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Magnetization dynamics in the normal and superconducting phases of UPd_2Al_3: II. Inferences on the nodal gap symmetry

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Abstract

This paper provides an analysis of neutron inelastic scattering experiments on single crystals of UPd_2Al_3. The emphasis is on establishing robust general inferences on the joint antiferromagnetic–superconducting state that characterizes UPd_2Al_3 at low temperatures. A distinction is drawn between these conclusions and various theoretical perspectives of a more model-sensitive nature that have been raised in the literature.

1. Introduction

In this paper the focus is on the inferences that may be drawn from inelastic neutron-scattering data on the nature of the antiferromagnetic–superconducting state in UPd_2Al_3. In particular, the aim is to establish the scope, and limits, of global properties concerning the symmetry and magnitude of the superconducting energy gap, $\Delta(\mathbf{k})$, and the quasiparticle pairing potential from observed changes in spectral form on passing below T_{sc}. Extensive reference will hence be made to the experimental evidence presented in the preceding topical review [1] where the results, obtained from samples prepared and measured at independent institutes, point to the robust nature of the thermodynamic physical properties of this material. It is this underlying commonality that forms the backbone of the present analysis and gives credence to the conclusions drawn.

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Whilst verification through full quantitative calculations of the neutron scattering cross section is, to our knowledge, not feasible, it is possible to establish those signatures of the superconducting state that follow from general arguments and to differentiate these from conclusions of a more model-specific nature. Thus for example, detailed, band-structure and model-dependent results will not be given since such approaches have been extensively discussed elsewhere [2–10].

In particular it will be seen that whilst unique wavevector- and energy-dependent information on $\Delta(\mathbf{k})$ is forthcoming, little can be said about the pairing mechanism of the superconducting state. Indeed, with our present state of knowledge we feel unable to offer any firm conclusions on this point. Nevertheless, symmetry constraints can be placed that serve as a yardstick against which the various propositions may be measured. To commence, a brief review of the underlying assumptions and constraints implicit in any analysis is given.

2. Analysis of the results

2.1. Basic considerations

Long-wavelength probes are, to a good approximation, insensitive to the translation symmetry operations of the lattice and all periodically related repeat units respond in a similar manner. Conventional optical and microwave spectroscopy, together with transport and thermodynamic measurements of the superconducting state, fall in this class. Use of oriented monocrystalline samples in conjunction with polarization techniques may yield directional sensitivity in propitious cases; however, inferences on, for example, the energy gap are limited to its magnitude and crystallographic point group symmetry. The unique role of inelastic neutron scattering as a probe of $\Delta(\mathbf{k})$ lies in its simultaneous wavevector and energy selectivity on the atomic and thermal scales, respectively. This sensitivity to the translation operations of the lattice brings to light the primary role of the space group symmetry of $\Delta(\mathbf{k})$ and permits one to extract previously inaccessible information.

Experimental results, as discussed in the preceding topical review [1] have established the electronic (magnetic) origin of the anomalous scattering observed below T_{sc}. The present work considers the inferences that may be drawn from such data involving the interaction of the neutron with both the condensate and the strongly correlated electronic quasiparticles of the superconducting state. To enable progress, an analysis of the spectral form of the generalized magnetic susceptibility is required.

2.2. Generalized magnetic susceptibility

In the following we examine a model dynamical susceptibility wherein the generic approach is to dissect the empirically determined $\chi_q(\omega)$ into two, or more, distinct components. A similar conceptual fragmentation, used for example in analyses of thermodynamic and μSR data, has been attributed to a 'dual' character of the 5f wavefunction. However, independent of detailed poles or resonances, there is only one magnetization correlation function. In cases of a multiple-peaked structure observed in frequency and/or wavevector, as observed in the response around $\mathbf{Q}_0$ (see the preceding topical review [1]), care must be taken in any decomposition to preserve the characteristic amplitude and phase correlations of the N-body state.

2.2.1. Primitive low-frequency–high-frequency model. A primitive model for such a structured response is to split the dynamical susceptibility into two distinct components, which are simply summed as incoherent contributions to generate a total response function:

$$\chi = \chi_1 + \chi_2. \tag{1}$$

At this level one may discuss independent contributions, attributed respectively to a low, χ_1, and a high, χ_2, energy part of χ. An initial analysis of this form in UPd_2Al_3, which highlighted the key *qualitative* changes observed on passing below T_{sc}, was presented by the JAERI group [11]. The strong renormalization in χ_1 inferred at T_{sc} is evidence for the *influence* of superconductivity on the magnetic response function. It is, however, not proof that the superconductivity is driven by the magnetic fluctuations.

2.2.2. Low-frequency–high-frequency coupling model. An ensuing level of sophistication is to take a coupling, generally of mean-field form, between the low- and high-frequency fragments of the full 5f-neutron-scattering *amplitude* with a fully dynamical (space-time retarded) coupling constant, $\bar{\lambda} = \bar{\lambda}'(q, \omega) + i\bar{\lambda}''(q, \omega)$, in an attempt to restore at least some of the principal correlations of the macrostate. However, in practice this method is normally approximated by replacing $\bar{\lambda}$ by a constant, λ, which is used in a direct calculation of the magnetic susceptibility, i.e. calculations at the level of probabilities (scattering cross sections). The field has an abundant literature with many, often equivalent, formalisms [12]. Even this minimal consideration may trigger profound modifications of spectral form. Principally, such modifications arise on account of a built-in positive feedback giving the net response a Stoner-like denominator that acts to enhance, preferentially, the low-frequency part with a concomitant renormalization of the effective low-energy line width, as can be seen by the following simple argument.

First, make a conceptual fragmentation of the magnetic system into low- and high-energy units, designated by M_1 and M_2, respectively, in which all internal interactions have been included. Then, with a mean-field coupling, λ, form $M_1 = \chi_1[H + \lambda M_2]$ and $M_2 = \chi_2[H + \lambda M_1]$ giving the total magnetization as $M = M_1 + M_2$ and susceptibility,

$$\chi = \frac{\chi_1 + \chi_2 + 2\lambda\chi_1\chi_2}{1 - \lambda^2\chi_1\chi_2}, \tag{2}$$

where χ_1 and χ_2 are the individual susceptibilities, λ the mean-field coupling and the primitive model of equation (1) is the $\lambda \to 0$ limit. At low frequencies the real parts of $\chi_{1,2}$ tend to a constant whilst the imaginary parts are proportional to the frequency. This, for the dissipative component of the total susceptibility as $\omega \to 0$, yields a denominator $1 - \lambda^2\mathrm{Re}\,[\chi_1]\mathrm{Re}\,[\chi_2]$. In contrast, at high frequencies the susceptibilities $\chi_{1,2}$ tend to zero and the denominator goes to unity. Thus, an increase in low-frequency response, ultimately driving a divergent response and transition of phase, can be incited through an augmented value of either of $\chi_{1,2}$ and/or the coupling constant. In the interest of simplicity it is often argued, as we do below, to keep the coupling constant local in space-time and temperature independent. Schematics of the response arising from such general coupling models for the normal antiferromagnetic state in UPd_2Al_3 are given in the left-hand panels of figure 1.

2.2.3. The low-energy susceptibility, χ_1, in the superconducting state. The fundamental problem facing any interpretation below T_{sc} is how to partition the scattered intensity between the excitations of the normal and condensate components. In the following we examine a general model of the dynamical susceptibility taking account of the phase coherence of the paired state on its symmetries and amplitude to resolve this dilemma. At the same time it is used to extract unique wavevector and energy-dependent information on the energy gap function.

The spin-susceptibility of excited quasiparticles below T_{sc} is modified by the effects of (i) superconducting phase coherence and (ii) the presence of a gap in the excitation spectrum. It is calculated in the following approximation for a singlet ground state as [13, 14] $\chi_1 = \chi_{qp} + \chi_c$,

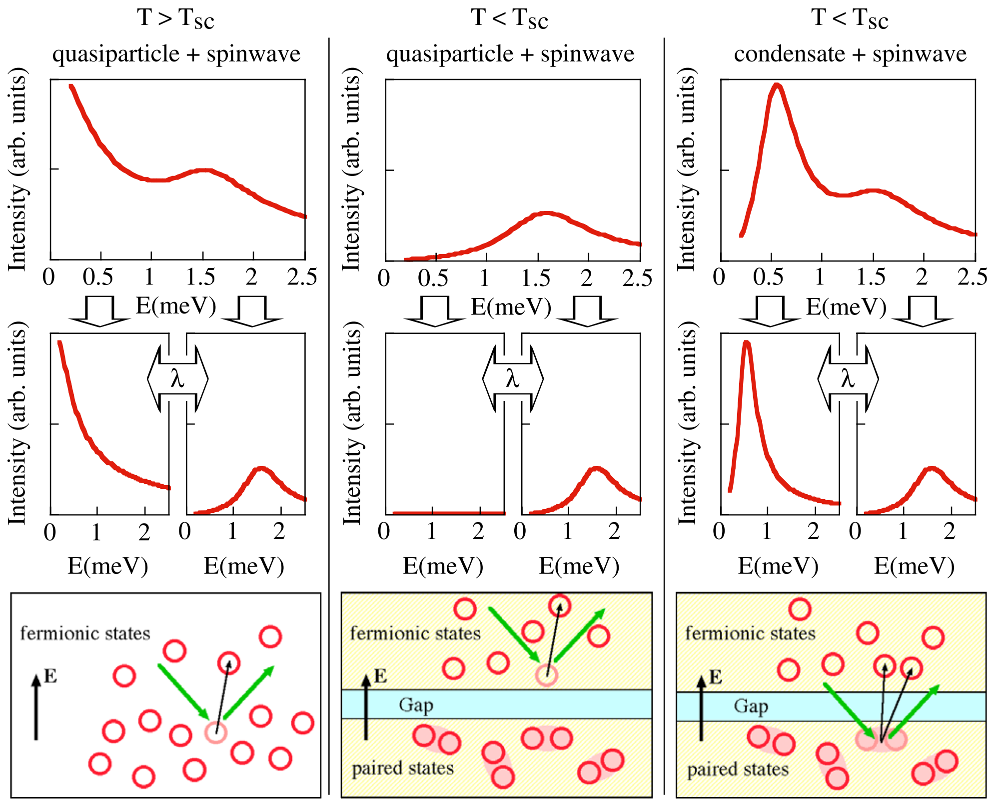

Figure 1. Left-hand panel: schematic of the response for $T > T_{sc}$ in the normal antiferromagnetic phase. The top frame illustrates the characteristic bi-modal scattering of UPd_2Al_3, which is shown in the middle two frames as a conceptual fragmentation into a coupled low- and high-energy response. The bottom frame is a cartoon representation of the response (thin [black] arrow) of the normal quasiparticle states ([red] hoops) to the neutron probe (thick [green] arrows). Central panel: the top frame is a schematic of response for $T < T_{sc}$ in the superconducting antiferromagnetic phase, shown in the middle frames as a conceptual fragmentation into a coupled normal quasiparticle low-and high-energy response. The vanishing magnitude of the quasielastic response occurs on account of both the gapping of the Fermi surface and the phase cancelling role from the antiferro-periodic nodal symmetry of the condensate on such excitations in the magnetic susceptibility (see equation (3)). The bottom frame is a schematic of the dynamic equilibrium between the paired and Fermion states. Normal quasiparticles, above the energy gap, are marked as red hoops and paired states, below energy gap, are designated as bound, overlapping hoops. The phase coherent condensate influences the possible excitation processes of the normal-state quasiparticles and such interference effects must be taken into account below T_{sc} in analysis of the quasiparticle spectra (designated by a [yellow] mesh). Right-hand panel: schematic of the response for $T < T_{sc}$ in the antiferromagnetic superconducting phase, shown in lower frames as conceptual fragmentation into a coupled condensate low-energy and normal quasiparticle high-energy response. The enhanced magnitude of the condensate response occurs on account of phasing role from the antiferro-periodic nodal gap symmetry on such excitations in the magnetic susceptibility. The bottom frame is a schematic of the dynamic equilibrium between the paired and Fermion states as in the central panel. As illustrated, direct excitation out of the condensate may occur. Again, the superconducting energy gap has a profound influence on the intensity of scattering at a given momentum and energy transfer, reflecting the symmetry and coherence of the wavefunction.

(This figure is in colour only in the electronic version) where the quasiparticle fraction is given by

$$\chi_{\mathrm{qp}}(\mathbf{q},\omega) = \sum_{\mathbf{k}} \frac{1}{2}\left[1 + \frac{\xi(\mathbf{k}+\mathbf{q})\xi(\mathbf{k}) + \cos[\Phi(\mathbf{q})]|\Delta(\mathbf{k}+\mathbf{q})||\Delta(\mathbf{k})|}{E(\mathbf{k}+\mathbf{q})E(\mathbf{k})}\right] \times \frac{f(\mathbf{k}+\mathbf{q}) - f(\mathbf{k})}{\omega - [E(\mathbf{k}+\mathbf{q}) - E(\mathbf{k})] + \mathrm{i}\Gamma} \tag{3a}$$

and the condensate by

$$\chi_{\mathrm{c}}(\mathbf{q},\omega) = \sum_{\mathbf{k}} \frac{1}{4}\left[1 - \frac{\xi(\mathbf{k}+\mathbf{q})\xi(\mathbf{k}) + \cos[\Phi(\mathbf{q})]|\Delta(\mathbf{k}+\mathbf{q})||\Delta(\mathbf{k})|}{E(\mathbf{k}+\mathbf{q})E(\mathbf{k})}\right] \times \frac{1 - f(\mathbf{k}+\mathbf{q}) - f(\mathbf{k})}{\omega - [E(\mathbf{k}+\mathbf{q}) + E(\mathbf{k})] + \mathrm{i}\Gamma} - \sum_{\mathbf{k}} \frac{1}{4}\left[1 - \frac{\xi(\mathbf{k}+\mathbf{q})\xi(\mathbf{k}) + \cos[\Phi(\mathbf{q})]|\Delta(\mathbf{k}+\mathbf{q})||\Delta(\mathbf{k})|}{E(\mathbf{k}+\mathbf{q})E(\mathbf{k})}\right] \times \frac{1 - f(\mathbf{k}+\mathbf{q}) - f(\mathbf{k})}{\omega + [E(\mathbf{k}+\mathbf{q}) + E(\mathbf{k})] + \mathrm{i}\Gamma}. \tag{3b}$$

Each element is the summation over the Brillouin zone of a product of a superconducting phase-coherence factor and a Lindhard style function. The normal-state quasiparticle fraction χ_{qp} arises from scattering between the quasiparticle levels, while χ_{c}, the condensate fraction, corresponds with the creation (in neutron energy loss) and condensation of quasiparticle pairs (in neutron energy gain). The notation is standard: $\xi(\mathbf{k}) = \varepsilon(\mathbf{k}) - \varepsilon_{\mathrm{F}}$ is the quasiparticle energy relative to the normal-state Fermi energy and $E(\mathbf{k}) = \sqrt{\xi(\mathbf{k})^2 + |\Delta(\mathbf{k})|^2}$ is the quasiparticle excitation energy above the superconducting state. The factor $\Phi(\mathbf{q})$ is the phase difference between $\Delta(\mathbf{k})$ and $\Delta(\mathbf{k}+\boldsymbol{q})$. It may be noted that the coherence function in equation (3) acts in an opposite sense on the normal and condensate contributions to the cross section. This excludes the simultaneous enhancement of the quasiparticle contribution and the condensate fraction to the scattering cross section.

2.2.4. Interpretation of the low-energy spectra on passing below T_{sc}. In this section we discuss how the observed changes to the low-energy spectra on entry to the superconducting phase have been understood within alternative scenarios. As will be seen, the minimal modifications to the response function (as given in equation (3)), implicit in the q-dependent theory of the phase-coherent state, are, in themselves, sufficient to understand the observations.

A key to understanding comes from figure 2, as well as figures 2–5 in the preceding topical review [1], which illustrates the scattering intensity around $\mathbf{Q}_0$. They show how the observed quasielastic scattering well inside the normal antiferromagnetic state decreases in proportion to $k_{\mathrm{B}}T$. This is an indication for the thermal response of a temperature-independent intrinsic susceptibility. The abrupt discontinuity in scattering intensity below T_{sc} where both the amplitude, which jumps to approximately twice that expected from the normal-state response as shown in figure 2, and the generic form, i.e. opening of a gap in response to a temperature dependence that no longer follows the simple $k_{\mathrm{B}}T$ law, are strong indicators that the superconducting ground state is having a profound influence on the magnetic response function.

Differences in inference on the physical nature and symmetry of the superconducting state then arise from attributing the measured changes in response on passing through T_{sc} either (i) purely to changes in the two, frequency-decomposed, normal-state components of χ and possibly the coupling constant, as schematized in the central panels of figure 1, or (ii) by taking

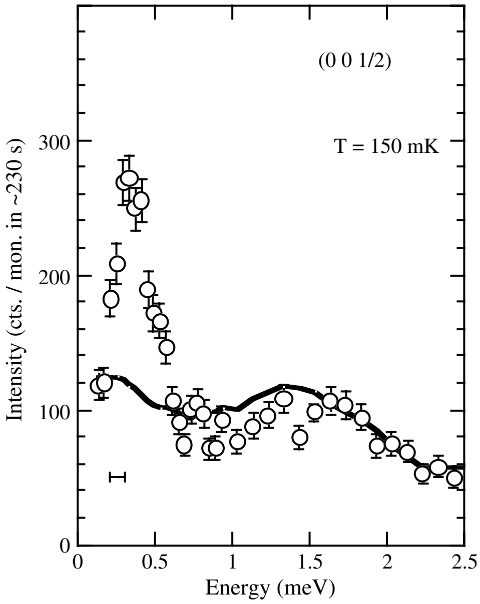

Figure 2. The scattering observed at $\mathbf{Q}_0$ in the superconducting state ($T = 150$ mK) as experimental points. Above T_{sc}, the low-energy spectrum appears as a quasielastic response together with a higher-energy, spin-wave-like, feature. The solid line is a smooth fit to 2.5 K data ($T > T_{sc}$) scaled by the Bose factor (with a constant background subtracted), the horizontal bar indicates the instrumental resolution. See figures 2 and 3 in the preceding topical review [1] for further details.

into account the role of excitations out of the developing superconducting ground state, as given in the right-hand panels of figure 1.

In the first scenario, the sole contribution to the cross section arises from quasiparticle excitations of the normal state. To have significant spectral weight at $\mathbf{Q}_0$ on passing below T_{sc} at low energy transfer would demand the presence of superconducting nodes commensurate with $\mathbf{Q}_0$ on sheets of the Fermi surface exhibiting a significant density of quasiparticle states. Such a situation is generally energetically unfavourable and is also at variance with available tunnelling data [2, 15]. The observation of an *enhanced* spectral weight in figure 2 also implies an additional concentration of the response in a wavevector or a frequency-amplification process, as discussed above. The 'tuning' options available are that either the spin wave (exciton) undergoes substantial changes in its amplitude, position and/or damping at these very low temperatures, or the low-energy component changes its characteristic amplitude and/or decay rate, and that an appropriate mixture, with or without changes in the (complex) coupling constant λ, is found. Finally, a condition for the 'inelastic' nature of the low-frequency response must be added, with such a scenario implying a new pole to have been generated in the normal-state response function.

Following the second option, the response at low frequencies and lowest temperatures is dominated by the condensate, as schematized in the right-hand panel of figure 1, with negligible contribution from the normal-state excitation spectrum. Such a picture is supported by the dramatic fall in heat capacity at temperatures well below T_{sc}, signalling a loss of normal-state quasiparticle excitations to the susceptibility, due to the opening of a gap on the high-state-density, strongly correlated sheets of the quasiparticle Fermi surface. The condensate now plays a key role *both* in structuring the magnetic response on account of its phase-coherent nature *and* in supplying an alternative channel of excitation.

In the debate between these two fundamental mechanisms we consider only the response of a singlet condensate since this is the symmetry compatible with the existing array of thermodynamic, transport and tunnelling data ([15–18] and references therein). Alternatives, based on spin-triplet pairing wavefunctions, which do not appear to be supported by thermodynamic and transport measurements, are not considered in detail here even though they may be capable of explaining some features of the neutron data [5, 16].

The myriad of possible analyses thus reduces to a choice between those two scenarios, and a discussion is given of each type. The models considered here encompass a mean-field coupling of two hypothetical components to χ with a simple, real, feedback parameter. The fundamental differences that remain are then the assumed dominance by excitations of the normal state both above and below T_{sc} [5, 16], whilst in [3, 19–22], at the lowest temperatures the major contribution at low energy transfer is from the condensate. The second point of divergence is the assertion that the nodal symmetry of the superconducting energy gap is considered as being determined purely by the local point group symmetry [5, 16], or that explicit account is to be taken of the underlying lattice symmetry and Fermi surface topology [3, 7, 19–22]. These primary choices then dictate the relative amplitude and phase symmetry of both the excitations of the normal-state quasiparticles and the excitations out of the condensate that form the basis of further analysis.

To commence an analysis with equations (3) we note that both the absence of strong thermal dilation effects and significant changes in magnetic moment on passing below T_{sc} indicate that the low-energy quasiparticle phasing, intrinsic to the condensed state, does not greatly alter either the lattice or the magnetic potentials which determine the Fermi surface[7]. Hence the spatial symmetries of excitation matrix elements, as expressed through the Lindhard functions, should not change. In particular, the resonant magnetic wavevector is anticipated to remain constant[8]. The important aspect of the cross section is the introduction of an energy gap in the denominator of the Lindhard sum for χ_c. The normal-state quasiparticle response χ_{qp} does not acquire the corresponding gap and is expected to remain quasielastic in form below T_{sc}[9].

2.3. *Symmetry of* $\Delta(\mathbf{k})$

2.3.1. Spatial symmetry of $\Delta(\mathbf{k})$. The partition of the measured response in (q, ω) space between normal and condensate excitations is most readily made on examination of the dynamical susceptibility for excitations with minimal energy. A semi-quantitative analysis illustrates the point. First we note that at low temperatures the Pauli principle restricts attention to those excitations of the condensate that involve quasiparticle states lying close to the Fermi surface, $(1 - f(\mathbf{k}+\mathbf{q}) - f(\mathbf{k})) \approx 1$. The contribution of the normal-state quasiparticle to the bare susceptibility becomes progressively weaker on lowering the temperature. Examination of the phase-coherence factor in χ reveals that, for excitations of the lowest energy, where the quasiparticle excitation energies $\xi(\mathbf{k}) = \xi(\mathbf{k}+\mathbf{q}) = 0$, the phase-coherence bracket reduces to $1 \pm \cos[\Phi(\mathbf{q})]$ for the normal and condensate fractions with the upper and lower sign, respectively. Hence, for a significant condensate response at wavevector $\mathbf{q}$, $\Delta(\mathbf{k}+\mathbf{q})$

[7] Differences in energy scale, $T_{sc} \ll T_N$, are insufficient in themselves to ensure immunity from the potential changes induced by phasing correlations; the issue must be established empirically.

[8] This point is experimentally verified in figure 12 of the preceding topical review [1], where no new response, significant on the scale of the emergent pole at $\mathbf{Q}_0$, arises below T_{sc} at low energy transfer.

[9] An alternative point of view is expressed in [16] where normal quasiparticle excitations appear to be responsible for the low-frequency pole below T_{sc}. The mechanism whereby the Lindhard function for normal-state excitations acquires a pole is not made explicit. This normal pole is, however, held to be a measure of $\Delta(\mathbf{k})$, subject to a strong coupling renormalization in position and width with a crystalline-electric-field (exciton) mode.

must be the negative of $\Delta(\mathbf{k})$, at least over a sizable portion of the zone. From the observation of an *inelastic* and *enhanced* scattering in the superconducting phase of UPd_2Al_3 around the antiferromagnetic reciprocal lattice vectors (i.e. $\mathbf{q} = \mathbf{Q}_0$ in equation (3)) the inferences are:

(i) that the dominant contribution arises from the condensate which
(ii) has a gap, $\Delta(\mathbf{k})$, displaying sign inversion on translation by $\mathbf{Q}_0$ over a major part of the zone. That is, the observed scattering suggests a spatially *antisymmetric* form of Δ be taken, $\Delta(\mathbf{k}) = -\Delta(\mathbf{k} + \mathbf{Q}_0)$ [3, 7, 19–22].

The sign difference between the coherence factor for excitations from the normal, $1 + \cos[\Phi(\mathbf{q})]$, and condensate, $1 - \cos[\Phi(\mathbf{q})]$, fractions is of further experimental importance. In antiferro-periodic symmetry, i.e. $\phi(\mathbf{q}) = \pi$ for $\mathbf{q} = \mathbf{Q}_{\mathrm{afm}}$, the coherence factor effectively *eliminates* normal state scattering at $\mathbf{Q}_0$, so no quasielastic response remains enabling a clear definition of the inelastic nature of the condensate response (see the central and right-hand panels of figure 1). In model systems having a jellium, $\mathbf{Q}_0 = 0$, or lattice periodic translation symmetry the phasing enhancement from the coherence factor reverses. In this case the susceptibility amplification has a maximum for the gapless quasiparticle excitations from the normal state and is small for excitations involving the condensate.

Expanding briefly on this point we recall that wavevectors $\mathbf{Q}_{\mathrm{mag}}$ spanning regions of high density of electronic states at the Fermi surface yield an enhanced susceptibility, and hence neutron cross section, through the Lindhard function. In order for experiments to benefit from this in the identification of $\Delta(\mathbf{k})$ below T_{sc}, the wavevector of maximal phasing of the condensate fraction must be commensurate with $\mathbf{Q}_{\mathrm{mag}}$. Maximizing $1 - \cos[\Phi(\mathbf{q})]$ requires $\phi(\mathbf{Q}_{\mathrm{mag}}) = \pi$. In a jellium approximation, or the presence of ferromagnetic correlations, this implies $\Delta(\mathbf{k}) = -\Delta(\mathbf{k} + \mathbf{G})$, where $\mathbf{G}$ is a vector of the reciprocal lattice, forcing $\Delta(\mathbf{k}) = 0$. Conversely, for a minimal period of $\Delta(\mathbf{k}) = \Delta(\mathbf{k} + \mathbf{G})$, in which case the condensate fraction gives zero response at the maxima of χ, a condensate may coexist with the ferromagnetic correlations. In this case the aspect of lattice translation invariance becomes trivial with the nodal symmetry of $\Delta(\mathbf{k})$ being determined by the crystallographic point group. As noted, in such materials the *normal* quasiparticle contribution is enhanced on passing below T_{sc}. This leads to the possible observation of a modification in the quasielastic intensity and lineshape on the energy scale of $\Delta(\mathbf{k})$. On account of the low energies involved, at the experimental level this may be seen as a weak change in the (ferro)magnetic Bragg peak intensity. Finally, in materials where the condensate phasing wavevector is incommensurate with that of the susceptibility, one anticipates only indistinct signs of the transition below T_{sc}.

2.3.2. Spectral form of $\Delta(\mathbf{k})$. The condensate response at $\mathbf{Q}_0$, under the constraint of a favourable phase-coherence symmetry and within the restriction that we consider only minimal excitation states having $\xi(\mathbf{k}) = 0$, is given by the imaginary part of the Pauli restricted summation $\sum_k^{\sim} 1/(\omega - 2|\Delta_k| + \mathrm{i}\Gamma)$. This is a sum of complex Lorentzian amplitudes centred at $2|\Delta_k|$ and of width Γ. In the case that $\Delta(\mathbf{k}) = -\Delta(\mathbf{k} + \mathbf{Q}_0)$ with $|\Delta_k|$ independent of $\mathbf{k}$, i.e. the square wave representation of the antiferro-periodic nodal gap state (previously referred to as the 'antiferromagnetic s-wave' state [3]) the response simplifies to a single pole centred at 2Δ of damping, Γ, related to the effective quasiparticle lifetime. The presence of a sizeable gap anisotropy, explicit in some models of the superconducting state [5, 16] would lead to interference over a sum of complex amplitudes giving a spectral form spread in energy and of diminished magnitude. The sharp profile of the condensate response in energy transfer observed in UPd_2Al_3 thus appears to favour the conjecture of an isotropic crystallographic point group symmetry of the energy gap over extended s- or d-wave variants [2–10, 16, 19–22] although detailed calculations are required in each case.

2.3.3. Spin symmetry of $\Delta(\mathbf{k})$. The deduced symmetry of the measured gap function, $\Delta(\mathbf{k}) = -\Delta(\mathbf{k} + \mathbf{Q}_0)$, implies $\Delta(\mathbf{k}) = \Delta(\mathbf{k} + \mathbf{G})$. As a consequence $\Delta(\mathbf{k})$ follows the translation symmetry of the crystallographic Brillouin zone, with the inference that the condensate wavefunction is not sensitive to the magnetic potential that defines the antiferromagnetic unit cell doubling along the hexagonal axis for the spin-polarized quasiparticles. This is, *a posteriori*, consistent with the spin-pairing symmetry underlying equation (3), and in agreement with available experimental data and detailed calculations based on the computed Fermi surface, that the gap function is a spin singlet [2, 7, 17, 18].

Nevertheless, theoretical approaches differ on this issue. Identifying three separate mechanisms of condensation based, respectively, on phonon exchange [24], spin-fluctuation exchange [2, 4, 23] or a novel crystalline-electric-field (CEF) exciton mode, as proposed in Sato *et al* [16] and Thalmeier [5], the results are that:

(i) A phonon-driven condensate will have an associated spin-singlet (even parity) state on account of the Pauli principle and spin invariance under phonon exchange unless higher-order spin–orbit driven effects are invoked. The resulting translation symmetry of the energy gap is that of the crystallographic Brillouin zone.

(ii) The spin-fluctuation condensate may achieve either spin-singlet or spin-triplet states on account of rotational invariance with a translation symmetry of the energy gap periodic in the crystallographic Brillouin zone.

(iii) The CEF-exciton mode of Sato *et al* [16] and Thalmeier [5] is of odd parity (i.e. spin-triplet symmetry) on account of coupling the Cooper spin pair with a local magnetic moment. Cast within the framework of the translation symmetry of the antiferromagnetic unit cell, the model is obliged to place the nodal region in the equatorial plane. In order to satisfy the observed antiferromagnetic repeat wavevector $\Delta(\mathbf{k})$ then requires the orthogonal phase symmetry, i.e. $\Delta(\mathbf{k}) \sim \sin(ck_z)$, where c is the lattice parameter and k_z is the z component of k [3], to that invoked above.

From (i) and (ii), and as a rather robust and general conclusion highlighted by Oppeneer and Varelogiannis [7] in their detailed calculations based on the computed Fermi surface in UPd_2Al_3, the observed symmetry of the gap function is *not* universally tied to any particular pairing mechanism. Thus, for example, Oppeneer and Varelogiannis show that either phonon or spin-fluctuation pairing are capable of producing both s-wave and d-wave condensates with the self-consistent computed gap in both cases following the symmetry of the crystallographic Brillouin zone in agreement with the general comments given above.

In a highly original analysis used to interpret a model cross section by Sato *et al* [16], and made explicit by Thalmeier [5], the basic ingredient is a fragmentation of the 5f shell into a local moment and itinerant state, with the local-moment dynamics described as an excitation of coupled ionic CEF levels. This yields a superconducting state of odd parity (i.e. spin-triplet symmetry) on account of coupling the Cooper spin pair with the magnetic moment, a conclusion apparently in contradiction with the inferences of available thermodynamic and transport data. A central role of the exciton mode in the vicinity of $\mathbf{Q}_0$ is ensured by the condition of its accidental degeneracy with the magnitude of the superconducting energy gap at each $T < T_{sc}$. This assumption appears to require both a substantial superconducting energy-gap maximum at the $\mathbf{Q}_0$ point in the zone and a change in nature of the normal-state response below T_{sc}, apparently at variance with, on the one hand, interpretation of tunnelling measurements [2, 15] and, on the other, the implications of equation (3). Additionally, since the neutron response arising from the condensate is given by a Lindhard sum over all $|\Delta(\mathbf{k})|$ (see equation (3) and the discussion in section 2.3.3), the implied variance of $|\Delta(\mathbf{k})|$ would be expected to result in a broad, weak spectral form as opposed to the strongly enhanced resonance observed in figure 2. The softening of the zone centre CEF level (exciton) on entering the superconducting phase required to generate sufficient feedback enhancement in the low-frequency mode of ~30% is certainly dramatic evidence of T_{sc} in this scenario given it is taken to represent the stable, (quasi)-localized, 5f component of the response. Further, the fundamental assumption of strongly localized 5f levels is difficult to reconcile with the lack of observation of CEF levels in the paramagnetic state [25] and the success of *ab initio* band-structure calculations using the delocalized LSDA approach to reproduce experimental Fermi surface areas, as measured by the de Haas–van Alphen (dHvA) effect, which are often taken as an indication that the 5f levels are largely delocalized [26, 27].

2.4. Tunnelling, Fermi surface topology and $|\Delta(\mathbf{k})|$

The interpretation of tunnelling data underlines the importance of the space group symmetry of the energy gap. As the neutron scattering response reveals, in UPd_2Al_3 the lattice symmetry appears to be determinant, enforcing nodes of the energy gap that are periodic within the Brillouin zone. The additional constraint of Fermi surface topology favours an energy gap with nodal structure in the vicinity of low-density quasiparticle states; a conclusion corroborated in UPd_2Al_3 by the rapid decrease of heat capacity below T_{sc} [17, 18]. Examination of the band structure in UPd_2Al_3 shows that the antiferro-periodic nodal gap state obeys this criterion with the high-state-density 'egg' Fermi surface sheet [26], as identified in both inelastic neutron scattering [19–22] and tunnelling experiments [2, 15], lying close to, but inside, the Brillouin zone boundary [3].

The available measures of $|\Delta|$, from both tunnelling and inelastic neutron scattering at the comparable point in the zone, thus support a gap function with nodes along $\boldsymbol{c}^*$. In other words, with minimal assumption it appears unnecessary to invoke a d-wave point-group character of the energy gap. Indeed, the totally symmetric, A_{1g}, s-wave point group, in conjunction with the Fermi surface as calculated, and substantiated through independent dHvA measurements [27], is able to account not only for the very good correlation in magnitude of the gap along the $\boldsymbol{c}^*$ axis, as determined by both tunnelling and neutron spectra, but also for the observation of many apparently anomalous thermodynamic and transport properties in this material [3].

2.5. Coupling schemes for superconductivity

Whilst valuable new information on the magnitude and symmetry of the energy gap is available, the use of inelastic neutron scattering to determine the coupling mechanism of the condensed state is speculative unless detailed quantitative calculations for the cross section are available. As a basis for discussion, one may ask such exercises to (i) yield a parity of the proposed superconducting paired state in agreement with that inferred from thermodynamic and transport data and (ii) offer a rationale for the appearance of an energy gap with (iii) an enhanced excitation below T_{sc} in the inelastic neutron-scattering spectra at (iv) *one* select point in the Brillouin zone, i.e. of defined translation symmetry, as demonstrated by the extensive mappings of $\chi(q, \omega)$ presented in the preceding topical review [1]. These four basic measures may then serve as a yardstick against which to compare the strikingly different model coupling mechanisms presented [2, 4–10].

3. Conclusion

The purpose of this paper has been to present the minimal requirements of any analysis of the low-temperature magnetic inelastic response in UPd_2Al_3. Inferences on the renormalization of electronic correlations in the superconducting state are made on the basis of differences in neutron-scattering spectral weight on passing below T_{sc}. In any approach, the critical influence of phase correlations that characterize the superconducting ground state on excitations of *both* the normal-state and condensate fractions must be taken into account.

In UPd_2Al_3 the observed differences permit robust inferences of the global gap symmetry, which embody the lattice translation invariance, in addition to estimates of its magnitude at selected points in the Brillouin zone. In this context it is of interest to note that the increasing range of antiferromagnetic quasielastic correlations to ∼100 Å on cooling to T_{sc} (figure 4 in [1]), approaches the estimated coherence length of the condensate [17, 18]. This suggests that the passage to the superconducting critical point may be foreshadowed in the response of the normal phase. When below T_{sc}, the quasielastic scattering apparently vanishes to be replaced by the gapped condensate response (figure 2).

Following the assumptions of the 'second scenario' discussed (right-hand panels of figure 1) the condensate plays a key role both in structuring the magnetic response and in providing an alternative excitation channel. The totally symmetric, A_{1g}, s-wave point group is able to account for a superconducting gap along the c^* axis, as observed by both tunnelling and neutron inelastic experiments, and for many apparently anomalous thermodynamic and transport properties [3].

On the other hand, the neutron data give little information about the detailed pairing mechanism itself [7]. Such inferences rapidly become *highly model sensitive*, both on account of theoretical approximation schemes adopted and numerical complexity in calculation. The situation is further aggravated in cases where the multi-modal nature of $\chi(\omega)$, with the inherent need for some decoupling approximation [3, 16, 19–22], gives parametric uncertainty in positions and widths of the modes.

Further progress will rely on improvements in the experimental technique, discoveries of other model systems and a deeper understanding of the generic problem of coupled order parameters.

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