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Ru NMR and NQR probes of the metamagnetic transition in $CeRu_2Si_2$

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We report ^{99}Ru NMR and NQR studies on $CeRu_2Si_2$ in a magnetic field (H) range of 0–15.5 T across the metamagnetic transition at $H_M=7.8$ T. From measurements of the hyperfine field, ΔH_{hf} at the Ru sites induced by the $4f$-spin polarization and the nuclear-spin lattice relaxation rate, $1/T_1$, three heavy-fermion (HF) regimes have been clarified. (1) For $0\leqslant H<3$ T, the HF state is formed below 8 K. (2) At $H_M=7.8$ T, $1/T_1T$ continues to increase upon cooling to 1.4 K, suggesting a very low characteristic energy. (3) In the polarized state above H_M, the $1/T_1T$ probing the quasiparticle excitation is dramatically suppressed at low T, accompanying a peak at high T. The crossover in the HF state takes place not only in the H variation but also in the T variation across the metamagnetic transition in $CeRu_2Si_2$. [S0163-1829(98)51518-4]

The intermetallic compound $CeRu_2Si_2$ crystallizes in the tetragonal $ThCr_2Si_2$-type structure. The T-linear coefficient in the specific heat (γ) amounts to 350 mJ/mol K^2, pointing to the formation for the heavy fermion (HF) state at low temperatures.[1] Although the ground state in $CeRu_2Si_2$ does not show any long-range order, various types of substitution experiments revealed that the antiferromagnetic (AF) ordering takes place, which suggests that $CeRu_2Si_2$ is close to the AF instability.[2] Among a rich variety of ground states realizing in heavy fermion compounds, the polarized magnetic phase of $CeRu_2Si_2$ is particularly interesting since a large continuous jump of the magnetization occurs at $H_M=7.8$ T.[3]

From systematic measurements such as susceptibility, thermal expansion,[4] and specific heat[5] under the magnetic field, it was shown that the effective mass is enhanced at H_M, whereas it is reduced for $H>H_M$. From these results, it was suggested that an application of magnetic field higher than H_M disturbed the HF state appreciably. Remarkably, an inelastic neutron-scattering experiment provided valuable informations about a possible evolution in the spin-fluctuation spectrum around H_M, pointing to the presence of two competing magnetic correlations.[6] A quasielastic magnetic response (q independent) is nearly independent of the field, whereas an inelastic response with incommensurate q vector $Q=(0.7,0.7,0)$ strongly depends on the field around H_M. This result suggests that the metamagnetic transition is associated with a drastic change of the magnetic correlations corresponding to the depression of AF intersite spin fluctuations and the emergence of ferromagnetic correlations above H_M. A high value reached by the magnetic polarization leads to a situation similar to ferromagnetism in this regular array of Ce ions.

In order to investigate the low-lying excitation in the HF state, the Si-NMR studies in $CeRu_2Si_2$ was reported for $H<H_M$.[7,8] The nuclear-spin lattice relaxation time (T_1) and the Knight shift (K) were measured down to 0.5 K under 1.2 T. From the T_1T=constant law and the T-independent Knight shift, it was claimed that the HF state was realized below 8 K and that the Kondo temperature was around 20 K in good agreement with the estimation from the quasielastic of neutron scattering linewidth.[7] A systematic NMR investigation is, however, not yet performed either above H_M nor in zero magnetic field. In this paper, we report the first Ru NQR and NMR studies in $CeRu_2Si_2$. A successful observation of Ru NQR signal has allowed us to obtain the magnetic property in zero magnetic field.

A polycrystalline sample of $CeRu_2Si_2$ was prepared from high-purity starting materials. The sample was crushed into powder with diameter less than 35 μm. Powder sample is preferentially aligned with the c axis parallel to the external field as reported in the Si NMR measurement.[7]

Figure 1 shows a field-swept Ru NMR spectrum for the oriented powder at 21.1 MHz and 4.2 K. The well-articulated shape is composed of four transitions from the electric quadrupole interaction for ^{99}Ru ($I=5/2$) and of a central transition for ^{101}Ru. The ^{99}Ru NQR originating from $\pm 3/2\leftrightarrow\pm 5/2$ transition was observed at 3.61 MHz with a full width at half maximum of 0.1 MHz. From both spectra, the quadrupole frequency and the asymmetry parameter of ^{99}Ru were obtained to be $\nu_Q\sim 1.75$ MHz and $\eta\sim 0$ with the principal axis along the c axis, respectively.

The magnetization (M) along the c axis in $CeRu_2Si_2$ is not linear for $H>H_M$ due to the polarized state through the metamagnetic transition. A detailed T variation of M for $H>H_M$ was obtained from the T dependence of the hyperfine field $\Delta H_{hf}=H_0-H_{res}$ at the Ru sites where $H_0=21.1$ MHz/$^{99}\gamma_n$ is the field which corresponds to $K=0$, and H_{res} is the resonance field. Figure 2 indicates the respective T dependence of $\Delta H_{hf\parallel}$ parallel to the c axis for various fields from 2.93 to 15.5 T together with $\Delta H_{hf\perp}$ perpendicular to the c axis at 10.7 T. The $\Delta H_{hf\parallel}$ at 2.93 T was obtained only in a T

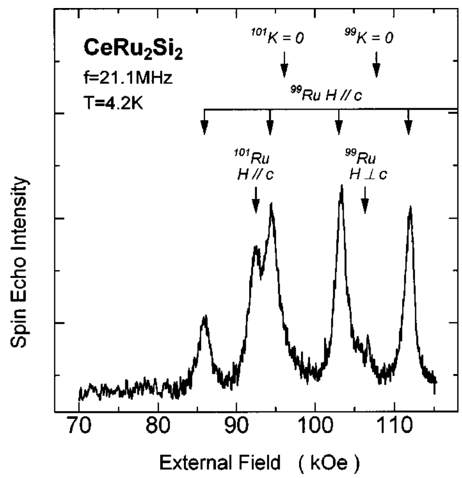

FIG. 1. Field-swept Ru NMR spectrum in the oriented powder of $CeRu_2Si_2$. Well-articulated spectrum originates from the electric quadrupole interaction for $I=5/2$ for two Ru isotopes. Upper down arrows denote each peak corresponding to the quadruple split transition of ^{99}Ru for $H\|c$ axis. The central transition of ^{101}Ru for $H\|c$ axis is only observable due to its larger nuclear quadrupole moment.

range of 1.4–4.2 K due to the weakness of NMR intensity. As seen in Fig. 2, the $\Delta H_{hf\|}$'s at 9.0 and 10.3 T in the high-field spin-polarized state continue to increase upon cooling down to 1.4 K, whereas the $\Delta H_{hf\|}$at 2.93 T, where the magnetization is proportional to the field, stays constant. The latter result is consistent with the previous Si Knight shift data $K_{\|}=\Delta H_{hf\|}/H_{res}$ at $\sim$1.1 T.

The $\Delta H_{hf\|}$ at the Ru sites induced by H_0 involves the $4f$ electron contribution through conduction-electron spin polarizations and the orbital contribution at the Ru sites and is hence expressed by

$$\Delta H_{hf}(H,T)=H_0-H_{res}$$
$$=A_{hf(orb)}\chi_{orb}H_{res}+A_{hf(4f)}M(H_0,T),$$

where χ_{orb} and $M(H_0,T)$ are the $4d$ orbital susceptibility and the magnetization per atom in units of μ_B, and $A_{hf(orb)}$

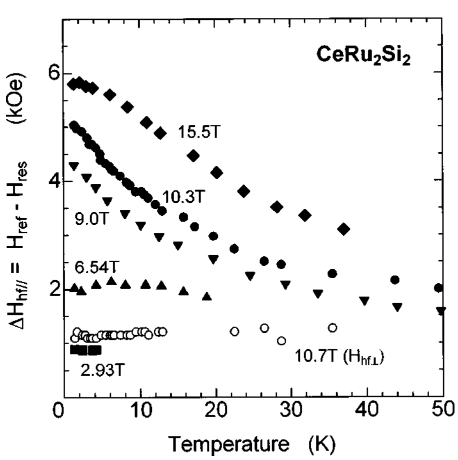

FIG. 2. Temperature dependence of the hyperfine field, $\Delta H_{hf\|}=H_0-H_{res}$ parallel to the c axis for various fields. Temperature dependence of $\Delta H_{hf\perp}$ perpendicular to the c axis at 10.7 T is also shown.

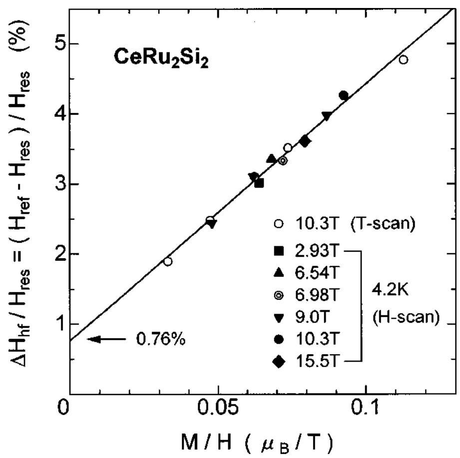

FIG. 3. $\Delta H_{hf\|}/H_{res}$ vs M/H_{res} plot. The magnetization data was referred from Ref. 3. Open circles and other marks correspond to the data obtained in the T variation under the constant H of 10.3 T and in the H variation at the constant T of 4.2 K, respectively.

and $A_{hf(4f)}$ are the corresponding hyperfine coupling constants, respectively. Since $K_{orb}=A_{hf(orb)}\chi_{orb}/\mu_B$, we obtain

$$\Delta H_{hf}=K_{orb}\ H_{res}+A_{hf(4f)}\ M$$
$$\Delta H_{hf}/H_{res}=K_{orb}+A_{hf(4f)}\ M/H_{res}.$$

$\Delta H_{hf}/H_{res}$ is plotted against M/H_{res} in Fig. 3 where the magnetization data reported by Haen *et al.* were used.[3] From a linear variation of $\Delta H_{hf\|}/H_{res}$ vs M/H_{res} as shown in Fig. 3, $K_{orb\|}$ and $A_{hf(4f)}$ are extracted to be 0.76% and 3.67 kOe/μ_B, respectively. The estimated $K_{orb\|}$ is almost the same as the Ru Knight shift in $LaRu_2Si_2$, 0.72%, indicating that $K_{orb\|}$ originates only from the Ru $4d$ contribution, but not from the Ce contribution. Since $\Delta H_{hf\perp}/H_{res}$ is comparable to $K_{orb\|}$, $\Delta H_{hf\perp}$ is not affected by the spin contribution from the $4f$ electrons. The positive and small value of $A_{hf(4f)}=3.67$(kOe/μ_B) is considered to originate from the Fermi contact interaction by $5s$ electron spin polarization induced through the hybridization with the $4f$ electrons. It should be noted that $\Delta H_{hf\|}/H_{res}$ vs M/H_{res} plots for the constant H of 10.3 T at various temperatures and for the constant T of 4.2 K under various magnetic fields is on a single line across the metamagnetic transition. The $A_{hf(4f)}$ does not change at all through the metamagnetic transition, indicating no drastic change in the electronic state as observed by magnetization,[9] specific heat[5] and transport measurements[10] in contrast to the large change observed in quantum oscillation experiments.[11,12] This apparent discrepancy may be directly linked to the microscopic formation of the quasiparticle bands and deserve now theoretical understandings.

Figure 4 shows the T dependence of the magnetization along the c axis divided by the external field, H, M/H obtained from the Ru NMR shift and the previous Si NMR data at $\sim$1.14 T from the Si NMR Knight shift with A_{hf}~2.21 (kOe/μ_B).[3] As indicated in the figure, the T dependence of M/H is remarkably different between the low and high field below 15 K, where a nonlinear response appears in the field-swept magnetization process. This temperature is denoted as the onset temperature, T_M for the emergence of the meta-

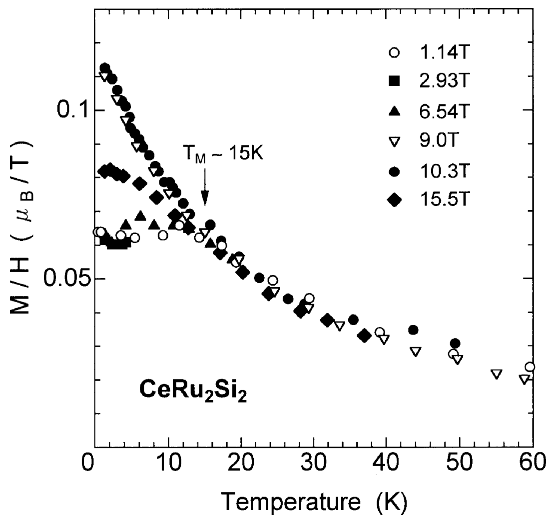

FIG. 4. Temperature dependence of the M/H deduced from the Ru and Si NMR shifts (see text).

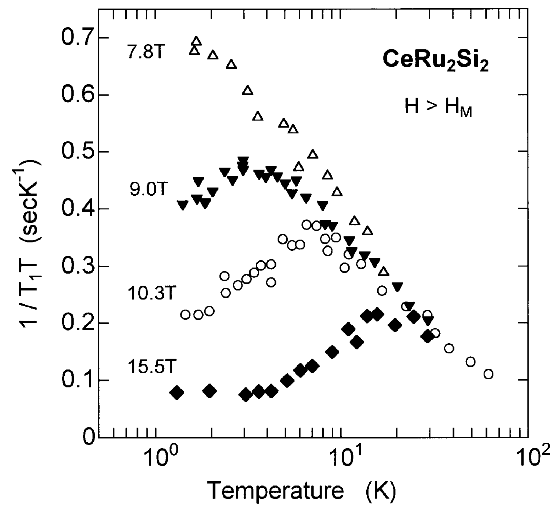

FIG. 6. Temperature dependence of $1/T_1T$ at H_M=7.8 T and 9.0, 10.3, and 15.5 T in the polarized state above and at H_M.

magnetic transition. The nearly T-independent behavior below 15 K in the low magnetic field regime is understood as a consequence of the formation of the HF state. Therefore, the monotonous increase below 15 K in the high magnetic field regime shows that the temperature population effect is huge in this polarized state above H_M.

The T dependence of nuclear-spin lattice relaxation rate divided by T, $1/T_1T$ below and above H_M are shown in Figs. 5 and 6, respectively. As seen in Fig. 5, $1/T_1T$'s in zero field and 2.93 T below 8 K stay constant. In the low-field regime, the experimental value of $^{99}(1/T_1TK_{iso}^2)=8.16\times10^3$ $(\mathrm{s\,K})^{-1}$ is comparable to the calculated one of $1/T_1TK^2 = \pi\hbar\ ^{99}\gamma_n^2 k_B/\mu_B^2 = 8.04\times10^3(\mathrm{s\,K})^{-1}$ based upon the quasiparticle Korringa relation. Here K_{iso} is estimated from $K_{iso}\sim K_{\parallel}/3$ since $K_{\perp}$ is negligible as mentioned above. The HF state below 8 K seems to be dominated by q-independent excitations since the phenomenological magnetic coherence length involves only a few lattice distance.[6] This fact is in good agreement with the analysis made in the formalism developed by Moriya,[13] where the parameter y_0, inversely proportional to the staggered susceptibility, is relatively large.[14]

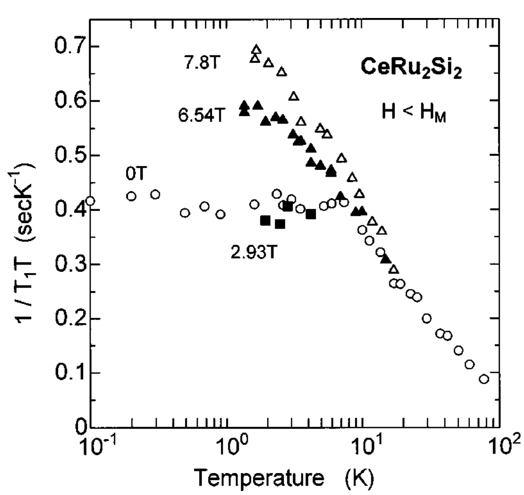

FIG. 5. Temperature dependence of $1/T_1T$ of Ru at 0, 2.93, and 6.54 T in the unpolarized state below and at H_M=7.8 T.

By contrast, $1/T_1T$ below 8 K increases with the increasing field and that at H_M=7.8 T continues to increase upon cooling as if there exists a critical anomaly almost at T=0 K. As the field increases further, $(T_1T)^{-1}$ is dramatically suppressed and a peak of $1/T_1T$ emerges around 4 K, 7 K, and 20 K for H=9, 10.3, and 15.5 T, respectively, as shown in Fig. 6. It should be noted that the T_1T=constant in the high-field regime holds below temperatures much lower than 8 K in the low-field regime. By using an effective density of states (DOS), $N_{eff}(E_F)$ at the Fermi level in the HF state, we may assume that $1/T_1T\propto N_{eff}(E_F)^2\propto\gamma^2$, where $\gamma=C/T$ is the T-linear coefficient in the the specific heat (C) at $T\to0$. In this context, the field-induced suppression in $1/T_1T$ under 7.8, 10.3, and 15.5 T at 1.4 K may be related with the reduction in $N_{eff}(E_F)$. $[N_{eff}(E_F,H)]/[N_{eff}(E_F,0)] = \sqrt{(1/T_1T)_H/(1/T_1T)_0}\sim1.30$, 0.73, and 0.44 at 1.4 K are compatible with $[N_{eff}(E_F,H)]/[N_{eff}(E_F,0)]=\gamma_H/\gamma_0\sim$ 1.27, 0.71, and 0.29 for H=8, 10, and 15.5 T at 1.5 K, respectively.[5] The apparent good scaling of $1/T_1T \propto N_{eff}(E_F)^2\propto\gamma^2$ reflects the fact that $CeRu_2Si_2$ never reaches a magnetic instability even at H_M since each quantity should have a different T dependence near the critical point.[13]

However, the emergence of a peak in $1/T_1T$ above H_M reflects the fact that the $4f$ state renormalized at the low T above H_M is quite different from the case in the unpolarized state. It is suggested that the HF bandwidth and DOS in the polarized state are much reduced as compared with those in the unpolarized state and at the metamagnetic transition, since most weight of $4f$ state in the HF state is considered to be transferred into the ferromagnetically polarized state well below the Fermi level.

Next the NMR result is compared with the recent results of inelastic neutron scattering experiments.[15] The nuclear relaxation study provides a first insight on the low-energy excitation which cannot be reached easily by inelastic neutron experiments. Recent development of the latter technique shows that the AF correlation with characteristic energy $(\sim k_BT_M)$ is replaced above H_M by a quasistatic long-range ferromagnetic correlation with a low-energy window ($<$4 K). This is in good agreement with the rapid decrease in

$1/T_1T$ implying a creation of a highly polarized state and with still the occurrence of a temperature dependent Knight shift. Let us stress that polarized neutron experiments, mainly sensitive to a change in the $4f$ magnetic form factor, shows also no indication of its modification through H_M in good agreement with the unchange of the hyperfine coupling constant.[15]

In conclusion, from the Ru NMR and NQR studies, the three HF states have been unraveled. In the low-field regime, the ordinary HF state below 8 K is evidenced from the quasiparticle Korringa relation. At $H_M = 7.8$ T, $1/T_1T$ continues to increase upon cooling to 1.4 K, suggesting a very low characteristic energy. The scaling of $1/T_1T \propto \gamma^2$ at low T across H_M reflects the fact that $CeRu_2Si_2$ never reaches a magnetic instability. In the high-field spin-polarized state, the size in $1/T_1T$ and the temperature where the T_1T=const law holds are much smaller than those in the unpolarized regime. Remarkably, the emergence of peak in $1/T_1T$ above H_M reflects the fact that the $4f$ state is renormalized in a quite different manner from the case in the unpolarized state. The crossover in the HF state in not only the H variation but also the T variation provides clues to elucidate the cause for the metamagnetic transition in $CeRu_2Si_2$.

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