Spatially Inhomogeneous Development of Antiferromagnetism in URu_2Si_2: Evidence from ^{29}Si NMR under Pressure

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(Received 22 January 2001; published 1 August 2001)

From ^{29}Si NMR study, we present evidence for spatially inhomogeneous development of antiferromagnetic (AF) ordering below $T_o = 17.5$ K in URu_2Si_2. In the pressure range between 3.0 and 8.3 kbar, we have observed the ^{29}Si NMR lines arising from the AF region as well as the previously observed ^{29}Si NMR line which correspond to the nonmagnetic region in the sample. The AF volume fraction is enhanced by applied pressure, whereas the magnitude of internal field at the Si site remains constant (910 Oe) up to 8.3 kbar. In the AF region, the ordered moment is about an order of magnitude larger than 0.03 μ_B/U.

DOI: 10.1103/PhysRevLett.87.087203 PACS numbers: 75.30.Mb, 75.50.Ee, 76.60.–k

The heavy-fermion compound URu_2Si_2 exhibits two successive phase transitions at 17.5 and 1.4 K. The transition at 1.4 K is recognized as an onset of unconventional superconductivity [1]. On the other hand, the phase transition at $T_o = 17.5$ K still remains mysterious for more than one decade. This phase transition at T_o is clearly discerned in the temperature (T) dependence of macroscopic quantities [1–3]. Remarkably, the T dependence of specific heat shows a clear λ-type anomaly at T_o. Associated with this phase transition, the energy gap opens over part of the Fermi surface, is reflected by an exponential T dependence of specific heat, electrical resistivity, and nuclear spin-lattice relaxation rate (T_1^{-1}) below T_o [1,2,4,5]. Below T_o, the development of a simple type-I antiferromagnetic (AF) ordering with 5 f magnetic moments along c axis has been revealed by the neutron diffraction (ND) [6]. The observed magnetic Bragg peak intensities are extremely weak, which has been interpreted as originating from a "tiny staggered moment" of only $0.03\mu_B$/U. However, this "tiny staggered moment" is hard to reconcile with large macroscopic anomalies observed at T_o. To explain this unusual phase transition at T_o, several scenarios have been proposed. Some theoretical approaches are based on the idea that macroscopic anomalies do not directly come from the AF transition, but rather from a transition associated with hidden order parameter [7–11]. Several papers have dealt with the possibility of the quadrupolar moments of U 5f electrons for the hidden order parameter [9–11]. However, no experimental evidence for quadrupolar ordering has been obtained up to now. In addition to a hidden order parameter hypothesis, there are other approaches that tiny staggered moments with highly reduced g values solely drive the phase transition at T_o [12–14].

Recently, the ND experiment performed under pressure up to 28 kbar has revealed that the staggered moment (μ) is strongly enhanced by applying pressure; the magnitude of μ increases linearly up to $0.25\mu_B$/U at 10 kbar and then shows a tendency to be saturated at 13 kbar [15]. At critical pressure ($P_C = 15$ kbar), the value of μ jumps to $0.4\mu_B$/U indicating an occurrence of the pressure-induced phase transition. The T dependence of μ, $\mu(T)$ changes its character at P_C; $\mu(T)$ is almost proportional to $\sqrt{T - T_o}$ below P_C, whereas $\mu(T)$ above P_C is similar to that of 3D Ising model. In spite of a remarkable enhancement of μ, the application of pressure has little effect on a macroscopic anomaly and causes only a slight increase in the value of T_o [16–18].

Interestingly, the muon spin resonance (μSR) results give two conflicting pictures with respect to the AF ordering in URu_2Si_2. Most of the μSR results have shown that the AF ordering with a tiny staggered moment develops homogeneously below T_o throughout the sample [19,20]. On the other hand, one μSR result has shown an inhomogeneous picture; below T_o, the zero-field μSR spectrum has a two-component structure which is well described by a sum of the signals arising from nonmagnetic and magnetic regions [21]. This μSR result suggests that the AF ordering develops at the most 10% of the sample at ambient pressure.

In order to identify the nature of unconventional AF ordering in URu_2Si_2, we performed ^{29}Si NMR experiments under pressure up to 8.3 kbar. The previous ^{29}Si NMR performed at ambient pressure could not detect any evidence for the AF ordering in URu_2Si_2 [4]. In this paper, we show definite evidence for coexistence of the AF region and the paramagnetic (PM)—region below T_o in URu_2Si_2.

The NMR measurements were carried out by the conventional spin-echo technique with a phase-coherent pulsed spectrometer. The detail of the sample preparation was given in a previous paper [15]. The measurements at ambient pressure were performed on a cylindrical single crystal of about 2 mm in diameter and 10 mm in length. As for the measurements in pressure, a polycrystalline sample, which was crushed into powder, was used. The powder sample was mixed with a small amount of Sn powder, of which superconducting transition was used for the determination of applied pressure. Subsequently, the sample mixed with Sn was embedded in staycast and then oriented under the H_{ex}; the microcrystals in a powder sample of URu_2Si_2 are easily oriented with the c axis parallel to the direction of the H_{ex} owing to the large anisotropy of susceptibility. Using this sample, we could measure the ^{29}Si NMR spectra for $H_{ex} \perp c$-axis as well as $H_{ex} \parallel c$-axis. The pressure was generated by a standard Cu-Be pressure cell with a 1:1 mixture of Flourinert 70 and 77 as a transmitting medium. To estimate pressure distribution $(\Delta P/P)$ in the prepared sample, we monitored T width of superconducting transition of Sn powder cast in staycast by means of ac susceptibility measurement. The evaluated $\Delta P/P$ is less than 5% even at $P = 8.3$ kbar, indicating that the inhomogeneity of pressure is negligibly small.

The ^{29}Si NMR spectra were measured under pressure up to 8.3 kbar by sweeping the frequency at $H_{ex} = 4.3$ T. As for ^{29}Si NMR, no quadrupolar effect is expected because of the nuclear spin $I = 1/2$ of ^{29}Si. Figure 1 presents the T evolution of the ^{29}Si NMR spectrum at 8.3 kbar for $H_{ex} \parallel c$ axis. As seen in Fig. 1, the single resonance line was observed at high T, whereas the additional resonance lines, which are symmetric with respect to the main resonance line, appeared below T_o. Note that there is no splitting of the resonance line for $H_{ex} \perp c$ axis. This fact indicates that the main resonance line and the two additional resonance lines correspond to the Si sites in which the internal field (H_{in}) is either zero or nonzero (i.e., parallel to the c axis), respectively. Therefore, the present ^{29}Si NMR result is consistent with the type-I AF spin structure previously determined by ND measurement [6]. Up to now, this AF ordering has been believed to develop uniformly throughout the sample at low T. On the other hand, the ^{29}Si NMR spectra show the resonance line arising from the PM region remains unambiguously below T_o and coexists with the resonance lines arising from the AF region. This gives direct evidence for the coexistence of the AF and the PM regions in the sample. As is clearly seen from Fig. 1, upon cooling, the main resonance line decreases drastically while the intensities of the additional resonance lines (i.e., the H_{in}-split resonance lines) increase, suggesting the AF region increases in volume. To quantify the development of volume fraction of the AF region, we measured the T variations of integrated intensities of the resonance lines arising from the AF region as well as the PM region at several pressures. As for the signal intensity, we performed the correction for spin-echo decay rate (T_2^{-1}) in the spin-echo method. Taking into consideration the Curie law of nuclear spin susceptibility, the values of the product of the spectral intensity and $T(I \times T)$ are plotted against T in Fig. 2(a). The $I \times T$ for the PM region, which is proportional to the PM volume fraction, significantly decreases below T_o with decreasing T, while the $I \times T$ for the AF region increases. These results obviously indicate that the AF region increases in volume at the expense of the PM region on cooling below T_o. At the T much lower than T_o, the PM region still remains in the sample. Figure 2(b) shows the T variation of H_{in} deduced from the H_{in}-split resonance lines at 8.3 kbar. The H_{in} increases rapidly just below T_o, and then keeps almost constant at low T. It should be noted that we could not determine the magnitude of H_{in} around T_o because of an extremely weak signal

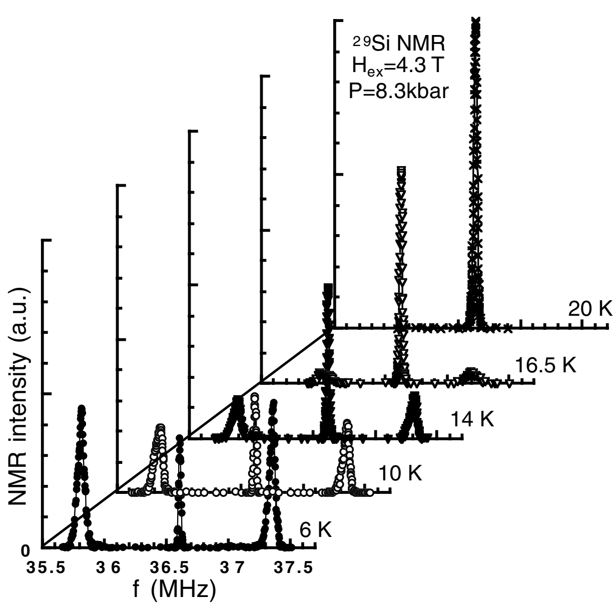

FIG. 1. T evolution of ^{29}Si NMR spectrum for $H_{ex} \parallel c$ axis at 8.3 kbar.

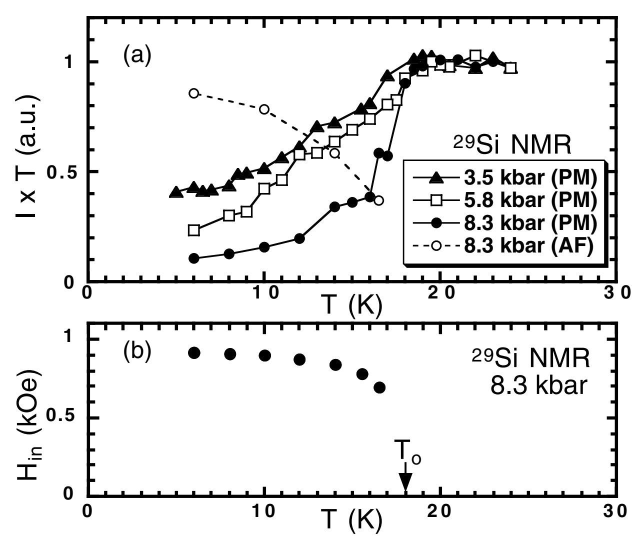

FIG. 2. (a) T dependences of $I \times T$ deduced from the ^{29}Si NMR arising from the PM and AF regions. (b) T dependence of H_{in} at 8.3 kbar. T_o indicated in the figure was determined from the T in which the $I \times T$ for the ^{29}Si NMR arising from the PM region starts to decrease markedly.

intensity arising from the AF region. At 6 K, the magnitude of H_{in} is obtained to be 915 Oe. We obtained the ^{29}Si NMR spectra between 3.0 and 8.3 kbar, which indicates that the H_{in} remains constant ($H_{in} = 910 \pm 10$ Oe) in this pressure range.

With comparing the results of the present ^{29}Si NMR and the previous ND, we now discuss the pressure effect on the AF ordering in URu_2Si_2. In Fig. 3, the H_{in} at Si-site in the AF region and the AF volume fraction taken at 6 K are plotted against pressure. The AF volume fractions (denoted by • in Fig. 3) were determined by comparing the integrated intensities of the ^{29}Si NMR resonance lines arising from the PM regions and the AF regions. Besides this plotting, Fig. 3 also shows the pressure variation of the AF volume fraction (denoted by ∘) estimated from the reduction in $I \times T$ for the ^{29}Si NMR arising from the PM region as compared to the value just above T_o. The values obtained by the two ways are almost the same, which confirms that we observed all the NMR signals arising from the Si site in the sample. For comparison, the pressure variation of integrated intensity of the (100) magnetic Bragg reflection (I_B) at 6 K is also displayed in the figure [15]. In general, the I_B is simply expressed by $I_B \propto \frac{V_D}{V_0}\mu^2$, where V_D and V_0 are the volumes of the domain and the sample, respectively. This expression means I_B depends on the AF volume fraction (i.e., $\frac{V_D}{V_0}$) as well as μ. The ^{29}Si NMR results indicate that the AF volume fraction increases with increasing pressure, whereas μ is constant up to 8.3 kbar. Hence, an enhancement of I_B by pressure is attributed to an increase in the AF volume fraction, not to an increase in the magnitude of μ. When μ is constant, the AF volume fraction is in proportion to I_B. As seen in the figure, the AF volume fractions obtained from the ^{29}Si NMR and the ND have almost the same pressure dependence up to 8.3 kbar. Assuming a homogeneous AF ordering throughout the sample, the ND result shows that the magnitude of μ increases linearly up to $0.25\mu_B$/U at 10 kbar and then tends to be saturated. Using the AF volume fractions obtained by the ^{29}Si NMR, the correct ordered moment is estimated to be about $0.3\mu_B$/U below P_C. However, the jump in the pressure dependence of I_B at P_C is difficult to explain only by the increase in the AF volume fraction, suggesting the discontinuous increase in magnitude of μ at P_C. With reference to the growth of μ upon cooling, I_B is reported to exhibit the weak T dependence below P_C, whereas the H_{in} at Si site increases very rapidly just below T_o. Therefore, the unconventional T dependence of I_B evidently originates not from an unusually slow saturation of μ, but from an increase in the AF volume fraction upon cooling. The quite similar T dependence of μ, as obtained in this study, has been found by a μSR experiment on a single crystalline URu_2Si_2 at ambient pressure [21].

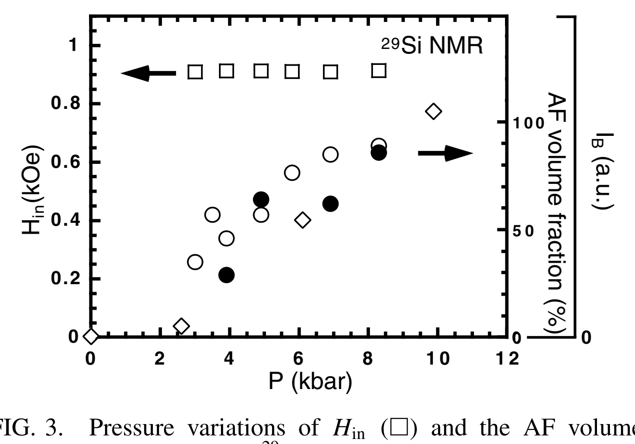

FIG. 3. Pressure variations of H_{in} (□) and the AF volume fraction (•, ∘) obtained by ^{29}Si NMR (see text). For comparison, the (100) magnetic Bragg intensity, I_B, data (◇) deduced from Ref. [15] are also plotted against pressure in the figure.

Next turning to the ND result at ambient pressure, it is clear from the present ^{29}Si NMR results that the small I_B comes not from the extremely small μ but from the reduced AF region in the sample. Actually, at ambient pressure, the $I \times T$ for the PM region is constant within the experimental uncertainty of about 10%. Because of the extremely small AF region, we have not directly observed the ^{29}Si NMR signal arising from the AF region for $H_{ex} \parallel c$-axis below 3.0 kbar. Thus, using the single crystalline sample, we measured preciously the ^{29}Si NMR resonance line arising from the PM region at 5.4 T to obtain indirect information on the AF order at ambient pressure. In Fig. 4 are shown the T dependences of the full-width at half-maximum (FWHM) of the observed resonance line for $H_{ex} \parallel c$ axis and $H_{ex} \perp c$ axis. As seen in the figure, the value of FWHM slightly increases below T_o indicating the appearance of additional H_{in} at Si site in the PM region. However, the increase in FWHM below T_o is ex-

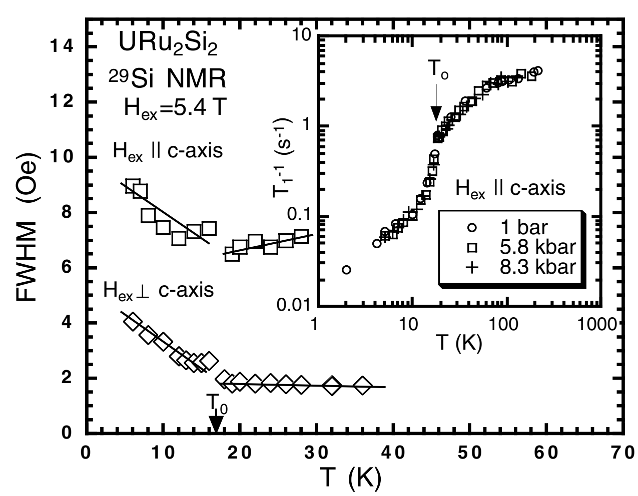

FIG. 4. T dependence of FWHM for the ^{29}Si NMR resonance line arising from the PM region at ambient pressure. The values for $H_{ex} \parallel c$ axis and $H_{ex} \perp c$ axis are denoted by □ and ◇, respectively. Lines are guides for the eye. The inset shows the T dependence of T_1^{-1} for $H_{ex} \parallel c$ axis at 5.8 and 8.3 kbar. The reported T_1^{-1} data at ambient pressure are also displayed in the figure. All the T_1^{-1} were obtained by using the ^{29}Si NMR signal arising from the PM region.

tremely small (about 2 Oe). This result indirectly shows an existence of the AF region in the sample. Since the $H_{\rm in}$ appears along the c axis at Si site in the AF region, the observed resonance line for $H_{\rm ex} \perp c$ axis is composed of the signals arising from the AF region as well as the PM region below T_o. In contrast, the observed resonance line for $H_{\rm ex} \parallel c$ axis is arising only from the PM region. Hence, the staggered moments in the AF tiny regions, which are distributed in a fairly uniform manner throughout the sample, must produce an inhomogeneous $H_{\rm in}$ at Si site in the PM region in order to give the observed broadening of the line. This result is associated with the finite correlation length of AF ordering observed by ND at ambient pressure [6,15].

Finally, let us discuss the origin of the phase transition at T_o. The present ^{29}Si NMR result indicates that the ordered moment is about an order of magnitude larger than $0.03\mu_B$/U in the AF region, which is inconsistent with the theoretical approach simply based on the "tiny staggered moments" with highly reduced g values. The inset of Fig. 4 shows the T dependence of T_1^{-1} measured by using the main resonance line at 5.8 and 8.3 kbar. In the figure, the reported T_1 data at ambient pressure are also plotted [4]. As seen in the figure, T_1^{-1} decreases drastically below T_o, which indicates that the energy gap opens partially on the Fermi surface associated with the phase transition at T_o. Furthermore, the T dependence of T_1^{-1} is almost pressure independent up to 8.3 kbar. Here, it should be emphasized that all the T_1^{-1} data in the figure were selectively measured by the ^{29}Si NMR signal arising from the PM region. Hence, it is obvious that a drastic decrease in T_1^{-1} below T_o is not ascribed only to the AF ordering of which the volume fraction is extremely small at ambient pressure. This indicates the existence of a phase transition associated with the hidden order parameter in the PM region. At ambient pressure, the macroscopic anomalies observed at T_o must originate predominantly from the transition associated with a hidden order parameter. The present ^{29}Si NMR results show that the AF region increases in volume at the expense of the PM region, in which a hidden order parameter develops, as pressure is increased. Consequently, these two types of order compete with each other for volume fraction below T_o. 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 the $5f^2$ configuration, the quadrupolar orderings and dipolar orderings intrinsically compete with each other owing to the incommutability of these operators [10,11,22]. The quadrupolar-ordering model qualitatively explains the present NMR results, although the mechanism by which the magnetic ground state is determined still remains unclear.

In conclusion, the ^{29}Si NMR performed in pressure up to 8.3 kbar has revealed a coexistence of the AF and PM regions below T_o in URu_2Si_2. The AF region increases in volume at the expense of the PM region on cooling. The AF volume fraction is enhanced by pressure, whereas the magnitude of μ is constant up to 8.3 kbar. Our ^{29}Si NMR results indicate that the weakness of the AF Bragg peak at ambient pressure originates not from the extremely small magnitude of μ, but from the smallness of the AF region in the sample. In the AF region, the ordered moment is about an order of magnitude larger than $0.03\mu_B$/U.

We thank S. Kawarazaki and F. J. Ohkawa for helpful discussions. This work was partly supported by a Grand-in-Aid from the Ministry of Education, Culture, Sports, Science and Technology of Japan. One of the authors (K. M.) has been supported by JSPS.

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