The correlation between the conduction electrons becomes stronger with decreasing field divergently. The HF state becomes unstable under lower fields and the NFL state without long-range order (phase I) appears. The low-temperature properties of the NFL are not consistent with any theoretical prediction. The appearance of NFL in phase I of $Ce_{0.5}La_{0.5}B_6$ may be related to the disappearance of the quadrupolar moments.

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[27] In a previous report [S. Nakamura, R. Sato, T. Nojima, M. Endo, N. Kimura, H. Aoki, and S. Kunii, Physica B **329-333**, 564 (2003)] we reported that the resistivity of $Ce_{0.5}La_{0.5}B_6$ for fields parallel to [110] showed the Fermi liquid behavior in phase I. However, in subsequent studies with more careful measurements we have found that the resistivity deviates from the Fermi liquid behavior at very low temperatures and under low magnetic fields also for $H\|[110]$.
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