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The appearance of homogeneous antiferromagnetism in URu_2Si_2 under high pressure: a ^{29}Si nuclear magnetic resonance study

K Matsuda[1,4], Y Kohori[1,5], T Kohara[1], H Amitsuka[2], K Kuwahara[2,4] and T Matsumoto[3]

[1] Division of Material Science, Graduate School of Science, Himeji Institute of Technology, Kamigori, Ako-gun, Hyogo 678-1297, Japan
[2] Division of Physics, Graduate School of Science, Hokkaido University, Sapporo 060-0810, Japan
[3] National Institute for Materials Science, Sengen, Tsukuba, Ibaraki 305-0047, Japan

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Abstract

We have investigated the low-temperature phase appearing below $T_o = 17.5$ K in URu_2Si_2 by means of ^{29}Si nuclear magnetic resonance (NMR) in a pressure range from 0 to 17.5 kbar across the pressure-induced phase transition at $P_c = 15$ kbar. At pressures below P_c, we have observed the ^{29}Si NMR lines arising from antiferromagnetic (AF) and paramagnetic (PM) regions in the sample, giving evidence for a phase-separated AF ordering below T_o. The AF region increases in volume fraction with increasing pressure up to P_c. In the PM region, the temperature-dependence of the nuclear spin–lattice relaxation rate at Si sites shows a rapid decrease below T_o, strongly suggesting the occurrence of a phase transition driven by a hidden order parameter. As applied pressure exceeds P_c, the AF ordering appears uniformly at T_o throughout the sample. In the pressure range from 0 to 17.5 kbar, the magnitude of the internal field at Si sites in the AF region remains constant (∼920 Oe), which indicates that the AF ordered moment is unchanged by pressure across P_c.

The heavy-fermion superconductor URu_2Si_2 has attracted much attention owing to its mysterious phase transition at $T_o = 17.5$ K. This phase transition is accompanied by appreciable anomalies in macroscopic quantities [1–3]. In particular, a large lambda-shaped anomaly is observed in the dependence on the temperature (T) of the specific heat at T_o. Associated with this phase transition, an energy gap opens over part of the Fermi surface

[4] Present address: Department of Physics, Graduate School of Science, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397, Japan.
[5] Present address: Department of Physics, Faculty of Science, Chiba University, Inage-ku, Chiba 263-8522, Japan.

below T_o. This is reflected by an exponential T-dependence of the specific heat, electrical resistivity, and nuclear spin–lattice relaxation rate (T_1^{-1}) [1, 4–6]. Below around T_o, neutron diffraction (ND) and x-ray magnetic scattering measurements have confirmed the development of a type-I antiferromagnetic (AF) ordering with U 5f magnetic moments along the c-axis [7, 8]. The observed AF Bragg peak intensities are extremely weak; they have been interpreted as originating from a 'tiny moment' of only $\sim$0.03 μ_B/U. However, with this 'tiny moment' it is hard to account for the observed macroscopic anomalies at T_o. This discrepancy has led to the suggestion that the macroscopic anomalies do not directly originate from an AF transition, but rather from a transition driven by a hidden order parameter. Some theoretical models have been proposed on the basis of this hidden order parameter hypothesis, though the hidden order parameter has not been elucidated experimentally so far [9–16]. Recent ND and nuclear magnetic resonance (NMR) measurements under pressure (P) have revealed a drastic pressure effect on the 'tiny moment' AF ordered state, giving a clue to how to settle the above-mentioned discrepancy [17, 18]. The ^{29}Si NMR measurements under pressure have found phase-separated AF ordering; between 3.0 and 8.3 kbar, the ^{29}Si NMR spectrum below T_o is a superposition of the signals arising from AF and paramagnetic (PM) regions in the sample. In the AF phase, the magnitude of the ordered moment remains constant in this pressure range while the volume fraction of the AF phase is enhanced by applied pressure. At ambient pressure, no NMR measurements have yet detected the AF phase because of its small volume fraction, although the μSR results obtained by Luke *et al* in 1994 [4, 5, 19, 20] gave evidence for the existence of an AF phase whose volume fraction is $\sim$10%. A pressure-dependent AF volume fraction is also supported by previous ND results, which indicate that the AF Bragg peak intensity is enhanced by pressure. The averaged magnitude of the AF moment, μ_{ND}, which is calculated on the assumption of a uniform AF ordering throughout the sample, is continuously increased from 0.02 μ_B/U (at ambient pressure) to 0.25 μ_B/U (at $\sim$10 kbar). Furthermore, the ND experiments have revealed a pressure-induced phase transition at $P_c = 15$ kbar. At P_c, the value of μ_{ND} shows a discontinuous jump from 0.23 to 0.40 μ_B/U without change in the AF spin structure. The pressure dependences of T_o and the lattice parameter a also show discontinuous change at P_c. This transition brings about a drastic change in the character of the T-dependence of μ_{ND}, $\mu_{ND}(T)$. The growth of $\mu_{ND}(T)$ is almost proportional to $\sqrt{T_o - T}$ below T_o, whereas $\mu_{ND}(T)$ above P_c is well described by a 3D Ising model. Although applying pressure considerably modifies the electronic state below T_o on a microscopic level, the macroscopic anomaly at T_o is almost unchanged by pressure [21–26].

In order to investigate the pressure-induced evolution of the magnetic properties of URu_2Si_2 from a microscopic point of view, we have performed ^{29}Si NMR measurements under pressure. Some results for below 8.3 kbar have already been reported [18]. In the present paper, we report new ^{29}Si NMR results for URu_2Si_2 over a wide pressure range up to 17.5 kbar, which covers pressure ranges on both sides of P_c. From the detailed measurements of the variations with pressure and T of the ^{29}Si NMR spectrum, we reveal that the system changes from a coexistence state of AF and PM regions below P_c to a uniform AF ordering state above P_c. Furthermore, our T_1^{-1}-results show that a drastic change takes place in the electronic state of the PM region at T_o, which provides evidence for a phase transition due to hidden order.

The pressure was generated by a nonmagnetic piston–cylinder-type pressure cell with a 1:1 mixture of Flourinert 70 and 77 as a transmitting medium. The applied pressure was determined by using the superconducting transition of Sn metal. The sample used was the same as that used in the earlier measurements; a powder sample was embedded in Stycast and then oriented under the field H_{ex} (see [18] for details). The measurements at ambient pressure were

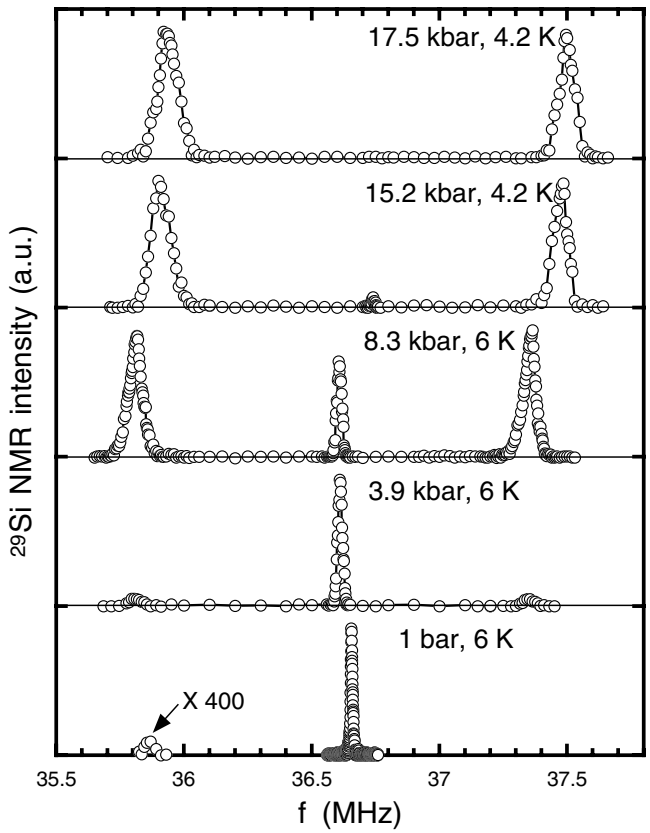

Figure 1. The pressure variation of the ^{29}Si NMR spectrum at low T. In the figure, previously reported data for 4.0 and 8.3 kbar are also shown for comparison. The small change in frequency of the central resonance line is due to the difference between the fields at which the spectra were taken. The present spectral measurements were carried out at a slightly higher field than the previous ones. At ambient pressure, we measured only the lower-frequency resonance line of the H_{in}-split lines because of the low intensity of the signal arising from the AF region.

performed on a cylindrical single crystal about 2 mm in diameter and 10 mm in length. The NMR measurements were carried out using a phase-coherent pulsed NMR spectrometer. The ^{29}Si NMR spectrum was obtained by plotting the spin-echo signal as a function of frequency at 4.3 T. In the measurements at ambient pressure, the technique of Fourier transforming the spin-echo signal was also employed because of the extreme narrowness of the resonance line arising from the single-crystalline sample.

In figure 1, we show the pressure variation of the ^{29}Si NMR spectrum for $H_{ex} \parallel c$-axis measured at low T well below T_o. In the pressure range $P < P_c$, each spectrum is composed of a central line and two satellite lines, which are symmetrically located with respect to the central line. Since ^{29}Si has nuclear spin $I = 1/2$, no quadrupolar effect contributes to the ^{29}Si NMR. Furthermore, in URu_2Si_2, there is crystallographically a site equivalent to Si. Therefore, the satellite lines arise from the Si site where the internal field (H_{in}) is either parallel or antiparallel to H_{ex}. That is, the satellite lines, hereafter referred to as H_{in}-split lines, correspond to the Si site in the AF region. On the other hand, the central line arises from Si site in the PM region. This assignment is confirmed by the experimental facts that the satellite lines are observed

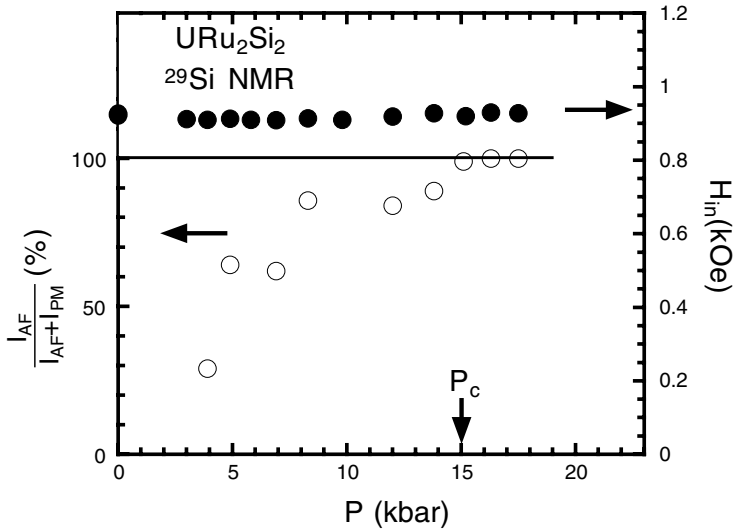

Figure 2. The pressure dependence of H_{in} and the normalized intensity of the H_{in}-split lines ($\frac{I_{AF}}{I_{AF}+I_{PM}}$).

only below T_o, and that no splitting of the resonance line is observed for $H_{ex} \perp c$-axis. As for the AF ordering, the results of the ^{29}Si NMR measurements are consistent with the type-I AF spin structure previously determined by means of ND. Hence, this NMR spectrum evidently indicates that the electronic phase separation into AF and PM regions occurs at low T in this system. In this pressure range $P < P_c$, as shown in the figure, the H_{in}-split lines are progressively enhanced by pressure, whereas the central line loses intensity. This indicates that the AF region volume fraction is enhanced with increasing pressure. It is remarkable that the H_{in}-split line has been detected at ambient pressure. This directly evidences that AF ordering with a 'static' nature takes place in an extremely small portion of the sample even at ambient pressure. Hence, this NMR result rules out a picture where the AF spin structure with 'tiny moments' develops uniformly throughout the sample at ambient pressure. At 17.5 kbar ($P > P_c$), the central line completely disappears, and the H_{in}-split lines alone are detectable at 4.2 K. Hence, the AF ordering develops uniformly throughout the sample at low T. From the spectrum, the magnitude of H_{in} at the Si site in the AF region was extracted. Figure 2 shows the pressure dependence of the extracted H_{in}. Over the whole pressure range up to 17.5 kbar, H_{in} remains constant ∼920 Oe, distinctly indicating that the magnitude of the AF ordered moment is unchanged by pressure. In order to provide quantitative information on the pressure-induced evolution of the AF ordering, we estimated the AF volume fractions (v_{AF}) from the ^{29}Si NMR spectra. In figure 2, the normalized intensity of the H_{in}-split lines ($\frac{I_{AF}}{I_{AF}+I_{PM}}$) is plotted as a function of pressure. Here, I_{AF} and I_{PM} represent the integrated intensities of the H_{in}-split lines and the central line, respectively. As for the signal intensity, we performed a correction for the spin-echo decay rate, T_2^{-1}, in the spin-echo method. As seen in the figure, $\frac{I_{AF}}{I_{AF}+I_{PM}}$ increases continuously with increasing pressure, and then reaches 100% around P_c. At ambient pressure, we cannot extract a reliable value of $\frac{I_{AF}}{I_{AF}+I_{PM}}$ owing to the extremely low intensity of the signal arising from the AF region.

Next, we deal with the T-variation of the ^{29}Si NMR spectrum measured at constant pressure. Figure 3(a) displays the ^{29}Si NMR spectra for $H_{ex} \parallel c$-axis obtained at 13.5 kbar,

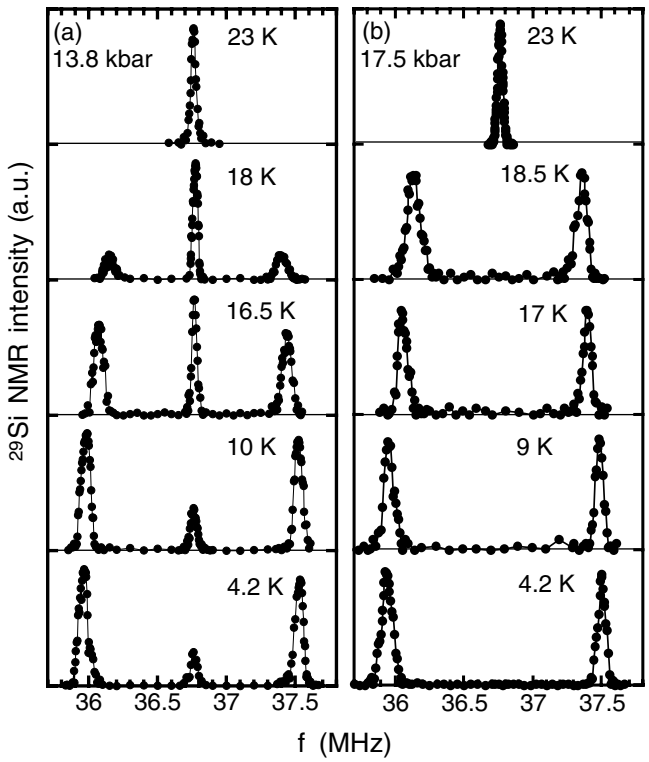

Figure 3. T-variations of the ^{29}Si NMR spectrum for (a) 13.8 kbar ($P < P_c$) and (b) 17.5 kbar ($P > P_c$).

slightly lower than P_c. A single resonance line is observed above T_o, whereas the H_{in}-split lines newly appear around T_o. As can be seen in the figure, the central line significantly decreases in intensity with decreasing T, whereas the H_{in}-split lines increase in intensity. This T-variation of the spectral intensity cannot be explained solely by the change in the T_2 of each line. In figure 4, the values of $v_{AF} = \frac{I_{AF}}{I_{AF}+I_{PM}}$, which are corrected for T_2, are plotted against T. As seen in the figure, $\frac{I_{AF}}{I_{AF}+I_{PM}}$ increases with decreasing T. This indicates that the AF ordering takes place in a portion of the sample around T_o and then increases in volume fraction with decreasing T. As previously reported for the same sample, the increase in AF volume fraction on cooling has been confirmed in the pressure range $P \leqslant 8.3$ kbar. Hence, this feature is common to the whole pressure range $P < P_c$. In figure 4, the T-dependence of the sum of spectral intensities, $I_{AF} + I_{PM}$, at 13.8 kbar is also shown. Taking into consideration the Curie law of nuclear spin susceptibility, the magnitude of $(I_{AF} + I_{PM})T$ is plotted against T. If the region undetectable by means of ^{29}Si NMR develops in the sample below T_o, the magnitude of $(I_{AF} + I_{PM})T$ could decrease below T_o. As can be seen in the figure, the magnitude of $(I_{AF} + I_{PM})T$ does not decrease significantly within our experimental accuracy. From the present measurements, therefore, we cannot confirm the existence of an undetectable region with volume fraction larger than $\sim$20%. As seen in figure 3, the resonance lines of the spectrum are well separated in the measured T-range. The sharp H_{in}-split lines, of which the full width at half-maximum is almost T-independent $\sim$60 Oe, reflect a uniform magnitude of the staggered moment in the AF region. The H_{in}-values at the Si site extracted from the spectra are also plotted in figure 4. H_{in} increases rapidly just below T_o, and then remains almost constant at low T. As applied

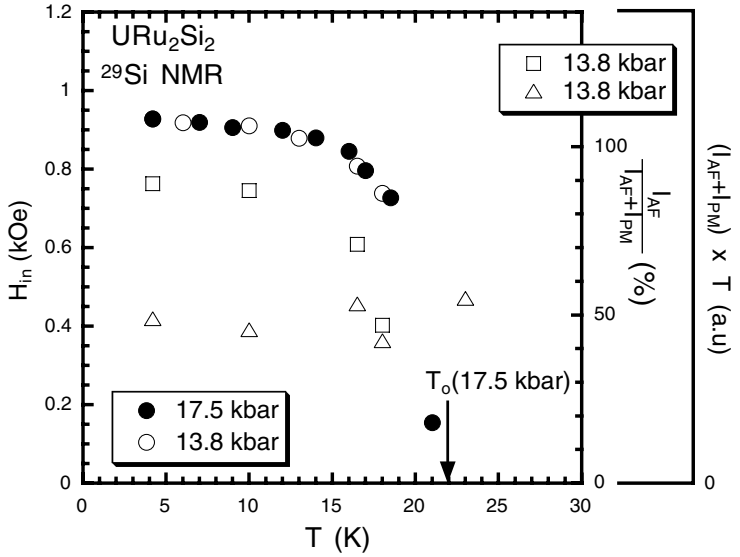

Figure 4. T-dependences of H_{in} (●, ○) and $v_{AF} = \frac{I_{AF}}{I_{AF}+I_{PM}}$ (□) extracted from ^{29}Si NMR spectra. In the figure, the magnitude of $(I_{AF} + I_{PM})T$ (△) is also plotted against T.

pressure exceeds $P_c = 15$ kbar, a drastic change occurs in the character of the T-variation of the ^{29}Si NMR spectrum. Figure 3(b) shows the ^{29}Si NMR spectra taken at 17.5 kbar. As seen in the figure, at T_o, a resonance line splits into two lines, namely H_{in}-split lines. The central line, which is commonly observed for $P < P_c$, completely disappears below T_o. This indicates that no PM region remains in the sample below T_o. Hence, the AF ordering occurs uniformly throughout the sample at 17.5 kbar ($P > P_c$), in contrast to the gradual increase in AF volume fraction on cooling for $P < P_c$. As seen in figure 4, the T-dependence of H_{in} at 17.5 kbar is the same as that obtained at 13.5 kbar.

Now, we compare the present ^{29}Si NMR results with those obtained by the previous ND measurements. According to the ND results for $P < P_C$, the T-variation of the AF Bragg intensity (I_B) shows an unusually slow saturation; I_B is proportional to $T_o - T$ over a wide T-range [17]. In this pressure range, the present ^{29}Si NMR measurements have revealed the electronic phase separation into AF and PM regions. In this case, the AF Bragg peak intensity is related to the AF volume fraction as well as the magnitude of the AF moment (μ_{AF}): $I_B \propto v_{AF}\mu_{AF}^2$. The magnitude of μ_{AF} increases rapidly just below T_o and then remains almost constant at low T, as is reflected by the T-dependence of H_{in} at Si sites. Hence, the unusually slow saturation of $I_B(T)$ originates rather from the gradual increase in the AF volume fraction on cooling. As for the pressure dependence of the AF ordered moment, the ^{29}Si NMR results indicate the magnitude of H_{in} to remain constant (∼920 Oe) across P_c up to 17.5 kbar. Thus, the enhancement of I_B by pressure comes from the increase in the AF volume fraction. At P_c, the pressure dependence of I_B shows a discontinuous increase with $I_B(15\text{ kbar})/I_B(13\text{ kbar}) \sim 2.5$. Using the relation $I_B \propto v_{AF}$ for the constant AF ordered moment, v_{AF} is estimated to be ∼40% at 13 kbar, just below P_c. On the other hand, the v_{AF} extracted from the ^{29}Si NMR spectrum does not show such a discontinuous increase at P_c, as can be seen in figure 2. One of the possible origins of the different v_{AF}-values is that our NMR may not detect all the ^{29}Si nuclei in the PM region owing to the short spin-echo decay times. Considering the limitation of the spectrometer that we employed, we expect T_2 at such Si sites to be shorter than 180 μs. In the region where U 5f electronic states have entered

a crossover regime toward the 'static' AF ordering, the spin-echo decays of ^{29}Si nuclei are expected to be governed by a fast T_1-process associated with a critical slowing down of U 5f moments. This gives rise to an overestimation of the AF volume fraction. Another possible explanation is that the sample used in the present study has a larger AF volume fraction than that in the previous ND measurements. Indeed, Luke *et al* [20] have reported a strong sample dependence of the AF volume fraction in URu_2Si_2 at ambient pressure, obtained from μSR measurements. In the present study, we employed a polycrystal annealed at 1000 °C for a week. The sample was crushed into powder in order to make rf pulses penetrate into the samples easily. The powdering of the bulk URu_2Si_2 may seriously affect the AF volume fraction of the sample under measurement. In order to explore the influence of the sample quality on v_{AF}, ^{29}Si NMR measurements on an as-grown sample are now in progress. In the pressure range $P > P_c$, H_{in} at the Si sites has almost the same T-dependence as that of the reported $\mu_{ND} = \sqrt{I_B}$ for $P > P_c$, which reflects the AF ordering taking place at T_o uniformly throughout the sample [17]. Furthermore, above P_C, the magnitudes of μ_{ND} as well as H_{in} at Si sites are constant regardless of pressure. Hence, a combination of the present ^{29}Si NMR and the previous ND results gives the pressure-independent AF ordered moment of $\mu_{AF} \sim 0.4\ \mu_B$/U. Using the hyperfine coupling constant $A_{hf} = 3.93$ kOe/μ_B obtained above T_o at ambient pressure, an estimation of the ordered moment gives $\sim$0.23 μ_B/U, which is somewhat smaller than $\mu_{AF} \sim 0.4\ \mu_B$/U [27]. This is associated with the symmetry of the Si site in the AF spin structure of this system. H_{in} at the Si site is predominantly produced through the transferred hyperfine interaction with the five nearest-neighbour AF moments, namely, the four AF moments antiparallel to H_{in} and the one AF moment parallel to H_{in}. Thus, it is not appropriate to determine the ordered moment by using A_{hf} obtained for the PM state above T_o.

We turn to the nuclear spin–lattice relaxation rate, T_1^{-1} of ^{29}Si. The measurements of T_1^{-1} were performed at 5.8, 8.3 ($P < P_c$), and 17.5 kbar ($P > P_c$). The T_1^{-1}-values obtained are plotted as a function of T in figure 5. Here, it should be emphasized that the ^{29}Si NMR can obtain selectively microscopic information from the AF and the PM regions, in contrast with the macroscopic measurements such as those of specific heat and electrical resistivity. In fact, the T_1^{-1}s of ^{29}Si in the PM and the AF regions (T_{1PM}^{-1} and T_{1AF}^{-1}, respectively) were measured by using the well-separated resonance lines arising from these regions. In the measurement T-range, the T-dependences of T_{1PM}^{-1} at 5.3 and 8.3 kbar are almost the same as the previous results obtained at ambient pressure. At high T, T_{1PM}^{-1} is nearly T-independent and proportional to T at low T below about 50 K. In the f-electron heavy-fermion systems, similar behaviour of T_1^{-1} is frequently observed when the electronic state of the f electrons is in a regime of crossover from the localized state at high T to the Fermi liquid state. Below T_o, T_{1PM}^{-1} decreases markedly and again varies proportionally to T with further decreasing T. This behaviour indicates a partial loss of the density of states due to the opening of an energy gap below T_o. Since T_{1PM}^{-1} is selectively measured for Si sites in the PM region, it is clear that the drastic decrease in T_{1PM}^{-1} does not directly originate from the AF ordering. Thus, this result evidences the occurrence of a phase transition due to a hidden order in the PM region. The T-dependence of T_{1PM}^{-1} is approximately reproduced by a simple expression:

$$T_1^{-1} = AT + B\exp\left(\frac{-\Delta}{k_B T}\right). \tag{1}$$

The first term represents the contribution from the residual density of states at the Fermi level and the second term that from the thermal excitations above the energy gap. By taking the parameters as $A = 1.2 \times 10^{-2}$ s^{-1} K^{-1}, $B = 940$ s^{-1} K^{-1}, and $\Delta/k_B = 140$ K, a fit with the T_{1PM}^{-1}-data at 5.8 kbar is obtained, as shown by a solid curve in the figure. Far below T_o, T_{1AF}^{-1}

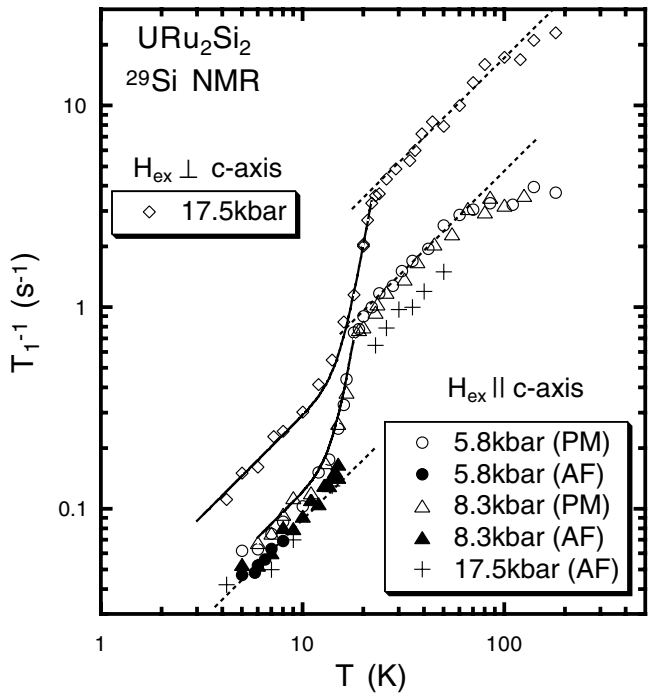

Figure 5. T-dependences of T_1^{-1} for ^{29}Si at 5.8, 8.3 ($P < P_c$), and 17.5 kbar ($P > P_c$). At 5.8 and 8.3 kbar, the T_1-measurements were performed for PM and AF regions by using central and H_{in}-split lines, respectively. The solid curves show the curves fitted using equation (1) given in the text. The dotted lines correspond to the relation $T_1^{-1} \propto T$.

was also measured by using the H_{in}-split line. We cannot obtain T_{1AF}^{-1} just below T_o because of the smallness of the signal arising from the AF region. In the measurement T-range, the T-dependences of T_{1AF}^{-1} at 5.8 and 8.3 kbar are the same within our experimental accuracy. The T_{1AF}^{-1} obtained is proportional to T far below T_o. The value of $\frac{1}{T_1T} \sim 9.5 \times 10^{-3}$ s^{-1} K^{-1} in the AF region is slightly smaller than $\frac{1}{T_1T} \sim 1.2 \times 10^{-2}$ s^{-1} K^{-1} in the PM region at low T. We compare the present T_1^{-1} and the previous specific heat (C) results. At T_o, the T-dependence of C shows a large lambda-shaped anomaly, which is almost unchanged by pressure [26]. In contrast with the T_1^{-1} of ^{29}Si, the reported C has contributions from both the AF and the PM regions. Since the AF volume fraction is extremely small at ambient pressure, the anomaly of C at T_o originates rather from the phase transition due to a hidden order in the PM region. As pressure is applied, the value of C/T at ∼2 K far below T_o is reported to be slightly decreased from 62 (at 1 bar) to 53 mJ K^{-2} mol^{-1} (at 6.3 kbar). Within the simple Fermi liquid description, we expect a relation $\frac{1}{T_1T} \propto N_{eff}(\varepsilon_F)^2 \propto \gamma^2$, where $\gamma = C/T$ is the T-linear coefficient in the C at $T \to 0$ and $N_{eff}(\varepsilon_F)$ the effective density of states at the Fermi level in the heavy-fermion state. Using $v_{AF} \sim 60\%$ obtained from the ^{29}Si NMR, the ratio $\gamma_{6.9\,\mathrm{kbar}}/\gamma_{1\,\mathrm{bar}}$ is crudely estimated as follows:

$$\frac{\gamma_{6.9\,\mathrm{kbar}}}{\gamma_{1\,\mathrm{bar}}} \sim \sqrt{\frac{(T_{1AF}T)^{-1}v_{AF} + (T_{1PM}T)^{-1}(1 - v_{AF})}{(T_{1PM}T)^{-1}}} \sim 0.93. \tag{2}$$

The extracted value is comparable to $\frac{(C/T)_{6.3\,\mathrm{kbar}}}{(C/T)_{1\,\mathrm{bar}}} \sim 0.85$. Therefore, the change in the magnitude of C/T under applied pressure could be explained by considering an increase in the AF region,

where the value of γ is slightly smaller than that in PM region. The T_1s for $H_{ex} \perp c$-axis as well as $H_{ex} \parallel c$-axis were measured at 17.5 kbar ($P > P_c$), where the AF ordering develops uniformly throughout the sample. The T_1^{-1} for $H_{ex} \perp c$-axis at 17.5 kbar is proportional to T between T_o and 80 K, and drops markedly below T_o. As indicated by the solid curve in figure 5, the T-dependence of T_1^{-1} for $H_{ex} \perp c$-axis below T_o is also fitted by equation (1), taking $A = 2.9 \times 10^{-2}$ s^{-1} K^{-1}, $B = 1500$ s^{-1} K^{-1}, and $\Delta/k_B = 140$ K. We have obtained the gap of $\Delta/k_B \sim 150$ K from the T-dependences of T_1^{-1} for ^{101}Ru as well as ^{29}Si at ambient pressure [4–6]. Although the ordering state below T_o at 17.5 kbar is quite different from that at ambient pressure, no significant change in magnitude of the gap was found from the T_1^{-1}-results at 17.5 kbar and ambient pressure. The results of electrical resistivity measurements also indicate that the energy gap is formed due to the phase transition at T_o even above P_c [22–24].

On the basis of the present ^{29}Si NMR results, we finally give a discussion of the nature of the low-T phase below T_o. In the pressure range $P < P_c$, the ^{29}Si NMR measurements have revealed the spatial separation of PM and AF regions in the sample below T_o. Furthermore, the T_1^{-1}-measurements detect the formation of an energy gap below T_o in the PM region. This result indicates the existence of a phase transition associated with a hidden order parameter in the PM region. The hidden order parameter is possibly due to a nonmagnetic degree of freedom other than the magnetic dipole of U 5f electrons. As for the hidden order, quite recently (2001), Bernal *et al* [19] found that an additional linewidth component (λ) of the ^{29}Si NMR line arising from the PM region appears below T_o at ambient pressure. The T-dependence of λ, which is independent of H_{ex}, is similar to that for a mean field theory. This result also suggests the existence of a hidden order in the PM region below T_o, although the microscopic origin of λ is not entirely clear. The present ^{29}Si NMR results show that with increasing pressure, the AF region increases in volume fraction at the expense of the PM region where a hidden order parameter develops. Furthermore, at a constant pressure, the AF volume fraction increases with decreasing T. The strong dependence of the AF volume fraction on pressure and T suggests that the AF and the hidden ordering states, which are almost energetically degenerate, compete with each other for volume fraction. On the other hand, no PM region remains below T_o in the pressure range $P > P_c$, which indicates that the entire system undergoes the AF transition at T_o. Until now, many competing theoretical models have been proposed for the hidden order in URu_2Si_2. As one of the possibilities for the hidden order parameter, some models have considered quadrupolar moments of U 5f electrons [9, 13, 14]. The model with a quadrupolar ordering in the singlet ground state (Γ_3 or Γ_4) of the $5f^2$ configuration explains well the observed macroscopic anomalies associated with hidden order at ambient pressure [9]. Recently, the free energy (F) has been calculated as a function of the molecular fields of quadrupoles (q) and dipoles (m), based on this model [28]. The calculated free energy surface $F(q, m)$ has double minima, which correspond to quadrupolar ordering and AF states, in $q - m$ space. This result suggests the possibility of a competition of quadrupolar and dipolar ordering states in this system. Besides the singlet ground state model, the doublet Γ_5 ground state model also gives a competition of quadrupolar and AF orderings [13, 29]. Although the quadrupolar ordering in URu_2Si_2 has not been confirmed experimentally, the quadrupolar ordering models can account for the phase-separated state observed by means of ^{29}Si NMR for $P < P_c$. In this scenario, the pressure-induced phase transition at P_c can be understood as a complete change in the order parameter of the phase transition to dipolar moments for $P > P_c$. This is qualitatively consistent with the present ^{29}Si NMR results which show the occurrence of an AF transition at T_o throughout the system for $P > P_c$.

In conclusion, ^{29}Si NMR measurements on URu_2Si_2 at pressures up to 17.5 kbar have revealed that the system changes from a state of coexistence of AF and PM regions below P_c to a uniform AF ordering state above P_c. In the AF region, the magnitude of the ordered

moment is constant ($\sim$0.4 μ_B/U) independently of the applied pressure up to 17.5 kbar. The rapid decrease of T_{1PM}^{-1} just below T_o shows that a drastic change takes place in the electronic state of the PM region at T_o. This indicates the occurrence of a phase transition due to a hidden order in the PM region.

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References

[1] Palstra T T M, Menovsky A A, van den Berg J, Dirkmaat A J, Kes P H, Nieuwenhuys G J and Mydosh J A 1985 *Phys. Rev. Lett.* **55** 2727
Schlabitz W, Baumann J, Pollit B, Rauchschwalbe U, Mayer H M, Ahlheim U and Bredl C D 1986 *Z. Phys.* B **62** 171
Maple M B, Chen J W, Dalichaouch Y, Kohara T, Rossel C, Torikachvili M S, McElfresh M W and Thompson J D 1986 *Phys. Rev. Lett.* **56** 185
[2] de Visser A, Kayzel F E, Menovsky A A, Franse J J M, van den Berg J and Nieuwenhuys G J 1986 *Phys. Rev.* B **34** 8168
[3] Miyako Y, Kawarazaki S, Amitsuka H, Paulsen C C and Hasselbach K 1991 *J. Appl. Phys.* **76** 5791
[4] Kohara T, Kohori Y, Asayama K, Kitaoka Y, Maple M B and Torikachvili M S 1986 *Solid State Commun.* **59** 603
[5] Kohori Y, Matsuda K and Kohara T 1996 *J. Phys. Soc. Japan* **65** 1083
[6] Matsuda K, Kohori Y and Kohara T 1996 *J. Phys. Soc. Japan* **65** 679
[7] Broholm C, Kjems J K, Buyers W J L, Matthews P, Palstra T T M, Menovsky A A and Mydosh J A 1987 *Phys. Rev. Lett.* **58** 1467
Broholm C, Lin H, Matthews P T, Mason T E, Buyers W J L, Collins M F, Menovsky A A, Mydosh J A and Kjems J K 1991 *Phys. Rev.* B **43** 12809
[8] Isaacs E D, McWhan D B, Kleiman R N, Bishop D J, Ice G E, Zschack P, Gaulin B D, Mason T E, Garrett J D and Buyers W J L 1990 *Phys. Rev. Lett.* **65** 3185
[9] Santini P and Amoretti G 1994 *Phys. Rev. Lett.* **73** 1027
Santini P 1998 *Phys. Rev.* B **57** 5191
[10] Barzykin V and Gor'kov L P 1995 *Phys. Rev. Lett.* **74** 3723
[11] Kasuya T 1997 *J. Phys. Soc. Japan* **66** 3348
[12] Ikeda H and Ohashi Y 1998 *Phys. Rev. Lett.* **81** 3723
[13] Ohkawa F J and Shimizu H 1999 *J. Phys.: Condens. Matter* **11** L519
[14] Tsuruta A, Kobayashi A, Matsuura T and Kuroda Y 2000 *J. Phys. Soc. Japan* **69** 663
[15] Shah N, Chandra P, Coleman P and Mydosh J A 2000 *Phys. Rev.* B **61** 564
[16] Chandra P, Coleman P and Mydosh J A 2002 *Physica* B **312/313** 397
Chandra P, Coleman P, Mydosh J A and Tripathi V 2002 *Nature* **417** 831
[17] Amitsuka H, Sato M, Metoki N, Yokoyama M, Kuwahara K, Sakakibara T, Morimoto H, Kawarazaki S, Miyako Y and Mydosh J A 1999 *Phys. Rev. Lett.* **83** 5114
Amitsuka H, Sato M, Metoki N, Yokoyama M, Kuwahara K, Sakakibara T, Morimoto H, Kawarazaki S, Miyako Y and Mydosh J A 2000 *J. Phys. Soc. Japan Suppl.* A **69** 5
[18] Matsuda K, Kohori Y, Kohara T, Kuwahara K and Amitsuka H 2001 *Phys. Rev. Lett.* **87** 087203
[19] Bernal O O, Rodrigues C, Martinez A, Lukefahr H G, MacLaughlin D E, Menovsky A A and Mydosh J A 2001 *Phys. Rev. Lett.* **87** 196402
[20] Luke G M, Keren A, Le L P, Uemura Y J, Wu W D, Bonn D, Taillefer L, Garrett J D and Ōnuki Y 1994 *Hyperfine Interact.* **85** 397
[21] Louis E, de Visser A, Menovsky A A and Franse J J M 1986 *Physica* B **144** 48
[22] McElfresh M W, Thompson J D, Willis J O, Maple M B, Kohara T and Torikachvili M S 1987 *Phys. Rev.* B **35** 43

[23] Ido M, Segawa Y, Amitsuka H and Miyako Y 1993 *J. Phys. Soc. Japan* **62** 2962
[24] Oomi G, Kagayama T, Onuki Y and Komatsubara T 1994 *Physica* B **199/200** 148
[25] Nishioka T, Mukai H, Nakamura S, Motoyama G, Ushida Y and Sato N K 2000 *J. Phys. Soc. Japan* **69** 2415
[26] Fisher R A, Kim S, Wu Y, Phillips N E, McElfresh M W, Torikachvili M S and Maple M B 1990 *Physica* B **163** 419
[27] Matsuda K, Kohori Y, Kohara T, Kuwahara K and Amitsuka H 2000 *Physica* B **281/282** 989
[28] Takahashi Y 2001 *J. Phys. Soc. Japan* **70** 2226
[29] Amitsuka H and Sakakibara T 1994 *J. Phys. Soc. Japan* **63** 736