Pressure-induced non-Fermi-liquid behavior in a heavy-fermion compound Ce$_7$Ni$_3$ around the antiferromagnetic instability

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Under increasing pressure, the Néel temperature of the heavy-fermion compound Ce$_7$Ni$_3$ ($T_N=1.9$ K for $P=0$) decreases and vanishes near $P_c\approx0.33$ GPa. Non-Fermi-liquid behavior appears at 0.4 GPa in both the specific heat and ac magnetic susceptibility, $C_m/T\sim-\ln T$ and $\chi_{ac}\sim(1-\alpha T^{1/2})$. 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 $0.33\leq P\leq0.75$ GPa. [S0163-1829(97)51502-5]

Heavy-fermion compounds have been the focus of intense investigation over the last decade. 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, $\gamma=\gamma/T\sim-\ln T$ and $\chi_{ac}\sim(1-\alpha T^{1/2})$. 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 $0.33\leq P\leq0.75$ GPa. [S0163-1829(97)51502-5]

Recently, Moriya and Takimoto have applied the self-consistent renormalization (SCR) theory of spin fluctuations to the heavy-fermion systems near the antiferromagnetic instability. It has been shown that the specific heat and resistivity exhibit the temperature variation of the NFL form, $C/T\sim-\ln T$ and $\rho\sim T^n$ ($n\approx1$) in a certain range of temperature. Kambe et al. have used this theory to analyze $C$ and $\rho$ of Ce$_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ (Refs. 7 and 8) and CeCu$_6\text{Au}_4$ and have shown that the NFL behavior is the consequence of antiferromagnetic spin fluctuations of 4$f$ electrons with characteristic energy much smaller than that in itinerant 3$d$-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. For the antiferromagnetic heavy-fermion alloy CeCu$_{5.7}\text{Au}_{0.3}$ ($T_N=0.49$ 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.

We have chosen Ce$_7$Ni$_3$, which is a heavy-fermion antiferromagnet with $T_N=1.9$ K.12–14 This compound crystallizes in the Th-Fe$_2$-type hexagonal structure with three nonequivalent sites for Ce atoms. 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 $\chi_{ac}$. 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$_7$Ni$_3$ 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$_7$Ni$_3$ and La$_3$Ni$_3$ 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 molecule wire were lapped together in an indium sheet.
By measuring the total heat capacity of Ce$_7$Ni$_3$(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 $T_c$. The electrical resistivity under pressure up to 1.5 GPa was measured by a dc four-terminal method in the range $0.35 \leq T \leq 300$ K. The measurement of ac magnetic susceptibility was performed in an ac field of 0.18 mT at 100 Hz using the Hartshorn bridge in the ranges $0.35 \leq T \leq 20$ K and $0 \leq P \leq 0.62$ GPa.

Figure 1(a) shows $C(T)$ of Ce$_7$Ni$_3$ and La$_7$Ni$_3$ at various pressures. For $P = 0$, a $\lambda$-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 $\chi_{ac}$, 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$_7$Ni$_3$. For this purpose, the value of $C$ for La$_7$Ni$_3$ 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 $\chi_{ac}$ 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, $\chi_{ac}$ becomes almost independent of temperature, again indicating the recovery of Fermi-liquid behavior. It is noteworthy that the value of $\chi_{ac}$ at 0.6 K is reduced by one order of magnitude in the measured pressure range.

The SCR spin fluctuations theory involving the following factors: 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 $J_Q = D q^2$, up to the effective Brillouin zone vector $\mathbf{q}_B$, 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 $T_s = D q_B^2/2$, while that in the energy space by $T_0 = T_s \Gamma_L / \pi$. The parameter $y_0$ is connected to $T_s$ and $\chi_{ac}$ through the relation $y_{0} = 1/[2 T_s \chi_{ac}(0)]$, and $y_0 = 0$ at the critical boundary. The static uniform susceptibility $\chi_u$ at $T = 0$ K is described as $\chi_u = 1/[2(1 - y_0) T_s]$. Hence, the observed decrease of $\chi_{ac}$ implies the increase of $y_0$ and/or $T_s$ 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 $y_0$, $\chi_c$, and $T_0$, where $\chi_c$ is the cutoff wave vector in units of $\mathbf{q}_B$. The solid lines in Fig. 1(b) are the results of fitting assuming $y_0 = 0$ for $0.33 \leq P \leq 0.54$ GPa, $y_0 = 0.02$ for $P = 0.62$ GPa and $y_0 = 0.1$ for $P > 0.72$ 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 $T \ll T_0$. 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...
far below \(T_0=13.5\) K. The Grüneisen parameter \(\Gamma_c=-\partial \ln \rho_0/\partial \ln V\) is estimated to be 220 around 0.4 GPa using the bulk modulus, \(B_0=25\) GPa.\(^{16}\) By contrast, in \(\text{Ce}_x\text{La}_{1-x}\text{Si}_2\) and \(\text{CeCu}_6-x\text{Au}_x\), \(T_0\) 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 \(\text{Ce}_7\text{Ni}_3\) has been reported in Ref. 16. The magnetic contribution to \(\rho(T)\) from \(4f\) electrons was estimated by using the relation, \(\rho_m=\rho(\text{Ce}_7\text{Ni}_3)-\rho(\text{La}_7\text{Ni}_3)\). 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 \(P=0.66\) GPa the relation \(\rho_m(T)-\rho_m(0)=A T^2\) 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

\[
\rho = r \left( \frac{\pi}{8 \nu_0^2} \right) \left( \frac{T}{T_0} \right)^2 = A T^2,
\]

where \(r\) is an adjustable parameter.\(^9\) The value of \(A\) is expected to diverge as the critical boundary is approached, i.e., \(\gamma_0\rightarrow 0\). This is what we observed in \(\text{Ce}_7\text{Ni}_3\) below 0.66 GPa. This fact supports the assumption of \(\gamma_0=0\) below 0.54 GPa for the analysis of specific heat. Furthermore, the extreme depression of \(A\) for \(P\approx 0.66\) GPa indicates the strong increase of \(\gamma_0\) 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 \(\text{Ce}_7\text{Ni}_3\) 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 \(\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2\) and \(\text{CeCu}_{6-x}\text{Au}_x\). Furthermore, the significant increase of \(T_A\) with decreasing the volume was suggested by the strong depression of \(\chi\) under pressure. To determine the pressure dependence of \(T_A\), inelastic neutron-scattering experiment under pressure is in progress.

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