Neutron Scattering Study of $URu_{2-x}Re_xSi_2$ with x=0.10: Driving Hidden Order Towards Quantum Criticality

T.J. Williams[1], G.M. Luke[1], W.J.L. Buyers[2], Z. Yamani[2], M.B. Maple[3], N. Butch[3]

[1] McMaster University, Hamilton, ON, Canada

[2] National Research Council, Canadian Neutron Beam Centre, Chalk River, ON, Canada

[3] University of California San Diego, La Jolla, CA, USA

In the field of strongly correlated heavy fermion systems, whose giant specific heat suggests massive electrons, one of the most puzzling long-standing issues is the nature of the hidden order below the large specific heat jump at T_0= 17 K in URu_2Si_2 [1,2]. A superconducting phase follows below 1.2 K. Despite much research [3-5] the order parameter remains unknown. The small antiferromagnetic moment of 0.03 μ_B that develops below 17 K cannot explain the large specific heat jump at this second order phase transition. Antiferromagnetism therefore cannot be the hidden order parameter. The system appears to have condensed into a new phase of matter for which the order parameter and associated symmetries differ from conventional expectations. In previous work we eliminated crystal fields and orbital currents as a source of hidden order [6]. We later found that the spins fluctuate above the 17 K transition in an incommensurate way centred on the wave vector (1 ± δ, 0, 0) with δ = 0.4 [7]. Emerging from that wave vector is a high-velocity cone of strongly damped gapless excitations that extend over a finite region of the Brillouin zone. In the precursor phase to hidden order we could then calculate that these gapless spin fluctuations give rise to a term in the specific heat that closely accounts for the magnitude of the giant specific heat linear in T and previously attributed to electrons only [7]. The specific heat jump at T_0 and its decrease below are now understood to arise from the formation of a spin gap below T_0. More recently the dynamic incommensurate spins have been interpreted as the response of an itinerant rather than local spin system and a connection made to a Fermi surface nesting with some similarities to that of chromium [8]. Nonetheless the symmetry of the actual order parameter that condenses remains hidden and unknown.

Another route to discovering the hidden order symmetry is to move away from the object of interest by applying pressure or chemical pressure by doping. External pressure has been recently found to cause commensurate condensation of a large antiferromagnetic moment of 0.3 μ_B and the collapse of the strong commensurate spin excitation [9]. The hidden order phase is lost. On the contrary the hidden order phase can be retained by applying negative chemical pressure. This is done by expanding the lattice constants by alloying with rhenium as discovered in the pioneering work of Dalichaouch *et al.*, [10] Bauer *et al.*, [11] have shown that Re replacement of Ru atoms in $URu_{2-x}Re_xSi_2$, reduces the hidden order transition from 17.5 K to 13 K for x=0.10. It also reduces the superconducting transition from 1.5 K to 0.23 K at x=0.01. Interestingly this represents a first step on the way to reaching a quantum critical point at where the hidden order gives over to ferromagnetism. The system exhibits non-Fermi-liquid behaviour for 0.15<x<0.6. The magnetic phase diagram of $URu_{2-x}Re_xSi_2$ as a function of doping level is shown in Figure 1.

The bulk magnetic critical behaviour in the ferromagnetic state has been previously studied, showing that the hidden order and ferromagnetic phase boundaries may actually meet at the critical concentration x=0.15 [12, 13]. This latter point implies that ferromagnetic fluctuations might affect the hidden order state.

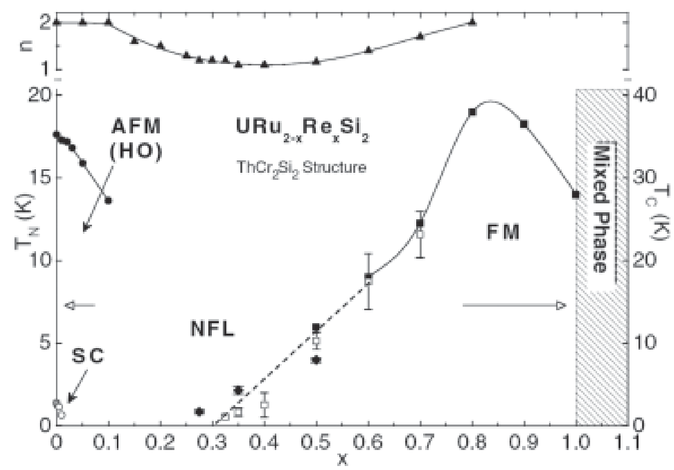

Fig. 1 *Magnetic phase diagram of $URu_{2-x}Re_xSi_2$ showing the antiferromagnetic (hidden order HO), superconducting, and ferromagnetic phases, from Ref. [11].*

Neutron scattering measurements were performed on the DUALSPEC spectrometer at the C5 beamline of the NRU reactor at Chalk River Laboratories. Single crystals of URu_2Si_2 and $URu_{1.9}Re_{0.1}Si_2$ were aligned in the (H 0 L) plane.

We find that there is no elastic AF scattering at the (1.6 0 1) scattering vector (Fig. 2(a)), nor at the equivalent wave vector (1.4 0 0) nor at the commensurate point (1 0 0). At the incommensurate wave vectors the scattering is inelastic and becomes strong for energy transfers above 0.5 THz, such as the scan at 0.7 THz in Fig. 2(b). This is also seen at the equivalent (1.4 0 0) point showing that adjacent layers are coupled antiferromagnetically.

The incommensurate spectrum therefore exhibits a spin gap similar to the gap found earlier in pure URu_2Si_2 at the same scattering vector [2]. However the increased spectral width shown in Fig. 3(a), and the lowering of the peak energy, shows that the incommensurate fluctuations are slowing. Since the

10% Re-doped sample is close to a quantum critical point, at which the hidden order phase vanishes, the slowing of the fluctuations may be due to the proximity to the transition.

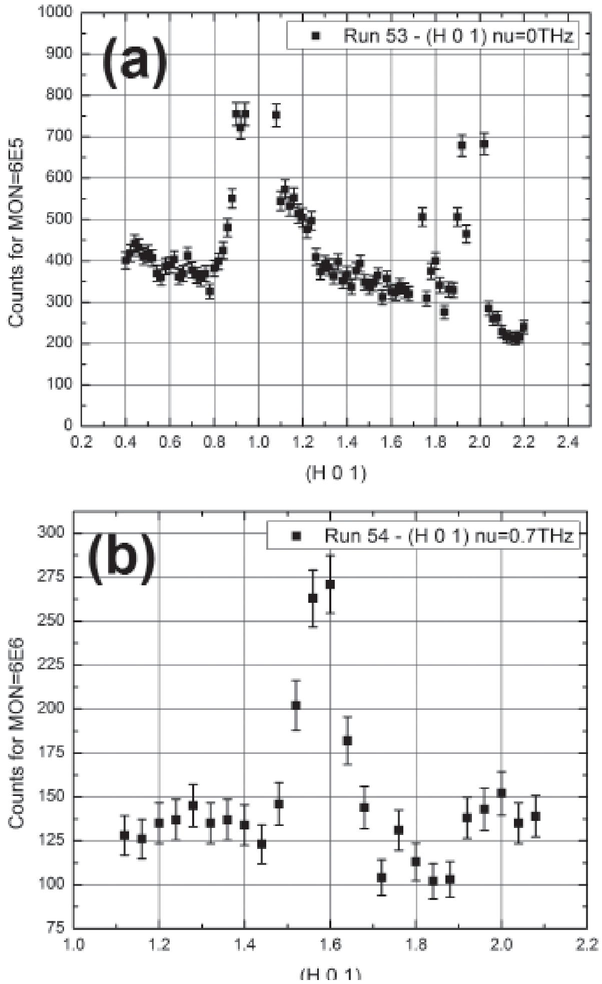

Fig. 2 *(a) (H 0 1) elastic scattering is absent at H=1.6 at 2.56 K. (b) (H 0 1) inelastic scattering for an energy transfer of 0.7 THz. Here we see the emergence of strong inelastic spin fluctuations at the incommensurate wave vector (1.6 0 1).*

Scans were performed at constant Q = (1.4 0 0), with energies ranging from 0 to 5 THz, and fit to a theoretical curve [14], given by:

$$\chi^{zz}(\vec{Q}^*,\omega) = |\Delta_{\vec{Q}^*}|^2 \int \frac{1}{\sqrt{E^2 - \Delta_{\vec{Q}^*}{}^2}} \frac{1}{\omega^2 - 4E^2}\, dE \qquad (1)$$

The theory is based on a spin resonance in the hidden order state, involving transitions between nested parts of the Fermi surface. The nesting vector is chosen to be $\vec{Q}^* = (1 \pm 0.4, 0, 0)$.

The fermion energies are assumed to rise quadratically above a hidden order gap Δ_{Q^*} allowing pairs of excitations to contribute to the dynamic susceptibility measured by neutron scattering [14].

Figure 3 shows the fits for the pure (b), and for the Re-doped system (a) for which Δ_{Q^*} = 0.29 THz, that is, about half the observed peak energy as expected for excitations of pairs. It can be seen that Eq. (1) fits well for the Re-doped sample, but not as well, surprisingly, for the parent compound for which the theory was designed. The greater damping required for the Re-doped system, and indeed the broadening visible in Fig. 3(a), suggests that the lifetime of the spin fluctuations is shorter in the Re-doped compound. Scattering from the rhenium impurities precedes or may be controlled by the approach to the quantum critical point.

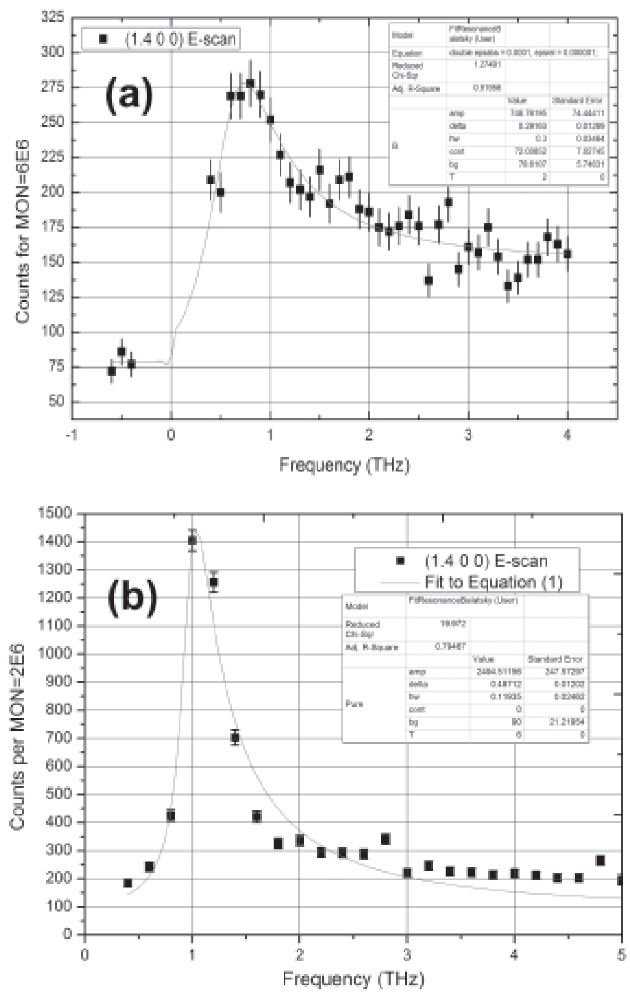

Fig. 3 *(a) The fit to the Balatsky theory [14] for the URh$_{1.9}$Re$_{0.1}$Si$_2$ sample. (b) The fit to the same equation in the pure URu$_2$Si$_2$ sample. The agreement is better for the Re-doped case.*

The commensurate fluctuations at (1 0 0) (not shown) also exhibit lower energy and larger damping for Re doping. As the temperature is increased, these commensurate fluctuations disappear much more quickly than do the incommensurate fluctuations at (1.4 0 0).

We conclude that the predominant behaviour that survives the approach to the quantum critical point is the robust cone of gapped incommensurate fluctuations, similar to the pure material. Both experiment and fits of the Balatsky model show that the lifetime of the spin fluctuations, or more likely the fermions from which they arise, is shorter in the Re-doped compound. The observation of a spin gap at (1 0 0) indicates that the hidden order phase survives at least half-way to the quantum critical point. Our neutron results therefore show that the effect of Re doping, in contrast to the antiferromagnetic enhancement and hidden order destruction by Rh doping [12,13], is to weaken but not destroy the hidden order on approach to the quantum critical point transition to ferromagnetism.

References

[1] C. Broholm et al., Phys. Rev. Lett. 58, 1467 (1987).
[2] C. Broholm et al., Phys. Rev. B 43, 12809 (1991).
[3] W.J.L. Buyers et al., Physica B 199&200, 95 (1994)
[4] T. E. Mason et al., J. Phys.: Condens. Matter 7, 5089 (1995).
[5] F. Bourdarot et al., Physica B 359-361, 986 (2005).
[6] C.R. Wiebe et al., Phys. Rev. B 69, 132418 (2004).
[7] C.R. Wiebe et al., Nature Physics 3, 96 (2007).
[8] J. Janik et al., J. Phys.: Condens. Matter 21, 192202 (2009)
[9] A. Villaume et al., J. Phys. Soc. Jap. 78, 053701 (2009)
[10] Y. Dalichaouch et al., Phys. Rev. B 39, 2423 (1989).
[11] E.D. Bauer et al., Phys. Rev. Lett. 94, 0464011 (2005).
[12] N.P. Butch and M.B. Maple. Phys. Rev. Lett. 103, 076404 (2009).
[13] N.P. Butch and M.B. Maple. J. Phys.: Condens. Matter 22, 164204 (2010).
[14] A.V. Balatsky et al., Phys. Rev. B 79, 214413 (2009).