The extent of quantum critical fluctuations into the heavy-fermion phase: A neutron scattering study of $Ce(Ru_{1-x}Fe_x)_2Ge_2$

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We present inelastic neutron scattering measurements on the heavy-fermion compound $Ce(Ru_{1-x}Fe_x)_2Ge_2$ ($x=0.87$). This composition is close to the quantum critical point (QCP) compound ($x=0.76$) in which it was observed that the decay of spin fluctuations could no longer be described by a simple exponential in time. Here we show that the relaxation mechanism for spin fluctuations exhibits a qualitatively similar momentum and temperature dependence in both compositions, possibly reflecting the underlying scaling law at the QCP. However, the details of the relaxation mechanism demonstrate that critical fluctuations are strongly damped along the composition axis. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1850338]

Metallic compounds that have been prepared to be on the verge of ordering magnetically at $T=0$ K, the quantum critical point (QCP), have been the subject of intense study during the past decade.[1,2] There is an ever growing list[1] of systems that show a departure from the predictions of Fermi-liquid theory regarding the low temperature behavior of specific heat, resistivity, and static susceptibility. Neutron scattering studies probing the microscopic origins of this non-Fermi-liquid (nFl) behavior revealed that some of these systems exhibit scaling:[3–6] the dynamic response of the system only depends on the ratio E/T of the energy E used to probe the system, and its temperature T.

These scaling relationships have led to the development of the local moment scenario[4,7,8] to describe the ordering process that takes place at the QCP. Even though experiments[6] on $Ce(Ru_{0.24}Fe_{0.76})_2Ge_2$ have since shown that this scenario is not borne out in all quantum critical systems, it captures many aspects of the response observed at the QCP. Si *et al.*[8] have proposed that the QCP is the point where local moments first remain present down to $T=0$ K because the magnetic fields associated with the fluctuations of the order parameter successfully impede the Kondo screening of the moments. Any change in interaction between the local moments and conduction electrons would either result in an ordered phase at finite temperatures, or in a nonmagnetic heavy-fermion phase. The response of the QCP-system to an external perturbation is captured in terms of a generalized Curie-Weiss law[4,8] in which E, T, magnetic field H are treated on equal footing with the momentum dependent Weiss temperature (as first observed in $CeCu_{5.9}Au_{0.1}$[4]). This scenario automatically leads to an accompanying divergence of the local susceptibility [observed in $CeCu_{5.9}Au_{0.1}$,[4] but not in $Ce(Ru_{0.24}Fe_{0.76})_2Ge_2$[6]] and to the appearance of E/T scaling laws (observed in multiple systems[3–6]). The local moment scenario should be contrasted to the spin density wave (SDW) scenario[9,10] where ordering is driven by an intrinsic instability of the electron fluid against electron–electron interactions, a scenario that does not allow for E/T scaling relationships to develop.

Recent experiments on $Ce(Ru_{1-x}Fe_x)_2Ge_2$ demonstrated[6] that QCP-systems exist in which the response shows characteristics of both scenarios. $Ce(Ru_{1-x}Fe_x)_2Ge_2$, prepared to be at the QCP ($x=0.76$), has been shown to be a unique system that evolves towards an incommensurate ordered phase in a manner that cannot be described by a single scenario. The dynamic response of this system is characterized by a modi-

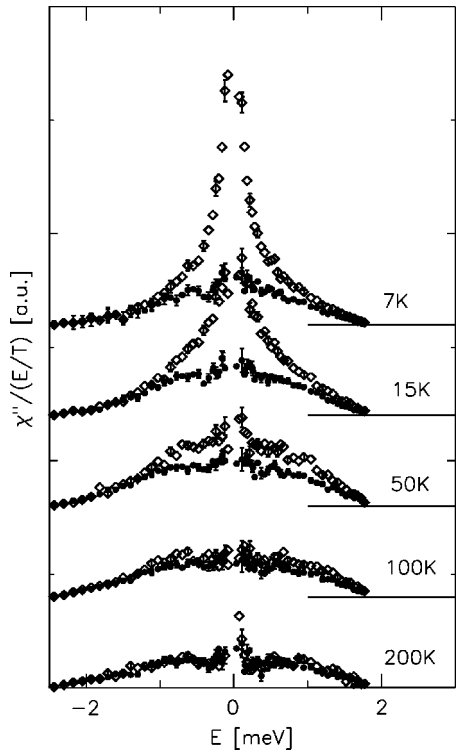

FIG. 1. Symmetrized imaginary part of the dynamic susceptibility $\chi''(q,E,T)/(E/T)$ for $q=0.4$ Å^{-1} for $Ce(Ru_{1-x}Fe_x)_2Ge_2$ ($x=0.76$ open symbols; $x=0.87$ filled circles). The various curves are offset for clarity, with zero intensity given by the horizontal lines on the right-hand side of the figure. The response for both concentrations is identical at $T=200$ K, but on cooling down the onset of critical fluctuations is clearly seen for the critical concentration.

fied Lorentzian line shape in energy and it exhibits E/T scaling, neither of which is predicted by the SDW scenario.[9,10] In addition, the local susceptibility was found to saturate[6] on approaching the QCP, while the exponents describing the dynamic and static evolution differed from each other at low temperatures, neither observation being consistent[8] with the local moment scenario. Instead, a change in intermoment coupling at the lowest temperatures appeared to drive the system to an ordered phase, somewhat similar to the changes in interaction strengths observed near the order–disorder phase transition in U_2Zn_{17}.[11]

Here we report on the dynamic response of $Ce(Ru_{1-x}Fe_x)_2Ge_2$ when prepared to be on the heavy-fermion (HF) side of the QCP. Comparison of the HF-response to the response at the QCP probes the role that concentration fluctuations play in the approach to ordering. Both the QCP sample and the HF sample were prepared using identical methods, measured under identical conditions and analyzed using identical data reduction procedures. Details are given elsewhere.[6] Microprobe measurements verified that the ~30 g polycrystalline samples displayed good homogeneity ($x=0.76\pm0.02$, and $x=0.87\pm0.02$). The neutron scattering experiments were carried out at the IN6 time-of-flight (TOF) spectrometer at the Institute Laue-Langevin. IN6 was operated with incident neutron wavelength λ of 5.12 Å, yielding an energy resolution of 0.1 meV (full width at half maximum). As an example of the fully corrected data, we show in Fig. 1 the magnetic scattering for both samples at neutron

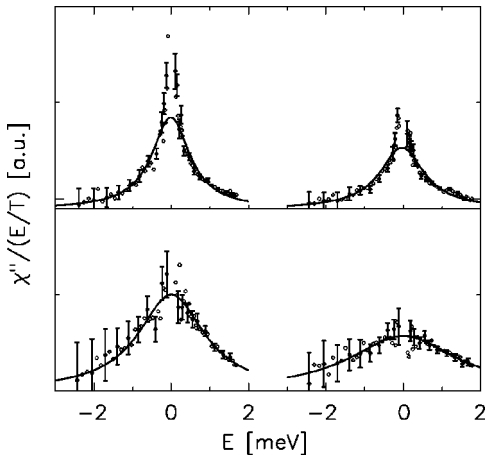

FIG. 2. Lorentzian line fits to the symmetrized dynamic susceptibility for $q=0.5$ Å^{-1} (left half of figure) and $q=1.4$ Å^{-1} (right) for the critical composition (top half), and for the HF composition (bottom half), both at 7 K. In contrast to the critical composition, no deviations from Lorentzian line shapes are observed in the HF compound; panel bottom halves do not share the same intensity scale.

momentum transfer $q=0.4$ Å^{-1} as a function of temperature T and neutron energy transfer E.

At $T=200$ K both compositions have the (expected) identical response of a local moment system with an unpaired cerium f electron: a broad distribution in E (Fig. 1) and q (not shown), reflecting the paramagnetic response of rapidly fluctuating magnetic moments. However, on lowering the temperature, marked differences in the magnetic response are observed between the two compositions (see Fig. 1): the onset of critical fluctuations is clearly observed for the critical composition[6] at $q=0.4$ Å^{-1}. Altogether, the fact that the observed response is so distinctly different between the two samples that were prepared using identical methods and that were of comparable homogeneity, rules out the possibility that the nFl response and unusual scaling behavior observed[6] in the sample at the QCP results from the polycrystallinity of the compound, from compositional inhomogeneities or from any other experimental artifact. It is especially important to rule out the possibility of compositional inhomogeneities contributing to a deviation from standard response in systems that are driven to a QCP by means of chemical[4,6] substitution.

In stark contrast to the sample prepared to be at the critical composition, we observed a Lorentzian line shape in the HF composition (Fig. 2) for all T and q values. This implies that in the HF-compound spin fluctuations decay according to a simple exponential in time. The spectral line width for the HF composition follows a linear T dependence [Fig. 3(a)], akin to the critical composition.[6] Even though the $T=0$ K value of the line width of the HF-composition greatly exceeds that of the critical composition [see Fig. 3(b)], the line width for both compositions shows a qualitatively similar q dependence, dropping sharply with decreasing q for $q<0.8$ Å^{-1}. This strongly suggests that in both compositions the decay mechanism of spin fluctuations is similar, and that the lifetimes of these fluctuations are solely determined by their distance from the QCP. This distance can be in temperature, wave number, energy transfer, or even composition. The

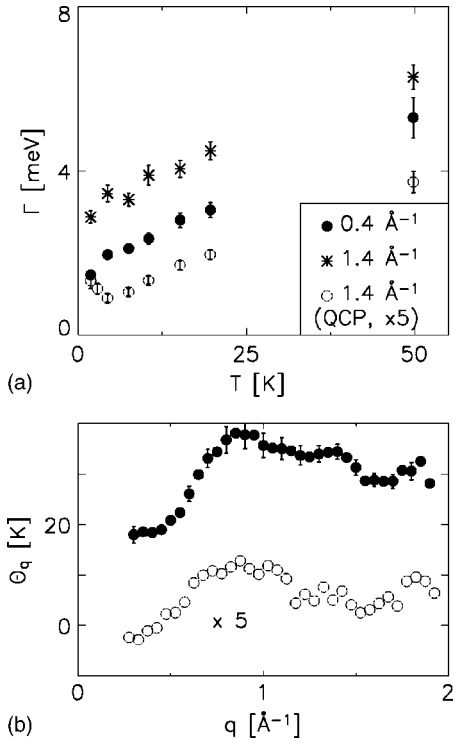

FIG. 3. Temperature dependence of the Lorentzian line width $\Gamma(T)$ for the HF sample (a). The line width follows a linear temperature dependence below 50 K, similar to the temperature dependence of the line width for the critical composition (Ref. 6) (open circles, non-Lorentzian line width multiplied by 5). We show the residual line width Θ_q obtained from a fit to $\Gamma(T)=\Theta_q+aT$ in part (b) (solid circles). The results for the critical composition are shown as open circles, multiplied by five. Note that both samples show a similar q dependence, suggesting that Θ_q can be interpreted as the distance to the QCP in q space.

fact that the line width is linear in temperature for both compositions, combined with the observed robustness of the q dependence, indicates that the generalized[4,8,12] Curie-Weiss law for the magnetic fluctuations observed in quantum critical[6] $Ce(Ru_{1-x}Fe_x)_2Ge_2$, in which temperature, momentum, and energy are treated on equal footing, can now be further generalized to also include composition.

In conclusion, we have shown that the two local moment systems $Ce(Ru_{1-x}Fe_x)_2Ge_2$ ($x=0.76$ and 0.87), which are indistinguishable at high temperatures, show a radically different response on cooling. The sample that is compositionally at the QCP displays critical fluctuations that cannot be described by standard theory, while the much weaker fluctuations in the heavy-fermion sample do not require a modified Lorentzian line shape. Thus, the critical fluctuations do not extend very far along the chemical composition axis. In both compositions, the lifetime of the spin fluctuations follows a linear temperature dependence and a qualitatively similar momentum dependence, indicating that the relaxation mechanism for these fluctuations is very similar for both compositions and that the decay rate is determined by the distance from the QCP in temperature, momentum, and composition. However, close to the QCP, spin fluctuations no longer decay according to a simple exponential in time, reflecting the dual role[13] of the conduction electrons in screening the local moments and in furnishing the intermoment coupling.

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