Ground state of a quantum critical system: Neutron scattering on Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$

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We present neutron scattering data on Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$ that show that magnetic ordering in the vicinity of a quantum critical point is restricted to short length scales even though all moments that are present have lined up with their neighbors. We argue that order is not limited by crystallographic defects but rather that quantum fluctuations disorder the system and dilute the magnetic moments to such an extent that a percolation network forms. The latter naturally explains how E/T scaling is possible in a system whose apparent dimensionality is above the upper critical dimension.

The quantum critical point (QCP) refers to the order-disorder phase transition that takes place at 0 K as a function of tuning parameter, such as using pressure or applying magnetic fields.[1,2] In a metal, this tuning changes the interaction between the atomic magnetic moments and the conduction electrons, driving a magnetically ordered state into a quantum-disordered state.[3] At the QCP, the interaction strength is critical: on the one hand, there is the tendency of the magnetic moments to order; on the other hand, the conduction electrons are now so strongly coupled to these moments that they become almost localized, thereby shielding the moments from each other. This competition manifests itself most strongly in these metals at the QCP where the response can no longer be described by standard Fermi-liquid theory. Some of the QCP systems exhibit E/T scaling,[4–8] the property that the response depends only on the ratio of probing energy E and temperature T. This E/T scaling is one of the most puzzling phenomena in these strongly correlated electron systems; with the possible exception[9] of CeCu$_{5.9}$Au$_{0.1}$, this scaling should be disallowed[1] since the dimensionality of these systems[5,7,8] is above the upper critical dimension, necessarily resulting in mean field behavior.

Understanding the state of matter near the QCP where Fermi-liquid theory fails might even be the starting point for explaining high-temperature superconductivity.[10] In order to understand the excited states of QCP systems that determine properties such as superconductivity, we must first characterize the ground state. In particular, we must determine the fate of the magnetic moments at the QCP. Fully shielded moments could lead to the formation[11,12] of a collective spin-density wave accompanied by magnetic fluctuations that decay exponentially in time, whereas vestiges of moments on the brink of ordering would lead to a divergent[9] local response. However, neither possibility could explain the response[7] of Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$, presumably because the response near the QCP is strongly influenced by disorder.[13] Here, we show that in quantum critical Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$ [$x=0.76$ (Ref. 7 and 14)], some local moments survive down

to 0 K and that they exhibit short-range incommensurate order (SRIO); this SRIO does not span the entire sample because of random quantum fluctuations. Our findings show how local random effects shape the response, and how the disorder associated with these fluctuations reconciles SRIO with E/T scaling.

We used an 11 g Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$ single crystal prepared to be at the QCP. Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$ crystallizes in the ThCr$_2$Si$_2$ structure,[14] with the magnetic cerium atoms forming a body-centered tetragonal lattice ($c/a=2.5$). A 4 cm long cylindrical crystal of ~7 mm diameter was grown using a floating zone furnace. We found the sample composition [using an electron probe microanalyzer (EPMA)] to be as intended with a few percentages of Ge-rich secondary phases.[15] The sample composition (normalized to 5 atoms/f.u.) is Ce$_{0.988}$(Ru$_{0.233}$Fe$_{0.777}$)$_2$Ge$_{1.993}$ at the top and Ce$_{0.996}$(Ru$_{0.261}$Fe$_{0.753}$)$_2$Ge$_{1.978}$ at the bottom. Thus,[7,14] the top is paramagnetic while the bottom is (just) in the ordered phase, consistent with the neutron scattering findings and the transport data. Specific heat data[15] showed the coefficient of the linear term γ at $T=0.3$ K to be 748 ± 5 mJ/mol K^2, and susceptibility data confirm that the moments are pointing along the easy axis (c axis).[15] Resistivity measurements[15] show the onset of coherence at $T_{coh}=15$ K.

We performed neutron scattering experiments to determine the ground state of Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$: the experiments were done using the HB3 (TRIAX) triple-axis spectrometers at Oak Ridge National Laboratory (Missouri Research Reactor), and the DCS time-of-flight spectrometer at the National Institute of Standards and Technology. The sample was mounted in the hhl-scattering plane on HB3 and DCS, and in the $h0l$ plane on TRIAX. The lowest temperature for the DCS (HB3/TRIAX) experiment was 0.4 K (1.56/4.4 K), using an incident neutron energy of 3.55 meV (14.7/13.7 meV), giving an energy resolution (full width at half maximum) of 0.10 meV (1.25/1.1 meV).

We show the magnetic scattering in Fig. 1. This scattering was separated from the incoherent nuclear scattering and
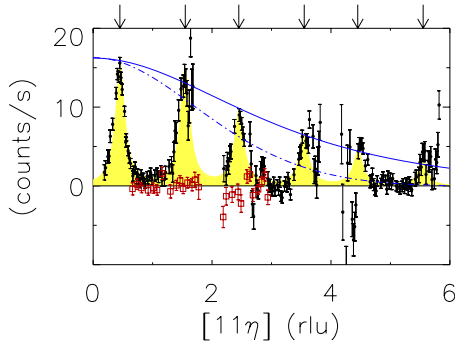
FIG. 1. (Color online) The onset of incommensurate ordering at low temperatures measured on HB3. There is a clear difference in elastic scattering between 2 and 56 K along $[11\eta]$ (black solid circles), whereas no difference is observed along $[00\eta]$ (light squares), showing that the moments are aligned along the c axis. The shaded area indicates the magnetic scattering. The gaps in the data points are caused by the $(1,1,2n)$ nuclear Bragg peaks and by the Al powder peaks. The solid line is the expected magnetic intensity based on the cross section for neutrons and the Ce f-electron form factor, (Ref. 22) the dotted line is calculated for Fe d-electrons. (Ref. 15). The arrows are at $(1,1,2n\pm0.45)$

weak background scattering by subtracting the signal at T =56 K. While this procedure slightly overestimates the background,[15] it does not complicate the interpretation. The experiments show that local Ce moments[16] persist down to the lowest temperatures. On lowering T, the magnetic intensity increases around the incommensurate positions $(n,n,2m\pm0.45)$, indicating that the system is on the verge of (conduction electron mediated) incommensurate order with propagation wave vector $(0,0,0.45)$. In agreement with poly-crystalline data,[7,16] we find that at low T the moment sizes and directions (up/down) have become correlated over a siz-able volume ($\sim2\times10^4$ Å^3 at T=2 K). The size of the or-dered moment is only $0.18\pm0.03\mu_B$/Ce ion (from compari-son with the nuclear scattering). Thus, the local moments are strongly Kondo shielded by the conduction electrons, as could already be inferred from the large γ value and the drop in ρ at T_{coh} when the screening clouds start to overlap. Next, we determine over what lengths the moments are correlated along the various symmetry directions.

The data show that the correlation lengths span an identi-cal number of magnetic moments along all directions, inde-pendent of the intermoment separation. In Figs. 2 and 3, we plot as a function of reciprocal lattice units (rlu) the net in-tensity measured along four directions at the ordering wave vector $(1,1,0.45)$, and the temperature evolution thereof. The spectra are described by Lorentzian line shapes of full width Γ in reciprocal space, corresponding to correlations that de-cay in real space as $\exp(-r/\xi)/r$, with $\Gamma=4\pi/\xi$. At 2 K, the spectral width Γ is 0.166 ± 0.003 rlu along $[00\eta]$ and 0.166 ± 0.004 rlu along $[\eta\eta0]$. For our body-centered struc-ture, both these numbers correspond to a correlation length spanning 12 magnetic moments. The spectral width (in rlu) along $[\eta\eta\eta]$ is found to be $2/3$ of the width measured along $[00\eta]$ (see Fig. 2). For a body-centered structure, this again corresponds to a correlation length spanning 12 moments.
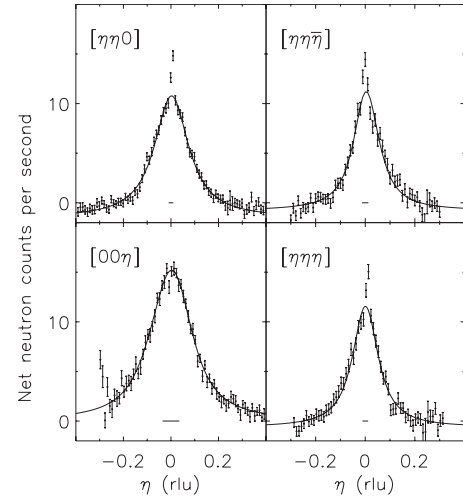

FIG. 2. Net elastic HB3 neutron scattering intensity near the ordering wave vector $(1,1,0.45)$ at T=2 K in four reciprocal space directions. The solid curves are fits to a Lorentzian line shape. The short horizontal lines at the origin of each figure denote the full resolution width determined at $(1,1,0)$.

This equivalence holds true for all measured temperatures [Fig. 3(a)]. While the data for $T>2$ K are not as accurate because of the difficulty in separating the magnetic from the nuclear scattering when the former becomes very broad, they suffice to rule out the possibility that the equivalence of the
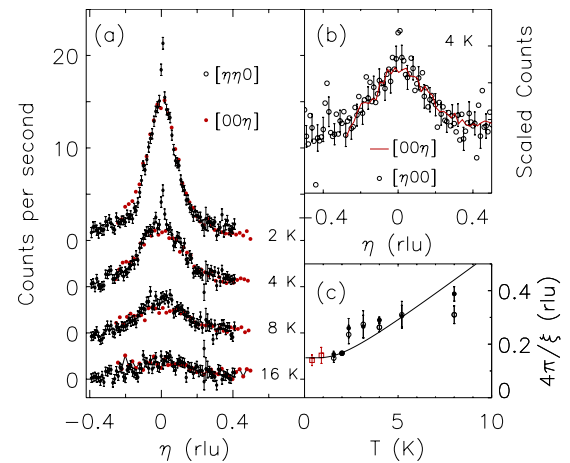

FIG. 3. (Color online) (a) The net magnetic scattering measured on HB3 along $[00\eta]$ (solid circles) and $[\eta\eta0]$ (open circles+error bars), centered on $(1,1,0.45)$. At all T, the FWHM in reciprocal space is the same along both directions (effective resolution is slightly worse along $[00\eta]$). For comparison, a constant background has been added to the $[\eta\eta0]$ data, and they have been multiplied by 1.3 to allow for a direct comparison between the two directions despite the sharp feature (Ref. 15) observed in the $[\eta\eta0]$ data. (b) Scaled 4K-HB3 data along $[00\eta]$ and 4.4K-TRIAX data [centered on $(1,0,1.45)$] along $[\eta00]$ show the full equivalence of the two directions. (c) The T-dependence of ξ. The HB3 data measured along $[\eta\eta0]$ ($[00\eta]$) are given by the open (solid) circles, the DCS data ($T<1$ K) by the open squares. The solid line is $\sim\coth(\Delta/T)$, with Δ=2.4 K.

correlation lengths at 2 K is an unlikely coincidence. Furthermore, TRIAX data comparing the $[\eta 00]$ and $[00\eta]$ directions [Fig. 3(b)] show these directions to also be fully equivalent ruling out all coincidence. We must therefore conclude that the correlation lengths are independent of the intermoment separation, even though the moments are 2.5 times further apart along the c direction than along the a direction. Finally, Fig. 2 also shows the sharp, ordered component (on top of the broader distribution) due to the Fe-poor phase at the bottom of the sample.[15] Since the overall contribution (the area under the curve) of this phase is small (3%), it does not affect our interpretation.

The DCS experiments show that ξ remains finite down to the lowest temperatures. While the DCS detectors are fixed in place so that measurements are along a curved path in hhl space, we can still get a reasonable estimate for $\xi(T)$ by assuming that the $[\eta\eta 0]$ and $[00\eta]$ directions are equivalent. The inferred correlation lengths are shown in Fig. 3(c) indicating that they remain finite down to $T=0$ K. At $T=0.4$ K, the width in *energy* of the fluctuations associated with ordering has become spectrometer resolution limited, implying that local order is maintained for at least 80 ps. Note that the HB3 resolution width exceeds the intrinsic linewidth of these fluctuations for temperatures below 20 K,[7] so that our elastic data, in fact, represent the energy integrated signal (ensuring that our reported correlation lengths are the true values).

From the fact that the correlation lengths span identical numbers of moments in all directions (contrary to what is expected for a SRIO system where the strength of the intermoment coupling varies with distance and direction[1]), and from the fact that these lengths saturate, we conclude that every Ce moment that can line up with its neighbors has indeed lined up. In this aspect, at the lowest T, the correlation lengths resemble those of a fully ordered system, yet long-range order has not been achieved. We argue that this reflects an intrinsic property of the system, rather than being an artifact of impurities or defects, or stacking faults associated with such disorder. First, the extent of the ordered regions actually *increases* with Ru doping; long-range order is achieved at a 1:4 doping level, well before Fe/Ru substitutional disorder is maximized. Second, crystallographic defects and stacking faults would result in a sampling over defect-limited ordered regions, which cannot yield the observed Lorentzian line shapes. Third, we actually observed (see Fig. 2) long-range order in the bottom (cutoff) 1 cm of our sample which was marginally richer in Ru than the rest of the sample; yet it was grown under identical circumstances. Thus, order cannot be limited by stacking faults or any other type of defect; it must reflect an intrinsic property of our system.

Because every Ce moment has lined up with its neighbors at the lowest T even though long-range order has not been achieved, we must conclude that not all Ce moments have neighboring moments. Since all moments are present at high T,[16] the magnetic moments must have been randomly diluted on cooling; some moments must have been shielded by the conduction electrons while other moments have survived. Nonidentical moments must originate from a nonconstant (in time or in space) overlap J of the Ce f-orbitals with the conduction electron states. Since the system is tuned to have
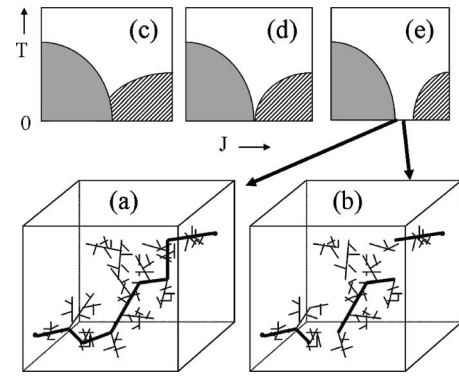

FIG. 4. The observed correlations associated with SRIO in our tetragonal system ($c/a=2.5$) span identical numbers of moments in all directions. This implies that some moments are missing and that a percolation network has formed [(a) and (b)], which in turn suggests a new phase diagram for our QCP system (e). In contrast to the spin-density wave scenario (Ref. 12) (c) and the local moment scenario (Ref. 9) (d), there is now a region in between the areas where the moments order (gray) and where they are fully shielded (striped), a region in which some moments survive down to 0 K.

a critical degree of overlap, even small changes will have noticeable effects.[3] A spatially varying overlap can originate from local lattice expansion and/or contraction caused by either an Fe or a larger Ru ion occupying a specific lattice site. A temporally varying overlap would be due to the zero-point motion of the cerium ions in their potential wells. Even though the amplitude of this motion is typically very small[17] [$\sim$0.1 Å at 16 K (Ref. 15)], in the vicinity of the QCP this motion will be large enough to upset the critical balance between moment shielding and moment surviving. Likely both effects conspire to give a nonconstant overlap.[15] Since the spatial variations caused by localized lattice expansion and/or contraction are identical to a pressure wave that has been frozen in, we refer to both temporal and spatial fluctuations as quantum fluctuations. Given that every moment that can line up with its neighbors has lined up (at least for 80 ps), we conclude that these quantum fluctuations have disordered the system, thereby preventing long-range order by locally diluting magnetic moments.

When moments are randomly removed from an ordered system, a percolation network emerges[18] (Fig. 4). Whether long-range order can be achieved depends on whether there is a path linking moments on either side of the sample via moments that have permanently survived or that are at least substantial enough to maintain the conduction electron polarization. At the percolation threshold, the lattice consists of long chains of interacting spins, the longest of which spans the entire sample [fat line in Fig. 4(a)]. Along this chain, the effective spatial dimensionality[18] has been reduced from $D=3$ to $D=2^-$, thereby lowering the system's dimensionality[1] to below the upper critical dimension and setting the stage[19] for E/T scaling. Our sample has ended up just below the percolation threshold, explaining why long-range order is absent and why the correlations span identical numbers of moments in all directions. Our results strongly suggest that for our system the phase diagram in Fig. 4(e) is appro-

priate, a diagram fundamentally different for the two main scenarios[9–12] outlined in the literature.

In conclusion, we have identified the ground state of $Ce(Ru_{1-x}Fe_x)_2Ge_2$, a member of a class of intensely studied 122-quantum critical systems.[2] On lowering the temperature, the local moments become increasingly more shielded by the conduction electrons. However, because of random fluctuations that change the overlap between the local electron states and the conduction electron states, some moments survive down to 0 K while others are fully shielded. The result is the random dilution of magnetic moments, leaving a percolation network. The experimental signature of such a network consists of magnetic correlations spanning equal numbers of moments in all directions. In retrospect, it could have been expected that systems prepared to have a critical degree of shielding would exhibit a local variation associated with this shielding, necessarily resulting in a dilution of the mag-

netic moments and the associated formation of a percolation network. It now remains to be established to what degree these quantum fluctuations are also present in other QCP systems and if temporal disorder plays a role in almost stoichiometric systems such as $CeNi_2Ge_2$,[20] $CeRu_2Si_2$,[21] UCu_4Pd,[4] and $CeCu_6$.[5] The fact that in URu_2Si_2 the extent (in Å) of the correlations also appears[23] to scale as c/a suggests that the latter might indeed be the case.

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