^{101}Ru nuclear magnetic resonance/nuclear quadrupole resonance (NMR/NQR) and ^{29}Si NMR studies of the metamagnetic-like transition in $CeRu_2Si_2$

2000 J. Phys.: Condens. Matter 12 2061

(http://iopscience.iop.org/0953-8984/12/9/310)

J. Phys.: Condens. Matter **12** (2000) 2061–2069. Printed in the UK PII: S0953-8984(00)09440-6

^{101}Ru nuclear magnetic resonance/nuclear quadrupole resonance (NMR/NQR) and ^{29}Si NMR studies of the metamagnetic-like transition in CeRu$_2$Si$_2$

K Matsuda†, Y Kohori†, T Kohara† and K Fujiwara‡

† Department of Material Science, Himeji Institute of Technology, Kamigori, Ako-gun, Hyogo 678-1297, Japan

‡ Department of Material Science, Shimane University, Nishikawatsu, Matsue-shi, Shimane 690-8504, Japan

Received 10 November 1999

Abstract. The metamagnetic-like transition at an external field (H_{ext}) ~7.7 T (H_M) in CeRu$_2$Si$_2$ has been probed by ^{101}Ru NMR/NQR and ^{29}Si NMR measurements in an external field up to 16.4 T. The nuclear spin–lattice relaxation rate (T_1^{-1}) of ^{101}Ru is field dependent at low temperatures (T). The value of $(T_1T)^{-1}$ remains T-independent below 7 K in zero field, whereas the value of $(T_1T)^{-1}$ at H_M continues to increase down to 1.4 K. At a field of 16.4 T (a field much higher than H_M), $(T_1T)^{-1}$ behaves independently of T again, as expected for the Fermi liquid state, where the value of $(T_1T)^{-1}$ is much smaller than that in zero field. The field dependence of the spin-echo decay rate (T_2^{-1}) measured at 4.2 K by means of NMR for both ^{101}Ru and ^{29}Si exhibits no anomaly near H_M, suggesting the absence of any enhancement of the ferromagnetic spin fluctuations.

1. Introduction

The cerium ternary intermetallic compound CeRu$_2$Si$_2$ possesses a tetragonal ThCr$_2$Si$_2$-type structure. In this compound, neither superconductivity nor long-range magnetic order has been observed down to 20 mK [1]. However, neutron scattering studies have revealed that incommensurate antiferromagnetic correlations develop at low temperatures (T) [2, 3], indicating that CeRu$_2$Si$_2$ is located in the vicinity of a magnetic instability point. The large electronic specific heat coefficient $\gamma \sim 350$ mJ mol^{-1} K^{-2} observed in zero field indicates that a Fermi liquid state with strongly interacting f electrons is realized at low T [4, 5]. The most remarkable feature of CeRu$_2$Si$_2$ is the metamagnetic-like transition observed at an external field (H_{ext}) ~7.7 T (H_M) for $\boldsymbol{H}_{ext} \parallel c$-axis in spite of the non-magnetic ground state [6, 7]. Much attention has been focused on the nature of this metamagnetic-like transition at which the following physical quantities exhibit anomalies. The effective masses obtained from the specific heat (C) and the susceptibility measurements exhibit an enhancement at around H_M and then decrease above H_M with increasing field [4, 5, 7]. At H_M, the results of susceptibility, C, Hall resistivity and magnetoresistance measurements display continuous behaviour [4, 5, 7–9]. On the other hand, the drastic changes in the Fermi surface and the effective masses around H_M obtained from de Haas–van Alphen (dHvA) effect measurements suggest that a complete change occurs in the 4f-electron nature from itinerant to localized at H_M [10]. According to the neutron scattering experiments [2, 3], the inelastic magnetic diffractions at the incommensurate wave vectors $\boldsymbol{q}$ = (0.3, 0.3, 0) and (0.3, 0, 0) disappear above H_M, while a q-independent contribution still remains. These neutron scattering experiments have shown the collapse of the antiferromagnetic correlations above H_M. In order to study this metamagnetic-like transition, some theoretical investigations have been carried out by taking into account a ferromagnetic exchange interaction [11–14]. A pseudo-gap (camel-back) structure in the density of states (DOS) of quasiparticles is assumed in a recent model [11], where the field-dependent exchange interaction and the magnetostriction effect play a crucial role in the metamagnetic anomaly. According to this model, the exchange interaction is antiferromagnetic in the low field, while it becomes ferromagnetic at around H_M. Indeed, a previous ^{29}Si nuclear magnetic resonance (NMR) experiment has shown that the spin-echo decay rate (T_2^{-1}) at the Si site is remarkably enhanced around H_M, which strongly suggests an enhancement of the ferromagnetic spin fluctuations [15]. On the other hand, a recent neutron scattering measurement has shown no short-range ferromagnetic fluctuations around H_M [3]. The magnetic property of 4f electrons above H_M is characteristic of a long-range ferromagnetic order induced by a field. Hence, it is important to obtain microscopic information on the ferromagnetic spin fluctuations not only at the Si site but also at the Ru site by means of NMR and nuclear quadrupole resonance (NQR). Quite recently, Ishida *et al* have obtained the unchanged hyperfine coupling constant from H_M and the field-dependent nuclear spin–lattice relaxation rate (T_1^{-1}) by means of ^{99}Ru NMR measurements in an external field up to 15.5 T [16]. In this paper, we discuss the results for T_2 measured by means of ^{101}Ru and ^{29}Si NMR together with the Knight shift (K) and T_1 in an external field up to 16.4 T; the studies mostly focused on the metamagnetic-like transition.

2. Experimental procedure

A polycrystalline sample of $CeRu_2Si_2$ was synthesized by arc melting stoichiometric amounts of the constituent elements (4N Ce, 4N Ru and 7N Si) in an argon atmosphere. The sample was annealed in an evacuated quartz tube for six days at 900 °C and then crushed into powder for use in the x-ray diffraction and NMR measurements. The NMR and NQR measurements were carried out by the conventional spin-echo technique with a phase-coherent pulsed spectrometer. A superconducting magnet with a maximum field of 17 T was used for the NMR measurements. The value of T_1 was measured by monitoring the recovery of the nuclear magnetization following the saturation pulse. The value of T_2 was obtained by monitoring the spin-echo amplitude, $I(2\tau)$, as a function of time (2τ) between the first pulse and the echo. In the NMR measurements at high field, microcrystals in the powder sample of $CeRu_2Si_2$ were aligned with the c-axis parallel to the external field due to the large anisotropy of the susceptibility.

3. Results and discussion

3.1. ^{101}Ru NMR/NQR spectra

Figure 1 shows the Ru NMR spectrum of $CeRu_2Si_2$ taken for aligned ($\boldsymbol{H}_{ext} \parallel c$-axis) microcrystals by sweeping the external field at the resonance frequency of 24.3 MHz. The spectrum obtained consists of a superposition of two equivalent spectra arising from the ^{99,101}Ru isotopes. Each NMR spectrum is composed of the centreline and the satellites induced by the first-order quadrupole effect. The T-dependence of K for $\boldsymbol{H}_{ext} \parallel c$-axis, measured from the centreline of ^{101}Ru, is plotted in figure 2. The value of K obtained at the field of 4.8 T (a field lower than H_M) has a broad maximum at around 10 K and then decreases slightly with decreasing T. On the other hand, the value of K obtained at the field of 10.7 T (a field higher than H_M) increases with decreasing T down to 4.2 K. The value of K is field independent above 20 K, where no metamagnetic-like transition is observed in the magnetization process. Using the

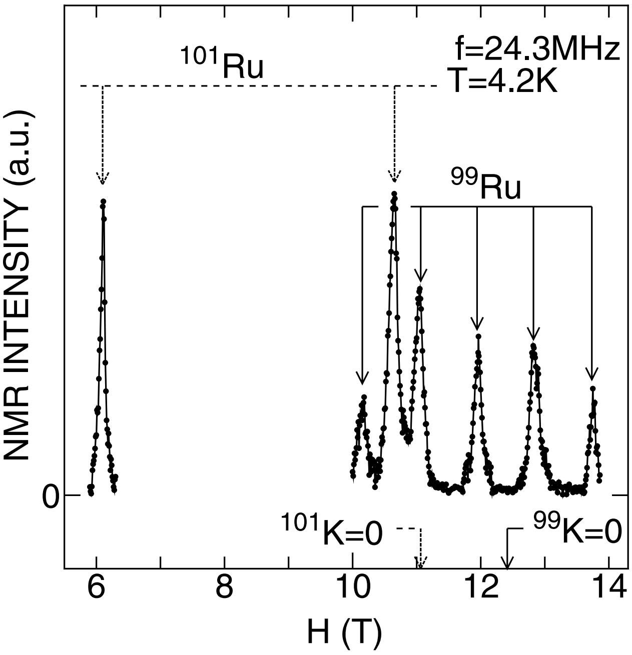

Figure 1. ^{99}Ru and ^{101}Ru NMR spectra taken at 4.2 K and 24.3 MHz with the *c*-axis parallel to the external field. The ^{99}Ru NMR spectrum consists of a centreline at around 12 T and four satellite lines. Due to the large nuclear quadrupole moment of ^{101}Ru, a centreline at around 10.7 T and a satellite line corresponding to the $-3/2 \Leftrightarrow -5/2$ transition were observed.

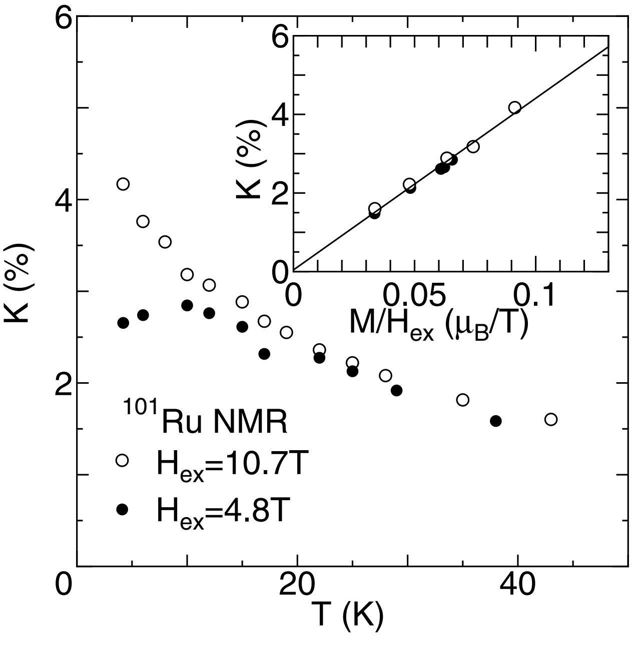

Figure 2. The T-dependence of the Knight shift of ^{101}Ru for external fields below and above H_M. In the inset of the figure, a plot of K versus M/H_{ex} is shown. Magnetization data extracted from reference [7] were used.

magnetization (M) data [6], K is plotted against M/H_{ex} in the inset of figure 2 with T as an implicit parameter. The fact that the Ks obtained at 4.8 and 10.7 T are on the same line as the hyperfine coupling constant (A_{hf}) of 4.3 kOe/μ_B suggests that there is no drastic change in the electronic structure on passing through H_M. This is consistent with the results of the magnetization and the magnetostriction measurements [7,8]. Our value of A_{hf} is slightly larger than the previous one (3.67 kOe/μ_B) obtained with external field variation at 4.2 K [16]. From the NMR spectrum, the magnitude of the electric quadrupole interaction parameter (ν_Q) of ^{101}Ru was deduced to be about 10 MHz. Indeed, two narrow ^{101}Ru NQR lines were observed near the resonance frequencies of 10.52 and 21.4 MHz at 4.2 K, as shown in figure 3. The lower- and the higher-frequency resonance lines of ^{101}Ru correspond to the transitions $\pm1/2 \Leftrightarrow \pm3/2$ and $\pm3/2 \Leftrightarrow \pm5/2$, respectively. From the NQR spectrum, ν_Q for ^{101}Ru and the asymmetry parameter of the electric field gradient (η) were precisely determined to be 10.52 MHz and 0, respectively. The NMR and the NQR spectra have shown that the direction of the electric field gradient is along the tetragonal c-axis.

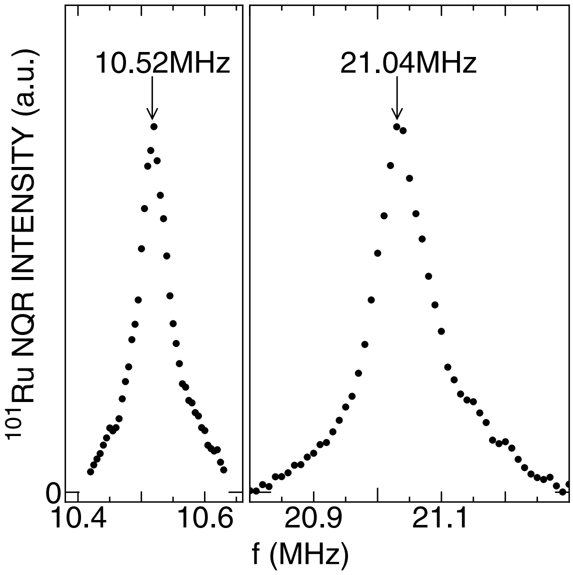

Figure 3. ^{101}Ru NQR spectra taken at 4.2 K. The lower- and the higher-frequency resonance lines arise from the transitions of $\pm1/2 \Leftrightarrow \pm3/2$ and $\pm3/2 \Leftrightarrow \pm5/2$, respectively.

3.2. Nuclear spin–lattice relaxation rate

T_1 in zero field was measured by means of ^{101}Ru NQR at the resonance frequency of 21.04 MHz. As for T_1 in the external field, the NMR signal arising from the $-3/2 \Leftrightarrow -5/2$ transition of the ^{101}Ru nuclear spins was used at 3.91, 7.69 and 12.4 T; we could distinguish the ^{101}Ru NMR signal from the ^{99}Ru one with complete confidence. The values of $(T_1T)^{-1}$ obtained in zero field and the external field up to 16.4 T are shown against T in figure 4. The (T_1^{-1})s measured by means of NMR and NQR are essentially equivalent, because the two (T_1^{-1})s are governed by magnetic fluctuations perpendicular to the same quantization direction, namely the c-axis. As can be seen in figure 4, $(T_1T)^{-1}$ in zero field remains T-independent below 7 K, indicating that the system is in the Fermi liquid state, which is consistent with the results of ^{29}Si NMR studies previously performed in low fields ($H_{ext} \ll H_M$) [17]. At 7.7 T, $(T_1T)^{-1}$

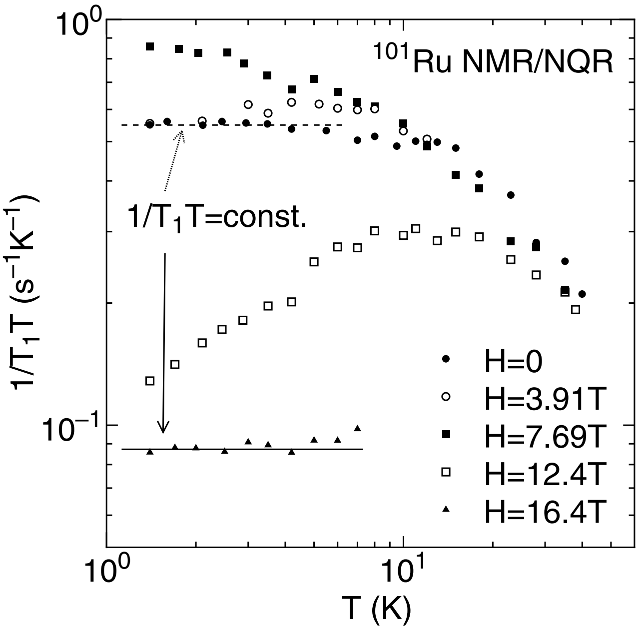

Figure 4. The T-dependence of $(T_1T)^{-1}$ in different fields applied along the c-axis.

increases gradually with decreasing T down to 1.4 K. By contrast, $(T_1T)^{-1}$ at 12.5 T has a broad maximum, and then decreases significantly at low T. At the field of 16.4 T (a field much higher than H_M), $(T_1T)^{-1}$ becomes T-independent again, as observed for zero magnetic field. However, the value of $(T_1T)^{-1}$ obtained at 16.4 T is much smaller than the value in zero field. Using the relation ($(T_1T)^{-1} \propto N(E_F)^2$) between the DOS at the Fermi level ($N(E_F)$) and T_1^{-1}, $N(E_F)$ at 16.4 T is found to be reduced to about 40% of the value in zero field. Our T-dependence of T_1^{-1} measured by means of ^{101}Ru NMR/NQR is in good agreement with the results in reference [13] measured using ^{99}Ru NMR/NQR over the whole field range.

Figure 5 shows the field dependence of T_1^{-1} taken at various temperatures T. At the high T of 35 K at which no metamagnetic-like transition is observed in the magnetization process, T_1^{-1} exhibits no anomaly near H_M and remains almost constant. The value of T_1^{-1} at 4.2 K has a weak maximum at around H_M and then decreases with increasing external field. With decreasing T below 4.2 K, a maximum of T_1^{-1} at around H_M becomes more pronounced. The decrease in T_1^{-1} in the field higher than H_M is associated with the reduction in $N(E_F)$. Similar field dependence is observed in the specific heat measurements, showing that C/T is enhanced near H_M and depressed above H_M [5]. The T-dependence of $(T_1T)^{-1/2}$ at 1.4 K is shown in the inset of figure 5 together with the previous data for C/T. The previous ^{29}Si NMR study has shown that the anisotropy of T_1 ($T_{1\parallel c}/T_{1\perp c} \sim 3$) is much smaller than that of the static susceptibility ($\chi_{\parallel c}/\chi_{\perp c} \sim 15$) at low T [6, 17]. In general, the nuclear spin–lattice relaxation is caused by the magnetic fluctuations via the hyperfine coupling. Strictly speaking, the hyperfine coupling constant should be expressed as a tensor. Although there is an ambiguity in the estimation of the hyperfine coupling tensor, the small anisotropy of T_1 implies that the off-diagonal elements of the hyperfine tensor contribute to T_1 for $\boldsymbol{H}_{ext} \parallel c$-axis. Hence, T_1 for $\boldsymbol{H}_{ext} \parallel c$-axis is not determined by the magnetic fluctuations perpendicular to the c-axis but is governed by those parallel to the c-axis via such a dipole–dipole coupling. In this case, both T_1 and C measured for $\boldsymbol{H}_{ext} \parallel c$-axis are governed by the magnetic fluctuations

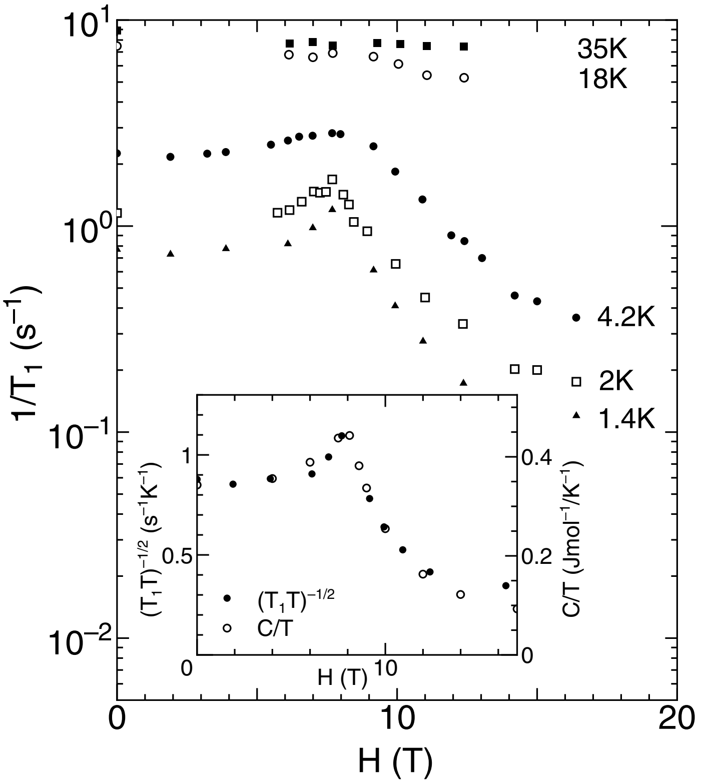

Figure 5. The field variation of T_1^{-1} at different temperatures. The field variation of $(T_1T)^{-1/2}$ at 1.4 K is shown in the inset of the figure, where C/T extracted from reference [4] is also plotted for comparison. The scale has been chosen to show that $(T_1T)^{-1/2}$ and C/T display the same field dependence below 5 T.

parallel to the c-axis. As shown in the figure, the $N(E_F)$ deduced from the NMR and that deduced from the specific heat ($C/T \propto N(E_F)$) measurements exhibit almost the same field dependence in the external field up to 15 T, which enables us to use a Fermi liquid description of this system. The decrease in the DOS at the Fermi level observed by means of NMR is associated with the drastic renormalization of the Fermi surface topology due to the change in the f-electron nature observed from the dHvA effect [10]. However, the field dependence of T_1^{-1} suggests that the renormalization effect does not occur in the narrow field range around H_M but decreases gradually above H_M.

3.3. Spin-echo decay rate

In the previous ^{29}Si NMR measurement, the value of T_2^{-1} at 4.2 K is strongly enhanced ($(T_2)_{2\mathrm{T}}/(T_2)_{10\mathrm{T}} \sim 80$) around 10 T, which is slightly larger than H_M [15]. As mentioned previously, the ferromagnetic exchange interaction reaching a maximum at H_M is considered to be one of the triggers for the metamagnetic-like transition in the model of the quasiparticle DOS with the camel-back structure [11]. Thus, this experimental result is interpreted as an enhancement of the ferromagnetic spin fluctuations parallel to the c-axis around H_M. In order to examine the enhancement of the ferromagnetic spin fluctuations at H_M, the T_2-measurements were performed using both ^{101}Ru and ^{29}Si NMR. When the magnetic fluctuations dominate the nuclear relaxations (the so-called T_1-process), T_1^{-1} and T_2^{-1} for $\boldsymbol{H}_{ext} \parallel c$-axis are related to the dynamical susceptibilities of electrons parallel and perpendicular to the c-axis, $\chi_\parallel(q,\omega)$ and $\chi_\perp(q,\omega)$, by

$$1/T_1 \propto \sum_q (A_{q\perp})^2 \operatorname{Im} \frac{\chi_\perp(q,\omega_0)}{\omega_0}$$

and

$$1/T_2 \propto (1/2) \sum_q (A_{q\perp})^2 \operatorname{Im} \frac{\chi_\perp(q,\omega_0)}{\omega_0} + \lim_{\omega\to 0} (A_{q\parallel})^2 \frac{\chi_\parallel(q,\omega)}{\omega}$$

respectively. Here $A_{q\parallel}$ and $A_{q\perp}$ are the hyperfine coupling constants parallel and perpendicular to the c-axis, respectively. According to the above expressions, T_1^{-1} is connected to the magnetic fluctuations perpendicular to the c-axis by the diagonal elements of the hyperfine tensor, and is indirectly connected to the fluctuations parallel to the c-axis by the off-diagonal elements of the hyperfine coupling tensor for this system, as discussed previously. However, T_2^{-1} is directly connected to the fluctuations parallel to the c-axis by the diagonal elements of the hyperfine coupling tensor. Therefore, T_2^{-1} is more sensitive to the ferromagnetic spin fluctuations around H_M than T_1^{-1}. In the nuclear spin–spin relaxation mechanism, there are large contributions from the interactions between the nuclear spins besides the T_1-process.

The inset of figure 6 indicates the spin-echo decay curves for ^{101}Ru and ^{29}Si NMR, which show nearly exponential behaviour over the whole external field range. The T_2-measurements for ^{101}Ru were performed with the NMR signal arising from the $-3/2 \Leftrightarrow -5/2$ transition of the ^{101}Ru nuclear spins. The value of T_2^{-1} was derived by fitting the function

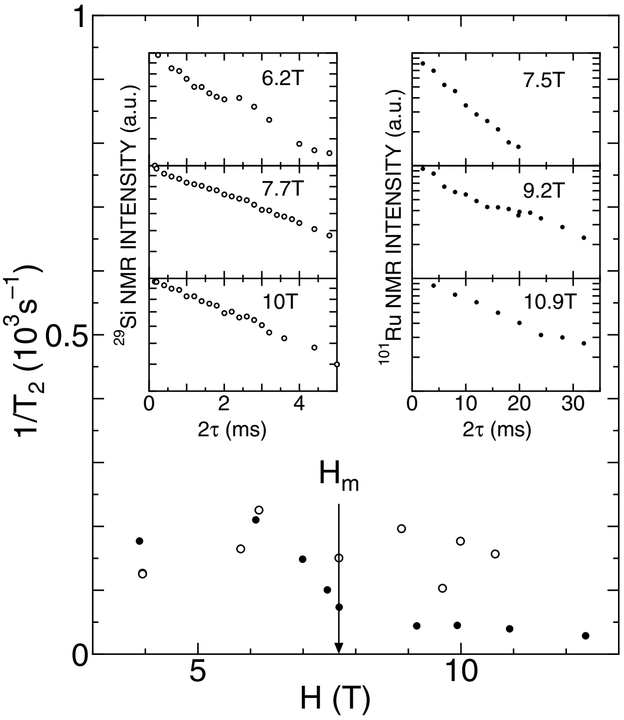

Figure 6. The field variation of T_2^{-1} for ^{101}Ru (solid circles) and ^{29}Si (open circles) at 4.2 K. In the inset of the figure, the spin-echo decay curves taken at different fields are shown.

$I(2\tau)/I_0 = \exp(-2\tau/T_2)$ to the measured data. Figure 6 shows the external field dependence of T_2^{-1} at 4.2 K. If the enhanced spin fluctuations contribute to the nuclear spin–spin relaxation, the value of $1/T_2$ could be enhanced around H_M. As can be seen in figure 6, however, T_2^{-1} for ^{101}Ru, $^{101}(T_2^{-1})$, has no anomaly near H_M and decreases monotonically with increasing field. The decrease in $^{101}(T_2^{-1})$ in the high field would be caused by the detuning of the ^{101}Ru nuclear spin–spin coupling due to the inhomogeneous broadening of the ^{101}Ru NMR spectrum line. Hence $^{101}(T_2^{-1})$ is dominated by the contributions from the interaction between the nuclear spins over the whole external field range. As can clearly be seen in the inset of figure 6, the spin-echo decay curve is nearly field independent over the whole field range. In our measurements, T_2^{-1} for ^{29}Si, $^{29}(T_2^{-1})$, also exhibits no enhancement near H_M, which is in contradiction to the previous result [15]. The field dependence of both $^{101}(T_2^{-1})$ and $^{29}(T_2^{-1})$ is well explained by just the contributions from the interaction between the nuclear spins. Even if ferromagnetic spin fluctuations develop around H_M, their effects on the nuclear spin–spin relaxation are negligibly small as compared with those from the interaction between the nuclear spins. As described previously, the nuclear spin–lattice relaxation for $\boldsymbol{H}_{ext} \parallel c$-axis has a contribution from the magnetic fluctuations parallel to the c-axis. Therefore, the ferromagnetic spin fluctuations should enhance the nuclear spin–lattice relaxation for $\boldsymbol{H}_{ext} \parallel c$-axis. However, the enhancement of the ferromagnetic spin fluctuations is not observed in the field dependence of T_1^{-1} of ^{101}Ru, as indicated in the inset of figure 5. The value of T_1^{-1} is slightly enhanced around H_M but only in proportion to C/T. The results of the ^{101}Ru and ^{29}Si NMR investigations are consistent with those from the neutron scattering measurements performed around the ferromagnetic zone centre in the high magnetic field, where no short-range ferromagnetic spin fluctuations were observed in the energy range of 0.4–10 meV around H_M [3]. The present NMR results strongly suggest the absence of any enhancement of the ferromagnetic fluctuations for such low energies, which are hard to detect in inelastic neutron scattering measurements.

4. Conclusions

The hyperfine coupling constant of ^{101}Ru is field independent, indicating that there is no drastic change in the electronic structure on passing through H_M. The value of T_1^{-1} for ^{101}Ru has a maximum at around H_M and then decreases with increasing field at low T; this is associated with the reduction of the DOS at the Fermi level for $H_{ex} > H_M$. The relation $(T_1T)^{-1/2} \propto C/T$ expected for the Fermi liquid state holds at 1.4 K over the whole field range up to 15 T. The field dependence of T_1^{-1} suggests that the effect of renormalization by the field decreases gradually above H_M. The value of $(T_1T)^{-1}$ is T-independent at 16.4 T below 5 K, as observed in zero field below 7 K. This result indicates that the renormalized Fermi liquid state, which has a smaller DOS than that in zero field, still exists in fields much higher than H_M. Furthermore, the field dependence of T_2^{-1} obtained from both ^{101}Ru and ^{29}Si NMR investigations shows no anomaly near H_M. We have shown the absence of any enhancement of the ferromagnetic spin fluctuations around H_M, which is consistent with the results of the inelastic neutron scattering experiments.

Acknowledgments

This work was partly supported by a Grant-in-Aid from the Ministry of Education, Science and Culture of Japan. One of the authors (KM) was supported by a Research Fellowship from the Japan Society for Promotion of Science for Young Scientists.

References

[1] Gupta L C, MacLaughlin D E, Tien Cheng, Godart C, Edwards M A and Parks R D 1983 *Phys. Rev.* B **28** 3673

[2] Rossat-Mignod J, Regnault L P, Jacoud J L, Vettier C, Lejay P, Flouquet J, Walker E, Jaccard D and Amato A 1988 *J. Magn. Magn. Mater.* **76+77** 376

[3] Raymond S, Regnault L P, Kambe S, Flouquet J and Lejay P 1998 *J. Phys.: Condens. Matter* **10** 2363

[4] Besnus M J, Kappler J P, Lehmann P and Mayer A 1985 *Solid State Commun.* **56** 779

[5] van der Meulen H P, de Visser A, Franse J J M, Berendschot T T J M, Perenboom J A A J, van Kempen H, Lacerda A, Lejay P and Flouquet J 1991 *Phys. Rev.* B **44** 814

[6] Haen P, Flouquet J, Lapierre P, Lejay P and Remenyi G 1987 *J. Low Temp. Phys.* **67** 391

[7] Sakakibara T, Tenya T, Matsuhira K, Mitamura H, Amitsuka H, Maezawa K and Onuki Y 1995 *Phys. Rev.* B **51** 12 030

[8] Kambe S, Suderow H, Flouquet J, Haen P and Lejay P 1995 *Solid State Commun.* **95** 449

[9] Kambe S, Flouquet J, Haen P and Lejay P 1996 *J. Low Temp. Phys.* **102** 447

[10] Aoki H, Uji S, Albbessard A K and Onuki Y 1993 *Phys. Rev. Lett.* **71** 2110

[11] Satoh H and Ohkawa F J 1998 *Phys. Rev.* B **57** 5891

[12] Ohkawa F J 1989 *Solid State Commun.* **71** 907

[13] Brenig W 1987 *Solid State Commun.* **64** 203

[14] Ueda K, Yamamoto K and Konno R 1990 *J. Magn. Magn. Mater.* **90+91** 419

[15] Takagi S, Ohama T, Kusumoto T, Yasuoka H and Onuki Y 1994 unpublished

[16] Ishida K, Kawasaki Y, Kitaoka Y, Asayama K and Flouquet J 1998 *Phys. Rev.* B **57** 11 054

[17] Kitaoka Y, Arimoto H, Kohori Y and Asayama K 1985 *J. Phys. Soc. Japan* **54** 3236