

NMR study of the electronic state in the dense Kondo compound CeNiAl₄

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²⁷Al NMR results are presented for a dense Kondo system CeNiAl₄ in the temperature range 3–300 K. The Knight shift K_{iso} and the spin-lattice relaxation rate T_1^{-1} exhibit a nonmagnetic behavior associated with a very low conduction electron density of states near the Fermi level. Both of them also reveal an insignificant role of 4*f* electrons in RKKY interactions in the range 50–300 K. However, below 50 K, 4*f* electrons participate in the excitation near the Fermi level and their extent of delocalization changes gradually down to 3 K. This is a consequence of a non-Fermi-liquid state below 50 K and contradicts the prediction of the existence of the heavy fermion state at low temperature from bulk measurements. Thus the present result gives a clear indication of the appearance of 4*f* moment compensation due to the predominance of the Kondo effect over RKKY interaction.

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I. INTRODUCTION

The 4*f* electrons of Ce or Yb compounds exhibit a variety of characteristics resulting in spin and valence fluctuations, spin and charge orderings, heavy fermions (HF's), Kondo insulators, and anisotropic superconductivity. The RKKY interaction and the Kondo effect are of basic importance to these magnetic properties.¹ When the Coulomb repulsive force between the 4*f* electrons at the same cerium site is strong, the freedom of the charge in the 4*f* electrons is suppressed and the freedom of the spin is retained, resulting in the localized 4*f* state. The degree of localization in the 4*f* electrons depends on the level of the 4*f* electrons E_f below the Fermi energy E_F , where a smaller E_f helps to enhance the localization. In the RKKY interaction, a localized spin of the 4*f* electrons leads to a spin polarization of the conduction electrons. This polarization interacts with another localized spin of the 4*f* electrons and creates an indirect magnetic interaction between the localized 4*f* spins. The strength of this RKKY interaction becomes stronger as the number of 4*f* electrons increases.

The impurity Kondo effect is understood as a bound state in which the localized spin of the 4*f* electrons is coupled antiferromagnetically with the spin of the conduction electrons forming a singlet state with a binding energy $k_B T_K$, where T_K is the Kondo temperature. The Kondo effect leads to the disappearance of the localized moment due to the formation of a spin-compensated Kondo cloud of conduction electrons around the impurity moment. In many Ce and Yb compounds, the 4*f* level E_f approaches the Fermi level, which brings about a hybridization between the 4*f* electrons and the conduction electrons. Strong hybridization leads to delocalization of the 4*f* electrons. The ground-state properties of the dense Kondo system, where the cerium ions are periodically aligned in the crystal, are interesting because at low temperatures the dense Kondo system is not a scattering state but a coherent state with a large effective mass of the conduction electron and a large electronic specific heat coefficient γ . The Fermi liquid state at low temperature is often called a HF state where the magnetic specific heat C_m of 4*f*

electrons is changed into electronic specific heat γT . While there is an adequate theoretical understanding of systems in the free electron limit or in the case of localized moments, systems having competing electron correlation and transfer effects can show unusual properties and are theoretically difficult to describe. It would be interesting to study the behavior of the HF materials close to the borderline from the magnetic, i.e., RKKY-interaction-dominated regime to the Kondo regime. In such a case, a non-Fermi-liquid behavior may appear due to the fact that the phase transition into a magnetically ordered state is pushed towards $T=0$ K.

Recently, among the many ternary-cerium-based intermetallic compounds, some new type of dense Kondo or valence fluctuating compounds have been found. For example, CeCu₂Si₂ (Refs. 2 and 3) is a superconducting heavy fermion system, whereas experimental evidence for heavy fermion band magnetism was obtained for Ce(Cu_{1-x}Ni_x)₂Ge₂ for $x=0.6$ and 0.7 .⁴ On the other hand, CeNiSn (Ref. 5) is an activation-type valence fluctuation system. It is therefore interesting to study such varieties of dense Kondo compounds in various ternary systems. CeNiAl₄ is a new member of the dense Kondo compounds ($T_K \approx 99$ K) in the valence fluctuation regime and is nonmagnetic down to 1.6 K, as suggested from electrical resistivity, specific heat, and magnetization measurements.^{6,7} The resistivity shows a broad maximum at around 120 K and is proportional to $-\ln T$ in the high-temperature regime around 200 K. The magnetization at 1.8 K at a field of 76.5 kOe is much smaller than that predicted theoretically. This may be explained by a strong mixing effect between the conduction electron and the 4*f* electron and/or by the crystalline electric field. On the other hand, the inverse of magnetic susceptibility follows the Curie-Weiss (CW) law in the range 100–300 K. Below 100 K, it deviates from the CW law and decreases sharply along all the three crystallographic axes.⁷ The reason for this is still not understood. The electronic specific heat coefficient γ_0 is estimated to be 175 mJ/mol K². From this it was suggested that CeNiAl₄ is in the HF state at low temperature. Furthermore, the entropy at 20 K reaches only 3.5 J/mol K which is rather smaller than $R \ln 2$. This reduction is considered to be due to the Kondo effect and/or valence fluctuation.

In order to understand the low-temperature electronic

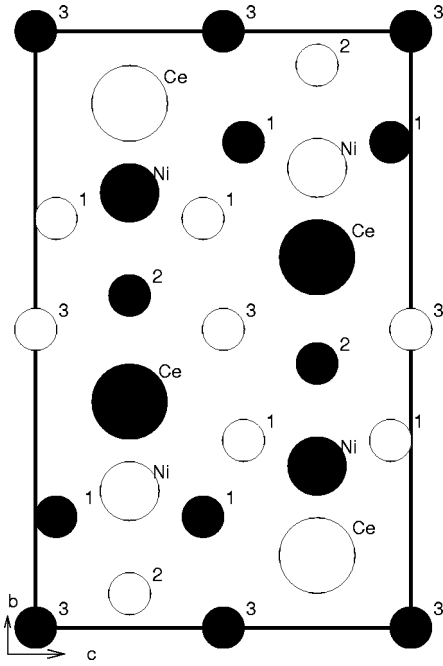


FIG. 1. Projection of the structure of CeNiAl_4 in the bc plane. The solid circles and the open circles show the atoms in the planes of $x=0$ and $x=0.5$, respectively. Al(1), Al(2), and Al(3) atoms are marked as 1, 2, and 3, respectively.

states, we have performed the detailed ^{27}Al nuclear magnetic resonance (NMR) studies in a polycrystalline sample of CeNiAl_4 in the temperature range 3–295 K. NMR is sensitive to the static and dynamic behavior of the magnetic environment of the nuclei and is therefore a useful tool for the study of exotic $4f$ magnetism.

II. EXPERIMENTAL RESULTS

The polycrystalline sample of CeNiAl_4 was prepared by repeated arc melting of the stoichiometric amounts of the constituent elements in an argon arc furnace and then subsequently annealed at 1100°C under vacuum for 3 days. The compound crystallizes in the orthorhombic YNiAl_4 structure⁸ with space group $Cmcm$ and $Z=4$ (Fig. 1). The unit cell contains three types of aluminum sites in the unit cell, viz., Al(1), Al(2), and Al(3). There are eight aluminum atoms in site 1 and four each to sites 2 and 3. X-ray diffraction patterns of the powder sample showed a single-phase compound belonging to the above space group.

^{27}Al NMR experiments were performed in a Bruker MSL100 pulse spectrometer at 24.2 MHz with a Varian V7400 electromagnet in the temperature range 3–295 K and at 78.29 MHz with a Bruker 7.04 T superconducting magnet at 100 and 295 K. In each case, temperature variation studies were made in an Oxford cryostat with ITC503 temperature controller. A homebuilt NMR probe with a silver rf coil was used to avoid spurious $^{63,65}\text{Cu}$ signals. The spectrum was recorded by applying a $\pi/2$ - τ - $\pi/2$ solid echo sequence. Knight shifts were measured with respect to the reference position (ν_R) of ^{27}Al resonance in AlCl_3 solution.

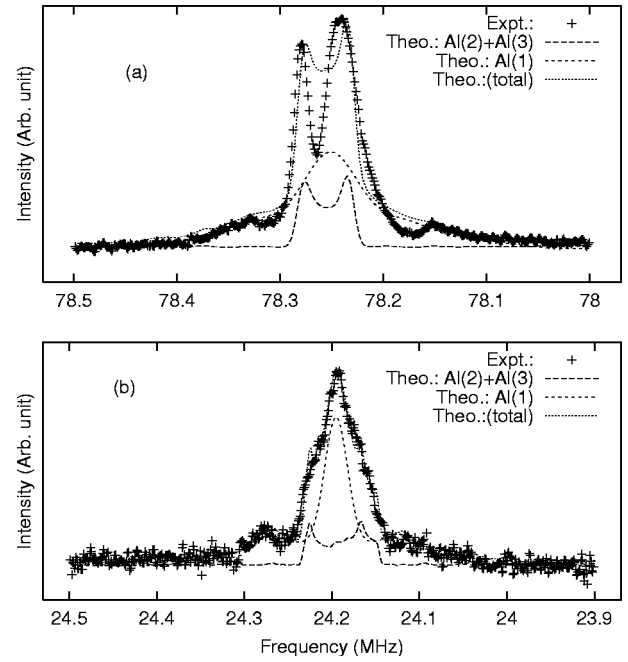


FIG. 2. ^{27}Al NMR spectra of CeNiAl_4 at 295 K: (a) $\mathbf{H}_0 = 7.04$ T and (b): $\mathbf{H}_0 = 2.17$ T.

A. Temperature variation of NMR spectra

Figure 2 shows a couple of ^{27}Al NMR spectra at 295 K at two widely different magnetic fields. It is seen that a single broad spectrum at 24.2 MHz [Fig. 2(b)] splits at 78.29 MHz [Fig. 2(a)]. The appearance of this splitting indicates that different types of aluminum sites experience unequal local magnetic fields which are more pronounced at higher frequency. Figure 3 shows some typical spectra at different temperatures at 24.2 MHz. The spectral feature changes from around 100 K. A splitting appears near the peak of the spectrum. This nature of the spectrum remains unaltered down to 50 K. Below this temperature, the splitting becomes less pro-

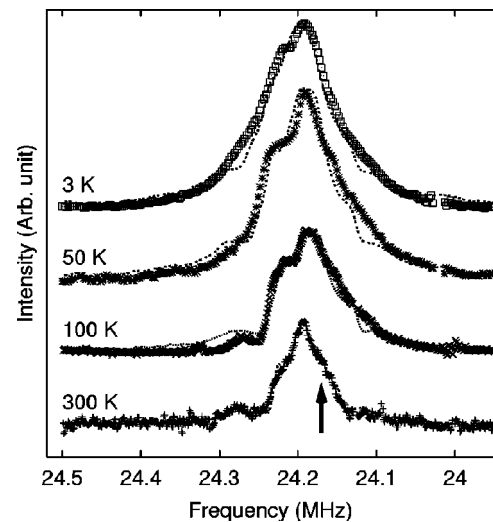


FIG. 3. Some typical ^{27}Al NMR spectra at 24.2 MHz at different temperatures. The dashed line indicates the theoretically fitted spectrum. \uparrow indicates reference position.

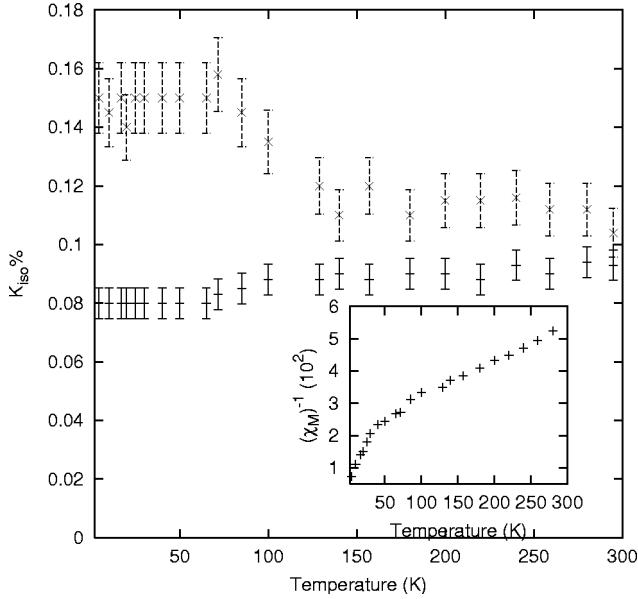


FIG. 4. Variation of K_{iso} with T at 24.2 MHz in CeNiAl_4 . \circ : Al(1). \times : Al(2) and Al(3). The inset shows the variation of $(\chi_M)^{-1}$ with T (Ref. 6).

nounced as a result of a considerable broadening of the overall linewidth. As there are three types of aluminum sites in the unit cell, the experimental line is a superposition of all three resonances. In order to extract the hyperfine interaction parameters, we have used the method of computer simulation. The validity of this procedure was checked by fitting the spectra at 295 K at two widely different resonance frequencies, 24.2 and 78.29 MHz, with almost the same set of parameters. Those which gave the best fitted spectra with the experimental line simultaneously at both frequencies were taken as the correct one, and that convention was followed for fitting the spectra down to 3 K. The experimental line was found to be a superposition of the central transition of the resonance lines of three types of aluminum sites. Figure 2 shows, along with the experimental lines, the theoretical lines as well as the constituent lines corresponding to the three types of Al sites. The constituent lines corresponding to Al(2) and Al(3) are found to be superimposed on each other in the temperature range 295–3 K and exhibit second-order quadrupolar splitting, whereas the same for Al(1) experiences a smaller quadrupolar interaction resulting in an unsplit line. The value of the intrinsic linewidth, 2β for the resonance line corresponding to Al(2) and Al(3) together, is found to be much smaller than that of Al(1). As a result, the resonance line of the former exhibits second-order quadrupolar splitting even at 78.29 MHz [Fig. 2(a)]. This is found to be consistent with respect to the Al-Al separation in the unit cell of CeNiAl_4 .⁸ The values of the quadrupolar interaction parameters at 295 K are $\nu_Q = 0.17 \pm 0.01$ MHz and $\eta = 0.1 \pm 0.02$ for the Al(1) site. Those for Al(2) and Al(3) are $\nu_Q = 1.12 \pm 0.02$ MHz and $\eta = 0.4 \pm 0.02$.

In Fig. 4, the isotropic part of the Knight shift, K_{iso} , obtained from fits to the measured NMR spectra is plotted as a function of temperature for different Al sites. In case of Al(1), K_{iso} is almost independent of temperature in the range

3–295 K. On the other hand, K_{iso} for sites Al(2) and Al(3) exhibit a small increment with lowering of temperature in the range 50–100 K, resulting in a splitting in the overall resonance line. Below this range, the magnitude of K_{iso} in each case remains temperature independent.

The effective magnetic field \mathbf{H} at the ^{27}Al nucleus in CeNiAl_4 may be written as

$$\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_c + \mathbf{H}_d^{4f} + \mathbf{H}_{\text{hf}}^{4f}, \quad (1)$$

where \mathbf{H}_0 is the applied magnetic field, \mathbf{H}_c is the contact hyperfine interaction due to purely conduction electrons, \mathbf{H}_d^{4f} is the dipolar field due to the Ce 4*f* electrons, and $\mathbf{H}_{\text{hf}}^{4f}$ is the contribution to the contact hyperfine field due to the *s-f* exchange. The \mathbf{H}_d^{4f} contributes mainly to the anisotropic part of the Knight shift. The calculated values of \mathbf{H}_d for all the Al sites were found to be negligibly small. So we will neglect this contribution to the measured Knight Shift. Therefore,

$$K_{\text{iso}}^{\text{tot}} = K_0 + K_{s-f}(T),$$

which reduces to

$$K_{\text{iso}}^{\text{tot}} = K_0 + \frac{zH_{\text{hf}}}{N_A\mu_B} \chi_M^f(T), \quad (2)$$

where K_0 is the shift due to *s* conduction electrons and $K_{s-f}(T)$ is due to *s-f* exchange interaction. H_{hf} is the transferred hyperfine field and is related to the transferred hyperfine coupling constant A_{hf} as $H_{\text{hf}} = A_{\text{hf}}/\gamma\hbar$, N is the Avogadro's number, μ_B is the Bohr magneton, χ_M^f is the Ce 4*f* electron contribution to the Bohr susceptibility, and z is the number of Ce ions bonded with aluminum nucleus. Thus as long as H_{hf} remains constant, $K_{\text{iso}}(T)$ should follow $\chi_M^f(T)$.

The inset of Fig. 4 shows the temperature dependence of the inverse of the molar susceptibility.⁶ Though χ^{-1} varies linearly with T in the range 50–295 K, K_{iso} for Al(1) does not follow this behavior. For Al(2) and Al(3), only a small temperature dependence of K_{iso} is discernable in the range 50–100 K. However, below 50 K, $(\chi_M)^{-1}$ decreases more rapidly with decreasing T . But such a behavior is again not followed in the K_{iso} vs T curve (Fig. 4). This indicates that a change in the hyperfine coupling constant might occur in this temperature region which cancels the effect of the enhancement of χ_M^f on ^{27}Al 's K_{iso} . The Knight shift result therefore suggests a modification of the hybridized 4*f*/ conduction electronic structure in the above temperature range, which alters H_{hf} . Such a type of effect was also observed in YbCuAl (Ref. 9) and CeSn_3 (Ref. 10) which exhibit Kondo behavior at low temperature and the rare-earth ions are in the fluctuating valence state. Thus the Knight shift result clearly reveals an insignificant role of the *s-f* exchange interaction as it does not produce an appreciable temperature-dependent hyperfine field at the Al nuclear site throughout the whole temperature range. It could happen because of the valence fluctuation of the Ce ion.

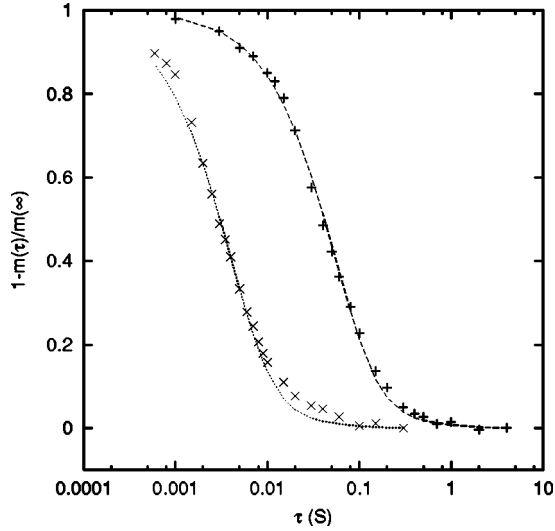


FIG. 5. Nuclear magnetization recovery $M(\tau)$ for ^{27}Al central transitions in CeNiAl_4 at 24.2 MHz. +: 4.4 K. x: 295 K.

B. Nuclear spin-lattice relaxation rates

^{27}Al spin-lattice relaxation rates $2W = T_1^{-1}$ were measured by saturating the central line only with a single $\pi/2$ pulse and monitoring the growth of the solid echo at variable delays.¹¹ T_1^{-1} was extracted from a fit to the data to the recovery law

$$\frac{M(\infty) - M(\tau)}{M(\infty)} = 0.029 \exp(-2W\tau) + 0.178 \exp(-12W\tau) + 0.794 \exp(-30W\tau), \quad (3)$$

which is obtained from the solution of the master equations in case of a magnetic relaxation of a nucleus with spin $I = 5/2$.¹² It is evident from Fig. 5 that the experimental recovery is reasonably reproduced by Eq. (3). Because of the overlapping of the constituent lines, the relaxation times of ^{27}Al in three different sites could not be measured separately, and so the measured T_1^{-1} represents an average over three Al sites. Moreover, the values of T_1^{-1} were nearly identical at the resonance frequencies of 24.2 and 78.29 MHz measured both at 100 and 295 K. This suggests that the f -electron spin fluctuation is much faster than the Larmor frequency of the ^{27}Al nucleus. This observation supports the recent x-ray photoelectron spectroscopy results in CeNiAl_4 .¹³

The temperature dependence of T_1^{-1} in CeNiAl_4 is presented in Fig. 6. The contributions to the measured T_1^{-1} may be written as

$$T_1^{-1} = T_{1K}^{-1} + T_{1f}^{-1}, \quad (4)$$

where the first term represents the Korringa contribution due to the contact interaction with conduction electrons and is given by

$$T_{1K}^{-1} = \pi \hbar^3 \gamma_e^2 \gamma_n^2 A_{\text{hf}}^2 N(E_F)^2 k_B T, \quad (5)$$

where A_{hf} is the transferred hyperfine coupling and $N(E_F)$ is the density of electronic states at the Fermi level. Hence for

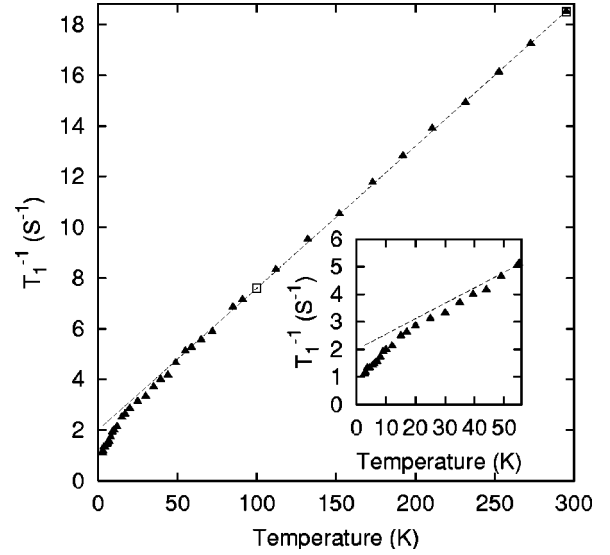


FIG. 6. Temperature dependence of ^{27}Al spin-lattice relaxation rate in CeNiAl_4 at (▲) 24.2 MHz and (□): 78.29 MHz. The dashed line corresponds to the equation mentioned in the text. The inset shows the magnified portion of the temperature region 3–50 K.

heavy fermion systems (HFS) the Korringa relaxation may also provide access to the hybridization-enhanced density of states at the Fermi level. The second term in Eq. (4) is due to spin fluctuations which will be transferred via RKKY interactions from the local moments to the site of the NMR nucleus. From the fluctuation dissipation theorem the contribution T_{1f}^{-1} for nuclear relaxation may be written as¹⁴

$$T_{1f}^{-1} = \frac{\gamma_n^2 k_B T}{2\mu_B^2} \sum_{\mathbf{Q}} |A_{\text{eff}}(\mathbf{Q})|^2 \frac{\text{Im} \chi(\mathbf{Q}, \omega_0, T)}{\omega_0}. \quad (6)$$

The effective coupling between the localized f moment and the nucleus at \mathbf{r} is given by

$$A_{\text{eff}}(\mathbf{Q}) = \sum_{\mathbf{j}} A_j(r) \exp(i\mathbf{Q} \cdot \mathbf{r}). \quad (7)$$

$A_{\text{eff}}(\mathbf{Q})$ will depend on the momentum transfer \mathbf{Q} and includes the coupling of the f spin to those of the conduction electrons and the form factor related to the hyperfine fields. If the electronic fluctuations are not spatially correlated, as is the case of CeNiAl_4 which does not order magnetically down to 1.6 K, the \mathbf{Q} -dependent part in Eq. (6) can be neglected. In this case Eq. (6) reduces to

$$T_{1f}^{-1} \propto \frac{k_B T}{\omega_0} \text{Im} \chi(\omega_0, T), \quad (8)$$

where ω_0 is the Larmor frequency. The absorptive part of the dynamic spin susceptibility can be written in a purely relaxational ansatz as

$$\text{Im} \chi = \chi_0 \frac{\omega \Gamma}{\omega^2 + \Gamma^2}, \quad (9)$$

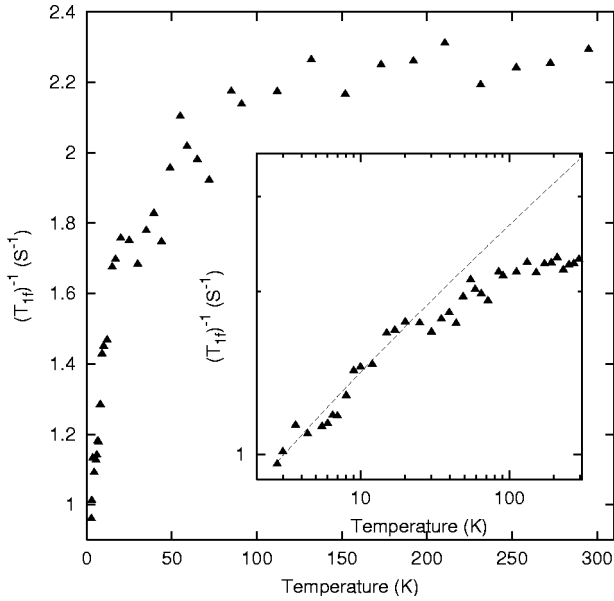


FIG. 7. Temperature dependence of Ce 4*f*-electron contribution to the relaxation rate in CeNiAl₄. The inset shows the same in logarithmic scale. The line indicates the logarithmic temperature dependence.

where Γ is the 4*f* moment relaxation rate and χ_0 is the static susceptibility. Taking into account that $\omega_0 \ll \Gamma$, Eq. (8) can be written as

$$T_{1f}^{-1} \propto k_B T \frac{\chi_0}{\Gamma_{\text{NMR}}}. \quad (10)$$

When both T_{1K}^{-1} and T_{1f}^{-1} are of comparable strength, the net relaxation rate would appear to be temperature independent.

From Fig. 6, it is seen that T_1^{-1} decreases linearly with T from 295 K down to 50 K following the relation $T_1^{-1} = 2.0 + 0.056T$. This indicates a weaker role of Ce 4*f* spin fluctuation on the ²⁷Al relaxation even above T_K compared to the purely Korringa-type conduction electron contribution. This finding also corroborates with the Knight shift result. The magnitude of the Korringa constant ($0.056 \text{ s}^{-1}\text{K}^{-1}$) is found to be order of magnitude smaller than other HF compounds like CeCu₂Si₂ ($0.6 \text{ s}^{-1}\text{K}^{-1}$)³ and YbAgCu₄ ($5.9 \text{ s}^{-1}\text{K}^{-1}$)¹⁵ though in these compounds, the Korringa constants were actually obtained from their lanthanum analogs. This suggests that the magnitude of $N(E_F)$ is much less in CeNiAl₄. Below 50 K the decreasing trend in T_1^{-1} is further enhanced down to 3 K.

By subtracting the Korringa contribution at each temperature from the measured T_1^{-1} , the contribution T_{1f}^{-1} was estimated. Figure 7 shows the variation of T_{1f}^{-1} with temperature. It is clearly seen that from 295 to 100 K, this contribution remains unaffected with temperature. From below 100 K, T_{1f}^{-1} starts to decrease slowly and from 50 K this decreasing trend is enhanced appreciably, indicating the onset of moment compensation. This result clearly shows that the Kondo effect predominates in this temperature range and therefore, suggests that CeNiAl₄ is a dense Kondo compound with T_K lying between 50 and 100 K. The inset of Fig. 7

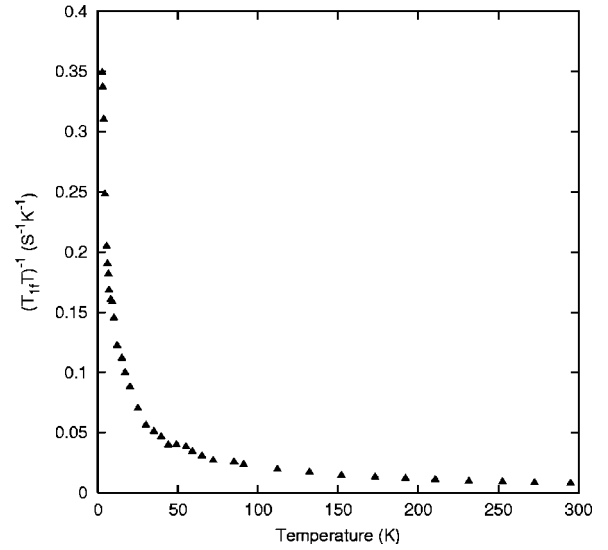


FIG. 8. Variation of the absorptive part of the dynamic susceptibility $\text{Im} \chi(\omega_0 T) \propto 1/T_{1f} T$ according to Eq. (8) in CeNiAl₄.

shows that T_{1f}^{-1} follows a power law behavior $T_{1f}^{-1} = CT^\alpha$ with $\alpha = 0.24$ and $C = -0.3$, in the temperature range 3–20 K. The value of α is significantly below unity, which is the value expected for a Fermi liquid. From the specific heat measurement it was suggested from the value of the Sommerfeld coefficient at $T = 0$ K (γ_0) that CeNiAl₄ was in the HF state. If it is indeed the case, then $(T_{1f} T)^{-1}$ should be constant as is expected for a pure heavy Fermi liquid. In Fig. 8, $(T_{1f} T)^{-1}$ is plotted as a function of T . It is seen that within the range 295–50 K, $(T_{1f} T)^{-1}$ is almost constant, indicating a negligible role of Ce 4*f* spin fluctuation on Al nuclear relaxation. Whereas below 50 K, it increases sharply without following a Korringa relation as shown in Eq. (4). This suggests that the 4*f* electrons participate in the excitation near the Fermi level and that the extent of delocalization changes gradually with decreasing temperature. Thus the present result clearly shows the non-Fermi-liquid behavior in the Kondo regime of CeNiAl₄. According to Eq. (6), $(T_{1f} T)^{-1}$ corresponds to the imaginary part of the dynamic susceptibility. It is to be noted that in the same temperature region, i.e., 3–50 K, where $\text{Im} \chi(\omega, T)$ increases sharply, the static susceptibility (χ_0), which is χ_M^f in Eq. (2) also showed a similar behavior. This suggests that the increase in χ_0 below 50 K is an intrinsic feature. Using the experimental values of T_{1f} and the static susceptibilities of CeNiAl₄ we calculated the magnetic relaxation rate Γ_{NMR} on the basis of Eq. (10). The variation of Γ_{NMR} with T is shown in Fig. 9. In the range 50–295 K, Γ_{NMR} closely follows a square root of T dependence as theoretically predicted by Cox *et al.*¹⁶ The prefactor 0.015 depends on the degeneracy of the ground multiplet and on $(\sqrt{T_K})^{-1}$. It is seen that below 50 K, there is a tendency of the experimental results to deviate from \sqrt{T} dependence. For $T < T_K$, a temperature-independent contribution is expected theoretically for $S = 1/2$ Anderson model.¹⁶

Theoretically non-Fermi-liquid behavior is expected to occur close to a quantum phase transition,¹⁷ but it can also

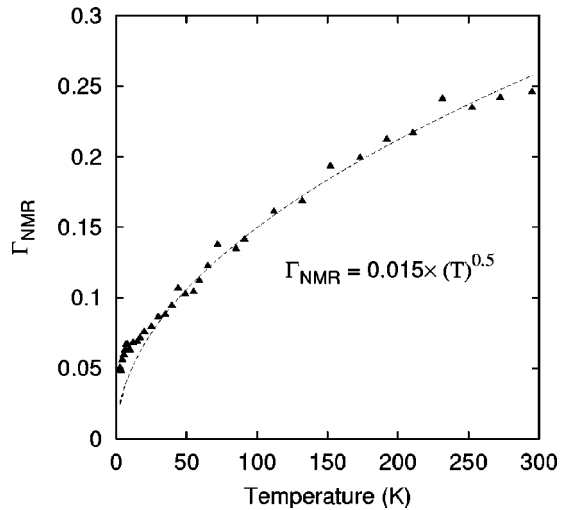


FIG. 9. Magnetic relaxation rate in CeNiAl₄ derived from NMR measurements $\Gamma_{\text{NMR}} \propto T_1 T \chi_0$. The dashed line is calculated using the relation shown in figure.

occur in multichannel Kondo systems¹⁸ or in impurity systems.¹⁹ In disordered Kondo systems the low-temperature thermodynamics also deviates from a Fermi liquid behavior and is characterized by a diverging susceptibility and specific heat rising faster than T , a behavior which is indeed observed below 9 K in C/T and below 50 K in the magnetic susceptibility behavior in CeNiAl₄.

III. CONCLUSION

We have presented the results of a detailed ²⁷Al NMR study in the dense Kondo system CeNiAl₄. The temperature

dependence of the line shape and the spin-lattice relaxation rate, T_1^{-1} were measured in the range 3–295 K. Both Knight shift and the T_1^{-1} clearly indicate that this compound is non-magnetic with very low conduction electron density of states near the Fermi level. The Knight shift result suggests a modification of the hybridized 4f/ conduction electronic structure below 50 K. On the other hand, the behavior of T_1^{-1} clearly reveal a negligible role of Ce 4f-electron spin fluctuations in the range 50–300 K. Whereas, below 50 K, 4f electrons participate in the excitation near the Fermi level and the extent of delocalization changes gradually below 50 K down to 3 K. These results clearly indicate a non-Fermi-liquid behavior in the temperature range and thus contradict the prediction of a heavy fermion ground state from the large value of γ in specific heat. Thus in this case the enhancement of γ at low temperature could be due to strong electron-electron correlations, which, however, do not lead to a heavy Fermi liquid state at low temperature. The present result gives a clear indication of the appearance of a 4f-moment compensation due to predominance of the Kondo effect below 50 K.

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- ¹Y. Onuki, T. Goto, and T. Kasuya, in *Materials Science and Technology*, edited by K. H. J. Buschow (VCH, Weinheim, 1992), Vol. 3A, p. 545.
- ²G. R. Stewart, *Rev. Mod. Phys.* **56**, 755 (1984).
- ³F. Steglich, A. Aarts, C. D. Bredle, W. Lieke, D. Meschede, W. Franz, and H. Schfer, *Phys. Rev. Lett.* **43**, 1892 (1979).
- ⁴N. Buttgen, R. Bohmer, A. Krimmel, and A. Loidl, *Phys. Rev. B* **53**, 5557 (1996).
- ⁵M. Kyogaku, Y. Kitaoka, H. Nakamura, K. Asayama, T. Takabatake, F. Teshima, and F. Fuji, *J. Phys. Soc. Jpn.* **59**, 1728 (1990).
- ⁶T. Mizushima, Y. Ishikawa, A. Maeda, K. Oyabe, K. Mori, K. Sato, and K. Kamigaki, *J. Phys. Soc. Jpn.* **60**, 753 (1991).
- ⁷T. Mizushima, Y. Ishikawa, K. Oyabe, K. Mori, and J. Sakurai, *Physica B* **186**, 457 (1993).
- ⁸Y. M. Rykhal, O. S. Zarechnyuk, and Y. P. Yarmolyuk, *Sov. Phys. Crystallogr.* **17**, 453 (1972).
- ⁹D. E. MacLaughlin, F. R. de Boer, J. Bijvoet, P. F. de Chatel, and

W. C. Mattens, *J. Appl. Phys.* **50**, 2094 (1979).

- ¹⁰S. K. Maik, R. Vijayraghavan, S. K. Garg, and R. J. Ripmeester, *Phys. Status Solidi B* **68**, 399 (1975).
- ¹¹E. Fukushima and S. B. W. Roeder, *Experimental Pulse NMR* (Addison-Wesley, Reading, MA, 1981).
- ¹²E. R. Andrew and D. P. Tunstall, *Proc. Phys. Soc. London* **78**, 1 (1961).
- ¹³K. Kashiwakura *et al.*, *J. Phys. Soc. Jpn.* **69**, 3095 (2000).
- ¹⁴C. H. Pennington and C. P. Slichter, *Physical Properties of High Temperature Superconductors* (World Scientific, Singapore, 1990), Vol. II.
- ¹⁵H. Nakamura, K. Nakijama, Y. Kitaoka, K. Asayama, K. Yoshimura, and T. Nitta, *Physica B* **171**, 238 (1990).
- ¹⁶D. L. Cox, N. E. Bickers, and J. W. Wilkins, *J. Appl. Phys.* **57**, 3166 (1985).
- ¹⁷A. J. Millis, *Phys. Rev. B* **48**, 7183 (1993).
- ¹⁸P. Schlottmann and P. D. Sacramento, *Adv. Phys.* **42**, 641 (1993).
- ¹⁹R. N. Bhatt and D. S. Fisher, *Phys. Rev. Lett.* **68**, 3072 (1992).