Antiferromagnetic Correlations in Strongly Valence Fluctuating CeIrSn

Y. Shimura,[1] A. Wörl,[2] M. Sundermann,[3,4] S. Tsuda,[1] D. T. Adroja,[5,6,*] A. Bhattacharyya,[7] A. M. Strydom,[6] A. D. Hillier,[5] F. L. Pratt,[5] A. Gloskovskii,[4] A. Severing,[3,8] T. Onimaru,[1] P. Gegenwart,[2,†] and T. Takabatake[1]

[1]*Graduate School of Advanced Science and Engineering, Hiroshima University, Higashi-Hiroshima 739-8530, Japan*
[2]*Experimental Physics VI, Center for Electronic Correlations and Magnetism, University of Augsburg, 86159 Augsburg, Germany*
[3]*Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*
[4]*Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany*
[5]*ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot Oxon OX11 0QX, United Kingdom*
[6]*Highly Correlated Matter Research Group, Physics Department, University of Johannesburg, PO Box 524, Auckland Park 2006, South Africa*
[7]*Department of Physics, Ramakrishna Mission Vivekananda Educational and Research Institute, Belur Math, Howrah 711202, West Bengal, India*
[8]*Institute of Physics II, University of Cologne, 50937 Cologne, Germany*

(Received 8 October 2020; accepted 29 April 2021; published 27 May 2021)

CeIrSn with a quasikagome Ce lattice in the hexagonal basal plane is a strongly valence fluctuating compound, as we confirm by hard x-ray photoelectron spectroscopy and inelastic neutron scattering, with a high Kondo temperature of $T_K \sim 480$ K. We report a negative in-plane thermal expansion α/T below 2 K, which passes through a broad minimum near 0.75 K. Volume and a-axis magnetostriction for $B \| a$ are markedly negative at low fields and change sign before a sharp metamagnetic anomaly at 6 T. These behaviors are unexpected for Ce-based intermediate valence systems, which should feature positive expansivity. Rather they point towards antiferromagnetic correlations at very low temperatures. This is supported by muon spin relaxation measurements down to 0.1 K, which provide microscopic evidence for a broad distribution of internal magnetic fields. Comparison with isostructural CeRhSn suggests that these antiferromagnetic correlations emerging at $T \ll T_K$ result from geometrical frustration.

DOI: 10.1103/PhysRevLett.126.217202

Heavy fermion behavior in Ce- and Yb-based intermetallic compounds arises from the hybridization of localized $4f$ moments with conduction electrons. The Kondo interaction increases exponentially with the antiferromagnetic (AF) exchange J between the local $4f$ and conduction electron spins, $T_K \sim \exp(-1/J)$, while the indirect exchange coupling between the moments varies like $T_{\mathrm{RKKY}} \sim J^2$. For large J, T_K dominates and a paramagnetic Fermi liquid ground state is expected with all $4f$ moments bound in nonmagnetic singlets. For very large T_K, exceeding $\sim 10^2$ K, not only the spin but also the $4f$ charge fluctuates, giving rise to intermediate valence behavior [1]. This directly manifests itself in the volume, which undergoes characteristic changes as a function of temperature or magnetic field. Since nonmagnetic Ce^{4+} has a smaller ionic radius than magnetic Ce^{3+}, intermediate valence behavior results in positive thermal expansion, as in ordinary metals, and a positive magnetostriction due to the stabilization of the Ce^{3+} valence state by magnetic field [2,3]. In this Letter, we report a *negative* low-temperature thermal expansion and magnetostriction in the intermediate valent CeIrSn and provide microscopic evidence for AF correlations developing at temperatures 2 orders of magnitude below the large $T_K \sim 480$ K.

CeIrSn and its sister compound CeRhSn crystallize in the hexagonal ZrNiAl-type structure in which the Ce atoms form a quasikagome lattice in the basal plane [4–6]. The Kondo temperatures were estimated as $T_K \sim 240$ K (Rh) and $\sim$480 K (Ir), from the Kondo maximum found in electrical resistivity and thermopower [7]. These high Kondo temperatures classify the materials as valence fluctuating. Nevertheless, both compounds exhibit non-Fermi liquid behavior below $\sim$2 K [8,9]. For CeRhSn, AF spin fluctuations were detected by nuclear magnetic resonance measurements over a wide temperature range between 1.3 K and 200 K [10], while down to 0.05 K, muon spin rotation (μSR) experiments confirmed the absence of long-range magnetic order [11]. The magnetization shows a strong Ising-type anisotropy demonstrated by the magnetic susceptibility χ which exhibits $\chi_c/\chi_a \sim 10$. CeRhSn and CeIrSn feature anisotropic metamagnetic crossovers only for fields applied perpendicular to the (easy) c axis at $B_M = 3.5$ T below 0.5 K and 5.5 T below 2 K, respectively [9,12,13]. For metamagnetic crossovers in ordinary Kondo lattice systems, the energy scale of $\mu_B B_M$ should be comparable to $k_B T_K$, where μ_B and k_B are the Bohr magneton and the Boltzmann constant, respectively [14]. Strikingly, the B_M values of both materials are 2 orders

of magnitude smaller than expected from the valence fluctuation energy scales of 240 K (Rh) and 480 K (Ir).

The emergence of metamagnetic signatures deep inside the valence fluctuating regime implies magnetic correlations. Thermal expansion and magnetostriction have proven to be highly suitable thermodynamic probes to investigate this behavior. Since the expansion coefficient measures the pressure derivative of entropy, the entropy accumulation near a quantum critical point (QCP) results in a sign change of thermal expansion [15]. In particular, a characteristically negative thermal expansion is observed when the ground state is tuned through a magnetic QCP to an AF ordered state [16–18].

For CeRhSn, the linear thermal expansion coefficient along the a axis divided by temperature α_a/T divergently increases on cooling, while Fermi liquid behavior is realized along the c axis [13]. This indicates that the ground state is located in the vicinity of a QCP which is sensitive to uniaxial stress along the a axis but insensitive to c-axis stress. Indeed, a sign change and anomaly of α_a/T indicative of long-range magnetic order have been found at $\sim$0.4 K for a-axis uniaxial pressure $p_a \geq 0.06$ GPa [19]. The anisotropic quantum criticality and the formation of magnetic order for in-plane stress have been associated with the Kagome structure, whose frustration can be weakened by in-plane distortion. More direct evidence for geometrical frustration has been reported for the isostructural compound CePdAl, which displays partial magnetic order below $T_N = 2.7$ K [20,21] with only 2/3 of the Ce moments being involved down to 0.03 K. In the isostructural CePtPb, related residual spin dynamics inside the AF ordered phase has been detected by μSR measurements [22]. In this Letter, we prove that isostructural CeIrSn, despite being a strongly valence fluctuating Kondo lattice, features bulk AF correlations at temperatures below 2 K.

The strongly valence fluctuating ground state of CeIrSn is confirmed by hard x-ray photoelectron spectroscopy (HAXPES). For details on the setup, data treatment, and analysis see the Supplemental Material (SM) [23] and also Refs. [30–32]. Figure 1 displays the Ce3d core-level emission spectrum of CeIrSn (blue line). The data show the typical $4f^0$, $4f^1$, and $4f^2$ derived structures often observed in Ce intermetallic compounds. It is remarkable that the $4f^0$ spectral weight at 915 eV (see red arrow) is almost as strong as in the α-type cerium compound $CePd_3$ [31]. A combined full-multiplet and configuration-interaction analysis [33,34] provides an excellent simulation of the experimental spectrum. The red line displays the result taking into account resolution and lifetime broadening as well as some Mahan asymmetry (SM). The orange line shows the simulation with reduced broadening to better reveal the underlying multiplet structure of the three atomic configurations. The analysis indicates that the Ce ground state contains as much as 16% $4f^0$ configuration, thus,

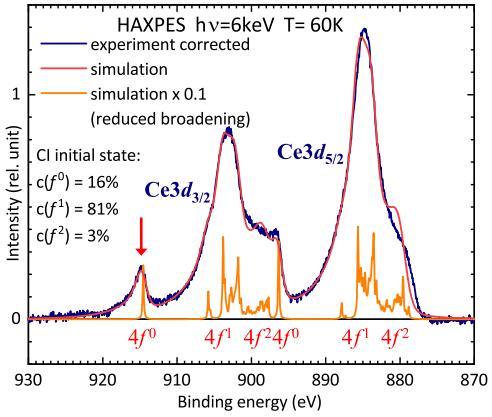

FIG. 1. Ce 3d core-level spectrum. Blue curve: as extracted from hard x-ray photoelectron spectroscopy (HAXPES); red curve: full-multiplet configuration-interaction simulation; orange curve: simulation with reduced broadening. The weights of the $4f^0$, $4f^1$, and $4f^2$ configurations in the ground state are also indicated.

demonstrating unambiguously the intermediate valent nature of CeIrSn. We would like to point out that the Ce3p core level spectrum can be described with the same parameters (SM). Inelastic neutron scattering (INS) reveals the absence of crystal-electric field excitations (SM), consistent with strong valence fluctuations evidenced by HAXPES.

Figure 2 displays the volume magnetostriction coefficient $\lambda_V = V^{-1}dV/dB$ for a magnetic field along the a axis at low temperatures, determined by high-resolution capacitive dilatometry [35] along three orthogonal directions (SM). At 0.05 K and 0.5 K, it exhibits a clear peak at $B_M = 6$ T which agrees with the metamagnetic crossover fields determined by dc magnetization $M(B)$ and ac susceptibility $\chi_{ac}(B)$ [9]. At 2 K, the peak is suppressed and only a broad shoulder remains. Interestingly, for fields below B_M, a negative λ_V is found which becomes more pronounced as temperature decreases. The Maxwell relation $(\partial V/\partial B)_P = -(\partial M/\partial P)_B$ indicates that the magnetization exhibits a

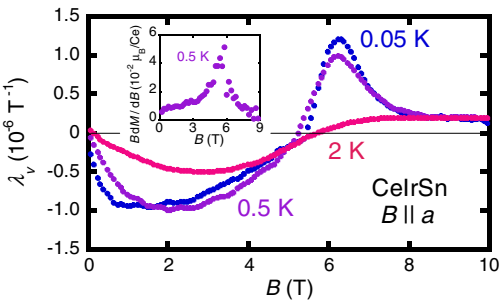

FIG. 2. Volume magnetostriction coefficient $\lambda_V = V^{-1}dV/dB$ for $B\|a$, determined from three independent linear magnetostriction measurements (SM). The inset shows the field variation of BdM/dB for $B\|a$ at 0.5 K, calculated from the dc magnetization data extracted from Ref. [9].

positive hydrostatic pressure dependence below B_M. This finding is in stark contrast to conventional Ce-based nonmagnetic valence fluctuating compounds, for which the application of hydrostatic pressure stabilizes the nonmagnetic Ce^{4+} state and therefore suppresses the magnetization [2]. As shown in the SM, the negative λ_V mainly arises from the largely negative longitudinal linear magnetostriction λ_a which indicates that uniaxial pressure along the a axis increases the magnetization for fields below 5 T.

The sign change of λ_V from negative to positive in the vicinity of B_M found in CeIrSn is unconventional among valence fluctuating and Kondo lattice systems with a metamagnetic crossover. A conventional example is $CeRu_2Si_2$ which exhibits a sharp metamagnetic crossover at $B_M = 7.7$ T [36], where λ_V is positive below and above B_M [37,38]. Furthermore, the field dependence of λ_V is related to the differential susceptibility $\chi(B) = dM(B)/dB$ by a single constant positive parameter Ω [39,40] as

$$\lambda_V(B) = \Omega B \chi. \tag{1}$$

The parameter $\Omega = -d\log B_M/d\log V = 1/\kappa_T(B_M)^{-1} \times (dB_M/dp)$ (κ_T: isothermal compressibility) quantifies the volume dependence of the metamagnetic transition field [41]. It is constant if the free energy scales with respect to B/B_M, implying that B_M is the dominating field scale. For Ce-based Kondo systems, the Kondo scale and thus also the metamagnetic transition field increase with pressure, giving rise to a positive scaling parameter Ω and, since the other factors in Eq. (1) are also positive, $\lambda_V > 0$. In contrast to Ce systems, the Kondo scale of Yb systems decreases under pressure and thus $\lambda_V < 0$ is expected. For instance, in $YbCu_5$ ($B_M = 17$ T) and $YbIr_2Zn_{20}$ ($B_M = 9.7$ T for $B \,||\, [100]$) λ_V remains negative below and above B_M and follows the magnetic susceptibility [14,42,43]. Generally, Eq. (1) requires that one dominating parameter governs the metamagnetic behavior.

The inset of Fig. 2 displays BdM/dB as a function of $B\|a$ for CeIrSn at 0.5 K calculated from the dc magnetization data in Ref. [9], which is positive in the whole field range. Thus, the sign change of λ_V below the metamagnetic transition indicates that the scaling is violated, in contrast to the conventional paramagnetic heavy-fermion systems described above. Indeed, the negative sign of λ_V at low fields is unusual for Ce-based systems and fully unexpected for an intermediate valence material with T_K as large as 480 K. Since magnetostriction is a volume sensitive probe, there is no way to associate this anomalous behavior with spurious secondary or impurity phases. The observed negative λ_V, through the Maxwell relation, indicates an increase of the magnetization with pressure. Such increase is typically associated with the suppression of AF order or AF correlations by pressure. In fact, a similar negative-positive sign change of λ_V at B_M was also observed in the Ce-based antiferromagnets $CeAl_2$ and $CeRh_2Si_2$ [44,45].

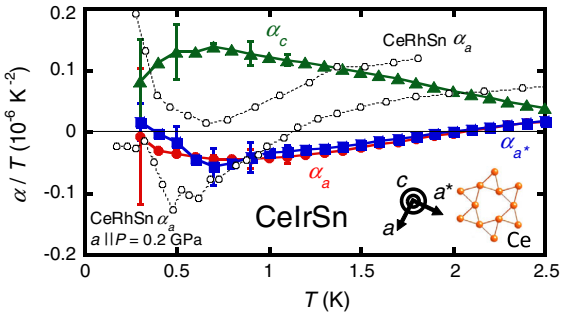

FIG. 3. Linear thermal-expansion coefficient as α/T of CeIrSn along the a (red circles), c (green triangles), and a^* (blue squares) axes at zero field. α_a/T data for CeRhSn at almost zero pressure and at 0.2 GPa applied along the a axis are extracted from Refs. [13,19].

Thus, the negative magnetostriction $\lambda_V < 0$ for CeIrSn suggests the development of cooperative magnetism at low fields and low temperatures. Even above B_M, the scaling of Eq. (1) does not work. In fact, it is only expected when the field energy equals the valence fluctuation energy, which will require high fields above 100 T for CeIrSn.

Figure 3 displays measurements of the linear thermal expansion $\alpha_i = L_i^{-1}dL_i/dT$ along the three main axes $i = a$, a^* and c (cf. the sketch) as α_i/T vs T at zero field. The in-plane expansivities are negative below 2 K and show a minimum at ~0.75 K, in contrast to a positive α_c/T. For comparison, we also plot α_a/T of CeRhSn at almost ambient pressure and under a uniaxial stress $P\|a$ of $P =$ 0.2 GPa [13,19]. Note that α/T of CeRhSn, measured at almost ambient pressure, diverges only along a, indicating that quantum criticality only couples to in-plane stress. This suggests quantum criticality driven by geometrical frustration and indeed the application of a distorting a-axis uniaxial stress, releases the non-Fermi liquid behavior and leads to a sign change of α at low temperatures.

As shown in Fig. 3, the α/T data of CeIrSn along the three directions do not diverge upon cooling to zero. This finding is in stark contrast to the low temperature divergence in α_a/T of CeRhSn at almost ambient pressure but qualitatively resembles the behavior found at a uniaxial stress of 0.2 GPa. The uniaxial stress induced discontinuous change of α_a/T in CeRhSn is attributed to magnetic ordering [19]. The negative in-plane thermal expansion without discontinuity observed in CeIrSn may thus indicate a short-range order. We recall that a negative thermal expansion manifests itself in Ce-based compounds such as $CeCu_{6-x}Au_x$ and $CeRhIn_{5-x}Sn_x$ when crossing the magnetic QCPs from a paramagnetic to an AF ordered state [16,17].

The anomalous negative in-plane thermal expansion and negative volume magnetostriction suggest that, despite its huge Kondo scale of 480 K, CeIrSn features a magnetic ground state. In order to obtain further microscopic information on the magnetic properties, we utilized μSR

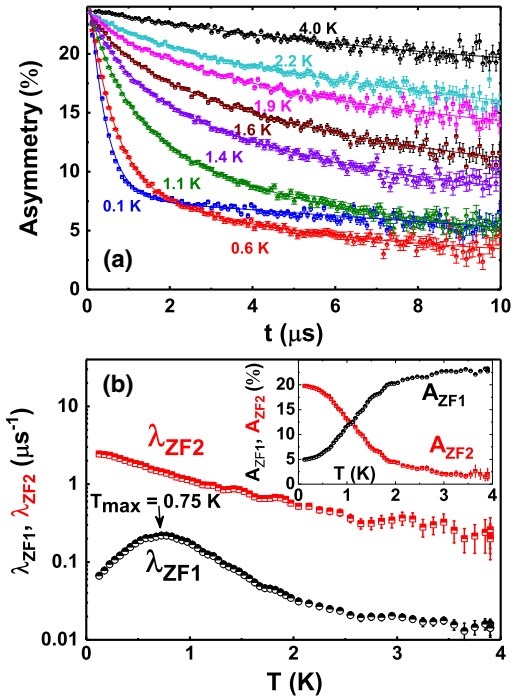

FIG. 4. (a) Zero field μSR spectra collected at the indicated temperatures. The solid curves are fits to the data by use of Eq. (2). (b) Temperature dependence of the slow (λ_{ZF1}) and the fast (λ_{ZF2}) relaxations. The inset shows the temperature dependence of the initial asymmetry of the slow relaxations (A_{ZF1}) and the fast one (A_{ZF2}).

measurements [46]. Figure 4(a) shows the ZF-μSR spectra measured at various temperatures ranging from 0.1 K to 4 K. The ZF-μSR data show neither loss of the initial asymmetry nor any sign of frequency oscillations down to 0.1 K. For a classical long-range magnetically ordered system below the magnetic transition temperature, ZF-μSR spectra exhibit either frequency oscillations for a small ordered moment or loss of initial muon asymmetry for a larger ordered moment (typically observed in ISIS-pulsed muon studies due to pulsed width of the muon beam). The ZF-μSR results indicate the presence of two components in the relaxation (Lorentzian form) below 2.5 K, while the spectra can be fitted with one slow component above 2.5 K.

The ZF-μSR spectra were fitted using various forms of the relaxation functions with one or two components. The best fits were obtained using Eq. (2) given below.

$$G_z(t) = A_{ZF1}e^{-\lambda_{ZF1}t} + A_{ZF2}e^{-\lambda_{ZF2}t} + A_{BG}, \qquad (2)$$

where A_{ZF1} (A_{ZF2}) and λ_{ZF1} (λ_{ZF2}) are the initial asymmetries and the electronic relaxation rates of the slow (fast) components, respectively. A_{BG} is the nonrelaxing background contribution from the silver sample holder, which was estimated to be 5% by fitting the 0.1 K spectra and was kept fixed. The results of the fit are presented in Fig. 4(b). At 4 K, the initial asymmetry (A_{ZF2}) of the fast relaxation is almost negligible (2%), while the one of the slow relaxation (A_{ZF1}) dominates (23%). With decreasing temperature, A_{ZF1} starts decreasing while A_{ZF2} starts increasing. After they cross near 1.2 K, the total asymmetry ($A_{ZF1} + A_{ZF2}$) remains temperature independent. At 0.1 K, A_{ZF2} is dominant (20%) and A_{ZF1} has smaller (5%) contribution. In contrast to the increase in the fast relaxation λ_{ZF2} on cooling, the slow relaxation component λ_{ZF1} exhibits a peak at ~0.75 K, which indicates the development of dynamical short-range magnetic correlations. Notably, this temperature agrees well with the temperature where α_a/T shows a minimum. The overall behavior of ZF-μSR spectra shows the presence of a very broad distribution of internal field without coherent precession of muon spins (see Fig. S6 in the SM). The Lorentzian relaxation function fits the μSR data of CeIrSn which is better than the stretch-exponential or root exponential functions used for a spin-glass system. The results of this analysis indicate that the ground state of CeIrSn is different from the classical spin-glass phase. These results are supported through the distribution of a broad internal field in our TF-μSR presented in the SM. The ZF-μSR spectra of CeRhSn exhibit only one Lorentzian component [11] with a weak anomaly near 1 K, while the asymmetry exhibits a similar temperature dependence as the one of A_{ZF2} in CeIrSn. Furthermore, the temperature dependences of the ZF- and TF-μSR asymmetries in CeIrSn indicate homogeneous magnetism without any phase separation. Thus, CeIrSn has two relaxation components. We note that a coexistence of magnetic order and magnetic fluctuating components was observed in the isostructural antiferromagnets CePdAl and CePtPb [20–22], likely caused by the frustrated quasikagome lattice. Since these CeTX systems including $Ce(Pd_xRh_{1-x})Sn$ ($0.75 > x > 0.20$) with the ZrNiAl-type structure exhibit AF ordering, we conjecture that the magnetic correlations in CeIrSn are also of AF type [12].

As discussed above, application of a-axis uniaxial stress, which evades frustration, has induced low-temperature magnetic ordering in CeRhSn [19]. Such a-axis stressed CeRhSn and CeIrSn share similar features such as a negative in-plane linear thermal expansion due to magnetic correlations. The negative magnetostriction along the a axis in CeIrSn also indicates that the magnetism is highly sensitive to in-plane uniaxial stress. Thus, the strong valence fluctuating material CeIrSn exhibits a highly uniaxial pressure-sensitive magnetism below 2 K. Reflecting the magnetic correlations, the specific heat divided by temperature $C(T)/T$ increases on cooling below ~4 K and tends to saturate around 2 K with a moderately large value of 80 mJ/K^2 mol, in spite of the strong valence fluctuations [9]. This characteristic temperature $T^* \sim 2$ K is 2 orders of magnitude smaller than $T_K \sim 480$ K. Emergence of magnetic correlations has been reported in various heavy-fermion metamagnets without long-range ordering like $CeRu_2Si_2$, for which, however, T^* is close to T_K [47]. The giant difference between T^* and T_K in CeIrSn indicates

that the origin of magnetic correlations is different from those in the conventional heavy-fermion metamagnets. The existence of such an exotic magnetic state deep inside the valence fluctuating regime may be caused by the competition between Kondo singlet formation and geometrical frustration in the ZrNiAl structure.

In this Letter, we have provided spectroscopic evidence for the strongly intermediate valent character of Ce in the quasikagome Kondo lattice CeIrSn, and reported anomalous negative in-plane linear thermal expansion below 2 K and negative volume magnetostriction at fields below a metamagnetic crossover that arises at energies 2 orders of magnitude below the bare Kondo scale. This behavior is unexpected for an ordinary intermediate valence state and points towards AF correlations, that are polarized beyond B_M. The ZF-μSR relaxation has two components. The relaxation rate of the slow component exhibits a peak around 0.75 K, while that of the fast component (developing below 2 K) saturates at this temperature, which coincides with the broad minimum of the negative a-axis thermal expansion. While μSR provides microscopic evidence for a broad distribution of quasistatic internal magnetic fields, thermal expansion and magnetostriction indicate that this is related to antiferromagnetism of bulk nature, incompatible with only a minor volume fraction. By comparison with isostructural CeRhSn, we conclude that magnetic frustration, resulting from the quasikagome structure, likely counteracts Kondo singlet formation even in this strongly intermediate valence material.

We would like to thank Stewart Parker and Rob Bewley for an enlightening discussion on the inelastic neutron scattering results. This work was supported by projects JSPS KAKENHI No. JP17K05545, No. JP15H05886, No. JP18KK0078, and No. JP18H01182 and by the German Research Foundation (DFG) via the Project No. 107745057 (TRR80). Y. S. thanks the Iketani Science and Technology Foundation (No. 0301080-A and No. 0321131-A) of Japanese funding support. D. T. A. would like to thank JSPS for the invitation fellowship and the Royal Society of London for the International exchange, UK-Japan, funding. A. B. would like to acknowledge the Department of Science and Technology (DST) India, for an Inspire Faculty Research Grant (DST/INSPIRE/04/2015/000169) and JNCASR for funding support. A. M. S. thanks the SA-NRF and the URC/FRC of UJ for financial assistance. We acknowledge the ISIS facility for beam time. We acknowledge DESY (Hamburg, Germany), a member of the Helmholtz Association HGF, for the provision of experimental facilities.

Y. S., A. W., and M. S. contributed equally to this work.

*devashibhai.adroja@stfc.ac.uk
†philipp.gegenwart@physik.uni-augsburg.de

[1] M. Loewenhaupt and K. Fischer, Valence fluctuation and heavy-fermion 4f systems, in *Handbook on the Physics and Chemistry of Rare Earths* (K.A. Gschneidner and Leroy Eyring, Amsterdam, 1993), Vol. 16, Ch. 105.
[2] H. Hafner, J. Magn. Magn. Mater. **47–48**, 299 (1985).
[3] A. Edelstein and N. Koon, Solid State Commun. **48**, 269 (1983).
[4] F. Hulliger, J. Alloys Compd. **196**, 225 (1993).
[5] B. Chevalier, C. Sebastian, and R. Pöttgen, Solid State Sci. **8**, 1000 (2006).
[6] R. Pöttgen and B. Chevalier, Z. Naturforsch. B **70**, 289 (2015).
[7] Y. Bando, T. Suemitsu, K. Takagi, H. Tokushima, Y. Echizen, K. Katoh, K. Umeo, Y. Maeda, and T. Takabatake, J. Alloys Compd. **313**, 1 (2000).
[8] M. S. Kim, Y. Echizen, K. Umeo, S. Kobayashi, M. Sera, P. S. Salamkha, O. L. Sologub, T. Takabatake, X. Chen, T. Tayama, T. Sakakibara, M. H. Jung, and M. B. Maple, Phys. Rev. B **68**, 054416 (2003).
[9] S. Tsuda, C. L. Yang, Y. Shimura, K. Umeo, H. Fukuoka, Y. Yamane, T. Onimaru, T. Takabatake, N. Kikugawa, T. Terashima, H. T. Hirose, S. Uji, S. Kittaka, and T. Sakakibara, Phys. Rev. B **98**, 155147 (2018).
[10] H. Tou, M. S. Kim, T. Takabatake, and M. Sera, Phys. Rev. B **70**, 100407(R) (2004).
[11] A. Schenck, F. N. Gygax, M. S. Kim, and T. Takabatake, J. Phys. Soc. Jpn. **73**, 3099 (2004).
[12] C. L. Yang, S. Tsuda, K. Umeo, Y. Yamane, T. Onimaru, T. Takabatake, N. Kikugawa, T. Terashima, and S. Uji, Phys. Rev. B **96**, 045139 (2017).
[13] Y. Tokiwa, C. Stingl, M.-S. Kim, T. Takabatake, and P. Gegenwart, Sci. Adv. **1**, e1500001 (2015).
[14] T. Takeuchi, S. Yasui, M. Toda, M. Matsushita, S. Yoshiuchi, M. Ohya, K. Katayama, Y. Hirose, N. Yoshitani, F. Honda, K. Sugiyama, M. Hagiwara, K. Kindo, E. Yamamoto, Y. Haga, T. Tanaka, Y. Kubo, R. Settai, and Y. Onuki, J. Phys. Soc. Jpn. **79**, 064609 (2010).
[15] M. Garst and A. Rosch, Phys. Rev. B **72**, 205129 (2005).
[16] J. G. Donath, F. Steglich, E. D. Bauer, F. Ronning, J. L. Sarrao, and P. Gegenwart, Europhys. Lett. **87**, 57011 (2009).
[17] K. Grube, L. Pintschovius, F. Weber, J.-P. Castellan, S. Zaum, S. Kuntz, P. Schweiss, O. Stockert, S. Bachus, Y. Shimura, V. Fritsch, and H. v. Löhneysen, Phys. Rev. Lett. **121**, 087203 (2018).
[18] R. Küchler, P. Gegenwart, J. Custers, O. Stockert, N. Caroca-Canales, C. Geibel, J. G. Sereni, and F. Steglich, Phys. Rev. Lett. **96**, 256403 (2006).
[19] R. Küchler, C. Stingl, Y. Tokiwa, M. S. Kim, T. Takabatake, and P. Gegenwart, Phys. Rev. B **96**, 241110(R) (2017).
[20] A. Oyamada, S. Maegawa, M. Nishiyama, H. Kitazawa, and Y. Isikawa, Phys. Rev. B **77**, 064432 (2008).
[21] A. Dönni, G. Ehlers, H. Maletta, P. Fischer, H. Kitazawa, and M. Zolliker, J. Phys. Condens. Matter **8**, 11213 (1996).
[22] A. C. Y. Fang, S. R. Dunsiger, A. Pal, K. Akintola, J. E. Sonier, and E. Mun, Phys. Rev. B **100**, 024404 (2019).
[23] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.126.217202 for experimental details, HAXPES, INS, Magnetostriction and TF-μSR, which includes Refs. [24–29].
[24] F. Pratt, Physica (Amsterdam) **289B–290B**, 710 (2000).

[25] A. D. Hillier, D. T. Adroja, P. Manuel, V. K. Anand, J. W. Taylor, K. A. McEwen, B. D. Rainford, and M. M. Koza, Phys. Rev. B **85**, 134405 (2012).
[26] A. P. Murani, A. Severing, and W. G. Marshall, Phys. Rev. B **53**, 2641 (1996).
[27] J. Jones and P. Hore, J. Magn. Reson. **92**, 276 (1991).
[28] D. S. Stephenson, Prog. NMR Spectrosc. **20**, 515 (1988).
[29] A. Maisuradze, A. Yaouanc, and P. D. de Reotier, J. Phys. Soc. Jpn. Conf. Proc. **21**, 011053 (2018).
[30] F. Strigari, M. Sundermann, Y. Muro, K. Yutani, T. Takabatake, K.-D. Tsuei, Y. Liao, A. Tanaka, P. Thalmeier, M. Haverkort, L. Tjeng, and A. Severing, J. Electron Spectrosc. Relat. Phenom. **199**, 56 (2015).
[31] M. Sundermann *et al.*, J. Electron Spectrosc. Relat. Phenom. **209**, 1 (2016).
[32] M. Sundermann, K. Chen, Y. Utsumi, Y.-H. Wu, K.-D. Tsuei, J. Haenel, A. Prokofiev, S. Paschen, A. Tanaka, L. H. Tjeng, and A. Severing, J. Phys. Conf. Ser. **807**, 022001 (2017).
[33] J. M. Imer and E. Wuilloud, Z. Phys. B **66**, 153 (1987).
[34] A. Tanaka and T. Jo, J. Phys. Soc. Jpn. **63**, 2788 (1994).
[35] R. Küchler, A. Wörl, P. Gegenwart, M. Berben, B. Bryant, and S. Wiedmann, Rev. Sci. Instrum. **88**, 083903 (2017).
[36] P. Haen, J. Flouquet, F. Lapierre, P. Lejay, and G. Remenyi, J. Low Temp. Phys. **67**, 391 (1987).
[37] A. Lacerda, A. de Visser, P. Haen, P. Lejay, and J. Flouquet, Phys. Rev. B **40**, 8759 (1989).
[38] A. de Visser, J. Flouquet, J. Franse, P. Haen, K. Hasselbach, A. Lacerda, and L. Taillefer, Physica (Amsterdam) **171B**, 190 (1991).
[39] L. Puech, J.-M. Mignot, P. Lejay, P. Haen, and J. Flouquet, J. Low Temp. Phys. **70**, 237 (1988).
[40] K. Matsuhira, T. Sakakibara, A. Nomachi, T. Tayama, K. Tenya, H. Amitsuka, K. Maezawa, and Y. Onuki, J. Phys. Soc. Jpn. **68**, 3402 (1999).
[41] A. B. Kaiser and P. Fulde, Phys. Rev. B **37**, 5357 (1988).
[42] N. Tsujii, H. Mitamura, T. Goto, K. Yoshimura, K. Kosuge, T. Terashima, T. Takamasu, H. Kitazawa, S. Kato, and G. Kido, Physica (Amsterdam) **294B–295B**, 284 (2001).
[43] Y. Shimura, T. Sakakibara, S. Yoshiuchi, F. Honda, R. Settai, and Y. Onuki, J. Phys. Soc. Jpn. **80**, SA051 (2011).
[44] E. Fawcett, V. Pluzhnikov, and H. Klimker, Phys. Rev. B **43**, 8531 (1991).
[45] O. Naito, T. Takeuchi, K. Kindo, Y. Tabata, and S. Kawarazaki, Physica (Amsterdam) **329B–333B**, 512 (2003).
[46] D. Adroja, C. Yang, T. Takabatake, A. Hillier, and A. Bhattacharyya, Geometrical frustration induced magnetism in single crystal of CeIrSn, STFC ISIS Neutron and Muon Source, https://doi.org/10.5286/ISIS.E.RB1810852 (2018).
[47] D. Aoki, W. Knafo, and I. Sheikin, C.R. Phys. **14**, 53 (2013).