Effect of Pressure on Tiny Antiferromagnetic Moment in the Heavy-Electron Compound URu_2Si_2

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We have performed elastic neutron-scattering experiments on the heavy-electron compound URu_2Si_2 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 μ_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 ~ 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.

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Antiferromagnetism due to extremely weak moments indicated in $CeCu_6$ [1], UPt_3 [2], and URu_2Si_2 [3] has been one of the most intriguing issues in heavy-fermion physics. URu_2Si_2 has received special attention because of its unique feature that the development of the tiny staggered moment μ_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^2 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 μ_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 μ_{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 μ_o, distinguishing it from T_o.

The crux of the problem will be how μ_o relates to the macroscopic anomalies. Recent high-field studies [10,23–25] have suggested that T_o and μ_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_2Si_2, 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 μ_o and causes a new phase transition.

A single-crystalline sample of URu_2Si_2 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 ^{4}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 encapsuled 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_2O_3 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.

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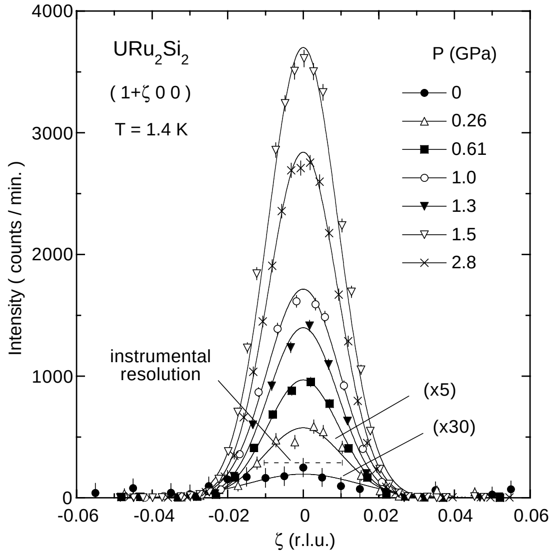

FIG. 1. Longitudinal scans of the antiferromagnetic Bragg peak (100) of URu_2Si_2 for several pressures.

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 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 ξ 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.

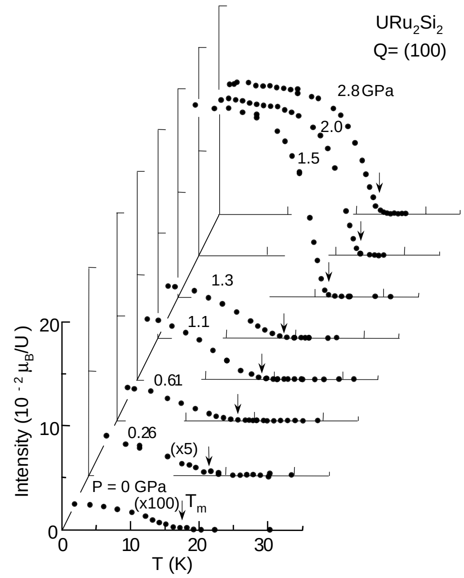

FIG. 2. Temperature dependence of the integrated intensity of the (100) magnetic Bragg reflection for various pressures.

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

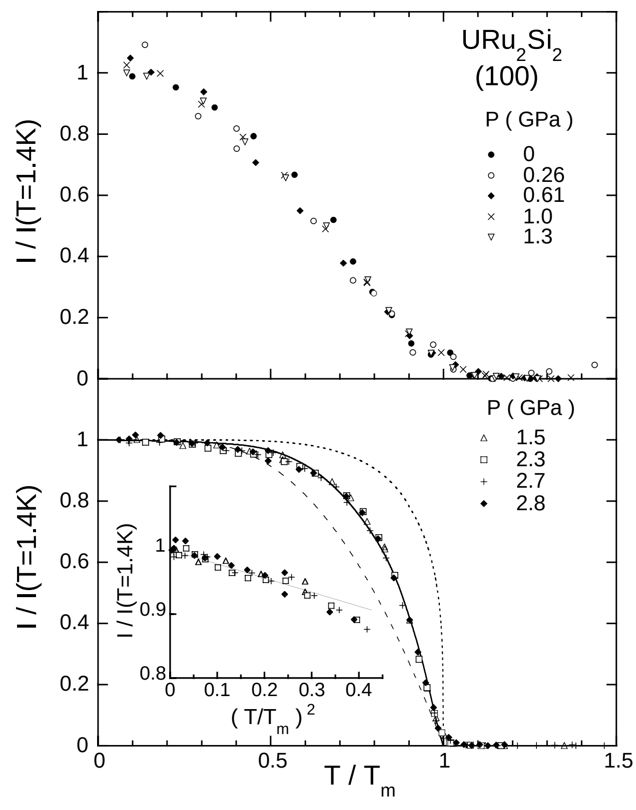

FIG. 3. Normalized intensities $I/I(1.4\ \text{K})$ plotted as a function of T/T_m for $P < P_c$ (top) and $P > P_c$ (bottom). Theoretical calculations based on 2D [40], 3D [35], and mean-field Ising models are also given by dotted, solid, and broken lines. The inset plots $I/I(1.4\ \text{K})$ versus $(T/T_m)^2$ 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 μ_o, T_m, and the lattice constant a. The magnitude of μ_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. μ_o at $P = 0$ is estimated to be about $0.017(3)\mu_B$, which is slightly smaller than the values [~ 0.02–$0.04\mu_B$] of previous studies [3,7,9,26], probably because of a difference in the selection of reference peaks. As pressure is applied, μ_o increases linearly at a rate $\sim 0.25\mu_B$/GPa, and shows a tendency to saturate at $P \sim$ 1.3 GPa. Around P_c, μ_o abruptly increases from $0.23\mu_B$ to $0.40\mu_B$, and then slightly decreases.

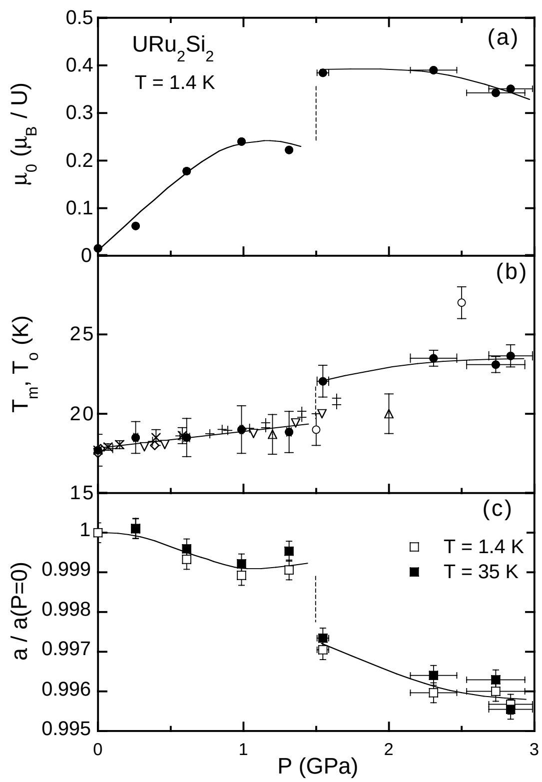

FIG. 4. Pressure variations of (a) staggered magnetic moment μ_o at 1.4 K; (b) the onset temperature T_m of the moment determined from this work (•) and the transition temperature T_o determined from resistivity (◇ [27], ▽ [28], △ [30], + [31], ∘ and [32]) and specific heat (× [29]); (c) the relative 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 $\delta T_m (\equiv T_m^+ - T_m^-)$ is shown by using error bars. The lines are guides to the eye.

In contrast to the strong variation of μ_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 ~ 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 ~ 2.8 K from a value (~ 19.2 K) extrapolated with the above fit. For $P > P_c$, T_m again gradually increases and reaches ~ 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 $-\partial \ln a/\partial P \sim 6.7 \times 10^{-4}$ GPa^{-1}. If the compression is isotropic, this yields an isothermal compressibility κ_T of 2×10^{-3} 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 μ_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 μ_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 μ_o with pressure.

The models that predict a hidden (primary) nondipolar order parameter ψ are divided into two branches according to whether ψ 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 α, β, and A are positive, and the dimensionless order parameters m and ψ vary in the range $0 \le m, \psi \le 1$ [37]. Minimization of $F^{(A)}$ with respect to m gives $m = -\delta\psi$, where $\delta \equiv \eta/2A$. The stability condition for ψ then yields $\psi^2 \sim \frac{\alpha}{2\beta}(T_o' - T)$, where $T_o' \sim T_o[1 + \mathcal{O}(\delta^2)]$. If $\mu_{\rm para} \sim 1.2\mu_B$ seen above T_o [10] corresponds to $m \sim 1$, then the observed increase in μ_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^2 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 ψ and m in type (B). For example, the models of quadrupolar order in the CEF singlets (Γ_3, Γ_4) [17] and a non-Kramers doublet (Γ_5) [18,19] both involve such magnetic instabilities. Interestingly, if the dipolar order takes place in the Γ_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_2Si_2 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.

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