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Possible Dynamic Nature of the Antiferromagnetic State in UPd_2Al_3

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A novel model of the antiferromagnetic ground state in UPd_2Al_3 is presented. The model, based on the observation that strongly correlated electron systems are prone to exhibit more than one point in the zone where the magnetic excitation spectrum is enhanced, is supported by direct inelastic neutron scattering data and offers insight into hitherto anomalous thermodynamic and transport behaviour. The dynamical aspect of the antiferromagnetic ground state may find wider application in other strongly correlated materials such as UPt_3 and URu_2Si_2 and its extension to an analogous model of multiple relaxation rates in ω space which offers an understanding of the behaviour of materials in the vicinity of a quantum critical point is noted.

KEYWORDS: UPd_2Al_3, antiferromagnetism

§1. Introduction

The interest in strongly correlated electron metals has been heightened by the discovery of the co-existence of magnetism and superconductivity in selected members of this class. In turn, the advent of strongly correlated superconducting materials has been of crucial importance in stimulating the debate that internal (dynamic) magnetic fields may play a critical role in the formation of novel superconducting phases. There exist examples based both on rare earth and actinide intermetallics with the implication of both antiferromagnetic, {$CeNi_2Ge_2$, $CeRh_2Si_2$, $CeIn_3$, $CeCu_2Ge_2$, UPt_3}[1–3] and, more recently, ferromagnetic polarisations, {Sr_2RuO_4, UGe_2}.[4,5] In almost all cases the inferred static moment on entering the superconducting phase is small. For example, in the cerium based antiferromagnetic metals, which become superconducting under external pressure, the temperature of the resistivity anomaly which correlates with $T_{\text{Néel}}$ at ambient pressure approaches zero concomitantly with the maximal induced superconducting transition temperature, T_{sc}. A similar situation pertains in situations where the superconducting phase is induced by alloying. Such materials typically have great sensitivity to composition and low superconducting transition temperatures reaching at most a few tenths of a Kelvin above absolute zero. In general these characteristics render them unsuitable candidates for detailed measurements of their dynamical magnetic response by neutron scattering or other techniques. An alternative group of materials with simultaneous antiferromagnetic order and a generally more robust superconducting phase, higher T_{sc}'s and less sensitivity to precise composition are presented by, for example, the Chevrel phase superconductors[6] and select rare earth borocarbides.[7] In these latter materials superconductivity and magnetism appear to be associated with individual groups of electronic states which constitute, at least in some analyses, two separate quantum fields; thermal and transport anomalies are then considered as arising from interactions between them.[8,9] As an apparently distinct case, UPd_2Al_3 exhibits a microscopic co-existence of magnetism, $T_{\text{Néel}}$ = 14.3 K, and superconductivity, T_{sc} = 2 K, with a long time average antiferromagnetic moment of 0.85 μ_B as determined by neutron scattering techniques. In this paper we suggest a novel view point on the antiferromagnetic nature of UPd_2Al_3 which may have rather wide and general implications. The discussion has been stimulated by preliminary results obtained on the spectrum of magnetic fluctuations concentrated around the wave vector, $\mathbf{Q}^*$ = (0.5,0,0.5), which is explicitly *not* equal to the observed antiferromagnetic ordering wave vector, $\mathbf{Q_o}$ = (0,0,0.5) where all wave vectors are expressed in reciprocal lattice units (rlu). Further experiments have been scheduled to refine these ideas.

The compound crystallises in the hexagonal $PrNi_2Al_3$ structure (space group *P6/mmm*) with lattice constants a = 5.350 Å and c = 4.185 Å at room temperature. Specific heat measurements reveal not only a strong enhancement of the linear term at low temperatures, characteristic of the heavy fermion state, but also two distinct anomalies associated with the antiferromagnetic and superconducting phase transitions respectively. The large specific heat jump at T_{sc}, ($\Delta C = 1.2\gamma T_{sc}$ (γ =140 mJ / mole K^2))[10] suggests that the superconducting ground state evolves out of interactions between quasiparticles located in strongly renormalised states in a low energy shell around the Fermi surface. Through previous work, principally inelastic neutron scattering by the group of Metoki at JAERI, Tokai, Japan,[11] and ourselves at Institut Laue Langevin (ILL), Grenoble, France in collaboration with Tohoku University[12–14] combined with tunnelling spectroscopy,[15] and stimulated by the extensive open literature on thermodynamic and transport measurements, an implication of the magnetic excitations in the stabilisation of the superconducting phase has been made.[11,13,15]

§2. Experimental data

In this section we discuss both the primary experimental data realised around $\mathbf{Q}^*$, which have stimulated the present view point on the antiferromagnetic nature of UPd_2Al_3, and corroborating evidence from thermodynamic and transport measurements reported in the literature. Whilst the general idea may have a wider application we bear in mind that spectral details, resulting from a more extensive temperature dependent mapping, will be presented in the near future.

The sample is as used in references 13. The experiments have been performed at ILL on both cold (IN14) and thermal (IN8) source spectrometers in the a* - c* scattering plane. The magnetic response, at $T = 0.2$ K, for energy transfers up to 5 meV and for wave vectors (a*,0,0.5) is given in Fig. 1. In the a* direction the magnetic intensity initially disperses away from $\mathbf{Q_o}$ in the manner of a strongly damped excitation, however around the (magnetic) zone boundary it is better described as an island of intensity centred around $\mathbf{Q^*}$ = (.5, 0, .5) with a peak in the energy response between 2 and 3 meV. This latter dynamic response, with a spatial scale of some 10's of lattice spacing along a and 20 along c, represents a damped oscillation of the magnetisation density with antiferromagnetic periodicity along both a and c. The observed magnetic structure has the moment $\mathbf{M}$ parallel to a and antiferromagnetically stacked along c with ordering wave vector $\mathbf{Q_o}$; this suggests that the nominal magnetic order (of a given domain) may be realised by islands of moments, arranged as in Fig. 2, oscillating between configuration 1 and 2 with wave vector $\mathbf{Q^*}$. This is the principal hypothesis of this paper: that a dynamic state (of wave vector $\mathbf{Q^*}$) may project a time average order parameter (at $\mathbf{Q_o}$). Of course in the context of itinerant magnetism this is not new, correlated electronic hopping ultimately yields the order parameter. Here the novelty arises in operating at a higher level with the slow, identifiable, dynamics of well defined moments. In Fig. 3 the thermal evolution of the scattering intensity at $\mathbf{Q^*}$ is given, at all temperatures measured energy scans in neutron energy loss reveal a broadly distributed line shape with intensity extending at least to 10 meV transfer.

This scenario is supported by the following observations concerning respectively (a) the direction of moment fluctuation {i}, (b) the wave vector of the fluctuations {ii - iv}, (c) the energy of the fluctuations {v - vi} and (d) the thermal evolution of the response {ii - viii}:

(i) neutron polarisation analysis at $\mathbf{Q_o}$ determines a significant fluctuating component of $\mathbf{M}$ in the $a-b$ plane.[14)]

(ii) the magnetic phase diagram established by neutron scattering[16)] indicates the preferential alignment of $\mathbf{M}$ along $\mathbf{a_1}$ and $\mathbf{a_2}$ for an applied field parallel to $\mathbf{a}$, i.e. the moments lie preferentially in directions consistent with those proposed for the (0.5,0,0.5) excitation. The phase diagram boundaries are supported by thermodynamic (χ)[17,18)] and transport studies.[19)] Extrapolation of thermodynamic measurements indicate that this $\mathbf{a_1}$-$\mathbf{a_2}$ phase, which exists at all studied temperatures below $T_{\text{Néel}}$ in an applied field, forms in zero applied field for 11 K $< T < T_{\text{Néel}}$.[17)]

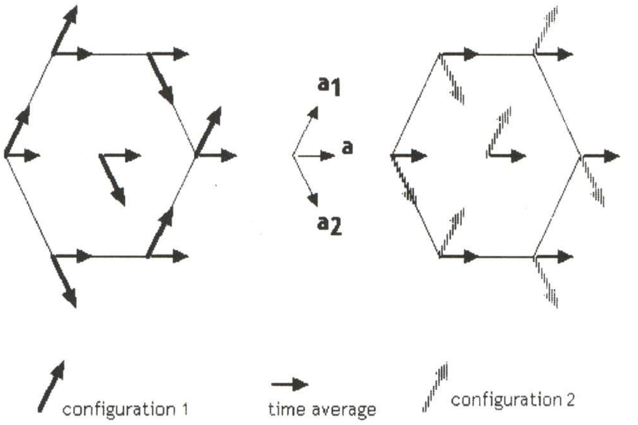

Fig. 2. The proposed model; moments fluctuate between configuration 1 and 2, and thereby account for both the dynamic $\mathbf{Q^*}$ response and the reduced, static, $\mathbf{Q_o}$ moment along $\mathbf{a}$.

(iii) an unusual metamagnetic transition below $T_{\text{Néel}}$ at 18 Tesla ($\mathbf{H}$ parallel $\mathbf{a}$) with a moment jump to $\sim 1.5\mu_B$.[20,21)] The transition smears out above the Néel temperature but remains as a broad feature up to 35 K where χ has its maximum.[22,23)]

(iv) bandstructure calculations give both a larger than observed magnetic moment (2 μ_B) and a lower energy to a $\mathbf{Q^*}$ (.5,0,.5) magnetic ordering wave vector than to the nominal ordering wave vector $\mathbf{Q_o}$ (0,0,.5).[24)] From (iii) and (iv) we estimate the oscillating moment to be $\sim$ 1.75 μ_B; given $\mathbf{a_1}$ and $\mathbf{a_2}$ subtend 120° this yields a time average moment of $\sim$ 0.8 μ_B along $\mathbf{a}$. On applying a field of sufficient strength the moment fluctuations may freeze in a given orientation (18 Tesla on 1.75 $\mu_B \sim k_B T_{\text{Néel}}$) enabling one see the full moment, explaining the unusual metamagnetic transition.

(v) the torsional stiffness for fluctuations in the $a-b$ plane falls by an order of magnitude above 10 K, and a plot of the low energy transfer scattering, at $\mathbf{Q_o}$ and $\hbar\omega = 0.2meV$, shows peaks at 11 K and 14 K.[14)] These dynamic signatures support observations indicating the existence of a quasi critical point at 11 K[17)] and the correlation of $\mathbf{Q^*}$ dynamics with the $\mathbf{Q_o}$ response. In this light we also recall the anomalous enhancement of $1/T_1$ in Pd^{105} NQR above 11 K.[25)]

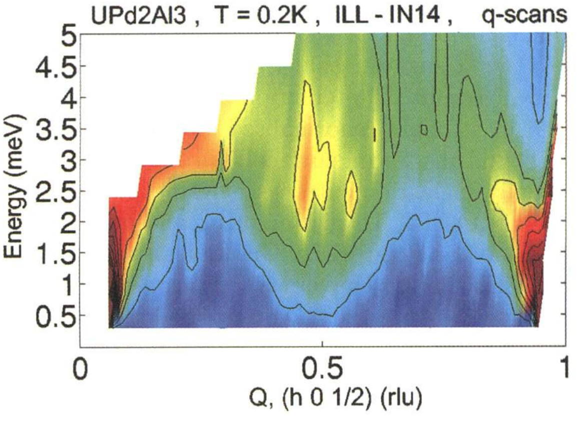

Fig. 1. The energy versus wave vector response in UPd_2Al_3 at 0.2 K measured along the (a*, 0, 0.5) axis on IN14 at the ILL. The intensity contours rise in equal steps from minimum (blue) to maximum (red). The distinct island of excess intensity around $\mathbf{Q^*}$ is evident.

(vi) the presence of a characteristic energy around 30 K - 40 K as evidenced in crystal field analyses,[17] from experimental anomalies in the heat capacity,[26] the susceptibility,[18] thermal expansion,[27] Knight shift Al^{27},[28] $1/T_1$ of Al^{27},[29] and the Knight shift Pd^{105}.[25]

(vii) the observation by time of flight neutron spectroscopy on a polycrystalline sample which reports,[30] at 25 K, a peak of the intensity at wave vectors near 1 $Å^{-1}$ centred at energy transfer $\sim$ 2.24 meV of width $\sim$ 0.75 meV when we note that $|\mathbf{Q}^*| = 0.97$ $Å^{-1}$.

(viii) resonant magnetic x-ray scattering data [unpublished] indicates an unusual pronounced tail of intensity at $\mathbf{Q_o}$ of steadily increasing width in q space extending at least to 25 K (i.e. approximately up to 2* $T_{\text{Néel}}$). The resonant nature of the probe gives sensitivity to magnetic correlations on the time scale of the uranium ion core hole lifetime 10^{-15} - 10^{-14} s. This supports the neutron data, Fig. 3, observations (vi, vii), and is consistent with there being persistent short range order above $T_{\text{Néel}}$ resulting from $\mathbf{Q}^*$ fluctuations.

§3. Discussion

To make the connection between the low frequency dynamics centred at $\mathbf{Q}^*$ which are broadly distributed in wave vector with the observation of static long range order at $\mathbf{Q_o}$ requires a further ingredient, spatial coherence. On cooling below $T_{\text{Néel}}$, a given $\mathbf{Q_o}$ domain arises from dynamic $\mathbf{Q}^*$ blocks which, remaining uncorrelated in their relative phase of fluctuation, form a spatially coherent unit stabilised by a self consistent mean field proportional to the time average moment. Whether there is a component of the fluctuating moment out of the $a-b$ plane and an effective precession about the **a** axis is, at present, unknown. The rise of spatial coherence gives a new dimension to the formation of antiferromagnetic order which may be viewed as a (spatial) condensation of (pre-existing) dynamically correlated regions. The analogy with, for example, some models of high-T_c superconductivity is evident. This extra degree of freedom gives a natural way to rationalise many of the thermodynamic and transport anomalies which persist to temperatures well in excess of $T_{\text{Néel}}$. The strongly reduced value of the ordered moment, which was previously not accounted for, may also be relevant in understanding the magnitude of the magnetic contribution to the experimental heat capacity at $T_{\text{Néel}}$. The entropy release is related to the excitation spectra and in no simple way to the nominal ordered moment, contributions to the entropy will arise from loss of spatial coherence of dynamic blocks in addition to the internal disordering of the moments.

The dissipation mechanism of the block oscillator dynamics is unknown, first indications are that it is roughly similar in magnitude at 1.5 K and 20 K, Fig. 3, and increases by 40 K. It is thus unlikely that an important intermediary role is played by the heavy quasiparticle density in coupling to the phonon heat bath; aside from coupling via a reservoir of normal electronic states, one possibility is that the magnetic dissipation arises directly from the distortion of the local, crystal field, environment by excitation of phonons. Above 40 K diminishing electronic correlations are reflected in the loss of natural

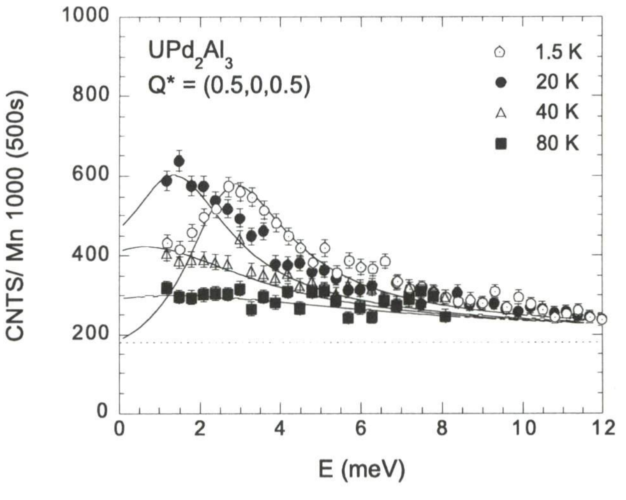

Fig.3. The $\mathbf{Q}^*$ response, measured on IN8 at ILL, which persists to at least 10 meV in energy and 80 K. The data below 1 meV have been suppressed since they fall within the (elastic) resolution volume. The response, which changes in character from damped inelastic to overdamped behaviour around 40 K, is modelled with the following parameters; **1.5 K**: $\chi \sim 205$ a.u., $\Gamma \sim$ 1.5 meV, $\omega_o \sim 2.4$ meV; **20 K**: $\chi \sim 205$ a.u., $\Gamma \sim 1.5$ meV, $\omega_o \sim$ 1.1 meV; **40 K**: $\chi \sim 102$ a.u., $\Gamma \sim 3$ meV, $\omega_o \sim 0$ meV; **80 K**: χ ~ 50 a.u., $\Gamma \sim 6$ meV, $\omega_o \sim 0$ meV. The dotted line represents the estimated background.

frequency of oscillation at $\mathbf{Q}^*$, Fig. 3, and by growth of a local moment phase as monitored, for example, through the developing Curie-Weiss like susceptibility.[23]

§4. Conclusion

The idea that the dynamical response at wave vector $\mathbf{Q}^*$ can provide a ground state time average moment at a nominal ordering wave vector $\mathbf{Q_o}$ may be useful in a first instance to understand some long standing anomalies in UPd_2Al_3. Evidence has been presented for this point of view based on neutron, thermodynamic and transport data. The model, which unites the $\mathbf{Q}^*$, $\mathbf{Q_o}$ response in a consistent manner, represents, respectively, the coherent oscillation of magnetisation density within a given block which is of random phase with respect to the other blocks in the domain, and a time averaged moment, the spatial coherence of which is maintained by the self consistent mean field.

Since it is generally known that strongly correlated electronic systems are prone to exhibit more than one point in the zone where the magnetic excitation spectrum is enhanced, this idea could have a more general relevance. Direct examples of heavy fermion superconductors with multiple soft spots in q include UPt_3 and URu_2Si_2; interestingly these materials have anomalous thermal properties including, for example, their heat capacities, and both exhibit strongly reduced 'static' moments as compared with their estimated dynamical fluctuations. The relative sensitivity of scattering experiments to the collapse of spatial coherence and the internal disordering of moments can lead to discrepancies between scattering and thermodynamic determinations of the value and form of the magnetic critical point. The widths and intensities of the magnetic peaks in URu_2Si_2[31,32] may, for example, be rationalised in this manner.

The dynamical model also strengthens the link between antiferromagnetic superconductors and other, non-ordering, strongly correlated superconducting systems such as the cerium based intermetallics or the high-T_c materials with their antiferromagnetic parentage and ferromagnetic superconductors such as UGe_2. In the latter compound, recent work[5] points directly to a critical role played by fluctuations at multiple soft spots in q space. In addition to the specific microscopic portrayal of these materials it will be of fundamental interest to construct a model of superconductivity based on such a dynamical (anti)-ferromagnetic state. In this latter context we note the, possibly accidental, similarity in spatial scale of the dynamic blocks and the superconducting coherence length in UPd_2Al_3. Finally, as discussed elsewhere,[33] extending the key ideas of multiple soft spots in q space and coherence to include multiple relaxation rates in ω space, yields valuable insights into the fundamental dynamics of materials in the vicinity of a quantum critical point.

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