

UNIVERSIDADE ESTADUAL DE CAMPINAS
SISTEMA DE BIBLIOTECAS DA UNICAMP
REPOSITÓRIO DA PRODUÇÃO CIENTÍFICA E INTELLECTUAL DA UNICAMP

Versão do arquivo anexado / Version of attached file:

Versão do Editor / Published Version

Mais informações no site da editora / Further information on publisher's website:

<https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.112.236401>

DOI: 10.1103/PhysRevLett.112.236401

Direitos autorais / Publisher's copyright statement:

©2014 by American Physical Society. All rights reserved.

DIRETORIA DE TRATAMENTO DA INFORMAÇÃO

Cidade Universitária Zeferino Vaz Barão Geraldo

CEP 13083-970 – Campinas SP

Fone: (19) 3521-6493

<http://www.repositorio.unicamp.br>

Emergent Antiferromagnetism out of the “Hidden-Order” State in URu₂Si₂: High Magnetic Field Nuclear Magnetic Resonance to 40 T

H. Sakai,* Y. Tokunaga, and S. Kambe

Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan

R. R. Urbano

*Instituto de Física “Gleb Wataghin”, Universidade Estadual de Campinas, 13083-859 Campinas, SP, Brazil
and National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32310, USA*

M.-T. Suzuki†

CCSE, Japan Atomic Energy Agency, 5-1-5, Kashiwanoha, Kashiwa, Chiba 277-8587, Japan

P. L. Kuhns and A. P. Reyes

National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32310, USA

P. H. Tobash, F. Ronning, E. D. Bauer, and J. D. Thompson

Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

(Received 18 December 2013; revised manuscript received 10 April 2014; published 10 June 2014)

Very high field ²⁹Si-NMR measurements using a fully ²⁹Si-enriched URu₂Si₂ single crystal were carried out in order to microscopically investigate the “hidden order” (HO) state and adjacent magnetic phases in the high field limit. At the lowest measured temperature of 0.4 K, a clear anomaly reflecting a Fermi surface instability near 22 T inside the HO state is detected by the ²⁹Si shift, ²⁹K_c. Moreover, a strong enhancement of ²⁹K_c develops near a critical field $H_c \approx 35.6$ T, and the ²⁹Si-NMR signal disappears suddenly at H_c , indicating the total suppression of the HO state. Nevertheless, a weak and shifted ²⁹Si-NMR signal reappears for fields higher than H_c at 4.2 K, providing evidence for a magnetic structure within the magnetic phase caused by the Ising-type anisotropy of the uranium ordered moments.

DOI: [10.1103/PhysRevLett.112.236401](https://doi.org/10.1103/PhysRevLett.112.236401)

PACS numbers: 71.27.+a, 74.40.Kb, 76.60.-k

The so-called “hidden-order” (HO) state, which emerges in the URu₂Si₂ heavy-fermion superconductor below a zero field phase transition at $T_O = 17.5$ K, has posed a long-standing mystery, particularly because of the inconclusive nature of its order parameter [1].

URu₂Si₂ crystallizes in the tetragonal ThCr₂Si₂-type structure—one of the most common in heavy fermion compounds. Very recently, the fourfold rotational symmetry of this structure was suggested to be broken mesoscopically in the *c* plane within the HO phase [2]. Nonetheless, nuclear magnetic resonance (NMR) spectral analysis [3] revealed that the twofold anisotropy is much smaller than previously estimated from magnetic susceptibility measurements on a tiny single crystal. Moreover, hydrostatic pressure of about 1 GPa transforms the HO state into a simple antiferromagnetic (AFM) phase with propagation vector $\mathbf{Q}_1 = (1, 0, 0)$ and magnetic moment of $0.4 \mu_B/U$ [4]. Two well-split NMR lines are observed in this commensurate (C-) AFM phase confirming this propagation wave vector, whereas only a single NMR line is seen in the HO phase [5].

Another very effective tuning parameter in this compound is magnetic field. The HO phase can be monotonically suppressed to $T \rightarrow 0$ at a critical magnetic field $H_c \approx 35.6$ T, above which new phases labeled as II, III,

and V appear, as shown in Fig. 1 [6,7]. This phase diagram was determined mainly by resistivity and Hall effect measurements. Recent thermal-expansion and magnetostriction experiments also confirm the phase boundaries between HO-II, II-III, and III-paramagnetic (PM) phases above 2 K [8].

Shubnikov–de Haas (SdH) experiments reveal an anomaly in the Hall resistivity at $H^* \sim 22$ T within the HO phase, which is accompanied by the emergence of a new Fermi surface above H^* [9]. On the other hand, pulsed field magnetization measurements are not sensitive to this anomaly at H^* and the lowest temperature data suggest that the system undergoes a first order metamagnetic phase transition at about 35 T [10,11]. Despite intense experimental efforts, microscopic investigations of URu₂Si₂ are lacking at high magnetic fields because of inherent experimental challenge. In order to unveil the intriguing magnetic anomaly at H^* and the nature of the novel phases emerging in the high field limit, we have performed ultra high magnetic field ²⁹Si-NMR experiments using a fully ²⁹Si-enriched URu₂Si₂ single crystal. In this Letter, we present evidence for an antiferromagnetic structure in the magnetic phase II caused by Ising-like local moments near H_c , which suddenly emerges from the itinerant HO state.

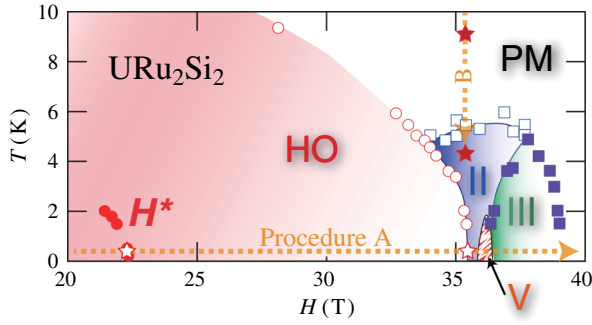


FIG. 1 (color online). H - T phase diagram for URu_2Si_2 for $H\parallel c$ [6,8,9]. The horizontal dotted arrow indicates experimental procedure (A) where the magnetic field was varied at constant 400 mK while carrying out the high field NMR measurements. The vertical dotted arrow indicates procedure (B) where the ^{29}Si -NMR signal was obtained varying the temperature near $H_c = 35.6$ T. The open stars represent H^* and H_c detected microscopically by NMR at 0.4 K. The solid stars represent both the NMR signal observed in the PM state and in phase II of URu_2Si_2 .

The NMR experiments were carried out using 30 T resistive (Bitter) and 45 T hybrid magnets at the National High Magnetic Field Laboratory in Tallahassee, Florida. The single crystals were enriched with 99.8% of the ^{29}Si isotope (nuclear spin $I = 1/2$ and gyromagnetic ratio $\gamma_N/2\pi = 8.4578$ MHz/T). The residual resistivity ratio of the sample was over 100, ensuring good crystallographic quality and no distribution of T_0 . A single crystal with dimensions $1 \times 1.5 \times 2$ mm³ was used. The pulse lengths and repetition time of RF pulses were chosen to be as short as possible because of the long relaxation time on the order of minutes at 400 mK [12]. The resulting nuclear magnetization was tipped less than $\sim 5^\circ$ from the fully recovered one. As illustrated by the dotted arrows in Fig. 1, the ^{29}Si -NMR signal was obtained following two experimental procedures: (A) the local frequency (ν_L) was swept stepwise under various fixed magnetic fields at a constant temperature of $T = 400$ mK, and (B) the external magnetic field was swept around 35.6 T at a constant ν_L for both $T = 9$ K and 4.2 K.

We begin with NMR data obtained following procedure (A). The ^{29}Si -NMR spectrum in the HO phase displays a single peak, as shown in Fig. 2(a), and there is only a slight change of shift at 22 T. On the other hand, the ^{29}Si -NMR spectrum suddenly disappears at $H_c = 35.6$ T, as shown in Figs. 2(b) and 2(c), with the value of H_c is reproduced approximately either by sweeping field up or down. This is in contrast to expectations for a second order transition, where the NMR spectrum should track the expected monotonic change in the shift. It is worth mentioning that there is no signature of critical slowing down that could increase the nuclear relaxation rates at this boundary. Therefore, these NMR data demonstrate that the HO phase does vanish microscopically at H_c and the

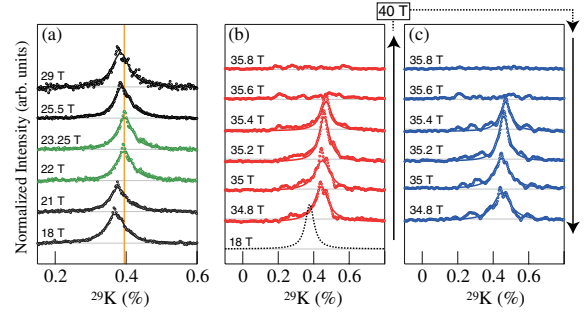


FIG. 2 (color online). (a) ^{29}Si NMR spectra of URu_2Si_2 at 0.4 K under several external magnetic fields along the c axis near $H^* = 22$ T. (b) and (c) show the spectra near $H_c = 35.6$ T, which were taken sweeping the applied magnetic field up to 40 T and down from it, respectively.

adjacent magnetic phase II emerges immediately above H_c with a very narrow field hysteresis.

As will be discussed, the ^{29}Si -NMR spectrum shifts and spreads out with a very peculiar shape in magnetic phase II at 4.2 K. This is the lowest temperature at which phase II was studied because the field range of phase II narrows appreciably at lower temperatures, giving way to the emergence of phases III and V, as shown in Fig. 1. The proximity of these phases creates strong fluctuations which in turn produce a wipe out effect of the ^{29}Si NMR spectrum at very low temperatures.

Figure 3(a) shows the ^{29}Si shift, $^{29}K_c$, at 400 mK plotted against the applied magnetic field. The Knight shift K is formally defined as $K = (H_0 - H_{\text{res}})/H_{\text{res}}$ with $H_0 = 2\pi\nu_L/\gamma_N$ and H_{res} is the resonant field. In general, K is proportional to the spin-component of the static susceptibility χ_{spin} via the hyperfine coupling constant A_{hf} , i.e., $K(T, H) = A_{\text{hf}}\chi(T, H)$. Although no spectral splitting

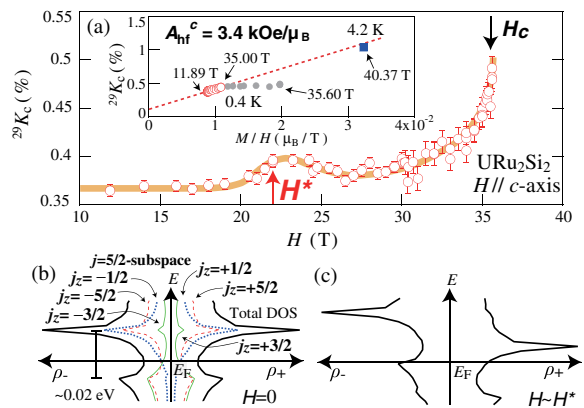


FIG. 3 (color online). (a) External magnetic field dependence of the NMR shift at 0.4 K along the c axis in URu_2Si_2 . The bold curve is a guide to the eye. The inset shows K - M/H plot. The magnetization data [11] in a pulsed field at 1.3 K are taken. (b) Total DOS (outer curves) for $H = 0$ calculated by a first principle FLAPW scheme. (c) Schematic picture of the total DOS for $H \sim H^*$.

and/or broadening of the ^{29}Si -NMR signal in the HO phase was observed, a small hump appears in $^{29}K_c$ around $H^* \sim 22$ T (see Fig. 3). $^{29}K_c$ increases rather strongly as the magnetic field is further increased towards $H_c \approx 35.6$ T. The K versus χ plot presented in the inset of Fig. 3(a) should show scaling of $^{29}K_c$ with the field dependence of the total susceptibility $\chi \equiv M/H$, which is calculated from magnetization data in a pulsed field at 1.3 K (Ref. [11]). The temperature dependence of $^{29}K_c$ and χ in the PM state below ~ 4 K is so gradual that the variation of χ between 1.3 and 0.4 K may be neglected. Indeed, despite the difference of temperatures in both experiments, $^{29}K_c$ is linearly proportional to M/H below ~ 35 T, as shown in the inset of Fig. 3(a). As seen in this inset, the value of $^{29}K_c$ obtained at 4.2 K in the field induced PM state at 40.37 T scales with the former linear behavior as suggested by the dashed line. A deviation from this behavior is seen only above ~ 35 T near H_c , which may be due to a slight change in the value of H_c between 0.4 and 1.3 K. Thus, the hyperfine coupling constant A_{hf}^c shows no significant change either in the HO phase or in the high-field PM state even at ~ 40 T. One may estimate $A_{\text{hf}}^c = 3.4$ kOe/ μ_B from the slope of the dashed line in the inset of Fig. 3, which is consistent with the reported value of 3.3–3.6 kOe/ μ_B obtained by ^{29}Si -NMR measurements in the PM state [12,13]. This is also in agreement with the conclusion from recent low-field ^{29}Si -NMR measurements [3,13] that there is no ordered moment. From a magnetic point of view, the HO phase can thus be viewed as a Pauli-PM-like state.

$^{29}K_c$ also is a fine measure of the density of states (DOS) at the Fermi surface, i.e., $K \propto \chi = \lambda g^* \mu_B^2 D(E_F)$, where λ is an enhancement factor dependent on the effective electronic mass, g^* is an effective g -factor, and $D(E_F)$ is the total DOS at the Fermi level. In the HO state, SdH experiments conclude that g^* of quasi particles is highly anisotropic, i.e., Ising-like with g^* being nearly zero when $H \parallel a$ [14,15]. Figure 3(b) shows the total DOS for $H = 0$ that is calculated by a first principle full-potential linearized augmented plane wave (FLAPW) scheme. Indeed, several band calculations for the PM state predict a total DOS where the Fermi level falls in a sharp minimum, and that the multiple Fermi surfaces are mainly composed of the total angular momentum $j = 5/2$ multiplet, with the $j = 7/2$ multiplet located more than ~ 1 eV from E_F due to the local crystalline electric field and the strong spin-orbit interaction [16,17]. When magnetic field is applied along the c axis, the Kramers degeneracies of f -states are lifted by the Zeeman effect, and the total DOS splits, as schematically illustrated in Fig. 3(c). We roughly estimate the peak-to-peak energy between peaks in DOS by a Zeeman energy $g^* j \mu_B H_c \sim 0.02$ eV, which is one order of magnitude smaller than ~ 0.17 eV in the PM band calculation [17]; such a difference would come from band renormalization in the HO state. Thus, the $j = 5/2$ states just near E_F produce an instability of the HO state at H_c , and the magnetic

moment from $j_z = \pm 5/2$ orbitals arises from an Ising-type anisotropy along the c axis.

In the following, we present the first observation of high field ^{29}Si -NMR spectrum in phase II of URu_2Si_2 . In order to carry out these experiments, we have opted for the experimental procedure illustrated by the vertical dotted arrow (B) in Fig. 1. The reason for this choice is that the ^{29}Si -NMR signal shifts abruptly when URu_2Si_2 undergoes a first order metamagnetic transition at H_c , right at the edge of the HO state [10,11]. So, field-swept NMR measurements were performed at constant frequency $\nu_L = 300.97$ MHz as the temperature was lowered from 9 K down to 4.2 K: a temperature where phase II covers the widest field range. The ^{29}Si -NMR spectra evolves smoothly across the transition at $T_{\text{II}} \approx 6$ K between the PM state and phase II, indicating its second order character. As shown in Fig. 4(a), the ^{29}Si -NMR signal changes remarkably from a single line in the PM state to a widely-spread spectrum within phase II. The latter is a characteristic spectral shape with a centered narrow peak and widely-spread shoulders with finite signal intensity on each side. The central line is considerably shifted to higher fields (lower $^{29}K_c$ values) when compared to the PM resonance position. This behavior indicates the emergence of AFM order in phase II.

Supposing that a long-ranged C-AFM order of Ising-type magnetic moments occurs with a propagation vector $\mathbf{Q}_1 = (1, 0, 0)$ under pressure, two well-separated NMR lines split by an internal field should be observed [5]. If the AFM order of XY-type moments occurs with $\mathbf{Q}_2 = (1/2, 1/2, 0)$ as observed in isostructural CePd_2Si_2 , the Si-NMR spectrum would show only a single line since the internal field cancels out at the Si sites [18]. Even if it was

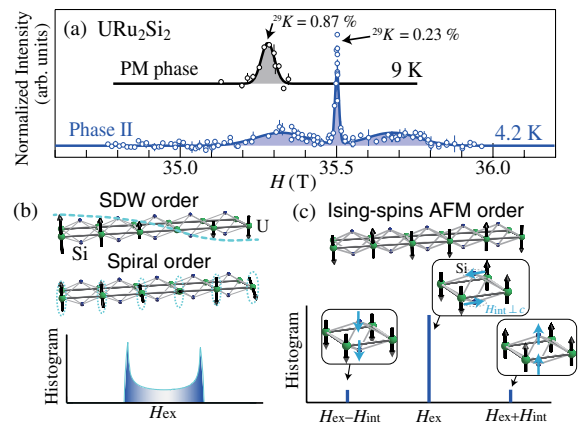


FIG. 4 (color online). (a) Field-swept ^{29}Si -NMR spectra at a constant frequency $\nu_L = 300.97$ MHz in the PM state at 9 K and in the phase II at 4.2 K. The shift ^{29}K values are indicated by arrows. Local magnetic environments for the Si sites and the histogram of the internal fields created at the Si sites are also shown to the external field (H_{ex}) for (b) IC-SDW and spiral AFM order and, (c) an $(\uparrow\downarrow)$ -AFM arrangement of Ising spins. The solid blue line (bold) drawn in (a) represents a simulation of the ^{29}Si -NMR spectrum at 4.2 K for the structure shown in (c).

Ising-like, only a small splitting would occur. In the rather complicated case of CeRh_2Si_2 [18,19] with $4q$ structure, which has $\mathbf{Q}_{2+} = (1/2, 1/2, 0)$ and $\mathbf{Q}_{2-} = (-1/2, 1/2, 0)$ with $\mathbf{Q}_3 = (1/2, 1/2, 1/2)$ -modulation [20], the ^{29}Si -NMR spectrum splits into two separated pairs due to the two internal fields $\pm H_{\text{int}}^{\text{A}}$ and $\pm H_{\text{int}}^{\text{B}}$. Now, if incommensurate (IC-) AFM order with any IC- \mathbf{Q}_{IC} is assumed to be realized, the NMR spectrum should display a “double-horn” shape, as illustrated in Fig. 4(b). Here, it should be noted that the “double-horn” pattern is unconstrained depending on whether the magnetic moments are ordered as an itinerant spin-density-wave (SDW) or a spiral order of local moments. In any case, the transferred internal field $H_{\text{int}}(i)$ at the i -th Si site should oscillate as $\propto \cos(2\pi\mathbf{Q}_{\text{IC}}\mathbf{r}_i)$, resulting in the “double-horn” histogram of H_{int} shown in Fig. 4(b). Thus, none of these cases can explain the characteristic spectral shape in the phase II of URu_2Si_2 , suggesting a necessity of new constraint.

An important hint comes from recent neutron diffraction studies of the expanded phase II of 4% Rh-doped URu_2Si_2 under pulsed magnetic field [21]. An *up-up-down* ($\uparrow\uparrow\downarrow$) ferrimagnetic structure characterized by the C-AFM vector $\mathbf{Q} = (2/3, 0, 0)$, with magnetic moments parallel to the c axis, has been suggested. The substitution of Ru by Rh atoms expands the area of phase II in the T - H phase diagram of $\text{U}(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$, although the HO state is completely quenched at 4% Rh-doping [7]. Therefore, accepting this result as a local constraint, we assume that the U spins in phase II of the pure URu_2Si_2 are ordered as Ising-like chains along the a -axis with equal spin magnitude, as shown in Fig. 4(c).

Without any lack of generality, one can consider magnetic order within a simple unit cell, where only the hyperfine coupling of the four nearest-neighbor (NN) U will contribute to each Si site. Figure 4(c) illustrates this situation. The local U-AFM structures can be sorted into three distinct local order patterns: (i) all four spins-up, (ii) all four spins-down, and (iii) two spins-up, two spins-down. In cases (i) and (ii), a finite c component of H_{int} from the U sites, $H_{\text{int}}^{\text{llc}}$, can be transferred to the Si site, whereas no projected H_{int} along the c direction is expected for case (iii), although a nonzero in-plane component $H_{\text{int}}^{\perp c}$ may exist. By applying this AFM model, the spectral shape histogram should display one centered line and two satellites, as shown in Fig. 4(c). If the next nearest neighbor (NNN) hyperfine couplings are also taken into account, a small splitting of the central resonance should be observed. Our results show no-splitting of the central peak, what may indicate that the NNN hyperfine contribution is negligible. [22]

A convolution of the histogram of Fig. 4(c) with inhomogeneous line broadening allows a reasonable fit to the high field ^{29}Si -NMR spectrum at 4.2 K within phase II of URu_2Si_2 , as shown in Fig. 4(a). The inhomogeneous linewidth of the peaks observed in the ^{29}Si -NMR spectrum is proportional to the distribution of the local

magnetic shift $^{29}K_c$, a direct consequence of an internal field distribution at the Si sites. The central resonance corresponds to the $H_{\text{int}}^{\perp c}$ component at the Si sites and displays a narrow linewidth, whereas the satellite peaks are broader because of the large contribution of $H_{\text{int}}^{\text{llc}}$ component. In addition, a distribution of \mathbf{Q} due to closeness to the T_{II} phase boundary, together with possible formation of domain structures from $\mathbf{Q} = (2/3, 0, 0)$ or $(0, 2/3, 0)$ AFM arrangements, could be plausible explanations for introducing an extra distribution in the histogram shown in Fig. 4(c). This scenario would be a natural consequence of the strong Ising anisotropy of χ . According to Ref. [12], the hyperfine coupling constant for ^{29}Si should be isotropic in URu_2Si_2 : $^{29}K_a$ is less than 0.1%. Indeed, we also found a nearly temperature- and field-independent $^{29}K_a \approx 0$ down to 400 mK and up to ~ 40 T for our enriched URu_2Si_2 single crystal. The interval $2H_{\text{int}}^c$ between split shoulders is ~ 0.4 T. By using the hyperfine coupling constant $A_{\text{hf}}^c = 3.4 \text{ kOe}/\mu_{\text{B}}$ from the PM and HO phases, we estimate an ordered moment of $\sim 0.6 \mu_{\text{B}}/\text{U}$. This value is in perfect agreement with the magnitude of the ordered moment determined by neutron diffraction measurements [21]. Thus, the observed high field ^{29}Si -NMR spectrum within phase II of URu_2Si_2 is consistent with the experimental data obtained in the expanded phase II of Rh-doped URu_2Si_2 . High field ^{29}Si NMR experiments also might shed new light on the nature of this expanded phase II with slight substitution of Rh for Ru in URu_2Si_2 [21] at temperatures and magnetic fields where the HO state is completely quenched.

In summary, ^{29}Si -NMR measurements up to 40 T are reported, for the first time, for a ^{29}Si enriched URu_2Si_2 high quality single crystal. From a microscopic point of view, the HO state ends at $H_c \approx 35.6$ T (at 0.4 K) after undergoing a first order transition with very narrow hysteretic behavior. $^{29}K_c$ in the HO phase is a fine measure of the bulk static susceptibility, which finds a subtle DOS anomaly around $H^* \sim 22$ T and an increase of DOS near H_c indicating a double peak structure in the total DOS of the HO state. Thus, the $^{29}K_c$ data reveal a nonmagnetic itinerant nature remaining in the HO state until fields very near H_c . On the other hand, an unusual ^{29}Si NMR spectrum is observed in the magnetic phase II right above the HO state. This NMR spectrum can be understood by AFM ordering of Ising moments along the a axis. These NMR experiments suggest that potential theories of the HO state in URu_2Si_2 should be seriously reviewed, in particular, accounting for the striking stability between the nonmagnetic HO phase and localized Ising moments in the adjacent phase II.

We thank H. Ikeda, K. Kuwahara, D. Aoki, and K. Sugiyama for valuable discussions. Work in Japan was supported by the Reimei Research Program of JAEA. Work at NHMFL was performed under the auspices of the NSF through the Cooperative Agreement No. DMR-0654118 and the State of Florida. R. R. U. also acknowledges

FAPESP (No. 2012/05903-6). Work at LANL was performed under the auspices of the U.S. DOE, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering.

*sakai.hironori@jaea.go.jp

†Present address: Center for Emergent Matter Science, RIKEN, 2-1, Hirosawa, Wako, Saitama 351-0198, Japan.

- [1] J. A. Mydosh and P. M. Oppeneer, *Rev. Mod. Phys.* **83**, 1301 (2011).
- [2] R. Okazaki, T. Shibauchi, H. J. Shi, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Ōnuki, H. Ikeda, and Y. Matsuda, *Science* **331**, 439 (2011).
- [3] S. Kambe, Y. Tokunaga, H. Sakai, T. D. Matsuda, Y. Haga, Z. Fisk, and R. E. Walstedt, *Phys. Rev. Lett.* **110**, 246406 (2013).
- [4] H. Amitsuka, M. Sato, N. Metoki, M. Yokoyama, K. Kuwahara, T. Sakakibara, H. Morimoto, S. Kawarazaki, Y. Miyako, and J. A. Mydosh, *Phys. Rev. Lett.* **83**, 5114 (1999).
- [5] K. Matsuda, Y. Kohori, T. Kohara, K. Kuwahara, and H. Amitsuka, *Phys. Rev. Lett.* **87**, 087203 (2001).
- [6] K. H. Kim, N. Harrison, M. Jaime, G. S. Boebinger, and J. A. Mydosh, *Phys. Rev. Lett.* **91**, 256401 (2003).
- [7] K. H. Kim, N. Harrison, H. Amitsuka, G. A. Jorge, M. Jaime, and J. A. Mydosh, *Phys. Rev. Lett.* **93**, 206402 (2004).
- [8] V. F. Correa, S. Francoual, M. Jaime, N. Harrison, T. P. Murphy, E. C. Palm, S. W. Tozer, A. H. Lacerda, P. A. Sharma, and J. A. Mydosh, *Phys. Rev. Lett.* **109**, 246405 (2012).
- [9] H. Shishido, K. Hashimoto, T. Shibauchi, T. Sasaki, H. Oizumi, N. Kobayashi, T. Takamasu, K. Takehana, Y. Imanaka, T. D. Matsuda, Y. Haga, Y. Ōnuki, and Y. Matsuda, *Phys. Rev. Lett.* **102**, 156403 (2009).
- [10] K. Sugiyama, H. Fuke, K. Kindo, K. Shimohata, A. A. Menovsky, J. A. Mydosh, and M. Date, *J. Phys. Soc. Jpn.* **59**, 3331 (1990).
- [11] K. Sugiyama, M. Nakashima, H. Ohkuni, K. Kindo, Y. Haga, T. Honma, E. Yamamoto, and Y. Ōnuki, *J. Phys. Soc. Jpn.* **68**, 3394 (1999).
- [12] Y. Kohori, K. Matsuda, and T. Kohara, *J. Phys. Soc. Jpn.* **65**, 1083 (1996).
- [13] S. Takagi, S. Ishihara, S. Saitoh, H. Sasaki, H. Tanida, M. Yokoyama, and H. Amitsuka, *J. Phys. Soc. Jpn.* **76**, 033708 (2007).
- [14] M. M. Altarawneh, N. Harrison, S. E. Sebastian, L. Balicas, P. H. Tobash, J. D. Thompson, F. Ronning, and E. D. Bauer, *Phys. Rev. Lett.* **106**, 146403 (2011).
- [15] M. M. Altarawneh, N. Harrison, G. Li, L. Balicas, P. H. Tobash, F. Ronning, and E. D. Bauer, *Phys. Rev. Lett.* **108**, 066407 (2012).
- [16] P. M. Oppeneer, J. Rusz, S. Elgazzar, M.-T. Suzuki, T. Durakiewicz, and J. A. Mydosh, *Phys. Rev. B* **82**, 205103 (2010).
- [17] H. Ikeda, M.-T. Suzuki, R. Arita, T. Takimoto, T. Shibauchi, and Y. Matsuda, *Nat. Phys.* **8**, 528 (2012).
- [18] Y. Kawasaki, K. Ishida, Y. Kitaoka, and K. Asayama, *Phys. Rev. B* **58**, 8634 (1998).
- [19] H. Sakai, Y. Tokunaga, S. Kambe, Y. Matsumoto, T. D. Matsuda, and Y. Haga, *J. Korean Phys. Soc.* **63**, 352 (2013).
- [20] S. Kawarazaki, M. Sato, Y. Miyako, N. Chigusa, K. Watanabe, N. Metoki, Y. Koike, and M. Nishi, *Phys. Rev. B* **61**, 4167 (2000).
- [21] K. Kuwahara, S. Yoshii, H. Nojiri, D. Aoki, W. Knafo, F. Duc, X. Fabrèges, G. W. Scheerer, P. Frings, G. L. J. A. Rikken, F. Bourdarot, L. P. Regnault, and J. Flouquet, *Phys. Rev. Lett.* **110**, 216406 (2013).
- [22] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.112.236401> for these histograms of transferred hyperfine fields created by the U moments at the Si sites.