

Magnetic short-range correlations and quantum critical scattering in the non-Fermi liquid regime of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ ($x=0.2-0.6$)

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The spin dynamics of uranium ions in the non-Fermi liquid compounds $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$, for $x=0.2$ to 0.6 , have been investigated using inelastic neutron scattering. The wave vector (q) dependence of the magnetic scattering provides evidence of short-range antiferromagnetic correlations at low temperatures for $x=0.2, 0.25$, but the scattering is nearly q independent at $x=0.35, 0.6$. The magnetic response, $\bar{S}(\omega)$, obtained from the q -independent part of neutron scattering, varies as $\omega^{-\alpha}$ with a composition-dependent exponent $\alpha=0.2-0.5$. The dynamic magnetic susceptibility $\chi''(q, \omega)$ of the q -independent part exhibits ω/T scaling for the energy transfer $\hbar\omega$ between 3.5 and 17 meV in the temperature (T) range of 5–300 K at all the compositions. This scaling, which indicates local quantum criticality, breaks down in the q range, $0.6-1.1 \text{ \AA}^{-1}$ at $x=0.2$ and 0.25 , that is dominated by short-range antiferromagnetic correlations. The appearance of power laws in the magnetic response measured by inelastic neutron scattering over a wide Re doping region indicates a disorder driven non-Fermi liquid mechanism for the low-temperature physical properties in these compounds.

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I. INTRODUCTION

The order parameters in classical phase transitions, which have been investigated in great detail in many systems, are driven by thermal fluctuations. Recent theories and experiments suggest a novel phase transition that can be driven by quantum fluctuations, when a nonthermal control parameter suppresses the magnetic-ordering temperature close to absolute zero.¹⁻⁷ New theories have been proposed to describe the electronic properties including the dynamics in the critical regime that is dominated by quantum fluctuations. Hertz has theoretically shown that a quantum mechanical system with a $T=0$ K phase transition exhibits strongly coupled static and dynamic critical behavior.¹ Varma *et al.*² showed that the phase transition or singularity at $T=0$ K also gives rise to interesting scaling behaviors. Phase transitions driven by quantum fluctuations or phase transitions near $T=0$ K are called quantum phase transitions.³⁻⁵ Experiments show that several transition metal and rare-earth-based metallic magnets exhibit a magnetic instability when the long range magnetic-ordering temperature is suppressed to zero Kelvin

through variation of a nonthermal control parameter δ such as chemical substitution, pressure, or magnetic field.⁵ The low-temperature magnetic, transport, and thermal properties of such compounds significantly differ from those of simple metals and other magnetic heavy-fermion compounds, whose properties are usually understood within Landau-Fermi liquid theory.⁶ Therefore, the anomalous electronic properties at a magnetic instability have been identified as non-Fermi liquid (NFL) behavior.⁷ The phase diagram in a magnetic system at the critical value of the control parameter δ is defined as a non-Fermi liquid quantum critical point (QCP).^{3,5} The NFL behavior is thought to arise from interactions involving the strong critical order-parameter fluctuations near the QCP.

Non-Fermi liquid behavior has been observed in the normal state of high-transition-temperature superconductors, ruthenates (e.g., CaRuO_3), pseudobinary alloys, and/or stoichiometric compounds of cerium, ytterbium, and uranium.^{4,5,7-11} Because NFL behavior is now common to phase diagrams of a wide variety of strongly correlated materials, theoretical understanding of its origin has gained vital importance. Now there is a large number of metallic materi-

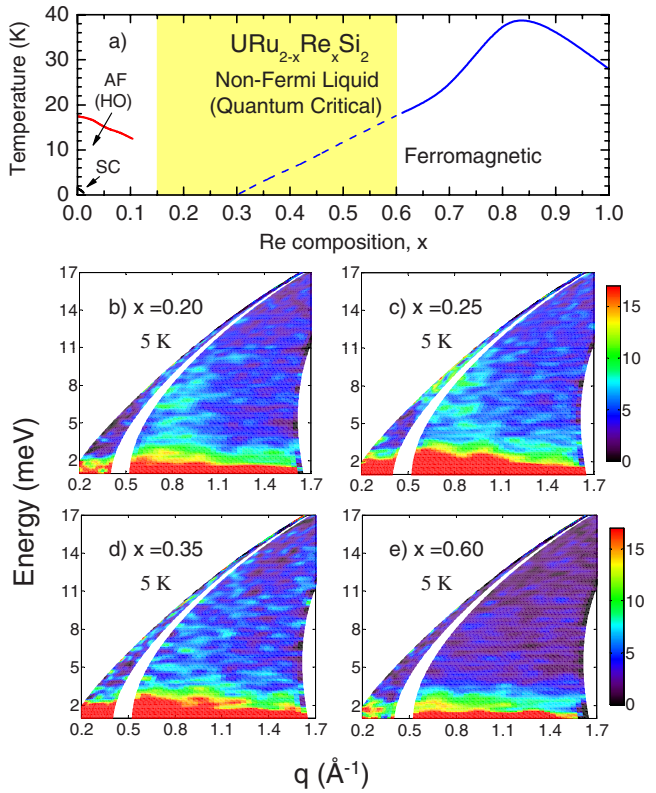


FIG. 1. (Color online) (a) Magnetic phase diagram of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$, consisting of superconducting (SC), antiferromagnetic (AF), and ferromagnetic phases. The shaded region indicates the quantum critical composition range with NFL behavior in electrical resistivity and specific heat. (b)–(e) Inelastic neutron scattering intensity map as a function of energy transfer and wave vector q , i.e., $S(q, \omega)$ (mb/meV/Str/f.u.) in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ for $x=0.2, 0.25, 0.35$, and 0.6 at 5 K . The phonon contribution, which is of the order of 10% – 20% of the total intensity in the q range of 0.5 – 1.7 \AA^{-1} , has been subtracted.

als that exhibits non-Fermi liquid behaviors in bulk transport, thermal, and magnetic measurements. Rapid progress on the theoretical front has led to several models of non-Fermi liquid behavior and different mechanisms of quantum criticality. Therefore, there is an urgent need for identifying the most suitable non-Fermi liquid model and the type of quantum criticality in a given material so that the new theories can be tested or new phenomena can be discovered. Experiments that probe the static order parameter and spin dynamical behavior in non-Fermi liquid materials, therefore, play an important role in resolving one of the key issues of quantum magnetism, i.e., the mechanisms of quantum phase transitions in materials near a magnetic instability.

Here, we focus on NFL behavior in $5f$ electron magnets. In recent work, a spectacular composition dependence of NFL scaling has been discovered in the magnetic, thermal, and transport properties of the pseudoternary compounds $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$.¹² The magnetic phase diagram of the system, presented in Fig. 1, shows that the antiferromagnetism [characterized by a small ordered U $5f$ moment and an apparent hidden order (HO) parameter] and superconductivity are rapidly suppressed with the substitution of Re for Ru.¹³ NFL

behavior, characterized by logarithmic temperature dependence of electronic specific-heat coefficient C/T and weak power laws in the temperature dependence of the electrical resistivity, appears for a wide range of Re concentrations $x=0.15$ to $x=0.6$ that includes a ferromagnetic quantum critical point at about $x_c=0.3$, beyond which ferromagnetic order develops and persists until $x\approx 1$. Alloys with x between 0.3 and 0.6 exhibit both ferromagnetic ordering and NFL behavior.¹² The low-temperature (below 5 K) dc magnetic susceptibility of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ alloys for $0.15\leq x\leq 0.3$, i.e., in the paramagnetic region of the phase diagram, also exhibits NFL behavior, characterized by power law temperature dependence $\chi_{\text{DC}}\propto T^{-n}$, where n is an exponent. The values of n vary from 0.2 at $x=0.2$ to 0.4 at $x=0.275$.¹² The exponent n cannot be reliably obtained from the above equation for $x\geq 0.3$ due to a strong contribution from the ferromagnetic phase to χ_{DC} . Fermi liquid behavior, which is observed for $x=0$ – 0.1 , is recovered at $x=0.8$.¹² The quantum critical region of the phase diagram occurs between an antiferromagnetic phase and a ferromagnetic phase. These features make $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ a special case when compared to UCu_4Pd , $\text{CeCu}_{5.9}\text{Au}_{0.1}$, and several other quantum critical magnets,^{5,14} in which the quantum critical regime appears at the boundary of a magnetically ordered phase and a paramagnetic (PM) phase.

Microscopic magnetic techniques such as nuclear magnetic resonance (NMR) and muon spin relaxation (μSR) indicate that in materials near quantum criticality, such as UCu_4Pd and $\text{CeCoGe}_{3-x}\text{Si}_x$ ($x=1.2$ and 1.5), the chemical disorder plays an important role in the electronic properties.^{15–17} Furthermore, these experiments indicate magnetic interactions and NFL mechanisms based on Kondo disorder model and Griffiths-McCoy phase.^{18–20} In the Kondo disorder model, NFL behavior is associated with the distribution of Kondo temperatures of the rare earth ions. On the other hand, in the Griffiths-McCoy phase model, the strong competition of Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction between f electrons²¹ and the Kondo interaction within a disordered and anisotropic phase leads to the formation of magnetic clusters. In this model, NFL behavior appears as a result of quantum mechanical tunneling between different excited states of magnetic clusters.

Neutron scattering experiments show that the dynamic susceptibility scales with the energy to temperature ratio, $\hbar\omega/T$, near the antiferromagnetic QCP in UCu_4Pd , $\text{CeCu}_{5.9}\text{Au}_{0.1}$, and $\text{Ce}(\text{Ru}_{1-x}\text{Fe}_x)_2\text{Ge}_2$,^{22–24} and spin-glass quantum critical point in $\text{Sc}_{1-x}\text{U}_x\text{Pd}_3$.²⁵ Given the unique features of the quantum critical region of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$, it would be desirable to know how the quantum critical behavior appears at the crossover from an antiferromagnetic (AFM) phase to a ferromagnetic (FM) phase. The increase in Ru site disorder from $x=0.2$ to 0.6 is expected to suppress antiferromagnetic correlations between uranium ions. Therefore, it is important to investigate how NFL scaling laws, particularly ω/T scaling, are affected by the proximity of two magnetic phases and increasing Ru site disorder in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ compounds. By comparing the temperature and composition dependence of the structure factor, measured by inelastic neutron scattering and by verifying the ω/T scaling at different wave vectors, energies, and alloy

compositions, we expect to learn about the origin of NFL behavior in these compounds. With this motivation, we have carried out an inelastic neutron scattering investigation of the spin dynamics of uranium ions in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ at four Re doping compositions $x=0.2, 0.25, 0.35,$ and 0.6 within the NFL regime. At all x , we observe ω/T scaling that is associated with a locally divergent magnetic response. However, there is evidence that the scaling breaks down at the wave vectors corresponding to short-range antiferromagnetic correlations, suggesting that a part of the magnetic response does not arise from quantum critical fluctuations.

This paper is organized as follows. The experimental methods are briefly described in Sec. II. Results and discussion are combined for all the alloys and presented in Sec. III. Sec. III is subdivided into two parts; wave vector dependent susceptibility is discussed first in Sec. III A followed by quantum critical scattering in Sec. III B. The conclusions of the present experimental work are presented in Sec. IV.

II. EXPERIMENT

The powder samples of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ with $x=0.2, 0.25, 0.35,$ and 0.6 and $\text{ThRu}_{1.8}\text{Re}_{0.2}\text{Si}_2$ were prepared by arc melting the constituent elements with purity better than 99.9% under argon atmosphere. Studies of $x=0.2$ and $x=0.6$ compositions were performed on samples consisting of approximately 10 g of granules roughly 2 mm in diameter, while 5 g of pellets were used in the case of $x=0.25$ and $x=0.35$. Inelastic neutron scattering measurements were performed in the temperature range 5–300 K, using the High Energy Transfer (HET) spectrometer at the ISIS pulsed neutron facility of Rutherford Appleton Laboratory, Oxfordshire, U.K. The incident energy $E_i=23$ meV of the neutrons was used with an energy resolution [full width at half maximum (FWHM)] of 1.4 meV at the elastic energy for the detector bank at 2.5 m. The phonon contribution to the scattering at low q was determined by two methods. In method 1, we used Monte Carlo simulations to calculate the low q phonon scattering based on the phonon scattering observed at high q . In method 2, we used the inelastic scattering data from the nonmagnetic reference compound $\text{ThRu}_{1.8}\text{Re}_{0.2}\text{Si}_2$ at similar temperatures. Both methods gave very similar results and were effective in separating the phonon contribution from the magnetic scattering of the $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ samples. Figure 1 shows the intensity contour maps obtained by method 1 for energy transfer (E) versus wave vector magnitude q for different x measured at 5 K. We would like to point out that the scattering intensity in the quasielastic region in the 1.5 to 2.5 meV energy range has some contribution due to multiple Bragg scattering from the sample and cryostat. These maps reveal that magnetic scattering in the $x=0.2$ and $x=0.25$ alloys is peaked in the q range of 0.6 – 1.1 \AA^{-1} . Further, the intensity contours indicate that the inelastic magnetic scattering from uranium has a strong composition dependence. The magnetic scattering is less q dependent at $x=0.35$ and becomes weaker in magnitude at $x=0.6$.

III. RESULTS AND DISCUSSION

A. Wave-vector-dependent susceptibility

The structure factor, $\bar{S}(q)$, obtained with method 1 by integrating the scattering intensity over the 5–15 meV energy

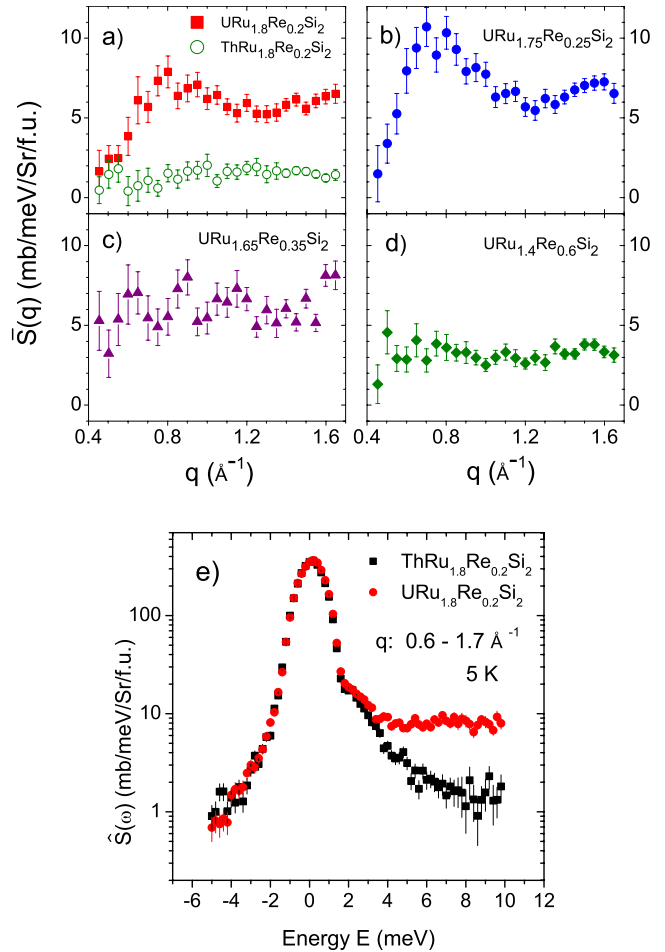


FIG. 2. (Color online) (a)-(d) Wave-vector-dependent structure factor $\bar{S}(q)$ in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ at four Re doping compositions $x=0.2, 0.25, 0.35,$ and 0.6 at 5 K. The peak at about 0.8 – 0.9 \AA^{-1} indicates short-range antiferromagnetic correlations. For a comparison, $\bar{S}(q)$ in $\text{ThRu}_{1.8}\text{Re}_{0.2}\text{Si}_2$ at 5 K is included in (a). (e) Parameter $\hat{S}(\omega)$ for the q range of 0.6 – 1.7 \AA^{-1} in $\text{URu}_{1.8}\text{Re}_{0.2}\text{Si}_2$ and $\text{ThRu}_{1.8}\text{Re}_{0.2}\text{Si}_2$, measured at 5 K, showing that the magnetic response from uranium $5f$ electrons is mainly in the inelastic scattering and the quasielastic scattering is negligible.

range, is shown for the four Re compositions in Fig. 2. A peak is observed around a q of 0.8 \AA^{-1} at $x=0.2$. This q is not the one associated with the long range antiferromagnetic order with (100) propagation vector that gives rise to a peak at $q=1.5 \text{ \AA}^{-1}$ in the parent compound URu_2Si_2 , but is the same as the q of the incommensurate propagation vector of $(0.6 \ 0 \ 0)$, where spin-wave-like excitations have been observed in URu_2Si_2 .^{26–29} A comparison of these data to those of the nonmagnetic reference compound $\text{ThRu}_{1.8}\text{Re}_{0.2}\text{Si}_2$ indicates that this peak is of magnetic origin [see Fig. 2(a)]. The existence of a peak at this value of q in the $x=0.2$ alloy is an indication of magnetic short-range correlations between uranium $5f$ electrons. This scattering peak is very broad and vanishes only above 100 K. The peak shifts to about 0.75 \AA^{-1} and becomes stronger at $x=0.25$, but is not present at $x=0.35$. Short-range correlations have also been observed in URu_2Si_2 up to 200 K.²⁶

The contribution of $5f$ electron spin fluctuations from paramagnetic uranium ions, i.e., low-energy spin fluctuations, is expected to appear as quasielastic magnetic scattering. If this contribution is present, then we can estimate the relaxation rate of $5f$ electrons, which in turn gives an idea of the time scale of spin fluctuations. We compared the quasielastic scattering in $\text{URu}_{1.8}\text{Re}_{0.2}\text{Si}_2$ with that of the nonmagnetic reference compound, $\text{ThRu}_{1.8}\text{Re}_{0.2}\text{Si}_2$, at different q values in the range of 0.6 to 1.7 \AA^{-1} . We have added the structure factors $S(q, \omega)$ at each of the measured q values in the q range of 0.6 to 1.7 \AA^{-1} to obtain a scattering parameter defined as $\hat{S}(\omega)$. Although this parameter may not be identical to the structure factor, it serves the purpose of comparing relative intensity of quasielastic scattering contributions in the two alloys because the scattering from both the alloys was measured under identical experimental conditions. Figure 2(e) compares the parameter $\hat{S}(\omega)$ in uranium and thorium alloys at $x=0.2$. We find that the quasielastic scattering contribution, below 2 meV, is almost the same in both the compounds, which means that there is no detectable magnetic contribution, within the resolution of the instrument from uranium $5f$ electrons to the quasielastic scattering, suggesting that paramagnetic spin fluctuations are either very weak or outside the region probed by quasielastic neutron scattering with $E_i=23$ meV on the HET spectrometer. This is understandable as most of the uranium ions tend to participate in antiferromagnetic or ferromagnetic type short-range magnetic order in the non-Fermi liquid regime.

The peak in the magnetic scattering has a strong temperature dependence between 5 and 300 K at $x=0.2$. To get an insight into the nature of magnetic correlations, we have converted the temperature-dependent magnetic structure factor $S(q, \omega)$ (obtained with method 1) to $\chi''(q, \omega)$ using the relation $\chi''(q, \omega)=[1-\exp(-\hbar\omega/k_B T)] S(q, \omega)$. Then the wave-vector-dependent susceptibility $\chi'(q)$ was obtained from Kramers-Kronig relations³⁰ by integrating the magnetic scattering intensity between 5 and 20 meV after correcting for the uranium (U^{4+}) magnetic form factor.^{26,31} The $4+$ valence state of uranium has been suggested from inelastic neutron scattering measurements in the parent compound, URu_2Si_2 .³² The valence state of uranium is also expected to be close to $4+$ in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ alloys. We choose the low energy cutoff at 5 meV to remove the contribution from nonmagnetic scattering. The susceptibility $\chi'(q, T)$, extracted from $S(q, \omega)$ at each temperature, is presented in Fig. 3, together with the best fit using a liquidlike structure factor used in the analysis of UCu_4Pd .^{33,34}

$$\chi'(q, T) = \chi'_{\text{loc}}(T) \left[1 + \sum_{i=1}^4 a_i(T) \sin(qr_i)/qr_i \right]. \quad (1)$$

The prefactor $\chi'_{\text{loc}}(T)$ represents the local magnetic susceptibility, which dominates the magnetic response at higher q . $\chi'(q, T)$ is modulated by the interionic exchange interactions, which are parameterized by $a_i=(S_0 \cdot S_i)$ ($i=1-4$), the expectation values of spin-spin interactions at U-U interatomic distances r_i ; $r_1=4.414$ \AA , $r_2=5.616$ \AA , $r_3=5.857$ \AA , and $r_4=8.113$ \AA . The magnitudes and the negative signs of a_1 and a_2 indicate that the nearest-neighbor exchange interaction is marginally antiferromagnetic and the next-nearest-neighbor

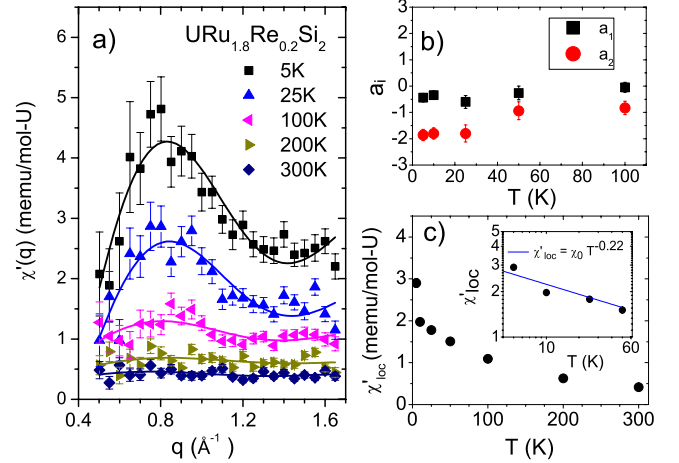


FIG. 3. (Color online) (a) Temperature and q dependence of χ' showing short-range magnetic order in $\text{URu}_{1.8}\text{Re}_{0.2}\text{Si}_2$. The solid lines are best fits by the liquid type structure factor, i.e., Eq. (1), described in the text. (b) The expectation values a_i for the first and the second near-neighbor U-U interatomic distances at $x=0.2$ as a function of temperature. (c) Temperature dependence of the χ'_{loc} extracted from Eq. (1) at $x=0.2$. Inset shows χ'_{loc} vs T in a log-log plot.

interaction is antiferromagnetic. The weakness of the nearest-neighbor exchange could result from a competition of ferromagnetic and antiferromagnetic interactions produced by anisotropic exchange. This is quite different from URu_2Si_2 , where the nearest-neighbor exchange interaction is ferromagnetic within the a - b plane in the antiferromagnetic structure with the (100) propagation vector.²⁶ Figure 3 also shows the temperature dependence of a_i and $\chi'_{\text{loc}}(T)$. $\chi'_{\text{loc}}(T)$ between 5 and 50 K could be fitted by a power law: $\chi'_{\text{loc}}(T) = \chi_0 T^{-\alpha}$, where $\chi_0 = 3.4(3) \cdot 10^{-3}$ emu/mol-U and exponent $\alpha = 0.22(2)$ [inset in Fig. 3(c)].

B. Quantum critical scattering

A further analysis of the magnetic scattering as a function of energy transfer gives an insight into the scaling laws and hence, the nature of the quantum phase transition. NFL scaling laws can be verified by investigating the energy ($\hbar\omega$) dependence of the dynamic structure factor $S(q, \omega)$. This can be done by fitting the individual constant q scans that can be generated from the contour plots to the expected form of $S(q, \omega)$. We have analyzed such constant q scans at different q values and found that there are two types of behaviors of the structure factor depending on the range of the q values: (i) An incoherent scattering region in which the structure factor is practically independent of q and (ii) a coherent scattering region in which the structure factor strongly depends on q (in the range of 0.6–1.1 \AA^{-1}) due to short-range antiferromagnetic correlations. We choose the q range that excludes short-range antiferromagnetic correlations, i.e., $q = 1.2-1.7$ \AA^{-1} at $x=(0.2, 0.25)$ and $q = 0.6-1.7$ \AA^{-1} at $x=(0.35, 0.6)$ for the analysis of structure factor in case (i). The scattering intensity is practically independent of q in this region. We have summed the scattering intensity over this q

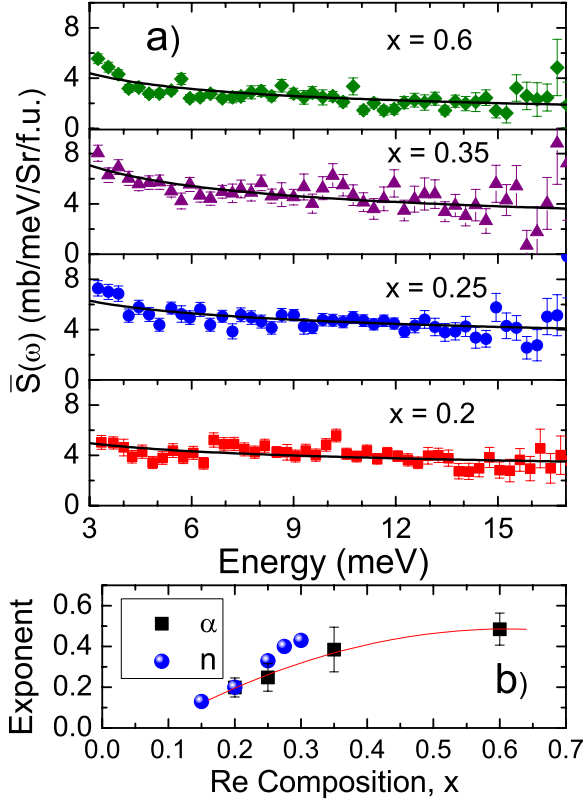


FIG. 4. (Color online) (a) Structure factor $\bar{S}(\omega)$ versus energy plots as a function of x in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$. The solid lines are best fits described in the text. (b) Composition dependence of exponents α from neutron scattering and n from dc susceptibility (Ref. 12).

range with equal weights and obtained a summed structure factor, denoted as $\bar{S}(\omega)$. Figure 4(a) shows a plot of $\bar{S}(\omega)$ at 5 K as a function of energy transfer $\hbar\omega$ for all x in the 3.5 to 17 meV energy range, together with the best fit by the scaling law:

$$\bar{S}(\omega) = S_0/(\hbar\omega)^\alpha, \quad (2)$$

where S_0 is a proportionality constant and α is the dynamic scaling exponent. α is found to be about 0.2 at $x=0.2$, 0.25 at $x=0.25$, 0.38 at $x=0.35$, and 0.5 at $x=0.6$, indicating a dependence on the Re composition of the alloy. These α values are comparable to the dc susceptibility exponent, showing that $\alpha \approx n$ for $x < 0.3$. The Kramers-Kronig relations, which can be used to relate $\chi''(q, \omega)$ measured by neutron scattering to the dc susceptibility $\chi'(q \rightarrow 0)$, predicts the equivalence of α and n if the magnetic response is q independent. Further, hyperscaling also implies $\alpha = n$.³⁵ The fractional values of the exponent α indicate that the quantum criticality in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ is driven by the critical fluctuations of U $5f$ local moments.³⁶⁻³⁹ The appearance of NFL behavior in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ for a wide range of Re doping concentrations, which also includes a ferromagnetic region, can be explained within the Griffiths-McCoy model, which predicts power-law behaviors of magnetic susceptibility at low temperatures on both sides of a ferromagnetic quantum critical point.¹⁸ The Ru site disorder plays a role as it modifies the crystal

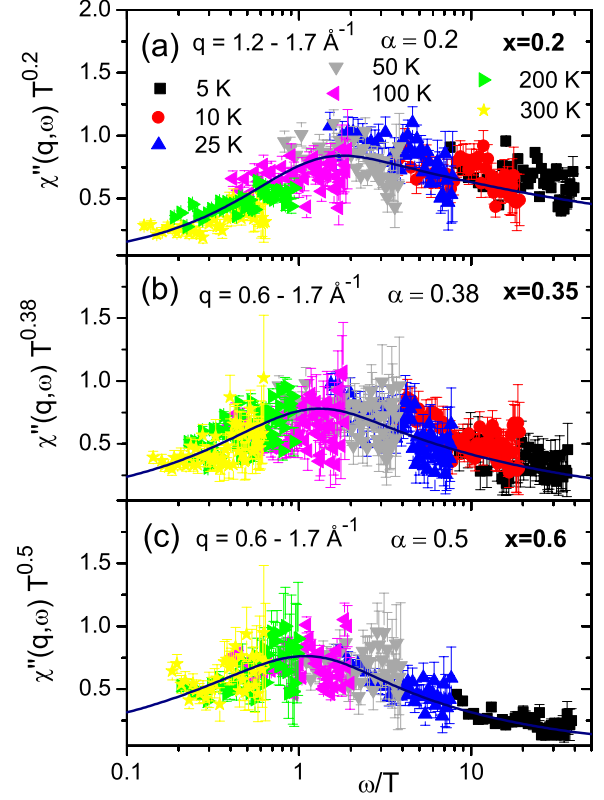


FIG. 5. (Color online) Semi-log plot of $\chi''(q, \omega) T^\alpha$ versus ω/T , showing the ω/T scaling at (a) $x=0.2$, (b) $x=0.35$, and (c) at $x=0.6$. The solid line is a fit by Eq. (3).

potential and increases the randomness in the intersite antiferromagnetic exchange interactions. The competition of antiferromagnetic exchange interaction and Kondo interaction in a site-disordered alloy system seems to help the formation of the Griffiths-McCoy phase, similar to the case in some other quantum critical rare earth compounds, such as $\text{CeCoGe}_{1.8}\text{Si}_{1.2}$.¹⁷ The exponent values from both bulk magnetic measurements¹² and our neutron scattering measurements support the disorder driven mechanism of non-Fermi liquid behavior within the Griffiths-McCoy model.

Theoretically, ω/T scaling is expected in the dynamic behavior of a system when there is a singularity at $T=0$ K.² This can be verified by plotting the energy dependence of $\chi''(q, \omega)$ at different temperatures. We have searched for such a scaling at three Re doping compositions: (i) at $x=0.2$, where antiferromagnetic correlations are dominant, (ii) at $x=0.35$, which is close to a QCP, and (ii) at $x=0.6$, where NFL behavior occurs in the ferromagnetic phase, by measuring $S(q, \omega)$ at six or seven temperatures in the range of 5 to 300 K. The scattering in the neutron energy loss side at all q for $x=0.35, 0.6$, and at high q for $x=0.2$, is found to exhibit ω/T scaling. The ω/T scaling also implies that $\chi''(q, \omega)$ measured at different temperatures can be collapsed onto a single universal curve defined by the relation²²

$$\chi''(q, \omega) T^\alpha = P(T/\omega)^\alpha \tanh(\omega/\beta T), \quad (3)$$

where P is a proportionality constant and β is the scale factor. We find that $\beta \approx 1$. Figure 5 [panels (a) to (c)] shows the

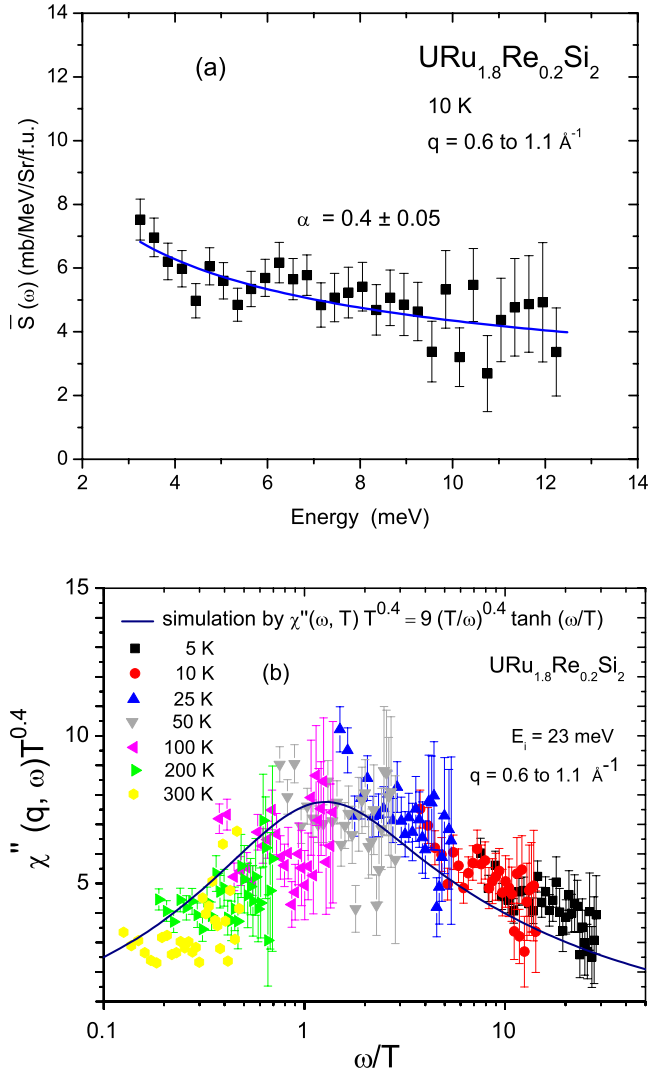


FIG. 6. (Color online) (a) Structure factor $\bar{S}(\omega)$ versus energy plot in $\text{URu}_{1.8}\text{Re}_{0.2}\text{Si}_2$ for the coherent part of the scattering, i.e., q covering the short-range order peak. The solid line is the best fit by Eq. (2). (b) Semi-log plot of $\chi''T^\alpha$ versus ω/T for the q range covering the short-range order peak in $x=0.2$ alloy, showing that ω/T scaling breaks down when the coherent part of the scattering is considered. The solid line is a simulation by the equation given in the figure with exponent $\alpha=0.4$.

plot of $\chi''(q, \omega)T^\alpha$ after normalizing with the scale factor P (5 at $x=0.2$, 10 at $x=0.35$, and 6.9 at $x=0.6$) as a function of ω/T for q ranges; (a) $x=0.2$ for $1.2 < q < 1.7 \text{ \AA}^{-1}$, which excludes the q range of antiferromagnetic short-range correlations, (b) $x=0.35$ for $0.6 < q < 1.7 \text{ \AA}^{-1}$ and (c) $x=0.6$ for $0.6 < q < 1.7 \text{ \AA}^{-1}$.

To analyze the structure factor for case (ii), we have chosen the q range that covers short-range antiferromagnetic correlations, i.e., $q=0.6-1.1 \text{ \AA}^{-1}$ at $x=0.2$. In this q range, the best fit to $\bar{S}(\omega)$ using Eq. (2), shown in Fig. 6(a), yielded the exponent $\alpha=0.4 \pm 0.05$. This value is twice as large as the value of α found in region (i). We used this exponent to plot $\chi''(q, \omega)T^\alpha$ as a function of ω/T in Fig. 6. We find that Eq. (3) does not yield a good fit to the experimental data. A simulation using Eq. (3) is plotted over Fig. 6 to compare the

data and the fit. Therefore, ω/T scaling defined by Eq. (3) is not clearly observed in the $x=0.2$ alloy, when the q range of $0.6-1.1 \text{ \AA}^{-1}$ is chosen; i.e., when the scattering is dominated by short-range antiferromagnetic correlations.

These results indicate that the NFL behavior in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ is associated with the ω/T scaling of the local (q -independent) susceptibility at all values of x , including, remarkably, the ferromagnetic phase at $x=0.6$. This is consistent with recent theoretical work, which shows that quantum critical fluctuations are predominantly local in character.^{36,37,39} However, the scaling breaks down in the q range, where the short-range antiferromagnetic correlations make a significant contribution to the magnetic response. Our results contrast with UCu_4Pd , in which the ω/T scaling is observed over the entire q range.³⁴ This suggests an important difference between quantum critical points at an AFM-FM boundary and an AFM-PM boundary. Whereas the entire magnetic response displays quantum critical scaling close to an AFM-PM transition, a fraction of the magnetic response is apparently not quantum critical close to an AFM-FM transition. This could be a signature of critical fluctuations near a quantum critical point when there are two competing order parameters.

Finally, we would like to note about the possible role of ferromagnetic correlations in the non-Fermi liquid region that coexists with ferromagnetic order. The $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ compounds with $x=0.35$ and $x=0.6$ are ferromagnetic at low temperatures. It is likely that ferromagnetic contribution to the scattering exists in these compounds at q values close to 0 \AA^{-1} . Therefore, the low energy fluctuations or low q fluctuations that have not been covered in our neutron scattering measurements may also be important. If these correlations are stronger, they may reveal another mechanism of non-Fermi liquid behavior at these two compositions. A ferromagnetic component in the scattering at such a low q region is hypothetically possible in the alloys with $x=0.2$ and $x=0.25$; however it is less likely because antiferromagnetic short-range correlations are relatively stronger at these compositions. Because our inelastic scattering experiments did not cover the lower q range between $0-0.2 \text{ \AA}^{-1}$, we cannot exclude the role of a ferromagnetic quantum critical point in the ω/T scaling observed in the wide Re doping composition range. Further investigations of these compounds using small-angle neutron scattering and inelastic neutron scattering in the low q region ($0-0.2 \text{ \AA}^{-1}$) at finite energy transfer would be desirable to find out if there is an additional mechanism for the approach to the quantum critical point in this system.

IV. CONCLUSION

In the present work, we have investigated the spin dynamics of $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$ ($x=0.2, 0.25, 0.3, \text{ and } 0.6$) compounds using inelastic neutron scattering. We observe NFL behavior associated with quantum critical scattering in both the antiferromagnetic and ferromagnetic sides of the quantum critical point ($x_c=0.3$) in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$. The divergent magnetic response that gives rise to the ω/T scaling over a wide Re doping region is associated with local q -independent fluctua-

tions, but there is evidence of a breakdown of quantum critical scaling at certain wave vectors where the antiferromagnetic correlations are dominant. These observations indicate that there is a common mechanism for non-Fermi liquid behavior in this system and the non-Fermi liquid scaling of the structure factor originates from q -independent (incoherent) inelastic neutron scattering. The composition dependence of the exponents can be qualitatively understood by considering the evolution of magnetic correlations from antiferromagnetic to ferromagnetic nature with the increase in Re doping. The Ru site disorder plays a role as it modifies the crystal potential and increases the randomness in the intersite antiferromagnetic exchange interactions. The power law scaling observed in neutron scattering and non-Fermi liquid behavior observed in the bulk physical properties indicate the disorder driven mechanism of non-Fermi liquid behavior within the Griffiths-McCoy model.

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- ¹J. A. Hertz, Phys. Rev. B **14**, 1165 (1976).
²C. M. Varma, P. B. Littlewood, S. Schmitt-Rink, E. Abrahams, and A. E. Ruckenstein, Phys. Rev. Lett. **63**, 1996 (1989).
³A. J. Millis, Phys. Rev. B **48**, 7183 (1993).
⁴S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, 1999).
⁵For a review, see G. R. Stewart, Rev. Mod. Phys. **73**, 797 (2001); P. Coleman, Ann. Henri Poincaré **4**, 1 (2003).
⁶L. D. Landau, Zh. Eksp. Teor. Fiz. **30**, 1058 (1956) [Sov. Phys. JETP **3**, 920 (1957)].
⁷M. B. Maple, C. L. Seaman, D. A. Gajewski, Y. Dalichaouch, V. B. Barbeta, M. C. de Andrade, H. A. Mook, H. G. Lukefahr, O. O. Bernal, and D. E. Maclaughlin, J. Low Temp. Phys. **95**, 225 (1994).
⁸L. Klein, L. Antognazza, T. H. Geballe, M. R. Beasley, and A. Kapitulnik, Phys. Rev. B **60**, 1448 (1999).
⁹J. Orenstein and A. J. Millis, Science **288**, 468 (2000).
¹⁰C. M. Varma, Z. Nussinov, and Wim van Saarloos, Phys. Rep. **361**, 267 (2002).
¹¹H. v. Löhneysen, A. Rosch, M. Vojta, and P. Wölfle, Rev. Mod. Phys. **79**, 1015 (2007).
¹²E. D. Bauer, V. S. Zapf, P.-C. Ho, N. P. Butch, E. J. Freeman, C. Sirvent, and M. B. Maple, Phys. Rev. Lett. **94**, 046401 (2005).
¹³Y. Dalichaouch, M. B. Maple, M. S. Torikachvili, and A. L. Giorgi, Phys. Rev. B **39**, 2423 (1989).
¹⁴O. Stockert, H. v. Löhneysen, A. Rosch, N. Pyka, and M. Loewenhaupt, Phys. Rev. Lett. **80**, 5627 (1998).
¹⁵O. O. Bernal, D. E. MacLaughlin, H. G. Lukefahr, and B. Andraka, Phys. Rev. Lett. **75**, 2023 (1995).
¹⁶O. O. Bernal, D. E. MacLaughlin, A. Amato, R. Feyerherm, F. N. Gygax, A. Schenck, R. H. Heffner, L. P. Le, G. J. Nieuwenhuys, B. Andraka, H. v. Löhneysen, O. Stockert, and H. R. Ott, Phys. Rev. B **54**, 13000 (1996).
¹⁷V. V. Krishnamurthy, K. Nagamine, I. Watanabe, K. Nishiyama, S. Ohira, M. Ishikawa, D. H. Eom, T. Ishikawa, and T. M. Briere, Phys. Rev. Lett. **88**, 046402 (2002).
¹⁸R. B. Griffiths, Phys. Rev. Lett. **23**, 17 (1969).
¹⁹E. Miranda, V. Dobrosavljevic, and G. Kotliar, Phys. Rev. Lett. **78**, 290 (1997).
²⁰A. H. Castro Neto, G. Castilla, and B. A. Jones, Phys. Rev. Lett. **81**, 3531 (1998).
²¹M. A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954); T. Kasuya, Prog. Theor. Phys. **16**, 45 (1956); K. Yosida, Phys. Rev. **106**, 893 (1957).
²²M. C. Aronson, R. Osborn, R. A. Robinson, J. W. Lynn, R. Chau, C. L. Seaman, and M. B. Maple, Phys. Rev. Lett. **75**, 725 (1995).
²³A. Schroder, G. Aeppli, E. Bucher, R. Ramazashvili, and P. Coleman, Phys. Rev. Lett. **80**, 5623 (1998).
²⁴W. Montfrooij, M. C. Aronson, B. D. Rainford, J. A. Mydosh, A. P. Murani, P. Haen, and T. Fukuhara, Phys. Rev. Lett. **91**, 087202 (2003).
²⁵S. D. Wilson, P. C. Dai, D. T. Adroja, S. H. Lee, J. H. Chung, J. W. Lynn, N. P. Butch, and M. B. Maple, Phys. Rev. Lett. **94**, 056402 (2005).
²⁶C. Broholm, J. K. Kjems, W. J. L. Buyers, P. Matthews, T. T. M. Palstra, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. Lett. **58**, 1467 (1987); C. Broholm, H. Lin, P. T. Matthews, T. E. Mason, W. J. L. Buyers, M. F. Collins, A. A. Menovsky, J. A. Mydosh, and J. K. Kjems, Phys. Rev. B **43**, 12809 (1991).
²⁷T. E. Mason, B. D. Gaulin, J. D. Garrett, Z. Tun, W. J. L. Buyers, and E. D. Isaacs, Phys. Rev. Lett. **65**, 3189 (1990).
²⁸F. Bourdarot, B. Fåk, K. Habicht, and K. Prokes, Phys. Rev. Lett. **90**, 067203 (2003).
²⁹C. R. Wiebe, J. A. Janik, G. J. MacDougall, G. M. Luke, J. D. Garrett, H. D. Zhou, Y.-J. Yo, L. Balicas, Y. Qiu, J. R. D. Copley, Z. Yamani, and W. J. L. Buyers, Nat. Phys. **3**, 96 (2007).
³⁰A. P. Murani, A. Severing, and W. G. Marshall, Phys. Rev. B **53**, 2641 (1996).
³¹J. Faber, Jr. and G. H. Lander, Phys. Rev. B **14**, 1151 (1976).
³²J. G. Park, K. A. McEwen, and M. J. Bull, Phys. Rev. B **66**, 094502 (2002).
³³B. D. Gaulin, J. S. Gardner, S. R. Dunsiger, Z. Tun, M. D. Lumsden, R. F. Kiefl, N. P. Raju, J. N. Reimers, and J. E. Greedan, Physica B **241-243**, 511 (1997).
³⁴M. C. Aronson, R. Osborn, R. Chau, M. B. Maple, B. D. Rain-

- ford, and A. P. Murani, Phys. Rev. Lett. **87**, 197205 (2001).
- ³⁵M. Vojta, N.-H. Tong, and R. Bulla, Phys. Rev. Lett. **94**, 070604 (2005).
- ³⁶P. Coleman, Physica B **259-261**, 353 (1999); P. Coleman, C. Pepin, Q. Si, and R. Ramazashvili, J. Phys.: Condens. Matter **13**, R723 (2001).
- ³⁷Q. Si, S. Rabello, K. Ingersent, and J. L. Smith, Nature (London) **413**, 804 (2001); Q. Si, S. Rabello, K. Ingersent, and J. L. Smith, Phys. Rev. B **68**, 115103 (2003).
- ³⁸D. R. Grempel and Qimiao Si, Phys. Rev. Lett. **91**, 026401 (2003).
- ³⁹Qimiao Si, Physica B Physica B **378-380**, 23 (2006).