Pressure-induced non-Fermi-liquid behavior in a heavy-fermion compound Ce_7Ni_3 around the antiferromagnetic instability

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Under increasing pressure, the Néel temperature of the heavy-fermion compound Ce_7Ni_3 (T_N=1.9 K for P=0) decreases and vanishes near $P_c \approx 0.33$ GPa. Non-Fermi-liquid behavior appears at 0.4 GPa in both the specific heat and ac magnetic susceptibilitiy, $C_m/T \sim -\ln T$ and $\chi_{ac} \propto (1-\alpha T^{1/2})$. Above 0.62 GPa, the normal Fermi-liquid state recovers, as indicated by the T independence of $C_m T$ and the T^2 dependence of the magnetic resistivity. The observed crossover with pressure is described by self-consistent renormalization theory of spin fluctuations (SF) in terms of the characteristic SF temperature T_0 which increases by a factor of 20 for $0.33 \leq P \leq 0.75$ GPa. [S0163-1829(97)51502-5]

Heavy-fermion compounds have been the focus of intense investigation over the last decade.[1] The low-temperature properties have been generally described within the framework of the conventional Fermi-liquid theory, while one observes huge values of the Sommerfeld coefficient, γ $[=C/T(T \rightarrow 0)]$, a Pauli-like spin susceptibility, χ, and the coefficient A of the T^2 dependence of electrical resistivity ρ with a relation, $\gamma \propto \chi \propto A^{1/2}$ (Ref. 1). Recently, non-Fermi-liquid (NFL) behavior, $C/T \sim -\ln T$, $\Delta\chi \propto -T^{1/2}$ and $\Delta\rho \propto -T$ has been reported for some U- and Ce-based alloys when the magnetic state is destroyed by the substitution of the constituent elements. A two-channel Kondo model was proposed to explain the NFL behavior, in U-based systems such as $U_{0.2}Y_{0.8}Pd_3$ (Ref. 2), $Th_{1-x}U_xRu_2Si_2$ ($x \leq 0.07$) (Ref. 3), and $U_{0.9}Th_{0.1}Be_{13}$ (Ref. 4). However, this model is not adequate to describe the NFL behavior observed in $CeCu_{5.9}Au_{0.1}$,[5] $CePtSi_{0.9}Ge_{0.1}$,[6] and $Ce_{1-x}La_xRu_2Si_2$ (Refs. 7 and 8) with orthorhombic or tetragonal site symmetry for Ce^{3+}. In both $CeCu_{1-x}Au_x$ and $CePtSi_{1-x}Ge_x$, the ground state changes from a nonmagnetic state to an antiferromagnetically ordered state near x_c=0.1, where the NFL behavior has been observed.

Recently, Moriya and Takimoto have applied the self-consistent renormalization (SCR) theory of spin fluctuations to the heavy-fermion systems near the antiferromagnetic instability.[9] It has been shown that the specific heat and resistivity exhibit the temperature variation of the NFL form, $C/T \propto -\ln T$ and $\rho \propto T^n (n \approx 1)$ in a certain range of temperature. Kambe *et al.* have used this theory to analyze C and ρ of $Ce_{1-x}La_xRu_2Si_2$ (Refs. 7 and 8) and $CeCu_{6-x}Au_x$,[8] and have shown that the NFL behavior is the consequence of antiferromagnetic spin fluctuations of $4f$ electrons with characteristic energy much smaller than that in itinerant $3d$-electron systems. They have pointed out further that the lattice disorder introduced by the alloying must be taken into account, because the SCR theory assumes a perfect lattice. Therefore, a systematic study of physical properties near the magnetic instability is desired on a heavy-fermion compound with an ordered crystal structure. In this respect, we should recall that weak magnetism is usually unstable against pressure.[10] For the antiferromagnetic heavy-fermion alloy $CeCu_{5.7}Au_{0.3}$ (T_N=0.49 K for P=0), the NFL behavior in $C(T)$ was observed at the critical pressure P_c=0.82 GPa where the Néel temperature T_N vanishes.[11]

We have chosen Ce_7Ni_3, which is a heavy-fermion antiferromagnet with T_N=1.9 K.[12–14] This compound crystallizes in the Th_7Fe_3-type hexagonal structure with three nonequivalent sites for Ce atoms.[15] Since one site and the other sites have trigonal and monoclinic symmetry, respectively, the two-channel Kondo effect is unlikely in this compound. In our previous work,[16] we found that the transition from magnetic to nonmagnetic state occurs at an extremely low pressure P_c=0.33 GPa from the measurement of ac magnetic susceptibility χ_{ac}. This low critical pressure enables us to study the whole transition from the critical regime to the Fermi liquid regime. In this paper, we report the observation of pressure-induced NFL behavior in Ce_7Ni_3 without alloying. Anomalous behaviors in $C(T)$, $\rho(T)$ and $\chi_{ac}(T)$ under high pressure will be interpreted in terms of the above-mentioned SCR theory.

Samples of Ce_7Ni_3 and La_7Ni_3 were prepared by arc melting under an argon atmosphere. From the slowly cooled ingot, small single crystals elongating along the hexagonal c axis have been obtained. The electron-probe microanalysis indicated no deviation in the stoichiometry larger than 1 at. % for the host phase and the presence of cerium oxide at approximately 1%. The heat capacity up to 0.75 GPa was measured using the ac method adapted for a high-pressure studies.[17] The sample, a thermometer of RuO_2 and a heater of moleculoy wire were lapped together in an indium sheet.

By measuring the total heat capacity of Ce_7Ni_3(3.22 mg) and the In sheet (20.31 mg), we calibrated both the pressure and the absolute value of $C(T)$; the former was determined from the known pressure dependence of the superconducting transition temperature $T_c(P)$ of In, and the latter from the jump of C at T_c.[18] The electrical resistivity under pressure up to 1.5 GPa was measured by a dc four-terminal method in the range $0.35 \leqslant T \leqslant 300$ K. The measurement of ac magnetic susceptibility was performed in an ac field of 0.18 mT at 100 Hz by using the Hartshorn bridge in the ranges $0.35 \leqslant T \leqslant 20$ K and $0 \leqslant P \leqslant 0.62$ GPa.

Figure 1(a) shows $C(T)$ of Ce_7Ni_3 and La_7Ni_3 at various pressures. For $P=0$, a λ-type anomaly appears at $T_N=1.9$ K. With increasing pressure, both the specific heat jump $\Delta C(T_N)$ and T_N decrease and vanish for $P=0.33$ GPa. The pressure dependence of T_N is consistent with that determined by the measurement of χ_{ac},[16] as shown in the inset of Fig. 2. The magnetic contribution to the specific heat C_m was estimated by the subtraction of C for La_7Ni_3. For this purpose, the value of C for La_7Ni_3 under pressures was estimated by the linear interpolation between the two values at 0 GPa and 0.69 GPa. Thus obtained, C_m/T is plotted in Fig. 1(b) as a function of ln$T$. At $P_c=0.33$ GPa, the C_m/T curve shows an upturn. At 0.38 GPa, however, C_m/T is proportional to $-\ln T$ over more than one decade in T, which is the NFL behavior. At a higher pressure of 0.54 GPa, C_m/T has a downward curvature below 4 K. Above 0.62 GPa, C_m/T is saturated at low temperatures, indicating the recovery of the normal Fermi-liquid state.

In order to confirm the transition from the NFL behavior to the Fermi-liquid behavior, we present in Fig. 2 the data of χ_{ac} vs $T^{1/2}$ at selected pressures between 0.40 GPa and 0.62 GPa. At 0.40 GPa, the NFL behavior, $\chi_{ac} \sim -T^{1/2}$, is observed only below 1 K, while at 0.49 GPa it is observed up to 5 K. At 0.62 GPa, χ_{ac} becomes almost independent of temperature, again indicating the recovery of Fermi-liquid behavior. It is noteworthy that the value of χ_{ac} at 0.6 K is reduced by one order of magnitude in the measured pressure range.

The SCR spin fluctuations theory involves the following factors:[9] the staggered susceptibility at 0 K, $\chi_Q(0)$ (Q is the antiferromagnetic ordering wave vector), the exchange energy J_Q [roughly of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction] with an assumed dispersion $J_Q - J_{Q+q} = Dq^2$ up to the effective Brillouin zone vector q_B, and the local dynamical susceptibility described as $\chi_L(\omega)=\chi_L/(1-i\omega/\Gamma_L)$. By combining these factors, the characteristic SF energy in the momentum space is given by $T_A = Dq_B^2/2$, while that in the energy space by $T_0 = T_A \Gamma_L \chi_L/\pi$. The parameter y_0 is connected to T_A and χ_Q through the relation $y_0 = 1/[2T_A\chi_Q(0)]$, and $y_0=0$ at the critical boundary. The static uniform susceptibility χ_n at $T \approx 0$ K is described as $\chi_n = 1/[2(1-y_0)T_A]$. Hence, the observed decrease of χ_{ac} implies the increase of y_0 and/or T_A with increasing pressure. The relation of $J_Q/T_A=1$ (Ref. 9) in turn suggests that pressure increases the RKKY interaction energy J_Q.

We now apply the SCR theory to describe the observed T dependence of C_m by using three parameters y_0, χ_c, and T_0, where χ_c is the cutoff wave vector in units of q_B.[9] The solid lines in Fig. 1(b) are the results of fitting assuming

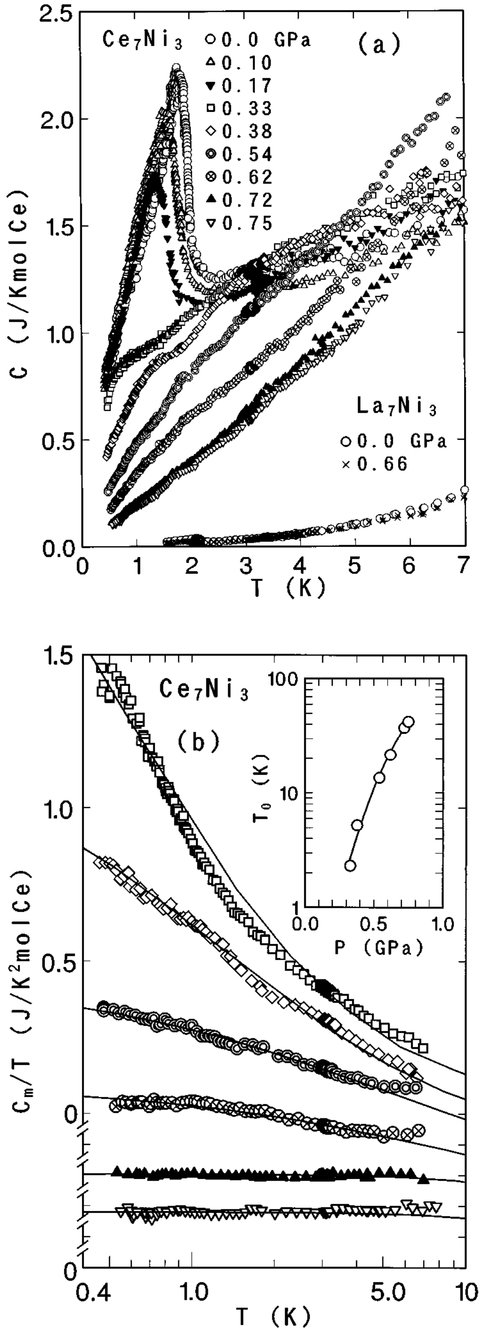

FIG. 1. (a) Temperature dependence of the specific heat of Ce_7Ni_3 and La_7Ni_3 at various pressures. (b) Magnetic specific heat divided by temperature C_m/T above 0.33 GPa as a function of ln$T$. Data for each P are shifted downward consecutively by 0.1 J/K^2 mol Ce for clarity. Solid lines indicate fits by the SCR theory (see text). The inset shows the pressure dependence of T_0, the characteristic temperature of spin fluctuations.

$y_0=0$ for $0.33 \leqslant P \leqslant 0.54$ GPa, $y_0=0.02$ for $P=0.62$ GPa and $y_0=0.1$ for $P \geqslant 0.72$ GPa. Thus obtained, T_0 increases strongly with pressure as shown in the inset of Fig. 1(b). At the critical boundary, C_m/T is expected to follow the form $C_m/T=\gamma-\beta T^{1/2}$ for $T \ll T_0$.[9] This form is not observed at $P=0.33$ and 0.38 GPa down to 0.5 K because this temperature is not sufficiently below T_0. At $P=0.54$ GPa, however, C_m/T follows the above form between 0.5 and 3 K, being

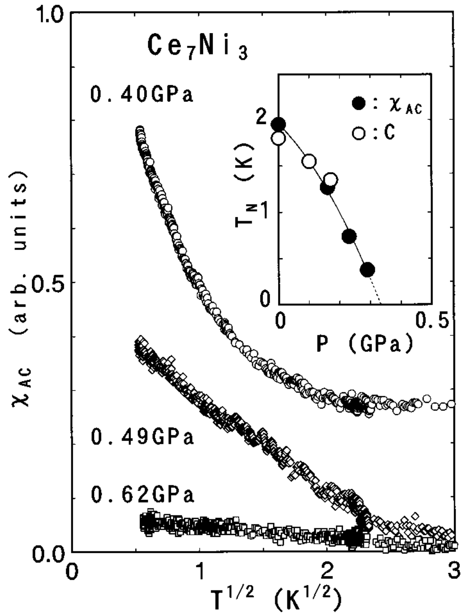

FIG. 2. ac magnetic susceptibility χ_{ac} of Ce_7Ni_3 as a function of $T^{1/2}$ under pressure between 0.40 GPa and 0.62 GPa. The inset shows the pressure dependence of T_N inferred from ac susceptibility (●) and specific heat (○) measurements.

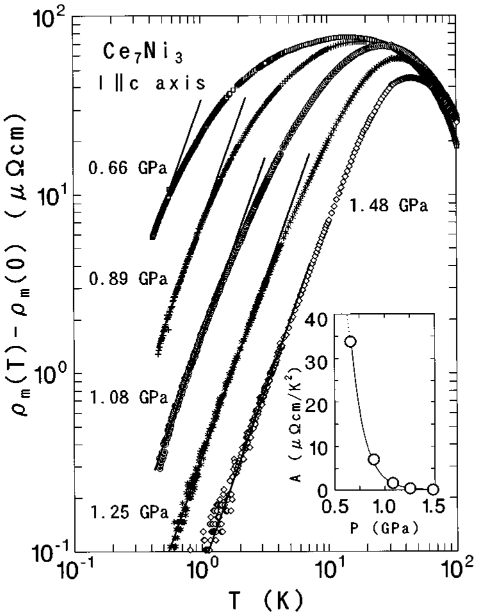

FIG. 3. Double-logarithmic plot of the magnetic contribution to electrical resistivity $\rho_m(T)-\rho_m(0)$ vs T for Ce_7Ni_3 at various pressures. Solid lines represent the form $\rho_m(T)-\rho_m(0)=AT^2$. The pressure dependence of A is shown in the inset.

far below $T_0=13.5$ K. The Grüneisen parameter $\Gamma_e=-\partial \ln T_0/\partial \ln V$ is estimated to be 220 around 0.4 GPa using the bulk modulus, $B_0=25$ GPa.[16] By contrast, in $Ce_xLa_xRu_2Si_2$ and $CeCu_{6-x}Au_x$, T_0 hardly changes near the critical boundary when the unit-cell volume is decreased by decreasing x.[19,20]

The pressure dependence of electrical resistivity along the c axis of Ce_7Ni_3 has been reported in Ref. 16. The magnetic contribution to $\rho(T)$ from $4f$ electrons was estimated by using the relation, $\rho_m=\rho(Ce_7Ni_3)-\rho(La_7Ni_3)$. Near the critical pressure $P=0.39$ GPa, $\rho_m(T)$ in the low-T range cannot be described by the power law.[16] For $P\geqslant 0.66$ GPa the relation $\rho_m(T)-\rho_m(0)=AT^2$ holds as indicated by straight lines of the double-logarithmic plot in Fig. 3. The range of T^2 dependence becomes wider and the coefficient A decreases strongly with pressure as shown in the inset of Fig. 3. This result is consistent with the enlargement of the temperature range of T-independent behavior in C_m/T above 0.62 GPa, and indicates that the Fermi-liquid state becomes stable in a larger range under pressure. According to the SCR theory, apart from the critical boundary, $\rho(T)$ is expressed as

$$\rho=r\left(\frac{\pi}{8y_0^{0.5}}\right)\left(\frac{T}{T_0}\right)^2=AT^2,$$

where r is an adjustable parameter.[9] The value of A is expected to diverge as the critical boundary is approached. i.e., $y_0\rightarrow 0$. This is what we observed in Ce_7Ni_3 below 0.66 GPa. This fact supports the assumption of $y_0=0$ below 0.54 GPa for the analysis of specific heat. Furthermore, the extreme depression of A for $P\geqslant 0.66$ GPa indicates the strong increase of y_0 and/or T_0, which is consistent with the result of $T_0(P)$ deduced from the specific heat.

In conclusion, we have found that Ce_7Ni_3 is the first example of the chemically ordered compound which shows non-Fermi-liquid behavior under pressure. The crossover from the NFL state to the normal Fermi-liquid state is described by the SCR theory of spin fluctuations. The strong dependence of T_0 on the volume distinguishes this system from the alloyed systems $Ce_{1-x}La_xRu_2Si_2$ and $CeCu_{6-x}Au_x$. Furthermore, the significant increase of T_A, with decreasing the volume was suggested by the strong depression of χ_{ac} under pressure. To determine the pressure dependence of T_A, inelastic neutron-scattering experiment under pressure is in progress.

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