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NMR Study of In-Plane Twofold Ordering in URu_2Si_2

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We report ^{29}Si NMR spectra and Knight shift measurements as a function of applied field orientation in the (001) basal plane of URu_2Si_2. Observed linewidth oscillations confirm the in-plane twofold ordered domain state observed in recent magnetic susceptibility measurements. Analysis of our linewidth data leads to estimate $\sim$0.4% for the twofold intrinsic (monodomain) susceptibility anisotropy in the basal plane, a value $\sim$15 times smaller than that obtained from recent susceptibility measurements.

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The heavy fermion compound URu_2Si_2 undergoes a second order phase transition [1–3] at $T_0 \sim 17.5$ K. Since the order parameter of the transition has not ever been clearly identified, such order has been termed ''hidden order'' [4]. Current understanding of the hidden order problem has recently been reviewed [5].

Owing to the tetragonal crystal structure of URu_2Si_2 (space group $I4/mmm$) [1], the physical properties of the disordered, i.e., paramagnetic, state of this compound exhibit fourfold rotational symmetry in the basal: (001) plane. In recent in-plane anisotropy measurements of the static magnetic susceptibility χ, the fourfold symmetry was reported to be spontaneously broken below T_0, and a twofold rotationally symmetric state was found to appear [6]. Since the symmetry breaking was observed without identifying its microscopic origin, this ordered phase is termed ''nematic'' for the moment; it has been interpreted with several models [7–10].

As a result of domain formation, the measurable twofold symmetric susceptibility depends strongly on sample size and can only be detected in very small (nearly monodomain) samples [6,11]. On the other hand, NMR results presented here do not depend on the sample and domain size, since NMR measurements probe the microscopic state at each nuclear site individually. The present NMR results are consistent with domain formation. While domains render the susceptibility and the NMR shift isotropic in the basal plane, in this Letter we show that a detailed analysis of ^{29}Si NMR linewidth data offers a quantitative measure of the twofold susceptibility effect. Our principal finding is that the intrinsic (monodomain) twofold susceptibility in our macroscopic single crystal sample is an order of magnitude less than that reported for far smaller monodomain samples by Okazaki *et al.* [6].

The low natural abundance ($\sim$4.7%) of the NMR isotope ^{29}Si ($I = 1/2$, gyromagnetic ratio $\gamma_{29}/2\pi =$ 845.77 Hz/Oe) has prevented highly accurate Si NMR measurements in URu_2Si_2 up to now. For the present study a single crystal sample has been prepared with the ^{29}Si isotope enriched to 53%, improving the NMR sensitivity by a factor $\sim$11. This makes it possible for the first time to resolve the in-plane magnetic anisotropy by means of NMR measurements. NMR spectra were obtained using fast-Fourier-transform of spin-echo signals at a fixed applied magnetic field. The NMR spectra were measured as a function of an applied field H rotated precisely in the (001) (basal) plane using a two-axis rotating sample stage. For this purpose a single crystal sample with almost perfect cylindrical shape ($1\,\mathrm{mm}\phi \perp [001] \times 3\,\mathrm{mm} \| [001]$), with narrow facets on the (110) planes, was used. Since the cross section of the sample in the basal plane is very nearly circular, no demagnetization effects need to be considered. High sample purity was confirmed by a residual resistivity $\sim 5\ \mu\Omega\,\mathrm{cm}$ [12].

We begin by formulating the NMR shift and susceptibility expected for the proposed twofold nematically ordered domain state [6] (Fig. 1). The local crystallographic symmetry of the Si site [13] in URu_2Si_2 is tetragonal 4 mm. In such a case there appear two types of magnetically orthorhombic domains, A and B. These domains have in-plane symmetry axes that are rotated by 90° relative to one another [6,11]. The susceptibilities for these domains may be written,

$$\chi_A(\theta) = \chi_\parallel \cos^2\theta + \chi_\perp \sin^2\theta + \chi_{\mathrm{orb}},$$
$$\chi_B(\theta) = \chi_A(\theta + 90^\circ) = \chi_\parallel \sin^2\theta + \chi_\perp \cos^2\theta + \chi_{\mathrm{orb}}, \quad (1)$$

respectively, where θ is the field angle relative to the twofold symmetry axis for A domains, which is found to be the crystalline [110] axis (for B domains it is $[1\bar{1}0]$) [6,11]. Recently, this domain structure has been confirmed directly in x-ray diffraction measurements [14]. The difference $\Delta\chi_{\mathrm{nem}} \equiv \chi_\perp - \chi_\parallel$ gives the magnitude of the twofold susceptibility. In Eq. (1) χ_{orb} is a θ and T-independent orbital term. In the following, we omit any further consideration of χ_{orb} or of the corresponding shift $K_{\mathrm{orb}} = A_{\mathrm{orb}}\chi_{\mathrm{orb}}$, since they play no role in our discussion.

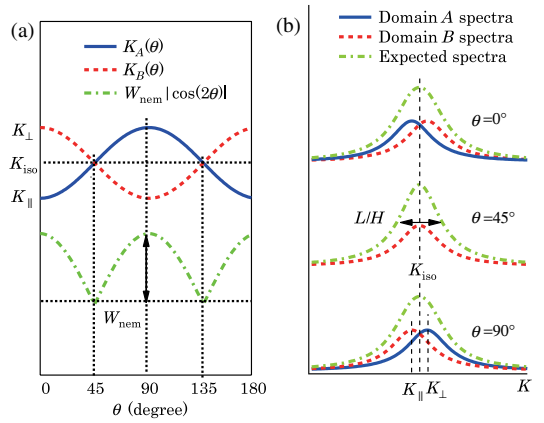

FIG. 1 (color online). (a) Blue (solid) and red (broken) lines show the angular dependence of Knight shifts $K_A(\theta)$ and $K_B(\theta)$ in Eq. (2). The net shift of the composite NMR line: K_{iso} is expected to be independent of θ in Eq. (3). The green (dotted) line represents the angular dependence of linewidth component $W_{nem}|\cos(2\theta)| = (H\Delta K_{nem} + W_{c2})|\cos(2\theta)|$ which exhibits characteristic sharp minima in Eq. (5). (b) Angular dependence expected for ^{29}Si NMR line broadening in the twofold ordered domain state. Blue (solid) and red (broken) lines represent spectra for domains A and B, respectively. At 45°, blue (solid) and red (broken) lines are identical. Green (dotted) lines indicate superposition of intensities from A and B domains that one might expect to observe experimentally. In practice the observed lines are always very nearly Lorentzian in form.

The ^{29}Si Knight shift $K(\theta)$ is defined as $K(\theta) \equiv (f_{res}(\theta) - f_0)/f_0$, where $f_{res}(\theta)$ is the observed resonance peak frequency and $f_0 \equiv \gamma_{29}H/2\pi$. To analyze the shift, we introduce the hyperfine coupling constant A giving [Fig. 1(a)],

$$K_A(\theta) = K_\parallel \cos^2\theta + K_\perp \sin^2\theta = K_{iso} - \frac{1}{2}\Delta K_{nem}\cos(2\theta),$$
$$K_B(\theta) = K_A(\theta + 90^\circ) = K_{iso} + \frac{1}{2}\Delta K_{nem}\cos(2\theta), \quad (2)$$

where $K_{\parallel,\perp} \equiv A\chi_{\parallel,\perp}$; $K_{iso} \equiv (1/2)(K_\parallel + K_\perp)$ is the isotropic shift; and $\Delta K_{nem} \equiv K_\perp - K_\parallel = A\Delta\chi_{nem}$ is the anisotropic shift. The torque magnetization data [6] give $\Delta\chi_{nem} > 0$. In the present case, $\Delta K_{nem} > 0$ as well, since $A > 0$ (see below). Assuming that the A and B domains each occupy half of the crystal and give ^{29}Si NMR lines of similar width and shape, the net shift of the composite line is estimated from Eq. (2) to be

$$K(\theta, T) = [K_A(\theta) + K_B(\theta)]/2 = K_{iso}. \quad (3)$$

Thus, $K(\theta, T)$ is expected to be independent of θ in the twofold domain state. Also, in the paramagnetic state where $\chi_\perp = \chi_\parallel$, $K(\theta, T)$ is found to be independent of θ at low fields [15].

It is clear from the foregoing that direct evidence for a twofold susceptibility in the domain state can only be obtained by analyzing ^{29}Si NMR linewidth data (see further details on this in the Supplemental Material [16]). In the paramagnetic state, linewidths are fourfold symmetric. Thus, the full linewidth at half maximum (FWHM) $L(\theta)$ may be expressed in units of magnetic field by

$$L_{para}(\theta) = 0.5W_{para}\cos(4\theta) + L_{c1}, \quad (4)$$

where the parameters W_{para} and L_{c1} are found to be independent of temperature (see the Supplemental Material [16] and below).

In the twofold domain state, one might expect a line splitting proportional to the difference $|K_A(\theta) - K_B(\theta)| = \Delta K_{nem}|\cos(2\theta)|$ [Figs. 1(a) and 1(b)]. Actually, because the Knight shift anisotropy ΔK_{nem} is estimated to be small in URu_2Si_2: $\Delta K_{nem} < 7 \times 10^{-3}\%$ at 5.4 T [17], the indicated shift anisotropy is expected to produce only a broadening effect. For line fitting purposes in the domain state, we add to Eq. (4) an oscillatory term $W_{nem}|\cos(2\theta)|$ plus a uniform broadening term L_{c2}, giving for a total width,

$$L_{nem}(\theta) = W_{nem}|\cos(2\theta)| + L_{c2} + L_{para}(\theta),$$
$$W_{nem} \equiv H\Delta K_{nem} + W_{c2}, \quad (5)$$

where in addition to the twofold shift term $H\Delta K_{nem}$, a field-independent oscillatory term W_{c2} also appears. As discussed in the Supplemental Material [16], we find actually that L_{c2} is independent of H, so that the field-dependent broadening in the domain state comes entirely from the twofold susceptibility and shift. Further, L_{c2} is thought to arise from weak but ordered magnetic moments in the hidden order state, an effect that may be connected to crystalline disorder; its slight anisotropy gives rise to the W_{c2} term. These terms are not expected to seriously affect measurements of ΔK_{nem}, which is our main focus in this Letter.

The rectified sinusoidal form of the W_{nem} term [Eq. (5)] exhibits sharp minima at $\theta = 45(1 + 2n)^\circ$ (n: integer), as shown in Fig. 1(a). These sharp minima, which have been found to occur widely in this study, are a special characteristic of the twofold domain state [18]. Such data confirm that the A and B domains are rotated by 90° relative to one another, since minima are observed with period of 90°, and that the A domain symmetry axis ∥ [110]. These findings serve to corroborate the results in Ref. [6].

It should be noted that the antiferromagnetic impurity phase [19] does not induce a fourfold linewidth at the Si site, since the antiferromagnetic ordered state of URu_2Si_2 induces an internal field along the [001] direction at the Si site. In addition, deviations from rotation about the [001] axis do not lead to any fourfold angular variation.

Figure 2(a) shows observed ^{29}Si NMR spectra at $H \sim 5.19$ T. The spectra have been fitted with Lorentzian functions, yielding values of $K(\theta)$ and $L(\theta)$. Here the nuclear dipole-dipole coupling contribution to the linewidth is estimated to be ~0.2 G, which is isotropic in the basal plane. The symmetric spectra in the ordered state indicate equal populations for the two domains. The measured

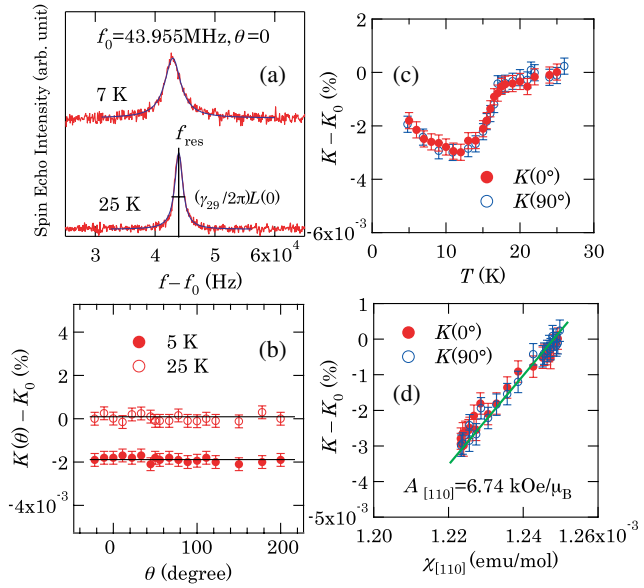

FIG. 2 (color online). (a) ^{29}Si NMR spectra for $\theta = 0^\circ$ ($H \parallel [110]$) in the paramagnetic (25 K) and ordered (7 K) states. Solid blue lines are fitted Lorentzian curves. Resonance peak frequencies and linewidths correspond to $f_{res}(0)$ and $(\gamma_{29}/2\pi)L(0)$, respectively. (b) Angular variation of the ^{29}Si Knight shift $K(\theta)$ at $T = 5$ K (filled circles) and 25 K (open circles). Solid lines are guides to the eye. Within experimental error the Knight shift has no angular dependence in either the paramagnetic (25 K) or ordered (5 K) state. (c) Comparison of the T dependence of the Knight shift at $\theta = 0^\circ$ (red closed circles) and $\theta = 90^\circ$ (blue open circles). $K_0 = 0.08\%$ is the Knight shift at $T = 25$ K. These two curves are identical within experimental error. (d) A K-χ plot of Knight shift versus static susceptibility. The plot is a straight line between 25 and 8 K; below 8 K it bends upward slightly. The solid green line is a least-squares fit of data between 25 and 8 K. The slope of the resulting line corresponds to a nuclear hyperfine coupling constant $A = 6.74$ kOe/μ_B.

linewidth ~1.5 kHz at 25 K in the present work is consistent with recently reported results on a single crystal sample [20], considering additional nuclear dipole-dipole broadening due to the enrichment of the ^{29}Si isotope. In previous results on an oriented powder sample [21], the linewidths in the paramagnetic and ordered states were much larger than in the present study, perhaps due to disorientation, defects, and impurities.

As shown in Fig. 2(b), no angular dependence of $K(\theta)$ is observed in either the paramagnetic or ordered states, consistent with the foregoing discussion [17,21].

The T dependences of $K(\theta = 0°)$ and $K(\theta = 90°)$ are presented in Fig. 2(c). The phase transition is clearly observed at T_0. For a wide temperature range in the paramagnetic and ordered states, the relation $K(0°) \cong K(90°)$ is confirmed, consistent with the fourfold local symmetry of the Si site in the paramagnetic state and with predictions for the twofold domain state.

A plot of the Knight shift versus the static susceptibility (K-χ plot) is presented in Fig. 2(d). Here we employ bulk data for χ measured for $H \parallel [110]$ in a large single crystal sample. In accord with the above discussion, χ in a multidomain sample was previously confirmed [6] to be independent of θ in both the paramagnetic and ordered states. Through the phase transition and down to ~8 K, the K-χ plot is linear within experimental error. This linearity indicates that the hyperfine coupling constant $A = 6.74 \pm 0.19$ kOe/μ_B does not change within error limits as one enters the ordered state. This behavior is also consistent with a previous result for $H \parallel [001]$ [22]. Below 8 K, K-χ bends upward slightly in the same manner for $K(0°)$ and $K(90°)$, meaning that the hyperfine coupling constant may be slightly modified [23].

Next, we discuss the linewidth results. The T and H dependences of $L(\theta)$ are consistent with reported results [20]. Thus, we focus on the angular dependence of $L(\theta)$ presented in Fig. 3(a). In the paramagnetic state, $L(\theta)$ is independent of T up to 70 K within experimental error. The linewidth also has a small fourfold amplitude as expected. The latter is a modulation of $L(\theta)$ driven by dipolar and pseudodipolar hyperfine couplings. The detailed modeling of these effects stemming from low-level crystalline disorder will be discussed elsewhere. As temperature decreases in the ordered state, the amplitude of the oscillating component increases. In particular, the sharp minima expected for the twofold domain state [see Eq. (5)] become pronounced at low temperatures. The appearance of oscillations with sharp minima at $\theta = 45^\circ$ is definite evidence for the presence of a susceptibility term having twofold symmetry axis ||[110] (for A domains) in this crystal. Values of W_{nem} deduced from linewidth data [Fig. 3(b)] then provide an estimate of a typical value for the two fold shift anisotropy ΔK_{nem} [18]. Solid lines for $L(\theta)$ are obtained using least-squares fits based on Eqs. (4) and (5). $L(\theta)$ is independent of T in the paramagnetic state, leading to T-independent values of W_{para} and L_{c1} based on Eq. (4) (see the Supplemental Material [16] for details). Using these values, W_{nem} and L_{c2} are then extracted using Eq. (5). The oscillations represented by W_{nem} are probably within the error bars of previous measurements [20].

Figure 3(b) shows the H dependence of W_{para} at $T = 25$ K and W_{nem} at 5 K. W_{nem} shows linear dependence vs H: $W_{nem} = (5.4 \pm 0.7) \times 10^{-6} H + (0.18 \pm 0.11)$ (H in units of Gauss), where the first and second terms correspond to $H\Delta K_{nem}$ and W_{c2}, respectively [see Eq. (5)]. Expressing ΔK_{nem} in terms of χ and A, one has,

$$\Delta K_{nem} = A \Delta \chi_{nem}. \quad (6)$$

The foregoing numbers then give for the monodomain twofold susceptibility anisotropy,

$$R \equiv \Delta\chi_{nem}/\chi_{\parallel} \sim (0.40 \pm 0.05)\% \quad \text{at 5 K}. \quad (7)$$

In the susceptibility study [6], the twofold anisotropy was found to depend strongly on sample size because of multidomain structure. Thus, R was found to be ~6% at 5 K in

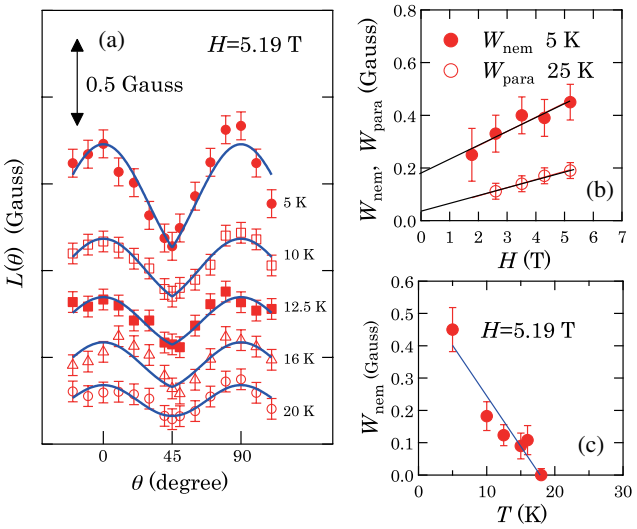

FIG. 3 (color online). (a) Angular variation of fitted Lorentzian linewidths $L(\theta)$ (red symbols) compared with calculated waveforms (blue lines). Curves are shown for a series of temperatures at $H = 5.19$ T. For each curve the vertical axis is offset for clarity. The solid lines are least-squares fits based on Eq. (5) in the ordered state and on Eq. (4) in the paramagnetic state. In the ordered state the characteristic sharp minimum appears around $\theta = 45^{\circ}$, and the resulting oscillation amplitude increases with decreasing T. Although results for angles ranging from $-20°$ to $110°$ are mainly presented here, the same angular dependence has been confirmed around $135°$ and $180°$ i.e., the sharp minima are observed at $\theta = 45(1 + 2n)°$ (n: integer). (b) The H dependence of the measured fourfold amplitude $W_{\rm para}$ is shown at 25 K (open circles) and that of $W_{\rm nem}$ is shown at 5 K (filled circles). The solid lines are least-squares linear fits to the data. (c) The T dependence of $W_{\rm nem}$ is plotted for $H = 5.19$ T. In the ordered state below T_0, $W_{\rm nem}$ begins to increase linearly. The solid line is a least-squares linear fit to the data.

the smallest (nearly monodomain) sample. A reorientation of domains may affect the estimate of R; however, such an effect is unlikely in the present case (see the Supplemental Material [16]). The present result [Eq. (7)], an order of magnitude smaller, calls into question the identification of $R \sim 6\%$ as the intrinsic value [6]. In NMR measurements, the measured portion of the sample depends on f_0 owing to RF skin-depth penetration effects. However, since $\Delta K_{\rm nem}$ is essentially independent of $f_0 = \gamma_{29} H/2\pi$, as shown in Fig. 3(b), our main findings remain unaffected by this, and thus are not surface sensitive.

The zero-field term from Fig. 3(b) is estimated to be $W_{c2} = 0.18 \pm 0.11$ G at $T = 5$ K, while $L_{c2} \sim 1$ G and is independent of field. These parameters indicate that very weak ordered moments appear in the hidden ordered state. A 1 G field at the Si site corresponds to a net neighboring U moment of $1/A \sim 1.5 \times 10^{-4}\,\mu_B$, which is 200 times smaller than the ordered moment $0.03\,\mu_B$ observed in neutron scattering measurements [24]. Further study is necessary to identify the physical origin of L_{c2} and W_{c2} (see the Supplemental Material [16]).

In the paramagnetic state at 25 K, $W_{\rm para}$ is expressed as $W_{\rm para} = (3.1 \pm 0.2) \times 10^{-6} H + (0.03 \pm 0.01)$ G, which is independent of T in the measured temperature range. The small value estimated for $W_{\rm para}$ at $H = 0$ is consistent with the result that the linewidth due to nuclear dipole-dipole coupling is very nearly isotropic in the basal plane.

The T dependence of $W_{\rm nem}$ is shown in Fig. 3(c). $W_{\rm nem}$ begins to increase below T_0 in the hidden order phase, as is indeed expected for the twofold nematically ordered domain state. The T dependence of $W_{\rm nem}$ below T_0 can be fitted with the simple form $W_{\rm nem} = 3.1 \times 10^{-2}(T_0 - T)$ Gauss at $H = 5.19$ T. This dominantly linear behavior suggests that both $\Delta K_{\rm nem}$ and W_{c2} have linear T dependence, which is roughly consistent with the previous result [6] $\Delta\chi_{\rm nem} \propto \{(T_0 - T) + 0.16(T_0 - T)^2\}$.

In conclusion, the present results establish an upper limit for the intrinsic susceptibility anisotropy in macroscopic single crystal samples of URu_2Si_2, namely, $R \sim 0.4\%$ at $T = 5$ K. Such a value is considerably less than the value $\sim$6% observed via direct susceptibility measurements [6] on a monodomain crystalline sample 5 orders of magnitude smaller in volume. Setting aside possible domain effects, the present result suggests that the intrinsic twofold anisotropy $\Delta\chi_{\rm nem}$ in high-quality crystals decreases with increasing sample size. On the other hand, other thermodynamic properties of the hidden order (e.g., T_0 [6] and the specific heat [25]) appear to be independent of sample size. These facts suggest that the twofold anisotropy, while not the primary order parameter which dominates thermodynamic properties, may be an accompanying order parameter which grows in magnitude with decreasing sample size for reasons not yet understood. Certainly the size dependence we note here needs further investigation.

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[1] T. T. M. Palstra, A. A. Menovsky, J. van den Berg, A. J. Dirkmaat, P. H. Kes, G. J. Nieuwenhuys, and J. A. Mydosh, Phys. Rev. Lett. **55**, 2727 (1985).
[2] W. Schlabitz, J. Baumann, B. Pollit, U. Rauchschwalbe, H. M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B **62**, 171 (1986).
[3] M. B. Maple, J. W. Chen, Y. Dalichaouch, T. Kohara, C. Rossel, and M. S. Torikachvili, M. W. McElfresh, and J. D. Thompson, Phys. Rev. Lett. **56**, 185 (1986).
[4] V. Tripathi, P. Chandra, and P. Coleman, Nat. Phys. **3**, 78 (2007).
[5] J. A. Mydosh and P. M. Oppeneer, Rev. Mod. Phys. **83**, 1301 (2011).

[6] R. Okazaki, T. Shibauchi, H. J. Shi, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, and Y. Matsuda, Science **331**, 439 (2011).
[7] S. Fujimoto, Phys. Rev. Lett. **106**, 196407 (2011).
[8] P. Thalmeier and T. Takimoto, Phys. Rev. B **83**, 165110 (2011).
[9] H. Ikeda, M.-T. Suzuki, R. Arita, T. Takimoto, T. Shibauchi, and Y. Matsuda, Nat. Phys. **8**, 528 (2012).
[10] J. G. Rau and H.-Y. Kee, Phys. Rev. B **85**, 245112 (2012).
[11] S. Tonegawa, K. Hashimoto, K. Ikada, Y.-H. Lin, H. Shishido, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, Y. Matsuda, and T. Shibauchi, Phys. Rev. Lett. **109**, 036401 (2012).
[12] T. D. Matsuda, E. Hassinger, D. Aoki, V. Taufour, G. Knebel, N. Tateiwa, E. Yamamoto, Y. Haga, Y. Onuki, Z. Fisk, and J. Flouquet, J. Phys. Soc. Jpn. **80**, 114710 (2011).
[13] H. Harima, K. Miyake, and J. Flouquet, J. Phys. Soc. Jpn. **79**, 033705 (2010).
[14] T. Shibauchi *et al.* (private communications). They have observed splittings of (880) diffraction peaks below T_0 using the synchrotron radiation x-ray source at SPring 8. This is similar to the iron-pnictide case; see, e.g., M. A. Tanatar, A. Kreyssig, S. Nandi, N. Ni, S. Bud'ko, P. Canfield, A. Goldman, and R. Prozorov, Phys. Rev. B **79**, 180508 (2009).
[15] A. Abragam, *Principles of Nuclear Magnetism* (Clarendon Press, Oxford, 1961), p. 204.
[16] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.110.246406 for details.
[17] K. Matsuda, Y. Kohori, T. Kohara, K. Kuwahara, and H. Amitsuka, Phys. Rev. Lett. **87**, 087203 (2001).
[18] A linewidth analysis showing how the Lorentzian linewidth term with W_{nem} arises out of the distribution of twofold shifts ΔK in the sample, where ΔK_{nem} is a characteristic value of such shifts, will be published elsewhere.
[19] H. Amitsuka, K. Matsuda, I. Kawasaki, K. Tenya, M. Yokoyama, C. Sekine, N. Tateiwa, T. C. Kobayashi, S. Kawarazaki, and H. Yoshizawa, J. Magn. Magn. Mater. **310**, 214 (2007).
[20] S. Takagi, S. Ishihara, M. Yokoyama, and H. Amitsuka, J. Phys. Soc. Jpn. **81**, 114710 (2012).
[21] O. O. Bernal, C. Rodrigues, A. Martinez, H. G. Lukefahr, D. E. MacLaughlin, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. Lett. **87**, 196402 (2001).
[22] S. Takagi, S. Ishihara, S. Saitoh, H. Sasaki, H. Tanida, M. Yokoyama, and H. Amitsuka, J. Phys. Soc. Jpn. **76**, 033708 (2007).
[23] Such behavior has been observed in many heavy fermion systems. In the present case, the hyperfine coupling may be slightly modified below the minimum of the K-χ plot. We distinguish two cases. First, it may be due to the formation of a heavy fermion state, as is suggested by an increase of specific heat C/T below $\sim$8 K. If K_{orb} (i.e., K extrapolated to $\chi_{[110]} = 0$ in the K-χ plot) remains invariant, the change in A below 8 K can be estimated as $\sim$1%. If the change of A pertains only to the small upturn in χ below 10 K, then it could be an order of magnitude greater than that. Because of this increase of A below 8 K, the point of W_{nem} at 5 K in Fig. 3(c) is a little bit above the solid line. For another example of these effects, see, e.g., H. Sakai, S.-H. Baek, S. E. Brown, F. Ronning, E. D. Bauer, and J. D. Thompson, Phys. Rev. B **82**, 020501 (2010).
[24] F. Bourdarot, N. Martin, S. Raymond, L.-P. Regnault, D. Aoki, V. Taufour, and J. Flouquet, Phys. Rev. B **84**, 184430 (2011).
[25] B. Fåk, C. Vettier, J. Flouquet, F. Bourdarot, S. Raymond, A. Verniere, P. Lejay, P. Boutrouille, N. Bernhoeft, S. T. Bramwell, R. A. Fischer, and N. E. Phillips, J. Magn. Magn. Mater. **154**, 339 (1996). Here it should be noted that sample quality for different sizes may not be the same.