Low-temperature specific heat and magnetic susceptibility near the pressure-induced quantum phase transition in Ce_7Ni_3

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The pressure-induced magnetic-nonmagnetic transition in a stoichiometric compound Ce_7Ni_3 was investigated using measurements of the specific heat C and the magnetic susceptibility χ on single crystals down to 0.07 K. The results show that the Néel temperature T_N decreases linearly with increasing pressure, and vanishes at $P_c = 0.39$ GPa. At 0.36 GPa $< P_c$, the C_m/T curve deviates strongly from $-\ln T$ dependence below 0.5 K, and exhibits a broad maximum around 0.15 K. At 0.43 GPa $> P_c$, while, on the other hand, a T-independent behavior appears below 0.2 K. These findings indicate that the $-\ln T$ dependence observed near P_c between 0.5 and 6 K is a crossover phenomenon to a Fermi-liquid ground state. However, the continuous increase of χ down to 0.09 K at 0.43 GPa is at variance with conventional Fermi-liquid theory. [S0163-1829(98)04542-1]

I. INTRODUCTION

In recent years, non-Fermi level (NFL) behavior in heavy-fermion alloys and compounds has received much attention. Anomalous temperature dependences of the specific heat C, magnetic susceptibility χ, and electrical resistivity ρ have been found: $C/T \propto -\ln T$, $\chi \propto (1-T^{1/2})$, and $\rho \propto T$.[1–10] This anomalous behavior has been attributed to distinct microscopic origins in different materials. A two-channel quadrupolar Kondo effect has been proposed to describe the NFL behavior in $U_{0.2}Y_{0.8}Pd_3$.[1] A distribution of Kondo temperatures T_K due to the disorder in $UCu_{5-x}Pd_x$ ($x = 1.0$ and 1.5), reproduced the anomalous temperature dependence of the specific heat and susceptibility above 0.3 K.[2] However a spin-glass transition was found for UCu_4Pd at lower temperatures.[3] The self-consistent renormalization (SCR) theory of spin fluctuations[4] has been applied to describe the specific heat and resistivity of $Ce_{1-x}La_xRu_2Si_2$ near the magnetic instability.[5] An antiferromagnetic phase transition suppressed to $T = 0$, i.e., a quantum phase transition, has been considered as the origin of the NFL behavior in some alloys, notably $CeCu_{6-x}Au_x$ (Ref. 6) and $CePtSi_{1-x}Ge_x$.[7] Suppression of antiferromagnetism in $CeCu_{6-x}Au_x$ for $x = 0.2$ and 0.3 has also been realized by applying pressure.[8] More recently, NFL behavior has been found even in chemically ordered compounds $CeNi_2Ge_2$,[9] $CeCu_2Si_2$ (Refs. 9 and 10), and Ce_7Ni_3.[11,12]

For Ce_7Ni_3, Sereni *et al.*[13] reported that an antiferromagnetically ordered state below $T_N = 1.8$ K coexists with a heavy-fermion state ($\gamma = 1.3$ J/K^2 mol Ce). This compound crystallizes in the hexagonal Th_7Fe_3-type structure and hence shows a uniaxial magnetic anisotropy.[14] The magnetization curve for $B \parallel c$ at 1.4 K exhibits a metamagnetic transition at 0.2 T.[15] The antiferromagnetism is suppressed under pressure and vanishes around 0.4 GPa, where NFL behavior appears in both the specific heat and magnetic susceptibility in the temperature range between 6 and 0.5 K.[11,12] This behavior is not expected to arise from a wide distribution of Kondo temperatures T_K, because Ce_7Ni_3 has a chemically ordered crystal structure. However, the Ce ions at the three nonequivalent sites in Ce_7Ni_3 might have a different T_K. A two-channel quadrupolar Kondo effect is also unlikely due to the trigonal and monoclinic symmetry of the Ce sites in this compound. Hence, only a quantum phase transition seems to be left to explain the NFL behavior of Ce_7Ni_3. In the previous work,[12] we have shown that the temperature dependence of *C/T* between 5 and 0.5 K at pressures above 0.4 GPa is reproduced well by SCR theory.[4] It is important to examine whether the $-\ln T$ dependence of *C/T* holds to lower temperature or *C/T* saturates to a constant value as is predicted by the SCR theory for three-dimensional antiferromagnets. In this paper, we report the results of the measurements of C and χ down to 0.07 K for single crystals of Ce_7Ni_3 under various pressures.

II. EXPERIMENTAL PROCEDURE

Single crystals of Ce_7Ni_3 were grown by a Czochralski method using a hot tungsten crucible in an rf induction furnace. As starting materials, we used high-purity Ce and Ni metals produced by Ames Laboratory and Johnson Matthey Ltd., respectively. The crystal orientation was determined by the backscattering Laue method.

The magnetization under pressures up to 1.2 GPa was measured by an extraction method with a clamp-type piston cylinder pressure cell at ISSP, University of Tokyo. The pressure cell was immersed in liquid ^{3}He and cooled down to 0.5 K. The detailed technique of the magnetization measurement is described elsewhere.[16]

The measurements of the heat capacity C and the ac susceptibility χ_{ac} under pressure have been done down to 0.07 K in a ^{3}He-^{4}He dilution refrigerator at the University of Karlsruhe. For these measurements, we used a standard heat-pulse and an ac mutual inductance method, respectively. A single-crystalline sample of ~100 mg with a cylindrical shape along the c axis was put into the Cu-Be pressure cell. We used a methanol-ethanol mixture as a pressure transmitting medium. The coils for the ac susceptibility measurements were mounted outside of the pressure cell, and were also used to measure the superconducting transition of a small Sn piece to determine the pressure. The frequency and amplitude of ac field were 129.5 Hz and less than 0.02 mT, respectively. The contributions of the pressure cell to C and χ_{ac} were determined in separate measurements without the sample and subtracted from the raw data. For the ac susceptibility the T dependence of the pressure cell between 0.09 and 3 K is small compared to that of the sample for most pressures. For $P=0.43$ GPa, with a weak T dependence of χ_{ac}, it amounts to 30%. Therefore although the subtraction yields only to arbitrary units for the sample contribution to χ_{ac}, the positions of the maxima are well defined. In order to facilitate a qualitative comparison for different pressures, a rough relative calibration is obtained by comparing the ac and dc susceptibility for $P=0$ and 0.11 GPa. The sample contribution to the total heat capacity (pressure cell, thermometer, heater, and sample) varies between 60 and 90% for temperatures between 0.1 and 3 K for $P=0.11$ GPa, and between 50 and 70% for $P=0.43$ GPa where the sample contribution is lowest. Taking into account the heat capacity error of 2%, the error for the specific heat is 6% maximally for $P=0.43$ GPa.

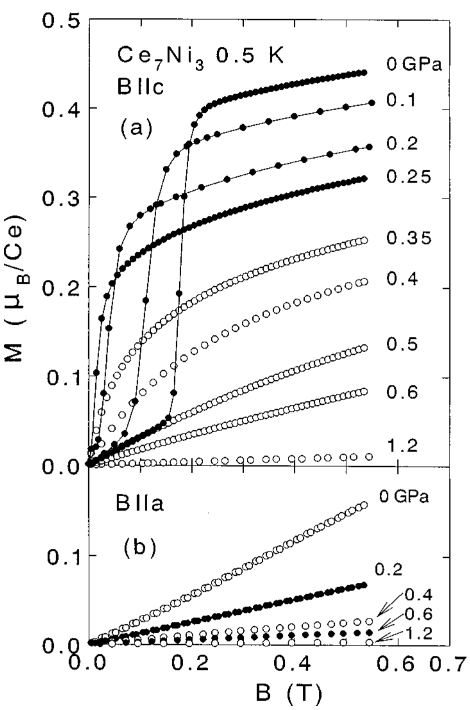

FIG. 1. Isothermal magnetization curves of Ce_7Ni_3 at 0.5 K for $B \| c$ and $B \| a$ under various pressures.

III. RESULTS AND DISCUSSION

The isothermal magnetization curves $M(B)$ at 0.5 K for $B \| c$ and $B \| a$ are shown in Fig. 1. At $P=0$, a sharp metamagnetic transition occurs at 0.18 T only for $B \| c$, which is in agreement with the previous observation.[15] With increasing pressure up to 0.25 GPa, the transition field B_c decreases at the rate of $dB_c/dP=-0.71$ GPa, and for 0.35 GPa, no metamagnetic transition was observed at 0.5 K. For $P \geqslant 0.35$ GPa, the magnetization curve is strongly depressed within creasing pressure, and the value of 0.55 T is only 0.011 μ_B/Ce for $P=1.2$ GPa. Likewise, $M(B \| a)$ decreases with pressure, albeit at a slower rate. The ratio $M(B \| c)/M(B \| a)$ at 0.55 T reaches a maximum of 7.7 at 0.4 GPa and decreases to 3.8 at 1.2 GPa. Thus, strong anisotropy survives in the paramagnetic state.

The magnetic susceptibility $\chi_{dc}=M/B$was determined in the low-field range where M is proportional to B. For $B \| c$ and $P \leqslant 0.25$ GPa, the magnitude of B was chosen in the range below B_c, and for $P \geqslant 0.35$ GPa, B was kept at 0.05 T. Figure 2 shows the temperature dependence of χ_{dc}. For $B \| c$, χ_{dc} at $P=0$ exhibits a peak at 1.9 K due to the antiferromagnetic transition. By contrast, $\chi_{dc}(B \| a)$ shows no anomaly at T_N and the temperature dependence levels off below 0.7 K. The previous specific heat and ac susceptibility data for P

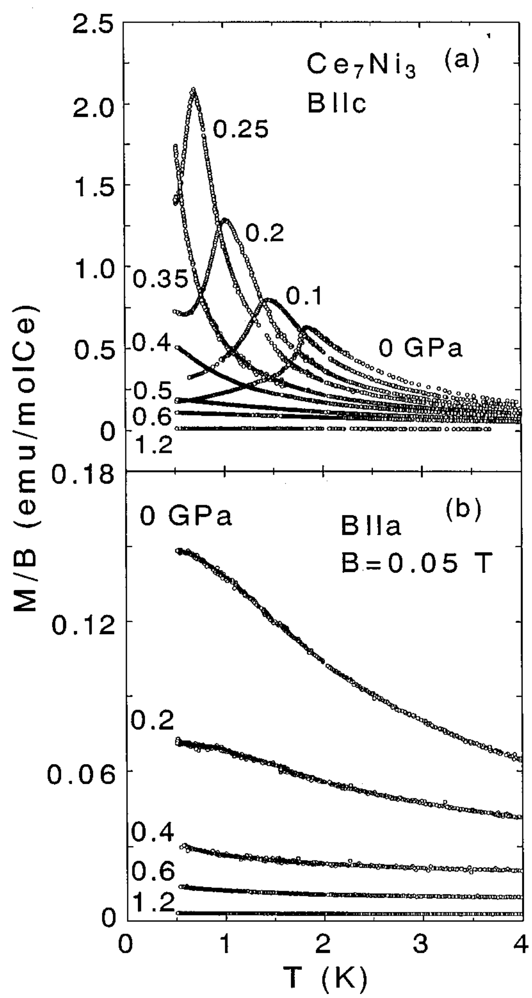

FIG. 2. Temperature dependence of the magnetic susceptibility M/B of Ce_7Ni_3 for $B \| c$ and $B \| a$ under various pressures.

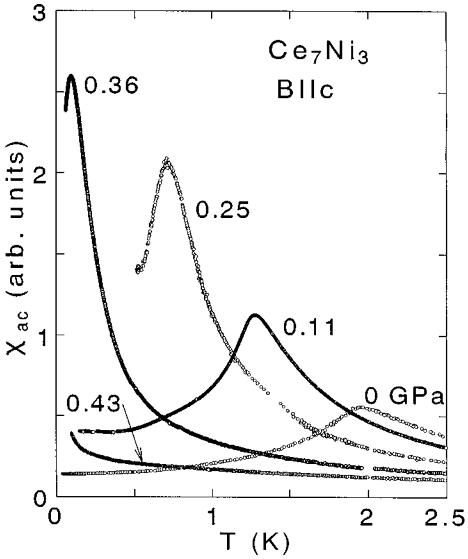

FIG. 3. Temperature dependence of χ_{ac} of Ce_7Ni_3 for $B \| c$. The data at 0.25 GPa represents $\chi_{dc}(T)$.

$=0$ suggested the presence of another magnetic transition at 0.6 K.[13] The saturated behavior in $\chi_{dc}(B\|a)$might be due to the onset of this transition. With increasing pressure up to 0.25 GPa, the peak height in χ_{dc} increases, but in the paramagnetic state for $P \geqslant 0.4$ GPa the low-temperature value of χ_{dc} decreases strongly. At 1.2 GPa, χ_{dc} for both $B\|c$ and $B\|a$ shows Pauli paramagnetic behavior with 0.01 and 0.003 emu/mol Ce, respectively.

The significant change in the magnetic susceptibility near the critical pressure has been further studied by the measurement of ac susceptibility χ_{ac} down to 0.07 K. The temperature dependences of χ_{ac} for $B\|c$ under various pressures are shown in Fig. 3, together with the data of χ_{dc} at 0.25 GPa which corroborates the results for $\chi_{ac}(T)$. For 0.36 GPa, $\chi_{ac}(T)$ exhibits a maximum at 0.096 K with the maximum value larger than that for 0.25 GPa. For 0.43 GPa, $\chi_{ac}(T)$ is much smaller and it gradually increases with decreasing temperature down to 0.09 K without any peak.

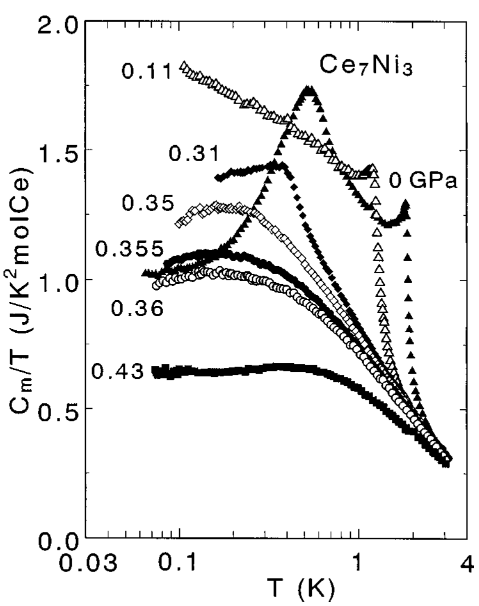

FIG. 4. Magnetic contribution to the specific heat C_m/T against $\ln T$ under various pressures.

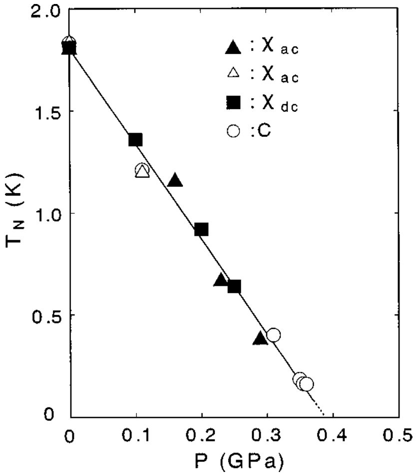

FIG. 5. Pressure dependence of the magnetic transition temperature T_N for Ce_7Ni_3 determined by the measurements of ac [▲ (Ref. 12), △] and dc susceptibility (■) and specific heat (○).

Figure 4 shows the magnetic contribution to the specific heat divided by temperature, C_m/T. The specific heat for La_7Ni_3 was subtracted as an estimate for the nonmagnetic contribution, which is less than 3% of *C/T* for Ce_7Ni_3 at 1 K.[11,12] The two magnetic transitions manifest themselves in the two maxima in the zero-pressure data at 1.8 and 0.6 K. At 0.11 GPa, the maximum at the higher temperature shifts to 1.2 K, whereas the other one disappears. At 0.35 GPa, *C/T* exhibits only a broad maximum centered at 0.2 K, which further broadens for 0.36 GPa. This broad maximum around 0.15 K can be ascribed to some type of magnetic transition because $\chi_{ac}(T)$ has a peak at 0.096 K as shown in Fig. 3. The behavior in both C_m/T and χ_{ac} is reminiscent of a spin-glass transition.[17] However, because a neutron diffraction study confirmed the chemically ordered structure of Th_7Fe_3 type,[18] the magnetic transition of this compound at 0.36 GPa appears unlikely to be a spin-glass transition. For 0.43 GPa, C_m/T levels off to a temperature independent value of 0.68 J/K^2 mol Ce below 0.2 K, indicating the recovery of the Fermi-liquid state with strongly enhanced effective electron mass. However, C_m/T exhibits a shallow maximum around 0.4 K. Since no anomaly appears in $\chi_{ac}(T)$ there, this maximum cannot be attributed to a magnetic transition.

The above results of *C*, χ_{dc}, and χ_{ac} allow us to determine the pressure dependence of T_N as shown in Fig. 5. We took the temperature of the maximum of $d(\chi T)/dT$ as T_N. For the specific heat, the position of the maximum of $C_m(T)$ is practically indistinguishable from those of C_m/T for $P \leqslant 0.31$ GPa. For larger *P*, a distinctive feature in $C_m(T)$ does not occur. However, a maximum in C_m/T is still observed up to 0.36 GPa. We therefore tentatively took these temperatures as an indication of magnetic order. With increasing pressure *P*, T_N decreases linearly and the relation $T_N = 1.81$ (K) $-4.65P$ (K/GPa) yields the critical pressure $P_c = 0.39$ GPa where $T_N \rightarrow 0$. The dependence of T_N near the quantum critical point was investigated theoretically by Millis using a renormalization group (RG) theory.[19] For an effective dimension $d=3$, the RG results confirm those of the self-consistent renormalization (SCR) theory of spin fluctuations.[4] The RG theory predicts that T_N varies as $|\delta - \delta_c|^{z/(z+1)}$ where δ and z are a relevant scaling variable and dynamical exponent, respectively. It was assumed that δ can be tuned by pressure, i.e., $|\delta - \delta_c| \sim |P - P_c|$. For the antiferromagnetic case with $z=2$, we expect that T_N varies as $(P_c - P)^{2/3}$. However, in certain systems, $CeCu_{5.7}Au_{0.3}$ (Ref. 8) and $CePd_2Si_2$,[20] the variation of T_N was found to be linear in $(P_c - P)$ as observed in the present case. This point requires further study. We note that $d=2$ for the critical fluctuations leads to a linear T_N dependence on P.[19,21]

We now discuss the specific heat of Ce_7Ni_3 in the vicinity of P_c. As mentioned above, the C_m/T curve at 0.36 GPa deviates strongly from the $-\ln T$ dependence below 0.5 K and exhibits a broad maximum. On the other hand, for $P = 0.43$ GPa, i.e., not far from the critical pressure $P_c = 0.39$ GPa, C_m/T is practically constant between 0.07 and 0.5 K, compatible with Fermi-liquid behavior. This behavior is in marked contrast to that of $CeCu_{6-x}Au_x$ under pressure. For $x=0.2$ and 0.3, an appreciable pressure range near P_c exists where *C/T* exhibits the characteristic $-\ln T$ dependence.[8] For Ce_7Ni_3, however, it is apparent from Fig. 4 that even for $P = P_c$, C_m/T is unlikely to show a $-\ln T$ dependence over a large temperature range as it might be bounded by the two curves for $P=0.36$ and 0.43 GPa.

We now turn to the shallow maximum in C_m/T around 0.4 K observed at 0.43 GPa. Recently, similar behavior in the specific heat of $CeNi_2Ge_2$ under magnetic field has been reported.[22] For zero field, *C/T* follows the form of $C/T = \gamma - \beta T^{1/2}$. In fields $B > 4$ T, *C/T* shows a broad maximum at around 1 K, below which *C/T* becomes independent of temperature. This maximum of *C/T* can be modeled by assuming a density of states (DOS) with a peak at the Fermi energy E_F, i.e., $D(\varepsilon) = D_0 - D_1|\varepsilon|^{1/2}$. This form of $D(\varepsilon)$ is consistent with the theory which takes the effect of spin fluctuations on the self-energy of quasiparticles into consideration.[23] By using this model, we could not reproduce the data of C_m/T at $P=0.43$ GPa for Ce_7Ni_3. A better fit was obtained by shifting the Fermi level above the peak of the DOS by 1.9 K, i.e., $D(\varepsilon) = D_0 - D_1|\varepsilon - \varepsilon_0|^{1/2}$, where $\varepsilon_F - \varepsilon_0 = 1.9$ K.

Although the recovery of a Fermi-liquid state at low temperatures below 0.2 K for $P=0.43$ GPa is suggested by the T-independent behavior of C_m/T, the continuous increase of χ_{ac} down to 0.09 K as shown in Fig. 3 is at variance with this interpretation. Such an increase cannot be ascribed to the effect of magnetic impurities in either the sample or the pressure cell, because χ_{ac} for $P=0.11$ GPa is constant down to 0.2 K after subtraction of the susceptibility of the empty pressure cell from the raw data. Figure 6 displays the double logarithmic plot of χ_{dc} vs T above 0.35 GPa. For $B \| c$, $\chi_{dc}(T)$ at 0.4 GPa does not follow a simple power law, and at 0.5 and 0.6 GPa the temperature dependence of χ_{dc} weakens towards low temperatures, suggesting the recovery of Fermi-liquid behavior. Interestingly, as shown in the lower part of Fig. 6, $\chi_{dc}(T)$ for $B \| a$ at both $P=0.4$ and 0.6 GPa also increases weakly with $\chi_{dc} \propto T^{-0.2}$ in the whole temperature range from 4 to 0.5 K.

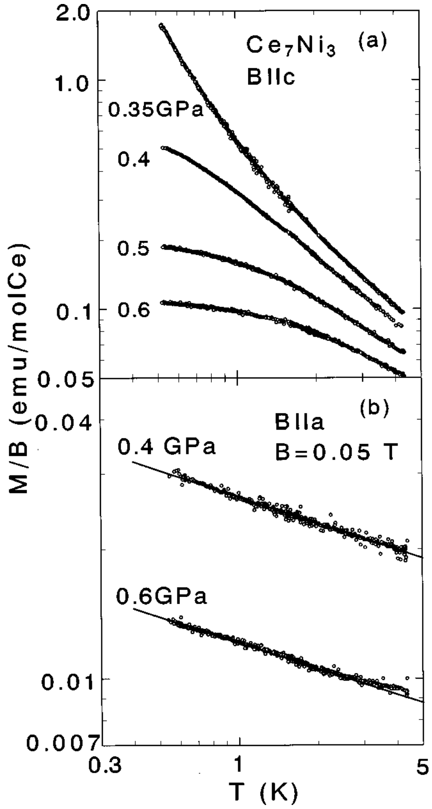

FIG. 6. Magnetic susceptibility *M/B* vs *T* on a double logarithmic plot at pressures above 0.35 GPa for Ce_7Ni_3. Solid lines are fits to a function $\chi \propto T^{-0.2}$.

In conclusion, we found that the $-\ln T$ dependence of C_m/T of Ce_7Ni_3 just above $P_c = 0.39$ GPa changes to a T-independent behavior, i.e., normal Fermi-liquid behavior at lower temperatures below 0.2 K. However, at $P=0.43$ GPa, both the broad maximum of C_m/T at 0.4 K and the continuous increase in $\chi(T)$ are not consistent with a conventional Fermi-liquid theory. These facts suggest that both $C(T)$ and $\chi(T)$ near P_c are strongly affected by spin fluctuations. In order to investigate the character of the spin fluctuations, neutron scattering experiments under pressure are in progress.

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