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New aspects on URu_2Si_2 and $CeTIn_5$ (T = Rh, Ir, Co) observed by high pressure NMR and NQR

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Abstract. NMR and NQR studies on two interesting systems (URu_2Si_2, $CeTIn_5$) were performed under high pressure. (1) URu_2Si_2: In the pressure range 3.0 to 8.3 kbar, we have observed new ^{29}Si NMR signals arising from the antiferromagnetic (AF) region besides the previously observed ^{29}Si NMR signals which come from the paramagnetic (PM) region in the sample. This gives definite evidence for spatially-inhomogeneous development of AF ordering below T_0 of 17.5 K. The volume fraction is enhanced by applied pressure, whereas the value of internal field (∼91 mT) remains constant up to 8.3 kbar. In the AF region, the ordered moment is about one order of magnitude larger than 0.03 μ_B. (2) $CeTIn_5$: The pressure and temperature (T) dependences of nuclear spin-lattice relaxation rate $1/T_1$ of ^{115}In in $CeTIn_5$ have shown that the superconductivity (SC) occurs close to an AF instability. From the T dependences of $1/T_1$ and Knight shift below T_c, $CeTIn_5$ has been found to exhibit non-*s* wave (probable *d* wave) SC with even parity and line nodes in the SC energy gap.

Keywords. URu_2Si_2; $CeTIn_5$; high pressure; NMR; NQR.

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1. Introduction

According to the recent studies on *f*-electron systems, most of the cerium and uranium compounds with heavy electron mass indicate unusual phenomena including magnetic and/or quadrupolar orderings, spin and/or valence fluctuations and anisotropic superconductivity by the application of pressure and substitution of constituent elements. Among them we performed NMR and NQR measurements on URu_2Si_2 and $CeTIn_5$ (T = Rh, Ir, Co) under high pressure for the purpose based on the following backgrounds.

[1]In collaboration with K Matsuda and Y Kohori (Himeji Inst. of Tech., Japan), H Amitsuka (Hokkaido Univ., Japan), E D Bauer and M B Maple (UC San Diego, USA) and J L Sarrao (LANL, USA)

1. URu_2Si_2: In the heavy-fermion compound URu_2Si_2, two successive phase transitions occur at 17.5 K and 1.4 K. The transition at 1.4 K is recognized as the onset of unconventional superconductivity (SC) [1]. All the basic properties (resistivity, specific heat, magnetization, thermal expansion) of URu_2Si_2 exhibit a clear mean-field-like transition at $T_0 = 17.5$ K independent of the sample quality. The neutron diffraction (ND), however, revealed that a simple type-I antiferromagnetic (AF) ordering with U moments develops along *c*-axis below T_0 and the observed extremely weak Bragg peak intensities are interpreted as originating from a 'tiny staggered moment' of only 0.03 μ_B/U [2]. Thus, unconventional phase transition at T_0 has remained a mystery for almost 15 years. Recently, the ND experiment performed under pressure up to 28 kbar has shown that the magnitude of the staggered moment (μ) increase linearly up to 0.25 μ_B/U at 10 kbar and then shows a tendency to be saturated at 13 kbar [3]. Since our previous ^{29}Si NMR carried out at ambient pressure could not detect any evidence for AF ordering [4], high pressure ^{29}Si NMR has been performed.

2. $CeTIn_5$: The occurrence of SC in strongly correlated *f*-electron systems has been an active area of research for several decades.

Since 1995, most AF Ce compounds such as $CePd_2Si_2$, $CeRh_2Si_2$, $CeIn_3$ and $CeNi_2Ge_2$ have been shown to become superconductors under high pressure [5]. Unfortunately, little knowledge of SC was revealed in these systems from NMR/NQR owing to the severe experimental conditions of lower superconducting transition temperature (T_c) and higher superconducting critical pressure (P_c). However, new pressure-induced superconductor $CeRhIn_5$ with higher T_c and lower P_c has recently been reported [6]. Besides $CeCoIn_5$ and $CeIrIn_5$ heavy-fermion superconductors, we have performed the ^{115}In NMR/NQR and ^{59}Co NMR on $CeTIn_5$ (T = Rh, Co, Ir) to examine the possibility of unusual SC mediated by AF spin fluctuations near AF instability.

2. Results and discussion

1. URu_2Si_2: The Si NMR spectra with no quadrupolar effect because of $I = 1/2$ for ^{29}Si, were measured under pressure up to 8.3 kbar by sweeping the frequency at $H_{ex} = 4.3$ T. Figure 1 presents the temperature (T) evolution of ^{29}Si NMR spectrum at 8.3 kbar for $H_{ex} \parallel c$-axis. As seen in this figure, just single resonance line was observed at high T, whereas two symmetric resonance lines with respect to the main one appeared below T_0. The result of no splitting for the resonance line for $H_{ex} \perp c$-axis (not shown in the figure) indicates that the internal field (H_{in}) at Si site is parallel to the *c*-axis. Up to now, this AF ordering has been believed to develop uniformly throughout the sample at low T. As is clearly seen from figure 1, the main resonance line decreases drastically upon cooling while the intensity of two H_{in}-split resonance lines increases, suggesting that the AF region increases in volume. At 6 K, the volume fraction of the AF region attains 85% of the whole volume. Figure 2a shows the T variation of H_{in} deduced from the H_{in}-split lines at 8.3 kbar.

The H_{in} increases rapidly below T_0, and keeps almost constant at low T. As seen in figure 2b, the estimated AF volume fraction at 6 K increases with increasing pressure whereas H_{in} (~ 91 mT) remains constant up to 8.3 bar. Assuming a homogeneous AF ordering throughout the sample, the ND results show that the μ value attains 0.25 μ_B/U at 10 kbar. Moreover, the correct ordered moment is estimated to be 0.3 μ_B/U below 8.3 kbar, using the AF volume fraction obtained by the Si NMR. The present Si NMR results show

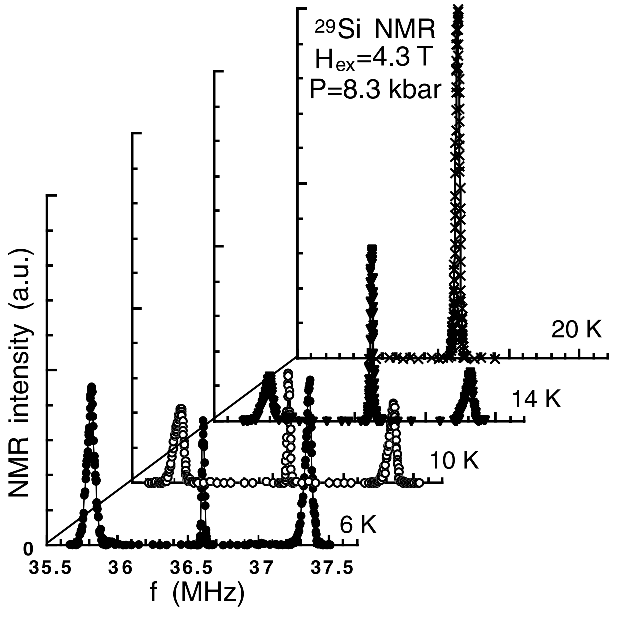

Figure 1. T evolution of ^{29}Si NMR spectrum for $H_{ex} \parallel c$-axis at 8.3 kbar.

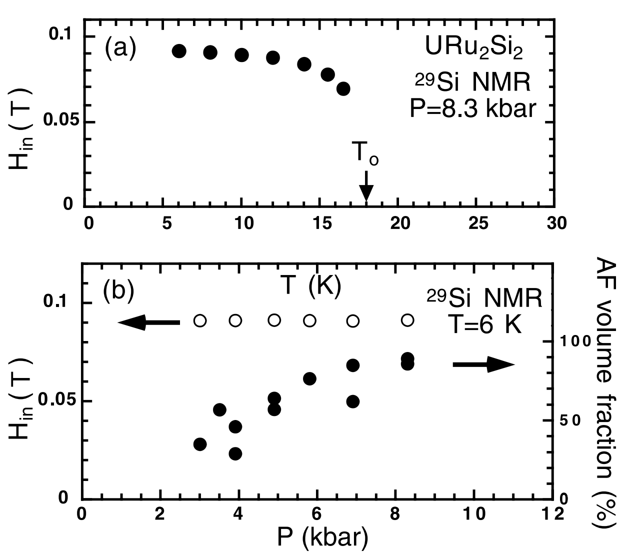

Figure 2. (**a**) T dependence of H_{in} at 8.3 kbar. (**b**) H_{in} and AF volume fraction vs. applied pressure.

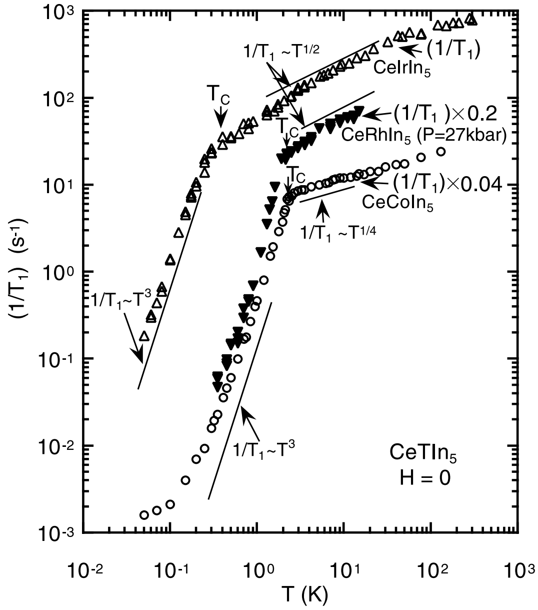

Figure 3. T dependence of $1/T_1$ in $CeTIn_5$.

that the AF region increases in volume with increasing applied pressure at the expense of paramagnetic (PM) region, where a hidden order parameter develops. Consequently, these two types of order compete with each other for volume fraction below T_0. As one of the possibilities for the hidden order parameters, there are some models based on quadrupolar ordering. In models with a doublet Γ_5 ground state of $5f^2$ configuration, the quadrupolar and dipolar orderings intrinsically compete with each other owing to the incommutability of these operators. The quadrupolar ordering model qualitatively explains the present NMR results, although the mechanism by which the magnetic ground state is determined remains unclear.

2. $CeTIn_5$: Distinct In NQR lines could be observed owing to huge quadrupole moment of ^{115}In ($I = 9/2$) in $CeTIn_5$ with a tetragonal $HoCoGa_5$ structure. Four 'allowed' transitions would be expected when asymmetric parameter η is small. Thus, two sets of sharp NQR signals (eight signals) corresponding to two crystallographically inequivalent In sites were observed in the paramagnetic T range (not shown in the figure).

Figure 3 shows the T dependence of nuclear spin-lattice relaxation rate ($1/T_1$) in $CeRhIn_5$ at $P = 27$ kbar, and $CeCoIn_5$ and $CeIrIn_5$ at ambient pressure. In the normal state $1/T_1$ varies close to $T^{1/2}$ in $CeRhIn_5$ and $CeIrIn_5$, and close to $T^{1/4}$ in $CeCoIn_5$, which are qualitatively explained by the spin fluctuations developed around the AF instability, following SCR theory by Ishigaki and Moriya [7]. Below T_c (2.3 K for $CeCoIn_5$, 0.3 K for $CeIrIn_5$, 2.2 K for $CeRhIn_5$ at $P = 27$ kbar), $1/T_1$ decreases nearly proportionally to T^3 at low T, indicating the energy gap with line nodes, which is reminiscent of relaxation behavior in many heavy-fermion superconductors such as UPt_3, URu_2Si_2, UPd_2Al_3 etc. Assuming a simple polar model $\Delta = \Delta_0 \cos\theta$, the calculated T_1 values reproduce well the

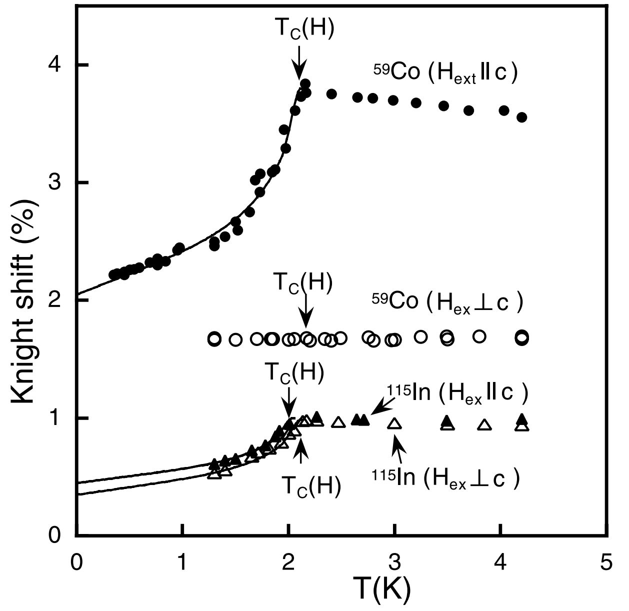

Figure 4. T dependence of Knight shifts ^{59}K and ^{115}K in $CeCoIn_5$. $T_c(H)$ stands for T_c under the external field applied for the NMR measurements.

experimental ones by evaluating tentatively $\Delta_0 = 10\ k_BT_c$, $8\ k_BT_c$ and $5\ k_BT_c$ for $CeCoIn_5$, $CeRhIn_5$ and $CeIrIn_5$, respectively.

Next, the T evolution of ^{115}In and ^{59}Co Knight shifts (abbreviated as ^{115}K and ^{59}K, respectively) below 4 K are shown in figure 4. As clearly seen in this figure, a large decrease of ^{115}K below T_c was observed for parallel and perpendicular directions to c-axis and a decrease of ^{59}K was observed only for parallel component, which has inferred the SC of even parity. No decrease of ^{59}K perpendicular to c-axis is expected mainly due to the large Co orbital contribution. The calculated T dependence of K by using the same energy gap value for T_1 analysis is also plotted in figure 2. The nearly T linear variation for K at very low T, which arises from the low energy excitation of quasi-particles in the superconducting state, has also clearly been measured. Thus, both T variations of $1/T_1$ and K are successfully explained by the polar model, indicating strongly the appearance of anisotropic even parity (probably d-wave) SC in $CeTIn_5$.

3. Summary

We observed ^{29}Si NMR signals arising from the AF region besides the previously observed ^{29}Si NMR signals which come from the PM region in the sample. This gives definite evidence for spatially-inhomogeneous development of AF ordering below T_0 of 17.5 K. The present Si NMR results indicate that the weakness of AF Bragg peak at ambient pressure originates not from the extremely small magnitude of magnetic moment, but from the smallness of the AF region in the sample. Next, in the normal state of $CeTIn_5$, the

T dependence of $1/T_1$ is well explained by the spin fluctuations developed around the AF instability (quantum critical point, QCP). Below T_c, the SC in $CeTIn_5$ is probably of d-wave.

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