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Magnetic fluctuations in heavy-fermion metals

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Abstract

Elastic and inelastic neutron scattering have been used to study the antiferromagnetic ordering and magnetic excitations of the U heavy-fermion superconductors UPd_2Al_3 and URu_2Si_2 above and below T_N. While both materials exhibit the coexistence of superconductivity and antiferromagnetic order, the nature of the antiferromagnetic order and magnetic fluctuations is qualitatively quite different. UPd_2Al_3 resembles a rare earth magnetic system with coupling of the 4f electrons to the conduction electrons manifested in a broadening of otherwise conventional spin wave excitations. This is in marked contrast to the unconventional antiferromagnetism found in URu_2Si_2.

Heavy-fermion compounds share a small energy scale derived from the d–f hybridization which is often described as a Kondo temperature. This is most dramatically reflected in a large electronic specific heat [1]. The low-temperature ground states exhibited by heavy fermion materials are particularly varied, with examples spanning nearly all of solid state physics including paramagnetic metals (e.g. $CeRu_2Si_2$ [2]), antiferromagnetically ordered metals (e.g. U_2Zn_{17} [4]), superconductors (e.g. $CeCu_2Si_2$ [5]), antiferromagnetically ordered superconductors (e.g. UPt_3 [6]), and even semiconductors (e.g. CeNiSn [7]). While it is useful to consider the differences in behaviour of these materials as arising from the competition between RKKY and Kondo interactions [4, 8] it is not possible to construct a schematic phase diagram common to all heavy-fermion materials. This is in contrast to high-temperature superconductors where one can describe the destruction of long-range antiferromagnetic order and eventual maximum in T_c with varying doping in terms of a universal phase diagram which is qualitatively the same for all high-T_c materials [9].

In this context it is useful to consider the properties of two compounds which do exhibit some similarities in their low-temperature behaviour. UPd_2Al_3 and URu_2Si_2 both undergo transitions to superconductivity from antiferromagnetically ordered states. Indeed, the resistivity, susceptibility, and specific heat for the two compounds are remarkably similar [10, 11]. Both have large specific-heat anomalies at the Néel transition which occurs at

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14.2 K in UPd_2Al_3 and 17.5 K in URu_2Si_2. The resistivity also displays signatures of the transition to antiferromagnetic order in the form of a knee for UPd_2Al_3 and a small peak for URu_2Si_2. Below their Neél temperatures, the Sommerfeld constants of the two compounds are both of order 100 mJ mol^{-1} K^{-2}. The superconducting transitions occur at 2 K and 1.3 K respectively, the former being the highest T_c of any heavy-fermion superconductor at ambient pressure.

Magnetic neutron scattering is a direct probe of the magnetic correlations, both static and dynamic, of heavy-fermion materials. The experiments reviewed in this paper were carried out using a variety of triple-axis spectrometers at the DR-3 reactor at Risø National Laboratory and the NRU reactor at Chalk River Laboratories [12–15]. The samples were single crystals grown by modified tri-arc Czochralski method (URu_2Si_2) and electron-beam zone refinement (UPd_2Al_3).

UPd_2Al_3 has a simple hexagonal structure belonging to the P6/mmm space group. Powder neutron diffraction has shown that the antiferromagnetically ordered state has magnetic moments of $\sim 0.85\mu_B$ aligned in ferromagnetic sheets in the hexagonal basal plane with the moment direction rotating by π in subsequent layers up the c-axis [16]. Fig. 1 shows the temperature dependence of the (0 0 $\frac{1}{2}$) magnetic Bragg reflection measured for a single crystal [14]. The Bragg intensity is proportional to the square of the sublattice magnetization. The line is a fit to the usual power law expression for a critical phase transition with an exponent $\beta = 0.33 \pm 0.02$, consistent with a phase transition of a three-dimensional XY system. The width of the magnetic Bragg reflections is resolution limited indicating the establishment of truly long-ranged antiferromagnetic order.

URu_2Si_2 has a body centred tetragonal crystal structure. Below 17.5 K, a tiny $0.04\mu_B$ ordered moment is aligned along the c-axis in ferromagnetic sheets with the moment direction alternating in subsequent sheets [3]. Fig. 2 shows the temperature dependence of the (1 0 0) Bragg reflection in zero magnetic field and for a magnetic field applied along the easy c direction [13]. In contrast to UPd_2Al_3, URu_2Si_2 does not exhibit critical behavior of the type associated with three-dimensional magnets with short-ranged interactions. The line in Fig. 2 is a fit to a mean-field form ($\beta = 0.5$) for the sublattice magnetization along with a contribution from the diverging staggered susceptibility (also with mean field exponents) above T_N. The latter is significant and is found in all samples (although its weight relative to the Bragg peak varies). It is always resolution limited in energy [17] which is not what is expected for paramagnetic critical scattering; however, this may be due to a combination of disorder and the induced-moment origins of the phase transition [3, 17, 18]. Although this procedure yields

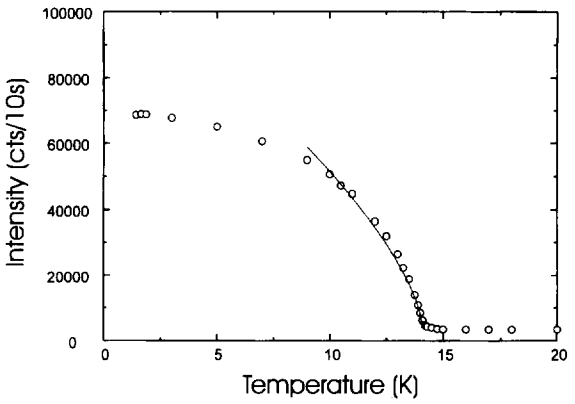

Fig. 1. Temperature dependence of the (0 0 $\frac{1}{2}$) magnetic Bragg reflection of UPd_2Al_3. The line is a fit to a power law with a critical exponent $\beta = 0.33 \pm 0.02$ (from Ref. [14]).

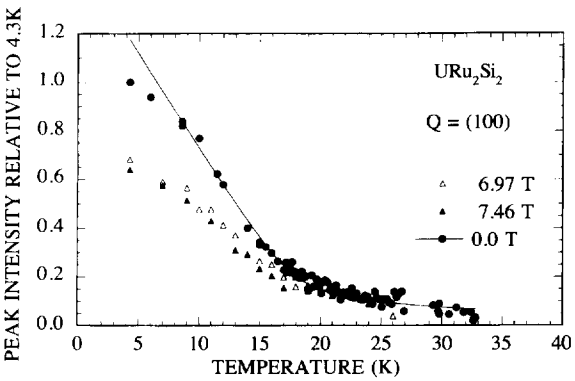

Fig. 2. Temperature dependence of the (1 0 0) magnetic Bragg reflection of URu_2Si_2. The line is a fit to the sum of an ordered Bragg peak and a diverging staggered susceptibility with mean field critical exponents (from Ref. [13]).

reasonable agreement with the observed temperature dependence it implies the critical region extends to very low temperatures since the evolution of the Bragg intensity is linear down to about 2 K. Furthermore, the magnetic order in URu_2Si_2 is never of infinite range. Even in the best case, it is limited to 450 Å along c and 200 Å in the basal plane [19].

The very small ordered moment, peculiar critical properties, and other behavior associated with magnetic ordering such as an anomaly in the nonlinear susceptibility have led to suggestions that the order parameter for the phase transition in URu_2Si_2 may not be the simple dipole magnetic moment observed in the neutron-scattering experiments [20, 21]. Symmetry arguments combined with polarized neutron measurements have ruled out possible

ordering involving higher angular momentum symmetry breaking fields [22].

One suggestion for exotic order is that two or three spins may be correlated on different sites, so as to be hidden at zero field. For such multispin order parameters [21] it is predicted that the application of a magnetic field should cause the emergence of a substantial ($\sim 1\mu_B$ in 10 T) staggered moment [23]. Fig. 3 shows the magnetic field dependence of the antiferromagnetic Bragg intensity for the three geometries necessary to test for symmetric spin tensor ordering [13]. In none of the cases is the expected increase in sublattice magnetization found. For a magnetic field applied along the easy c-axis there is a gradual reduction in the ordered moment, consistent with the expectations for a dipole induced moment transition, although surprisingly this is not accompanied by a corresponding reduction in T_N (see Fig. 2).

In addition to probing static antiferromagnetic order, neutron scattering can be used to observe magnetic excitations. The inelastic magnetic neutron scattering cross-section is proportional to the spin–spin correlation function which is in turn related, via the fluctuation dissipation theorem, to the momentum- and energy-dependent generalized susceptibility. Fig. 4 shows a series of constant-$\boldsymbol{Q}$ scans for UPd_2Al_3 for wave vectors displaced from $(0\,0\,\frac{1}{2})$ along the c-axis for $T = 4.2$ K. A clearly defined peak, indicative of a propagating spin-wave mode, is visible with linear dispersion moving away from the ordering wave vector. The width of the excitation is broader than the spectrometer resolution (0.35 meV). This lifetime broadening provides a measure of the f–d hybridization. We have mapped out the dispersion of the spin wave energy and damping along the high symmetry directions in UPd_2Al_3 [14,15]. The modes remain resonably well-defined along the c-axis, but the damping is substantially higher (relative to the excitation energy) within the basal plane. The zone-boundary energy is also lower in the ab plane despite a reasonably isotropic spin-wave velocity near $(0\,0\,\frac{1}{2})$. This is most likely due to the combined effects of frustration of nearest-neighbour antiferromagnetic interactions for a hexagonal lattice and long-ranged interactions. Most surprising is that the dispersion and polarization of the excitations near the antiferromagnetic zone centre show that the spin wave is a transverse acoustic mode, with no resolvable anisotropy gap, a feature which makes UPd_2Al_3 unique among all U compounds which typically have gapped or overdamped magnetic-excitation spectra. Measurements of the magnetic excitations as the phase transition is approached show the softening and diverging susceptibility expected for an order parameter with continous symmetry.

In URu_2Si_2 the $\boldsymbol{Q}$-dependence of the magnetic excitation spectrum has also been mapped out throughout

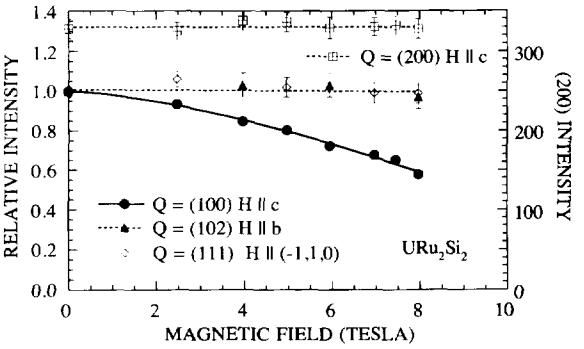

Fig. 3. Magnetic field dependence of the antiferromagnetic Bragg intensity for the three field orientations required to test for symmetric spin tensor ordering. Also shown is the field independent (2 0 0) nuclear reflection verifying mechanical stability of the sample (from Ref. [13]).

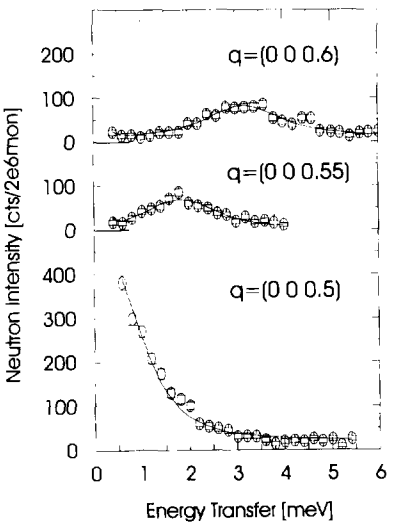

Fig. 4. Constant Q scans in UPd_2Al_3 at 4.2 K for $Q = (0, 0, \frac{1}{2})$, (0, 0, 0.55), and (0, 0, 0.6) (from Ref. [14]).

most of the zone [17]. In that case there is a large (~ 2 meV) gap at the ordering wave vector. The excitations are longitudinal (i.e. polarized along the moment direction) and qualitatively well described by a singlet–singlet model for the ground state and first excited state of the U ions. This also gives a means of understanding the very small ordered moment – a singlet ground-state system with induced-moment ordering can have an arbitrarily small ordered moment. Although qualitatively attractive this model cannot, at least at the mean-field level, quantitatively reconcile the observed magnetic excitation

spectrum with the $0.04\mu_B$ moment or the large specific-heat anomaly at T_N. As in UPd_2Al_3 there is significant damping of the magnetic excitations. In URu_2Si_2 this is most pronounced for wave vectors along the c-axis, parallel to the moment directions and the polarization of the excitations.

Given the unusual nature of the static magnetic order that develops in URu_2Si_2 it is of some interest to determine how the spin dynamics vary through the phase transition. If the antiferromagnetism is a real bulk phenomena then the ordering should be accompanied by a divergence in the real part of the susceptibility at the ordering wave vector. Constant $\boldsymbol{Q}$ scans at (1 0 0) for temperatures near T_N have been analysed to extract the excitation energy, damping, and the real part of the susceptibility (by Kramers–Kronig transform) [13]. The results are summarized in Fig. 5. The spin-wave frequency softens as expected on approaching the phase transition. It does not appear to vanish but that can be attributed to the finite resolution volume sampled in any real experiment. Most importantly, the real part of the susceptibility exhibits a cusp, consistent with the expected divergence and some experimental rounding. It should be stressed that a substantial fluctuating moment ($\sim 2.2\mu_B/U$) [3, 17] is associated with this excitation so the observation of critical behaviour cannot be attributed to some small volume effect at stacking faults or grain boundaries.

While there is no current theoretical explanation of the anomalous magnetic properties of URu_2Si_2, we believe that magnetic frustration plays a role. The likely origin of the frustration is the long range oscillatory nature of the RKKY interactions between U magnetic moments rather than geometrical lattice frustration [17]. The importance of frustration is clearly seen in the temperature dependence of a low frequency (0.4 meV) constant-energy scan along the (1 k 0) direction shown in Fig. 6 [12]. Near T_N the intensity is peaked near (1 0 0), consistent with the observed divergence in the susceptibility and eventual ordering at that wave vector. At higher temperatures, however, the response is dominated by another, not obviously commensurate wave vector (1, 0.4, 0), corresponding to a local minimum in the low temperature dispersion relation [17].

In conclusion, detailed studies of the structure and dynamics of magnetic ordering in UPd_2Al_3 and URu_2Si_2 have shown that despite the apparent similarities in the transport and thermodynamics of these materials the nature of magnetic ordering is quite different. UPd_2Al_3 exhibits relatively conventional magnetism, quite similar to that found in the metallic rare earths while the peculiarities of magnetic order in URu_2Si_2 are most likely due to a subtle interplay of frustration, singlet ground state magnetism, and possibly disorder.

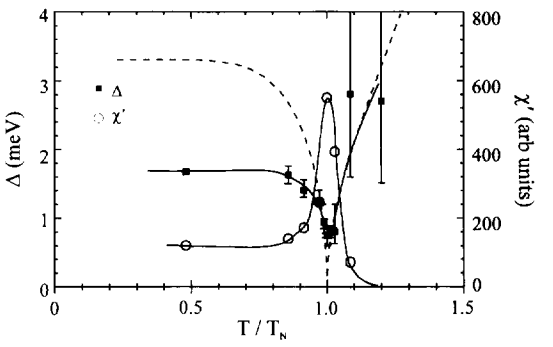

Fig. 5. Temperature dependence of the spin wave energy and real part of the susceptibility at the ordering wave vector of URu_2Si_2. The cusp in the susceptibility is evidence of a bulk dipolar transition. The details of the fits used to extract these parameters are described in Ref. [13].

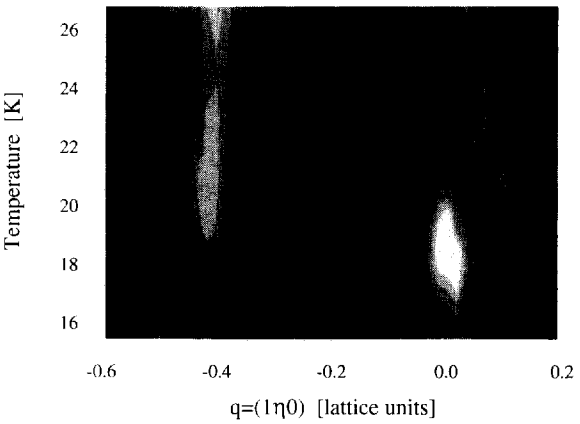

Fig. 6. Wave vector and temperature dependence of the intensity of magnetic scattering for 0.4 meV energy transfer in URu_2Si_2 with intensity mapped onto a linear grey scale, white being most intense. The competition between two different ordering wave vectors is evident as temperature is lowered (from Ref. [12]).

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