

Magnetic Structure of Phase II in $U(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$ Determined by Neutron Diffraction under Pulsed High Magnetic Fields

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We report neutron diffraction measurements on $U(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$ single crystal under pulsed high magnetic fields up to 30 T applied along the tetragonal c axis. The high-field experiments revealed that the field-induced phase II above 26 T corresponds to a commensurate up-up-down ferrimagnetic structure characterized by the wave vector $\mathbf{q} = (2/3, 0, 0)$ with the magnetic moments parallel to the c axis, which naturally explains the one-third magnetization plateau and the substantially changed Fermi surface in phase II. This a -axis modulated magnetic structure indicates that the phase II near the hidden order phase is closely related to the characteristic incommensurate magnetic fluctuations at $\mathbf{Q}_1 = (0.6, 0, 0)$ in the pure system URu_2Si_2 , in contrast to the pressure-induced antiferromagnetic order at $\mathbf{Q}_0 = (1, 0, 0)$.

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The hidden order (HO) below $T_0 = 17.5$ K in URu_2Si_2 is one of the most interesting topics in heavy-fermion systems. More than 25 years after the discovery of this system, the order parameter below T_0 is still unknown [1]. Near the HO phase, different phases can be induced by pressure and magnetic field. Pressure transposes the characteristic resonant magnetic fluctuations with $\mathbf{Q}_0 = (1, 0, 0)$ in the HO phase [2–4] to the static long-range antiferromagnetic (AFM) order associated with a large magnetic moment of $0.4\mu_B/U$ [5] where the fluctuations at \mathbf{Q}_0 vanish while other characteristic fluctuations at the incommensurate position $\mathbf{Q}_1 = (0.6, 0, 0)$ remain [6]. The duality between the HO phase and the AFM phase under pressure is well illustrated by the switch from HO to AFM order with a quasi-invariance of the Fermi surface (FS) [7] and also by the restoration of HO without mark of any metamagnetic transition [8]. On the other hand, magnetic field effects on the HO phase are more complicated. The detailed structure of the magnetic field—temperature (H - T) phase diagram has been constructed by recent high-field experiments [9–11]. When a field is applied along the tetragonal c axis, the HO phase disappears at 35 T, and field-induced phases (called phases II and III) subsequently appear between 35 and 40 T with a clear three-step metamagnetism [12–14] and substantial changes of the FS [15,16]. The existence of the field-induced phases just before entering in the polarized paramagnetic phase suggests a competition between the HO and these unknown magnetic order parameters. Thus, the determination of their magnetic structures may constitute an important piece to understand the nature of HO, but there is still insufficient information about the field-induced phases due to the difficulty to

perform neutron scattering experiments under strong magnetic fields.

In this Letter, we present the magnetic structure of phase II in the 4% Rh-doped URu_2Si_2 determined by neutron diffraction under pulsed high magnetic fields up to 30 T. The complicated H - T phase diagram of the pure compound is then reduced for $U(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$ to a single field-induced phase with the critical field $\mu_0 H_C = 26$ T, which is experimentally identified as phase II by the x dependence of the H - T phase diagram in $U(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ [17–21]. We prove that the magnetic structure of phase II is the ferrimagnetic structure characterized by the wave vector $\mathbf{q} = (2/3, 0, 0)$. The determined translational symmetry provides a solid basis for the FS calculations of phase II as well as an indication of the close relation between phase II and the incommensurate magnetic fluctuations at \mathbf{Q}_1 in pure URu_2Si_2 .

URu_2Si_2 belongs to the large family of rare-earth or actinide-based 1-2-2 intermetallic compounds, which crystallize in the body-centered tetragonal ThCr_2Si_2 structure (space group $I4/mmm$). A single crystal of $U(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$ was grown by the Czochralski method in a tetra-arc furnace. A nearly cylindrical shaped single crystal with a diameter of 3.5 mm and 5 mm long along the c axis was cut by a spark cutter. Resistivity, specific heat, and magnetic susceptibility measurements showed no HO transition. The residual resistivity ratio of the sample was 11 and its Sommerfeld coefficient was 80 mJ/mol K^2 . The characteristic metamagnetic transitions of the single crystal were confirmed by a magnetization measurement under pulsed magnetic fields of 30 msec rise and 120 msec fall performed at Laboratoire National des Champs Magnétiques Intenses, Toulouse, as shown in Fig. 1, before

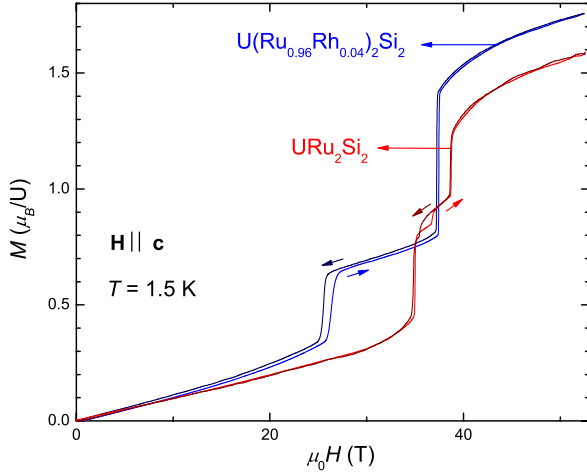


FIG. 1 (color online). Magnetization of $\text{U}(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$ at 1.5 K (blue line). The magnetic field is parallel to the tetragonal c axis. The magnetization of pure URu_2Si_2 is also shown (red line).

doing the neutron diffraction experiments. The magnetization in phase II equals approximately one third of the saturated value above 50 T, once a linear contribution of Pauli or Van Vleck paramagnetism has been subtracted from the magnetization. Neutron diffraction experiments were performed using the thermal neutron three-axis spectrometer IN22 located at the Institut Laue-Langevin, Grenoble. Incident neutrons of wavelength $\lambda = 1.53 \text{ \AA}$ were selected using a pyrolytic graphite (PG) monochromator. After the monochromator, a PG filter was placed for reducing the higher-order contamination of neutrons. The scattered neutrons were detected in the two-axis mode without an analyzer. The high-field experiments were done in two scattering geometries. In the first setting, the sample was set to a small magnet coil, and it was mounted in the ^4He cryostat with the c and a axes in the horizontal scattering plane for measuring at $(h0l)$ positions. Since scattering angles were limited to less than about 30° inside the magnet, the sample was aligned by measuring the $(2, 0, 0)$ nuclear Bragg peak using $\lambda/2$ neutrons without the PG filter. At this $(1, 0, 0)$ position, the wave vector resolution was 0.046 r.l.u. at full width at half maximum along the a axis and the mosaic spread of the sample was 0.5° . The absence of the AFM reflection at \mathbf{Q}_0 in zero magnetic field [20] was also confirmed by measuring rocking curves with and without the PG filter. In the second setting, the sample was rotated by 45° with respect to the cylindrical axis and was mounted with the c and $[110]$ axes in the horizontal scattering plane for measuring at the $(hh0)$ positions. In both settings, the pulsed magnetic fields were applied parallel to the cylindrical axis of the crystal, which was tilted by less than $\alpha \sim 5^\circ$ from the c axis. The slight tilting of the magnetic field from the c axis does not affect the critical field H_C because the critical fields show a $1/\cos\alpha$ dependence [13]. The pulsed measurements were

done more than 100–150 times at about 10-minute intervals, keeping the rotation angle of the sample and the scattering angle fixed for each reflection. Since the magnet partially shades the neutron flight path, in order to compare the intensity at different reflections, the volume fractions of the sample exposed to neutrons were estimated assuming a simple circular cylindrical shape of the sample and neglecting the beam divergence of neutrons. Details of the experimental setup of high-field neutron diffraction measurements are described in Ref. [22].

$\text{U}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$ has a strong uniaxial magnetic anisotropy with its easy axis along the c axis, which is a local single U -site property [23], and its phase II is characterized by a “one-third” magnetization. Based on these two experimental facts, the magnetic structure of phase II is expected to be a ferrimagnetic structure tripling the unit cell, with the magnetic moments parallel to the c axis. However, many ferrimagnetic arrangements with different \mathbf{q} vectors could explain the one-third state. Thus, focusing our attention on \mathbf{q} vectors along the high symmetry directions $[0,0,1]$, $[1,0,0]$, and $[1,1,0]$, we searched for magnetic reflections related to the phase II. In fact, such magnetic structures have already been reported in several 1-2-2 compounds: a magnetic structure modulated along the c axis with $\mathbf{q} = (0, 0, 2/3)$ in UNi_2Si_2 [24] and UPd_2Si_2 [25–27] and a magnetic structure modulated along the $[1,1,0]$ direction with $\mathbf{q} = (1/3, 1/3, 0)$ in the La-doped CeRu_2Si_2 [28,29]. On the other hand, an a -axis modulated structure with the magnetic moments parallel to the c axis has been reported only in cases of incommensurate structure, for example, in $(\text{Ce}, \text{La})\text{Ru}_2\text{Si}_2$ [28,29] and RRu_2Si_2 ($R = \text{Tb}$ and Dy) [30].

At the wave vectors $(1, 0, 1/3)$ corresponding to $\mathbf{q} = (0, 0, 2/3)$ and $(1/3, 1/3, 0)$, no magnetic peak was observed in phase II within the detection limit. In addition, no peak was detected below 30 T at $\mathbf{Q}_0 = (1, 0, 0)$, at which AFM order is induced under pressure in URu_2Si_2 [5]. This suggests that effects of magnetic field on HO are different from those of pressure. On the other hand, we have found a clear magnetic reflection at $(2/3, 0, 0)$ above $\mu_0 H_C = 26 \text{ T}$. Figure 2(a) shows the time dependence of magnetic field and neutron counts at $(2/3, 0, 0)$ at 1.7 K, where neutron counts are normalized by the time binning and the number of pulses. The times of 3.8 and 5.5 msec in Fig. 2 correspond to the critical field H_C . The neutron counts clearly increase above H_C , indicating that the signal comes from the magnetic order parameter of the phase II characterized by the wave vector $\mathbf{q} = (2/3, 0, 0)$. To confirm this, we have measured at the wave vector $(4/3, 0, 0)$, which is equivalent to $\mathbf{q} = (2/3, 0, 0)$. The neutron counts at $(4/3, 0, 0)$ also increase above H_C , as shown in Fig. 2(b), and their intensity above H_C is weaker than that at $(2/3, 0, 0)$, indicating that these peaks are of magnetic origin. This gives direct evidence that the magnetic structure of phase II is modulated along the a axis,

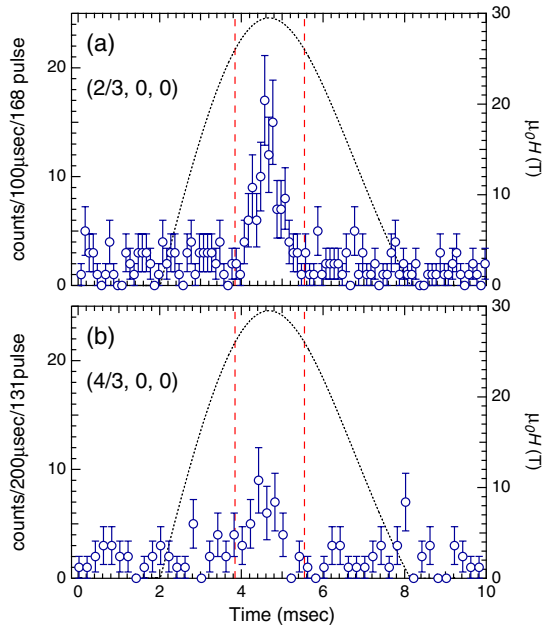


FIG. 2 (color online). Time dependence of magnetic field (dotted line) and neutron counts (open circles) at the $(2/3, 0, 0)$ and $(4/3, 0, 0)$ magnetic Bragg peaks at 1.7 K. The vertical red dashed lines denote the times corresponding to the critical field $\mu_0 H_C = 26$ T.

with a period of three unit cells. Figure 3 shows the magnetic field dependence of the intensity at the $(2/3, 0, 0)$ and $(4/3, 0, 0)$ magnetic Bragg peaks and at the $(1,1,0)$ nuclear Bragg peak. The data correspond to an average of the neutron intensity measured with increasing and decreasing fields. The intensity at $(2/3, 0, 0)$ and $(4/3, 0, 0)$ increases around H_C . This rapid increase of intensity is consistent with the first-order nature of the field-induced phase transition. However, the slope above H_C is somewhat gradual compared to the sharp jump of the bulk magnetization. This may be due to an unavoidable magnetic field gradient inside the small magnet. At $(1,1,0)$, which has a small nuclear structure factor sensitive to ferromagnetic signals, the intensity slightly enhances around H_C . The result is reasonable if the magnetic structure has a squared up form; indeed, the third harmonics of $\mathbf{q} = (2/3, 0, 0)$ coincides with a period of the lattice.

From the experimental results, the most likely magnetic structure is a collinear structure with a squared up form, i.e., the so-called up-up-down ferrimagnetic structure. In this case, there should be two domains, that is, $\mathbf{q} = (2/3, 0, 0)$ and $\mathbf{q} = (0, 2/3, 0)$. Taking into account the domains, the magnitude of the magnetic moment μ is estimated to be $0.6 \pm 0.1 \mu_B/U$ by the ratio of peak intensity between $(2/3, 0, 0)$ and $(1,1,0)$, where the U^{4+} magnetic form factor with the dipole approximation is used [31,32]. One-third of this magnetic moment μ agrees with the jump of the bulk magnetization $0.25 \mu_B/U$. Moreover, using $\mu = 0.6 \mu_B/U$, the intensity at $(1,1,0)$ is calculated

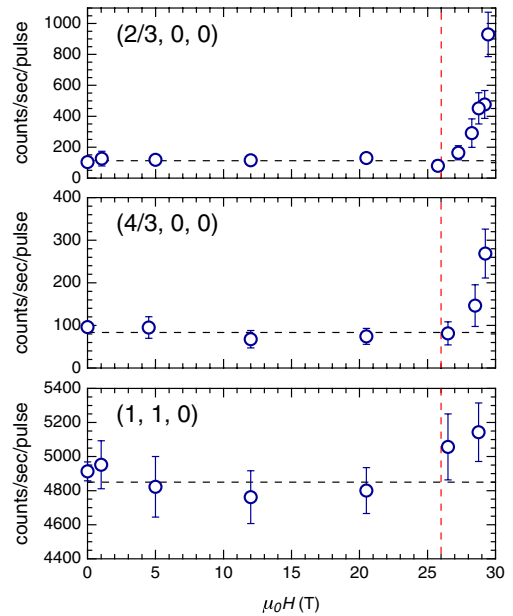


FIG. 3 (color online). Magnetic field dependence of the intensity at the $(2/3, 0, 0)$ and $(4/3, 0, 0)$ magnetic Bragg peaks and at the $(1,1,0)$ nuclear Bragg peak at 1.7 K. The intensity of neutron counts is averaged over increasing and decreasing fields. The vertical red dashed lines correspond to the critical field $\mu_0 H_C = 26$ T. The horizontal dashed lines represent the background intensity.

to be about 150 counts/sec, which also agrees with the data, where both domains contribute to the ferromagnetic component. As other possible magnetic structures, we might have to consider a collinear structure with a sine wave form and a noncollinear structure with magnetic moments canted from the c axis. However, the former can be excluded because this sine modulated structure is inconsistent with the observed enhancement of the $(1,1,0)$ nuclear peak intensity as well as the one-third magnetization. The latter is also unlikely because of the strong Ising anisotropy in this system, although the possibility of such double \mathbf{q} magnetic structure cannot be excluded only by the present data. Therefore, we conclude that the magnetic structure of phase II of $U(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$ is the ferrimagnetic structure with $\mathbf{q} = (2/3, 0, 0)$ and the magnetic moments aligned along the c axis, as schematically shown in Fig. 4. To our knowledge, this magnetic structure is reported for the first time in a rare-earth or actinide-based 1-2-2 compound.

The determined commensurate wave vector $\mathbf{q} = (2/3, 0, 0)$ is very close to the incommensurate wave vector $\mathbf{Q}_1 = (0.6, 0, 0)$. In the pure system URu_2Si_2 , quasielastic magnetic fluctuations at \mathbf{Q}_1 with the itinerant character of the $5f$ electrons in the paramagnetic phase drastically change into well-defined resonant excitations at 4 meV in the HO phase [2–4]. Inelastic neutron scattering experiments under magnetic fields up to 17 T reported that the energy gap of low-energy excitation at \mathbf{Q}_1 slightly

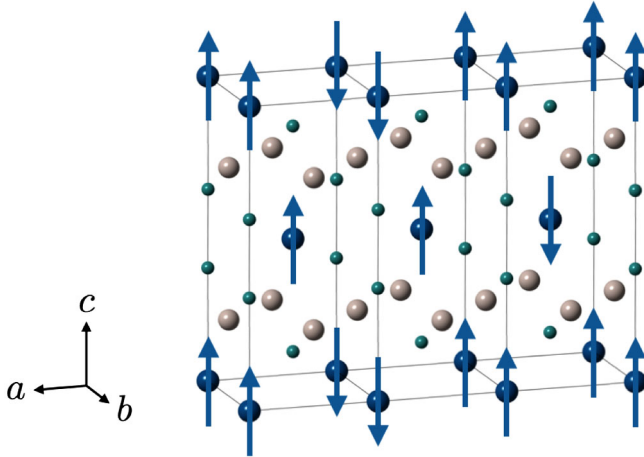


FIG. 4 (color online). Magnetic structure of the field-induced phase II of $U(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$.

decreases, while the gap at \mathbf{Q}_0 increases with increasing fields in the HO phase [33]. Interestingly, the extrapolated values of the two energy gaps cross at a field close to the critical field 35 T. If the incommensurate fluctuations at \mathbf{Q}_1 become the strongest of the whole Brillouin zone around the critical magnetic field, they may be changed by strong magnetic fields into static long-range magnetic order with the commensurate wave vector locked into the nearest rational value with respect to a period of the lattice, that is, $\mathbf{q} = (2/3, 0, 0)$. Since it is common in many magnetic materials that incommensurate magnetic fluctuations are locked into a lattice through magnetoelastic coupling, there might be no doubt that the a -axis modulated structure is related to the incommensurate magnetic fluctuations at \mathbf{Q}_1 . Further experiments would be needed to test if, in Rh-doped as well as in the pure URu_2Si_2 compounds, the development of magnetic order at $\mathbf{q} = (2/3, 0, 0)$ could be a consequence of a field-induced transfer of weight from the magnetic fluctuations at \mathbf{Q}_1 . However, it is not yet possible to perform INS experiments above 17 T [33], which limits the possibilities to test this hypothesis. A similar situation is reported in another heavy-fermion system: the La-doped CeRu_2Si_2 [28,29]. In this system, static AFM order with the commensurate wave vector $\mathbf{q} = (1/3, 1/3, 0)$, which is close to one of the three incommensurate wave vectors of magnetic fluctuations in pure CeRu_2Si_2 [34,35], is induced under magnetic fields. Indeed, it is interesting that two different phases with large magnetic moments induced by pressure and magnetic field near the HO phase are closely associated with the underlying commensurate and incommensurate magnetic fluctuations, respectively, in pure URu_2Si_2 . Furthermore, the different magnetic structures induced by pressure and magnetic field are consistent with the contrastive FS: the unchanged FS under pressure [7] and the substantially changed FS under strong magnetic fields [15,16]. At least, the present result clearly indicates that the translational symmetry of the HO is not identical to that of phase II.

It should be noted that the magnetic structure of phase II is the same structure as that previously proposed from magnetization measurements [13]. In Ref. [13], following the arguments based on a localized picture of the $5f$ electrons, the three-step metamagnetic transitions in URu_2Si_2 were explained as the energy level crossing among different states described by a simple effective spin Hamiltonian, where exchange constants between U ions within third neighbors were assumed to be all antiferromagnetic. In fact, the AFM interactions are qualitatively consistent with those at short distances derived from analyzing the INS spectra in the HO phase [2]. The inter-site AFM interactions within third neighbors could describe the magnetic structures of the field-induced phases in URu_2Si_2 effectively. This possibility is expected to be examined by future high-field experiments.

In summary, for the first time we directly observed the magnetic order parameter of the field-induced phase II of $U(\text{Ru}_{0.96}\text{Rh}_{0.04})_2\text{Si}_2$ by neutron diffraction under pulsed high magnetic fields up to 30 T. The magnetic structure of phase II is the ferrimagnetic structure with $\mathbf{q} = (2/3, 0, 0)$, indicating the close relation between phase II and the characteristic incommensurate magnetic fluctuations at $\mathbf{Q}_1 = (0.6, 0, 0)$. The determined translational symmetry of phase II is key information on its feedback on the electronic structure. The FS in phase II must be governed by a new Brillouin zone quite distinct from that related to a lattice doubling in the pressure-induced AFM phase characterized by $\mathbf{Q}_0 = (1, 0, 0)$ and the unidentified spatial periodicity in the HO. Possible theories of HO should be checked for the stability between the HO phase and this well-identified field-induced AFM phase, taking into account all the physical ingredients of URu_2Si_2 , notably its FS evolution.

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