

## Effect of Pressure on Tiny Antiferromagnetic Moment in the Heavy-Electron Compound URu<sub>2</sub>Si<sub>2</sub>

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(Received 6 July 1999)

We have performed elastic neutron-scattering experiments on the heavy-electron compound URu<sub>2</sub>Si<sub>2</sub> for pressure  $P$  up to 2.8 GPa. We have found that the antiferromagnetic (100) Bragg reflection below  $T_m \sim 17.5$  K is strongly enhanced by applying pressure. For  $P < 1.1$  GPa, the staggered moment  $\mu_o$  at 1.4 K increases linearly from  $\sim 0.017(3)\mu_B$  to  $\sim 0.25(2)\mu_B$ , while  $T_m$  increases slightly at a rate  $\sim 1$  K/GPa, roughly following the transition temperature  $T_o$  determined from macroscopic anomalies. We have also observed a sharp phase transition at  $P_c \sim 1.5$  GPa, above which a 3D-Ising type of antiferromagnetic phase ( $\mu_o \sim 0.4\mu_B$ ) appears with a slightly reduced lattice constant.

PACS numbers: 75.30.Mb, 75.25.+z, 75.50.Ee

Antiferromagnetism due to extremely weak moments indicated in CeCu<sub>6</sub> [1], UPt<sub>3</sub> [2], and URu<sub>2</sub>Si<sub>2</sub> [3] has been one of the most intriguing issues in heavy-fermion physics. URu<sub>2</sub>Si<sub>2</sub> has received special attention because of its unique feature that the development of the tiny staggered moment  $\mu_o$  is accompanied by significant anomalies in bulk properties [4–6]. In particular, specific heat shows a large jump ( $\Delta C/T_o \sim 300$  mJ/K<sup>2</sup> mol) at  $T_o = 17.5$  K, which evidences  $5f$  electrons to undergo a phase transition [4,5]. Microscopic studies of neutron scattering [3,7,8] and x-ray magnetic scattering [9] have revealed an ordered array of  $5f$  magnetic dipoles along the tetragonal  $c$  axis with a wave vector  $Q = (100)$  developing below  $T_o$ . The magnitude of  $\mu_o$  is found to be  $0.02$ – $0.04\mu_B$ , which however is roughly 50 times smaller than that of the fluctuating moment ( $\mu_{\text{para}} \sim 1.2\mu_B$ ) above  $T_o$  [10]. This large reduction of the  $5f$  moment below  $T_o$  is apparently unreconciled with the large  $C(T)$  anomaly, if  $\mu_{\text{para}}$  is simply regarded as a full moment [11].

To solve the discrepancy, various ideas have been proposed, which can be classified into two groups: (i) the transition is uniquely caused by magnetic dipoles with highly reduced  $g$  values [12–14]; (ii) there is hidden order of a nondipolar degree of freedom [15–22]. The models of the second group ascribe the tiny moment to side effects, such as secondary order, dynamical fluctuations and coincidental order of a parasitic phase. Each of the dipolar states may have its own energy scale, and to take account of this possibility we define  $T_m$  as the onset temperature of  $\mu_o$ , distinguishing it from  $T_o$ .

The crux of the problem will be how  $\mu_o$  relates to the macroscopic anomalies. Recent high-field studies [10,23–25] have suggested that  $T_o$  and  $\mu_o$  are not scaled by a unique function of field. In addition, the comparison of  $T_o$  and  $T_m$  for the same sample has suggested that  $T_m$

becomes lower than  $T_o$  in the absence of annealing [26]. In this Letter we have studied the influence of pressure on the tiny moment of URu<sub>2</sub>Si<sub>2</sub>, for the first time, by means of elastic neutron scattering. Previous measurements of resistivity and specific heat in  $P$  up to 8 GPa have shown that the ordered phase is slightly stabilized by pressure, with a rate of  $dT_o/dP \sim 1.3$  K/GPa [27–32]. We now show that pressure dramatically increases  $\mu_o$  and causes a new phase transition.

A single-crystalline sample of URu<sub>2</sub>Si<sub>2</sub> was grown by the Czochralski technique in a tri-arc furnace. The crystal was shaped in a cylinder along the  $c$  axis with approximate dimensions 5 mm diameter by 8 mm long, and vacuum annealed at 1000 °C for one week. Pressure was applied by means of a barrel-shaped piston cylinder device [33] at room temperature, which was then cooled in a <sup>4</sup>He cryostat for temperatures between 1.4 and 300 K. A solution of Fluorinert 70 and 77 (Sumitomo 3M Co. Ltd., Tokyo) of equal ratio served as the quasihydrostatic pressure transmitting medium. The pressure was monitored by measuring the lattice constant of NaCl, which was encapsulated together with the sample.

The elastic neutron-scattering experiments were performed on the triple-axis spectrometer TAS-1 at the JRR-3M reactor of Japan Atomic Energy Research Institute. Pyrolytic graphite PG(002) crystals were used for monochromating and analyzing the neutron beam with a wavelength  $\lambda = 2.3551$  Å. We used a 40'–80'–40'–80' horizontal collimation, and double 4-cm-thick pyrolytic graphite (PG) filters as well as a 4-cm-thick Al<sub>2</sub>O<sub>3</sub> filter to reduce higher-order contamination. The scans were performed in the ( $hk0$ ) scattering plane, particularly on the antiferromagnetic Bragg reflections (100) and (210), and on the nuclear ones (200), (020), and (110). The lattice constant  $a$  of our sample at 1.4 K at ambient pressure is  $4.13(1)$  Å.

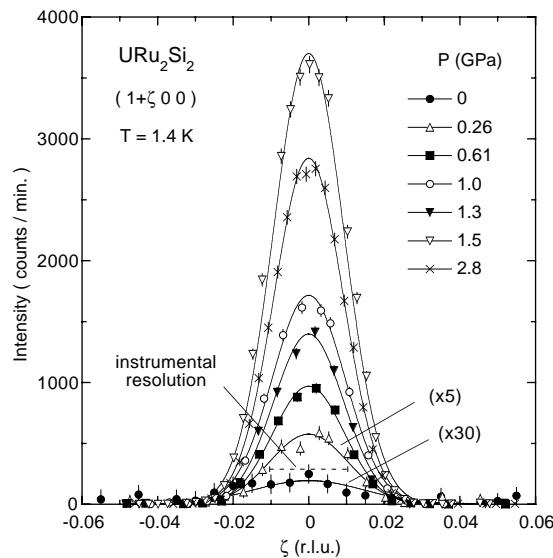


FIG. 1. Longitudinal scans of the antiferromagnetic Bragg peak (100) of  $\text{URu}_2\text{Si}_2$  for several pressures.

Figure 1 shows the pressure variations of elastic scans at 1.4 K along the  $a^*$  direction through the forbidden nuclear (100) Bragg peak. The instrumental background and the higher-order contributions of nuclear reflections were determined by scans at 35 K and subtracted from the data. The (100) reflection develops rapidly as pressure is applied. No other peaks were found in a survey along the principle axes of the first Brillouin zone; in addition, the intensities of (100) and (210) reflections follow the  $|Q|$  dependence expected from the  $\text{U}^{4+}$  magnetic form factor [34] by taking the polarization factor unity. These ensure that the type-I antiferromagnetic structure at  $P = 0$  is unchanged by the application of pressure.

The widths (FWHM) of the (100) peaks for  $P = 0$  and 0.26 GPa are significantly larger than the instrumental resolution [ $\sim 0.021(1)$  reciprocal-lattice units], which was determined from  $\lambda/2$  reflections at (200). From the best fit to the data by a Lorentzian function convoluted with the Gaussian resolution function, the correlation length  $\xi$  along the  $a^*$  direction is estimated to be about 180 Å at  $P = 0$  and 280 Å at 0.26 GPa. For the higher pressures  $P \geq 0.61$  GPa, the simple fits give  $\xi > 10^3$  Å, indicating that the line shapes are resolution limited.

The temperature dependence of the integrated intensity  $I(T)$  at (100) varies significantly as  $P$  traverses 1.5 GPa ( $\equiv P_c$ ) (Fig. 2). For  $P < P_c$ , the onset of  $I(T)$  is not sharp:  $I(T)$  gradually develops at a temperature  $T_m^+$ , which is higher than  $T_o$ , and shows a  $T$ -linear behavior below a lower temperature  $T_m^-$ . Here we empirically define the “antiferromagnetic transition” temperature  $T_m$  by the midpoint of  $T_m^+$  and  $T_m^-$ . The range of the rounding,  $\delta T_m \equiv T_m^+ - T_m^-$ , is estimated to be 2–3 K, which is too wide to be usual critical scattering. Above  $P_c$ , on the other hand, the transition becomes sharper ( $\delta T_m < 2$  K), accompanied by an abrupt increase in  $T_m$  at  $P_c$ .

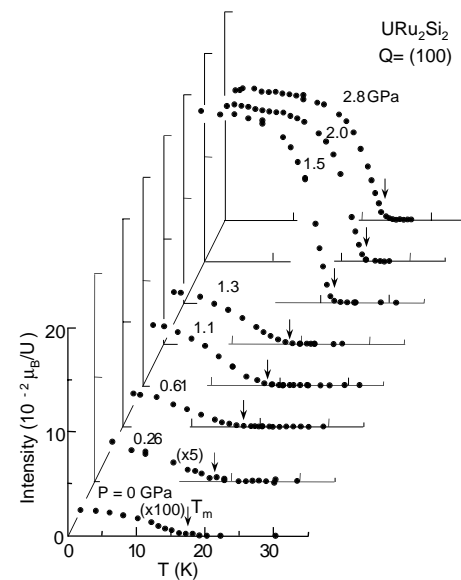


FIG. 2. Temperature dependence of the integrated intensity of the (100) magnetic Bragg reflection for various pressures.

If  $I(T)$  is normalized to its value at 1.4 K, it scales with  $T/T_m$  for various pressures on each side of  $P_c$  (Fig. 3). This indicates that two homogeneously ordered phases are separated by a (probably first-order) phase transition at

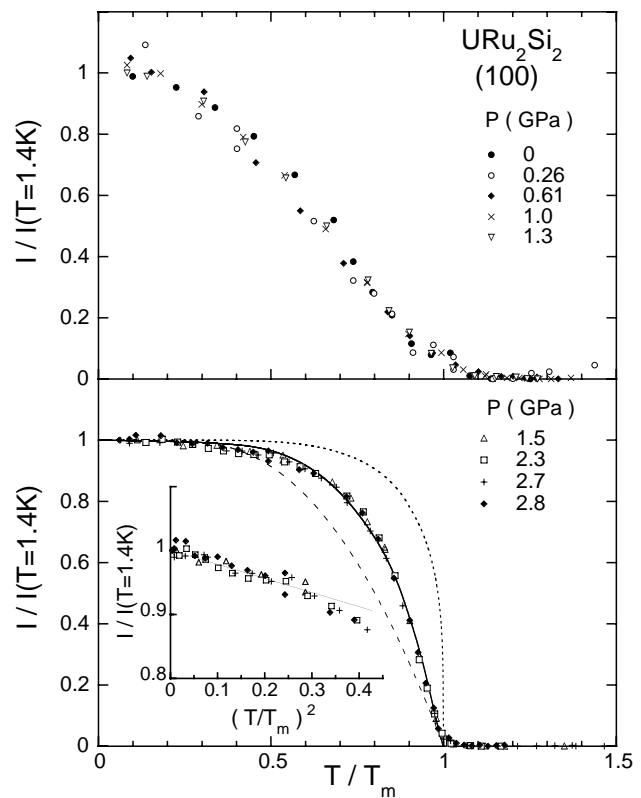


FIG. 3. Normalized intensities  $I/I(1.4\text{ K})$  plotted as a function of  $T/T_m$  for  $P < P_c$  (top) and  $P > P_c$  (bottom). Theoretical calculations based on 2D [40], 3D [35], and mean-field Ising models are also given by dotted, solid, and broken lines. The inset plots  $I/I(1.4\text{ K})$  versus  $(T/T_m)^2$  at low temperatures. The thin line is a guide to the eye.

$P_c$ . The growth of  $I(T)$  for  $P < P_c$  is much weaker than that expected for the mean-field Ising model, showing an unusually slow saturation of the staggered moment. On the other hand, the overall feature of  $I(T)$  for  $P > P_c$  is approximately described by a 3D Ising model [35]. In the low temperature range  $T/T_m < 0.5$ , however,  $I(T)$  rather follows a  $T^2$  function (the inset of Fig. 3), indicating a presence of gapless collective excitations [36].

In Fig. 4, we plot the pressure dependence of  $\mu_o$ ,  $T_m$ , and the lattice constant  $a$ . The magnitude of  $\mu_o$  at 1.4 K is obtained through the normalization of the integrated intensity at (100) with respect to the weak nuclear Bragg peak at (110). The variation of the (110) intensity with pressure is small ( $<5\%$ ) and independent of the crystal mosaic, so that the influence of extinction on this reference peak is negligible.  $\mu_o$  at  $P = 0$  is estimated to be about  $0.017(3)\mu_B$ , which is slightly smaller than the values [ $\sim 0.02\text{--}0.04\mu_B$ ] of previous studies [3,7,9,26], probably because of a difference in the selection of reference peaks. As pressure is applied,  $\mu_o$  increases linearly at a rate  $\sim 0.25\mu_B/\text{GPa}$ , and shows a tendency to saturate at  $P \sim 1.3$  GPa. Around  $P_c$ ,  $\mu_o$  abruptly increases from  $0.23\mu_B$  to  $0.40\mu_B$ , and then slightly decreases.

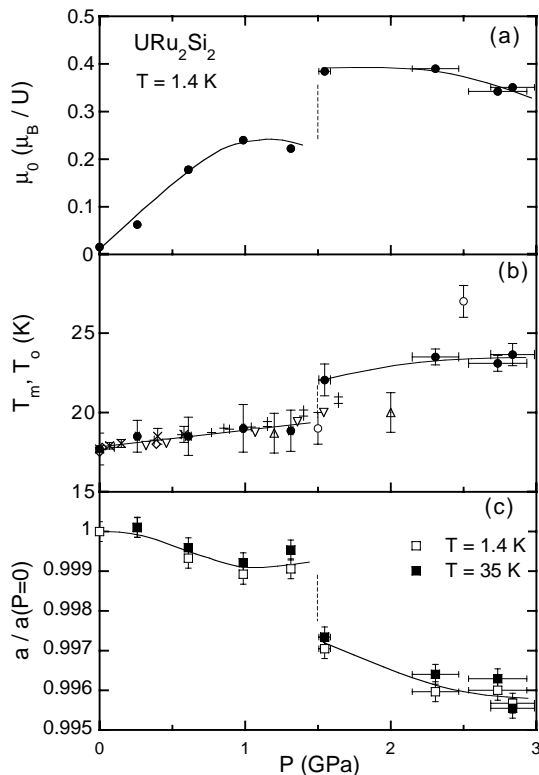


FIG. 4. Pressure variations of (a) staggered magnetic moment  $\mu_o$  at 1.4 K; (b) the onset temperature  $T_m$  of the moment determined from this work ( $\bullet$ ) and the transition temperature  $T_o$  determined from resistivity ( $\diamond$  [27],  $\nabla$  [28],  $\triangle$  [30],  $+$  [31],  $\circ$  and [32]) and specific heat ( $\times$  [29]); (c) the relative lattice constant  $a(P)/a(0)$  at 1.4 and 35 K.  $T_m$  is defined by  $(T_m^+ + T_m^-)/2$  (see the text), and the range  $\delta T_m (= T_m^+ - T_m^-)$  is shown by using error bars. The lines are guides to the eye.

In contrast to the strong variation of  $\mu_o$ ,  $T_m$  shows a slight increase from 17.7 to 18.9 K, as  $P$  is increased from 0 to 1.3 GPa. A simple linear fit of  $T_m$  in this range yields a rate  $\sim 1.0$  K/GPa, which roughly follows the reported  $P$  variations of  $T_o$ . Upon further compression,  $T_m$  jumps to 22 K at  $P_c$ , showing a spring of  $\sim 2.8$  K from a value ( $\sim 19.2$  K) extrapolated with the above fit. For  $P > P_c$ ,  $T_m$  again gradually increases and reaches  $\sim 23.5$  K around 2.8 GPa. The pressure dependence of  $T_o$  in this range is less clear, and the few available data points deviate from the behavior of  $T_m$ ; see Fig. 4(b).

The lattice constant  $a$ , which is determined from the scans at (200), decreases slightly under pressure [Fig. 4(c)]. From a linear fit of  $a$  at 1.4 K for  $P < P_c$ , we derive  $-\partial \ln a / \partial P \sim 6.7 \times 10^{-4} \text{ GPa}^{-1}$ . If the compression is isotropic, this yields an isothermal compressibility  $\kappa_T$  of  $2 \times 10^{-3} \text{ GPa}^{-1}$ , which is about 4 times smaller than what was previously estimated from the compressibilities of the constituent elements [6]. Around  $P_c$ , the lattice shrinks with a discontinuous change of  $-\Delta a/a \sim 0.2\%$ . Assuming again the isotropic compression, we evaluate  $\Delta \ln V / \Delta \mu_o \sim -0.04 \mu_B^{-1}$  and  $\Delta \ln T_m / \Delta \ln V \sim -27$  associated with this transition. Note that a similar lattice anomaly at  $P_c$  is observed at 35 K, much higher than  $T_o$ . This implies that the system has another energy scale characteristic of the volume shrinkage in the paramagnetic region. We have confirmed the absence of any lowering in the crystal symmetry at  $P_c$  within the detectability limit of  $|a - b|/a \sim 0.05\%$  and  $\cos^{-1}(\hat{a} \cdot \hat{b}) \sim 2'$ . The  $c$  axis is perpendicular to the scattering plane and cannot be measured in the present experimental configuration. Precise x-ray measurements under high pressure in an extended  $T$  range are now in progress.

The remarkable contrast between  $\mu_o$  and  $T_m$  below  $P_c$  offers a test to the various theoretical scenarios for the 17.5 K transition. Let us first examine the possibility of a single transition at  $T_m (= T_o)$  due to magnetic dipoles. In general,  $T_m$  is derived from exchange interactions, and is independent of  $g$ . Therefore, the weak variations of  $T_m$  with pressure will be compatible with the ten-times increase of  $\mu_o (= g\mu_B m_o)$ , only if  $g$  is sensitive to pressure. The existing theories along this line explain the reduction of  $g$  by assuming crystalline-electric-field (CEF) effects with low-lying singlets [12], and further by combining such with quantum spin fluctuations [13,14]. To account for the  $P$  increase of  $\mu_o$ , the characteristic energies of these effects should be reduced under pressure. Previous macroscopic studies however suggest opposite tendencies: the resistivity maximum shifts to higher temperatures [28,30–32] and the low- $T$  susceptibility decreases as  $P$  increases [27]. The simple application of those models is thus unlikely to explain the behavior of  $\mu_o$  with pressure.

The models that predict a hidden (primary) nondipolar order parameter  $\psi$  are divided into two branches according

to whether  $\psi$  is odd (A) or even (B) under time reversal [37]. The polarized neutron scattering has confirmed that the reflections arise purely from magnetic dipoles [8]. For each branch, therefore, secondary order has been proposed as a possible solution of the tiny moment. The Landau free energy for type (A) is given as

$$F^{(A)} = -\alpha(T_o - T)\psi^2 + \beta\psi^4 + Am^2 - \eta m\psi, \quad (1)$$

where  $\alpha$ ,  $\beta$ , and  $A$  are positive, and the dimensionless order parameters  $m$  and  $\psi$  vary in the range  $0 \leq m, \psi \leq 1$  [37]. Minimization of  $F^{(A)}$  with respect to  $m$  gives  $m = -\delta\psi$ , where  $\delta \equiv \eta/2A$ . The stability condition for  $\psi$  then yields  $\psi^2 \sim \frac{\alpha}{2\beta}(T_o' - T)$ , where  $T_o' \sim T_o[1 + \mathcal{O}(\delta^2)]$ . If  $\mu_{\text{para}} \sim 1.2\mu_B$  seen above  $T_o$  [10] corresponds to  $m \sim 1$ , then the observed increase in  $\mu_o$  gives  $dT_o/dP \sim T_o dm_o^2/dP \sim 0.8$  K/GPa, which is in good agreement with the experimental results ( $\sim 1.3$  K/GPa) [27–32].

In type (B), the simplest free energy invariant under time reversal [37,38] must take the form

$$F^{(B)} = -\alpha(T_o - T)\psi^2 + \beta\psi^4 + a(T_m - T)m^2 + bm^4 - \zeta m^2\psi^2. \quad (2)$$

The continuous secondary order does not affect  $T_o$ , but enhances  $C(T)$  at  $T_m$  as  $\Delta C/T_m \sim Nk_B m_o^2/T_m$ . In the same way as in type (A), we obtain  $d(\Delta C/T_m)/dP \sim 20$  mJ/K<sup>2</sup> mol GPa, when  $T_m \sim T_o$ . This cancels out with the  $P$  increase in  $T_o$ , resulting in a roughly  $P$ -independent jump in  $C(T)$ . This is consistent with previous  $C(T)$  studies up to 0.6 GPa [29], in which  $\Delta C_m/T_o$  is nearly constant, if entropy balance is considered. Note that in type (B)  $T_m$  can in principle differ from  $T_o$ , which could also be consistent with the annealing effects [26].

The phase transition at  $P_c$  might be understood as a switching between  $\psi$  and  $m$  in type (B). For example, the models of quadrupolar order in the CEF singlets ( $\Gamma_3, \Gamma_4$ ) [17] and a non-Kramers doublet ( $\Gamma_5$ ) [18,19] both involve such magnetic instabilities. Interestingly, if the dipolar order takes place in the  $\Gamma_5$  state, it will be accompanied by the disappearance of magnon excitations, since the nature of excitations changes from a dipolar origin to a quadrupolar one [19]. Our preliminary results of inelastic neutron scattering support this possibility [39].

In conclusion, we have shown that the staggered magnetic moment associated with the 17.5 K transition in URu<sub>2</sub>Si<sub>2</sub> is significantly enhanced by pressure. In contrast to the ten-times increase of the dipole moment, the transition temperature is insensitive to pressure. This feature is consistent with the hidden-order hypotheses. We have also found that the system undergoes a pressure-induced phase transition at around 1.5 GPa, evolving into a well-behaved magnetic phase.

We are grateful to T. Osakabe, T. Honma, and Y. Ōnuki for technical supports. One of us (H.A.) also thanks F.J. Ohkawa for helpful discussions. This work was partly supported by the JAERI-JRR3M Collaborative Research Program, and by a Grant-in-Aid for Scientific

Research from the Ministry of Education, Science, Sports and Culture of Japan.

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