

Magnetisation dynamics in the normal and condensate phases of UPd₂Al₃

I: Neutron inelastic scattering results

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Abstract:

This paper, I, presents new results from neutron inelastic scattering experiments performed by two independent groups on different single crystals of UPd₂Al₃. The focus is on the experimental position whilst the sequel, II, advances theoretical perspectives. We present a detailed and complete characterisation of the magnetisation dynamics in UPd₂Al₃ as measured by neutron inelastic scattering primarily in the form of extensive surveys in energy-momentum space under a wide range of experimental conditions, and put our observations in context with the selected fraction of experimental data that has been previously published. In this way we emphasize the commonality and robust nature of the data which indicate the intricate nature of the dynamic magnetic susceptibility of this material. We argue that the presence of a strong quasielastic magnetic signal and relatively high superconducting transition temperature, $T_{sc} \sim 1.8$ K, opens a window on the low energy magnetic density dynamics. This yields unique insight into the formation and symmetries of the low temperature ground state which exhibits a microscopic coexistence of antiferromagnetism and superconductivity making UPd₂Al₃ one of the most accessible heavy-fermion superconductors that can be fully characterised by neutron spectroscopy.

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1. INTRODUCTION

The co-existence of magnetism and superconductivity continues to attract the attention of the condensed matter community. It is of particular interest to establish whether the superconducting state is stabilised via a dynamic deformation of the lattice, magnetic or other electronic potential. For both high- T_c and heavy-fermion superconducting materials the discussion has been, and still is, extremely controversial. Three fundamental questions arise: First, is it meaningful to discuss superconductivity and magnetism as two separate phenomena or are they joint manifestations of a novel low temperature ground state? Second, on assuming some reduction of the two aspects may be made, what are the symmetries of the order parameters and finally, can one identify coupling mechanisms that maintain the broken symmetry of the appropriate wave function?

Of the materials that are known to exhibit both ordered magnetism *and* superconductivity, the compound UPd_2Al_3 has an especially interesting place. Initially investigated by C. Geibel and collaborators¹ it has the following favourable properties. First, a simple atomic structure, hexagonal space group P6/mmm ($a = 5.350 \text{ \AA}$, $c = 4.185 \text{ \AA}$), and the possibility to grow stoichiometric, bulk superconducting single crystals of ~ 2 -3 g. Second, a simple antiferromagnetic structure, $T_N = 14.3 \text{ K}$, with ferromagnetic sheets of uranium moments parallel to $[1\ 0\ 0]$ stacked in alternating directions $+ - + -$ along the hexagonal c -axis, see **Fig. 1**, giving an antiferromagnetic wave vector $\mathbf{Q}_0 = (0\ 0\ 1/2)$ reciprocal lattice units (rlu) [2, 3]. Third, superconductivity coexists with antiferromagnetic order below a relatively high temperature of $\sim 1.8 \text{ K}$ giving an energy scale accessible to modern high resolution neutron spectrometers. From the large specific heat and concomitant jump at T_{sc} of $\Delta C = 1.2 \gamma T_{sc}$ ($\gamma = 140 \text{ mJ/mol-K}^2$) [1] it has been suggested that the superconducting ground state evolves out of interactions between heavy quasiparticles at the Fermi surface. Finally, UPd_2Al_3 possesses a set of intriguing physical properties amongst which number, a significant uranium moment $\sim 0.85 \mu_B$ [2, 3, 4], the absence of a Hebel-Slichter peak,⁵ a T^3 dependence of the nuclear-spin relaxation time, T_1 [6], and the power-law behaviour of the specific heat,⁷ all of which have prompted suggestions of unconventional superconductivity.

Of the many techniques available to characterise the spectral magnetic response of this system, neutron inelastic scattering is one of the most powerful giving information on the electronic and nuclear dynamics over temporal (10^{-13} to 10^{-10} s) and spatial ($\sim 400 \text{ \AA}$) scales ideally suited to investigation of both magnetic and superconducting phenomena. A general

formalism, based on linear response theory, relating the cross section to the dissipative component of the magnetic susceptibility ($\text{Im } \chi$), exists for the scattering of the neutron against stable thermodynamic states.⁸ Within its domain of validity, this enables the inference of direct microscopic information on the dynamic evolution of the magnetic quasiparticle–hole excitation spectra in correlated magnetic macrostates. In the superconducting state the response is modified by the dynamical restrictions imposed by the phase correlated condensate.⁹ In addition to the contribution from the quasiparticle-hole excitations of the normal state, the neutron may also couple directly to the superconducting ground state via transitions associated with excitation/condensation of Cooper pairs. As with the normal state excitations, the amplitude of the response depends on the space-time symmetry of the condensate, and, in favourable circumstances, one may observe its signatures through its contribution to the magnetic excitation spectrum. As we shall see, in UPd_2Al_3 this is indeed the case.

Attempts to examine other heavy-fermion superconductors, e.g. UPt_3 [Ref. 10], URu_2Si_2 [Ref. 11, 12], UBe_{13} [Ref. 13], UNi_2Al_3 [Ref. 14], by neutron inelastic scattering have all been hampered by the difficulty that the quasielastic, dynamic, correlations are weak. In the case of ferromagnetic superconductors such as UGe_2 [Ref. 15] relevant experiments to access the superconducting ground state would have to be performed under substantial pressures (10 ~ 15 kbars) and low temperatures $T_{\text{sc}} \sim 0.2$ K. Similar temperature restrictions in the recently discovered ambient pressure systems, ZrZn_2 [Ref. 16] and URhGe [Ref. 17], make neutron inelastic scattering experiments difficult from the viewpoint of the temperatures needed as well as the extremely high resolution required to access fluctuations on the scale of T_{sc} (~ 20 μeV). These problems are compounded by the intrinsic problem of the separation of nuclear and magnetic contributions to the cross section at the ferromagnetic position. Thus, although inelastic scattering has been observed from these materials it cannot be correlated in a simple manner with the dynamics of the changing thermodynamic macrostates involved.

It is the specific combination of physical properties that make a neutron inelastic scattering investigation of the normal to superconducting transition in UPd_2Al_3 possible on account of a dominant *quasielastic* contribution to the magnetisation autocorrelation function at low energies. This opens an experimental window, via high resolution neutron inelastic scattering, on the low energy dynamics that play a key role both in the formation of the antiferromagnetic heavy-fermion state and the simultaneous antiferromagnetic-superconducting ground state.

Résumé of previous work using neutron inelastic scattering:

The first neutron inelastic scattering work on single crystals was at Risø National Laboratory where broad excitations with a strong dispersion along the c^* ($[0\ 0\ 1]$) axis up to ~ 8 meV at the magnetic zone boundary (where the full width half maximum (*fwhm*) is ~ 9 meV) were reported.¹⁸ In the basal plane strongly damped excitations were found, with poles and widths of similar extent, increasing up to ~ 4 meV [18]. These studies, carried out with 0.3 meV resolution (*fwhm*), found no low energy gap in the excitations at the magnetic zone centre, \mathbf{Q}_0 , and no change when the material became superconducting. However, since the energy resolution was on the scale of ~ 3 K, it is perhaps not surprising that no effect was observed below T_{sc} .

Work on polycrystalline material at the ISIS spallation source by Krimmel *et al.*¹⁹ then followed giving an overview of the inelastic response function up to ~ 20 meV. This study gives no evidence for a discrete crystal field level scheme and the principle results of these experiments were, that (a) over the studied range of wave vectors a broad quasielastic contribution was present in the scattering at all measured temperatures with a *fwhm* of 9.8 meV at $T = 25$ K and 22.8 meV at 150 K, and (b) at $T = 25$ K a strong maximum in the scattered intensity with an energy transfer ~ 2.2 meV at $|\mathbf{Q}| \sim 1 \text{ \AA}^{-1}$ was identified.

Experiments on single crystals were made by the Tohoku University group using the JRR-3M research reactor (JAERI, Tokai) [N. Aso, PhD-thesis, Tohoku Univ. 1996; unpublished] which motivated higher-resolution experiments at the Institut Laue Langevin, Grenoble (ILL) in 1996 [Ref. 20]. Around this period a parallel effort started by the group at the Advanced Science Research Centre in JAERI, Tokai, Japan [Refs. 21–22]. Over the following years several papers have been published concentrating on the magnetic response in the vicinity of \mathbf{Q}_0 , the magnetic zone centre, including polarisation analysis, temperature and field dependent studies. This has resulted in a disparate literature, masking rather than highlighting the fundamental importance and remarkable degree of experimental agreement between data collected on different samples by independent experimental groups. A point of much interest has been the exploitation of initial results obtained by Metoki *et al.*²³ with high energy resolution techniques to resolve the significant intensity around a second characteristic wave vector, $\mathbf{Q}^* = (1/2\ 0\ 1/2)$; this aspect, investigated in more detail at the ILL and Paul Scherrer Institute (PSI), led to an alternative perspective on the origin of the $\mathbf{Q}_0 = (0\ 0\ 1/2)$ Bragg peaks²⁴.

In parallel with the experimental program, theoretical efforts have been underway to understand the rather unusual effects reported. Early approaches by Sato *et al.*^{20,25} were followed by the analyses of Bernhoeft *et al.*²⁶⁻³⁰ which yielded, for the first time to our knowledge from neutron inelastic scattering, the symmetry of the energy gap based on the role of the phase coherence intrinsic to the superconducting macrostate. More recently Sato *et al.*³¹ have published an alternative interpretation of the *same* data building on some aspects of the interpretation given in Refs 26-30. Whilst further work³² on tunnelling into carefully prepared films supports the interpretations drawn in Refs. 26-30, various other conclusions on the energy gap symmetry, together with more general remarks about the potential driving the superconductivity³¹⁻³⁵ have also appeared.

In view of the general interest generated by the data from these experiments, which arises from their rich information content with respect to the superconducting energy gap symmetry and magnitude, further experiments using cold and thermal three-axis spectrometers were recently performed. This paper provides a comprehensive coverage of the current experimental situation. Important new data is presented mainly in the form of extensive surveys in energy-momentum space under a wide range of experimental conditions. All comparable data presented are consistent between experiments performed on independent samples at JAERI, ILL, and PSI. In instances where selected parts of data sets have been published previously the original references, wherein more experimental information can be found, are cited.²⁰⁻³⁰

To avoid confounding the data, which stand alone, with possible interpretations, the analytic reduction of the results is deferred to Part II wherein the focus is on the phase coherence and lattice periodicity symmetries and constraints which need to be respected in any given approach. It is hoped that the combination of papers, I and II, may stimulate an interaction between theoretical modelling and further experiments in developing an understanding of the antiferromagnetic superconducting state.

2. EXPERIMENTAL RESULTS

The experiments have been performed on two different samples at JEARI, ILL and PSI respectively. The samples with a nominal composition of $\text{UPd}_{2.02}\text{Al}_{3.03}$ were grown from a melt of high purity elements by the Czochralski method^{37,38}. The crystals have a typical mass ~ 2.5 g, are cylindrical in shape and show a mosaic spread of about 1 degree. Both samples exhibit a superconducting transition at ~ 1.9 K.

A Overview of the effects around the magnetic zone centre $\mathbf{Q}_0 = (0\ 0\ 1/2)$

The trends with reducing temperature at \mathbf{Q}_0 , and the region close to \mathbf{Q}_0 along the c^* direction, are shown in **Fig. 2** [Ref. 26]. The data are taken at fixed $k_f = 1.15 \text{ \AA}^{-1}$ with an energy resolution ~ 0.09 meV (*fwhm*) at the elastic position. A region of this width is excluded in Fig. 2; this removes both the coherent and incoherent elastic scattering and, for this reason, the emerging Bragg peak below T_N does not appear in the figure. Further information on the scale of the Brillouin zone along c^* is given in Figs. 12-14.

A global survey of the dispersion parallel to the hexagonal axis in the vicinity of the antiferromagnetic wave vector \mathbf{Q}_0 at fixed temperature is afforded by the contour plots of Fig. 2, whilst **Fig. 3** gives an overview of the inelastic intensity as a function of temperature at \mathbf{Q}_0 . Inspection of the figures reveals the following effects. On entering the normal antiferromagnetic state at 14 K the response is dominantly quasielastic with a maximum around the incipient antiferromagnetic wave vector. As the temperature is reduced below $\sim T_N/2$ the quasielastic scattering is increasingly concentrated around the point \mathbf{Q}_0 and, over this same temperature range in the normal state, the quasielastic intensity scales approximately with $k_B T$ (Fig. 3) indicating the intrinsic cross section to be more or less temperature independent. In addition to the low energy response at \mathbf{Q}_0 , highlighted via the colour scheme of Figs. 2 and 3, on cooling below $T_N/2$ a distinct, all be it broad, inelastic feature, variously nominated as a spin wave¹⁸ or exciton mode,³¹ appears. For temperatures below 2.5 K a characteristic energy maximum is seen around \mathbf{Q}_0 at ~ 1.5 meV with a *fwhm* of ~ 0.4 meV. For wave vectors, q (where the reduced wave vector, \mathbf{q} , is defined in terms of the scattering vector, \mathbf{Q} , by $\mathbf{Q} = \mathbf{Q}_0 + \mathbf{q}$ and $q = |\mathbf{q}|$), above ~ 0.1 rlu this mode decays into weak space-time correlations of feeble amplitude, see also Figs. 12-14 and [Ref. 18]. In the superconducting phase below $T_{sc}/2$ the low energy, $E < 1.4$ meV, response in the vicinity of \mathbf{Q}_0 is dramatically renormalised. A strong magnetic pole, seen as the diffuse orange area in fig. 3, develops at an energy transfer of about $E \sim 0.4$ meV.

Some of the scans used to compile the contour plots in Figs. 2 and 3 are given in **Figs. 4 and 5** as a function of temperature at \mathbf{Q}_0 and at 0.15 K and 1.8 K as a function of q_l (i.e. along c^*) and q_h (in the basal plane), respectively. The left hand panel of Fig. 4 gives the evolution of the low energy peak in the superconducting phase. The resonant pole remains on heating to ~ 1 K with little change in its amplitude, width or position in accordance with it being a quantum excitation; on further heating the pole collapses becoming indistinguishable from a quasielastic response for $T > 1.75$ K [Ref. 30]. The high frequency, inelastic, part of the excitation spectrum at ~ 1.5 meV for $T < T_{sc}$ is essentially temperature independent below 2.5 K ($T_{sc} \sim 1.8$ K). At higher temperatures, in the normal state, the pole softens (Fig. 3), whilst the quasielastic response rapidly broadens in energy scale to become the dominant feature as shown in Fig. 4, right hand panel.

The effective dispersions of the low and high energy features, at temperatures $T \ll T_{sc}$ and $T \sim T_{sc}$, parallel to the hexagonal axis in the vicinity of \mathbf{Q}_0 , are given in Fig. 5. Both low and high energy modes disperse strongly at 0.15 K with the concomitant collapse of the low frequency amplitude. Whereas, for $T \sim T_{sc}$ (1.8 K), see Fig. 5 (right hand panel), the low energy excitation changes to a quasielastic response of falling amplitude as q_l diverges from \mathbf{Q}_0 , the higher energy inelastic mode appears to retain its form and disperses essentially as at 0.15 K. At an energy transfer of 0.4 meV, corresponding with the peak of the response in the superconducting phase, the relative normal and superfluid state spatial extent of the correlations along c^* may be inferred from the q -scans shown in **Fig. 6**. The widths in the normal and superconducting states correspond to a length scale in real space of ~ 100 Å. Thus one surmises that the slow ($\sim 10^{-11}$ s) antiferromagnetic magnetic correlations, which change strongly on passing below T_{sc} , arise from regions of ~ 100 Å in extent along the hexagonal axis in both the normal and superconducting phases. This figure also demonstrates that, at the given energy transfer, there is no other response in the c^* direction, see also Fig 12.

At comparable temperatures the thermal evolution in the normal state along the hexagonal axis ($0\ 0\ q_l$) and in the hexagonal plane ($q_h\ 0\ 1/2$) are given in **Fig. 7**. The striking feature is the rapid fall off in quasielastic intensity on moving away from \mathbf{Q}_0 , whilst the inelastic (spin wave or exciton) feature around 1.5 meV continues unabated to at least $q_h \sim 0.08$ rlu with a weak dispersion. On heating to $\sim T_N$ the quasielastic scattering remains as the only discernable feature and becomes more extended in q -space (i.e. shorter range in real space).

The nature of the scattering close to \mathbf{Q}_0 is such that, without modelling the magnetic response function, the separation of quasielastic and propagating components is not without ambiguity, see II. As a simplest model independent approach, which implicitly ignores all coupling and damping effects, **Fig. 8** gives the intensity maxima as observed at $T = 2$ K in the form of a dispersion relation. At small values of q_l and q_h two components are resolved, whilst above ~ 0.05 rlu away from \mathbf{Q}_0 , as indicated in the figure, the quasielastic response collapses leaving a distinct dispersive mode with a stiffness differing by $\sim 50\%$ in the two directions.

Given the strong, qualitative, change in character of the low energy response on passing below T_{sc} , Figs. 2 and 3, it is important to establish the nature of the peak occurring for $T \ll T_{sc}$. To this end careful polarisation analysis, **Fig. 9**, has been carried out at \mathbf{Q}_0 at both $k_f = 1.15 \text{ \AA}^{-1}$ and 1.3 \AA^{-1} . Components, longitudinal and transverse with respect to the ordered moment, I_L and I_T , have been extracted from the observed spin flip and non-spin flip intensities I_{SF} and I_{NSF} using the following formulae:

$$I_{SF} = I_T R_{NSF} + I_L R_{SF} + B$$

$$I_{NSF} = I_T R_{SF} + I_L R_{NSF} + B$$

with

$$B = \text{const.} : \quad \text{Background, constant in all scans (6cts/1000mn)}$$

$$R_{NSF} = \frac{I_{NSF \text{ elastic}}}{I_{SF \text{ elastic}} + I_{NSF \text{ elastic}}} : \quad \text{ratio of non-spin flip to total magnetic Bragg intensity}$$

$$R_{SF} = \frac{I_{SF \text{ elastic}}}{I_{SF \text{ elastic}} + I_{NSF \text{ elastic}}} : \quad \text{ratio of spin flip to total magnetic Bragg intensity}$$

giving:

$$I_L = \frac{(I_{SF} R_{SF} - I_{NSF} R_{NSF})}{R_{SF} - R_{NSF}} - B$$

$$I_T = \frac{(I_{NSF} R_{SF} - I_{SF} R_{NSF})}{R_{SF} - R_{NSF}} - B$$

These results establish that the entire dynamical response, i.e. low energy inelastic or quasielastic and high energy inelastic, is predominately spin reversing (i.e. time asymmetric), *transversely polarised* to the magnetic moment and, taken with the $\mathbf{Q} \times \mathbf{M}$ selection of the neutron dipole cross section, polarised in the hexagonal basal plane.²⁶ A longitudinal contribution is not observed below 10 K. This eliminates the possibility that the quasielastic

response of the normal state is destroyed on entering the superconducting phase and replaced by, for example, a phononic contribution. One infers that the dynamical fluctuations observed here, like the time-averaged moment, are confined to the basal plane.

The contour plots in Fig. 2, and the temperature dependence of $\chi''(Q_0, \omega)$ given in Fig. 3, clearly highlight the change in the low energy spectral form on passing below T_{sc} with the *inelastic* signature of the superconducting state being *qualitatively* different from the quasielastic response in the normal state. Thus, whilst the slow correlations, Fig. 6, are approximately constant in spatial extent, remain transversely polarised, Fig. 9, and strongly focused around \mathbf{Q}_0 , the internal dynamics rearrange with the evolution of an excitation gap in the magnetic response. However, in contrast with the thermally excited quasielastic scattering of the normal state, this emergent, inelastic response lies significantly above $k_B T$, indicative of quantum excitation. It exhibits a strong dispersion in the vicinity of \mathbf{Q}_0 both along the hexagonal axis and in the basal plane as displayed in Fig. 5.

It has been noted in Fig. 3 that for $T_{sc} < T < \sim T_N/2$ the dynamical susceptibility is approximately constant at \mathbf{Q}_0 . This enables a comparison to be made between the calculated response expected on cooling to low temperature and that measured. The result is given in Fig. 4 (bottom curve on left hand side), where the solid line giving the anticipated low temperature behaviour clearly is at variance with the data at low energy transfer both in form and amplitude. As discussed in Part II, the excess intensity in the low temperature experimental data has been taken as a strong indicator for an association of this mode with the superconducting gap function.

That the low energy inelastic feature is related to the superfluid state is further substantiated by its progressive quenching both on heating at zero field, Figs. 3 and 4, and, at $T = 0.4$ K, under an applied magnetic field, **Fig. 10**. The collapse in both pole position and intensity of the low energy inelastic feature, centred around 0.4 meV at $B = 0$ T, around B_{c2} ($= 3.6$ T) is strong support for the origin of the low energy inelastic signal being the excitation of quasiparticles out of the paired superconducting ground state. However, whilst both heating above T_{sc} and magnetic fields greater than B_{c2} suppress the peak, differences remain. Notably, on heating at zero field, the inelastic response of the condensate is stable to $T_{sc}/2$ and then replaced by a quasielastic component³⁰. The behaviour under field is less clear on account of the available resolution and is currently subject of further investigation. The data and magnetic field contour plot of Fig. 10 for $T \ll T_{sc}$ suggest that the collapse of the condensate response starts at the lowest fields and is almost complete for $B \sim B_{c2}/2$.

Despite the dramatic changes in the low energy excitation spectrum in the superconducting phase, the higher energy peak at 1.5 meV transfer differs little in its presentation from that in the normal state, see Fig. 4. It remains transversely polarised, Fig. 8, and focused around \mathbf{Q}_0 with a strong dispersion both along and perpendicular to the hexagonal axis, Fig. 5. Inferences based on this high energy inelastic feature depend critically on the model used and in some scenarios may lead to quantitative changes in the inferred energy pole and width below T_{sc} .³¹ Such details, however, are not robust features of the data analysis. They depend *sensitively* upon the modelling and are crucially dependent on the fact that all features both above and below T_{sc} are on the scale of the experimental resolution in q . Any meaningful parameterisation must include the evident dispersion and fit all data under a given thermodynamic condition simultaneously.

Finally, the response measured in different Brillouin zones scales uniformly at all measured energy transfers as shown in **Fig. 11**, where the dispersion parallel to the hexagonal axis in the vicinity of \mathbf{Q}_0 is presented in both the $(0\ 0\ q_i)$ and $(1\ 0\ q_i)$ zones. One may therefore attribute the *entire* response to states of similar spatial localisation. From its extent in momentum space one concludes the states to be spatially compact. The global intensity reduction by a factor of ~ 2.2 between the two zones is consistent with the uranium form factor and indicates that the observed intensity may be attributed to magnetic polarised (uranium) states of $5f$ symmetry.²⁷ This, together with the results of polarisation analysis and the field dependent studies refutes any suggestion that a magnetic quasielastic component disappears to be replaced by an inelastic phononic or charge density contribution that becomes prominent below T_{sc} .

To summarise, the magnetic response close to \mathbf{Q}_0 comprises: (i) a quasi-elastic response or a very low energy pole which are indistinguishable within the available energy resolution at all temperatures $T > \sim T_{sc}/2$, (ii) an inelastic (spinwave or exciton) response in both the normal and superconducting antiferromagnetically ordered states $T < T_N$ and (iii) the dramatic growth of a dominant inelastic feature at energies ~ 0.4 meV in the superconducting phase temperatures below $T_{sc}/2$.

B Overview of effects across the Brillouin zone:

The response across the Brillouin zone at 0.15 K for energy transfers up to of 4 meV ($k_f = 1.3 \text{ \AA}^{-1}$) is shown in **Fig. 12**. The strong localisation of scattering around \mathbf{Q}_0 in the $(0\ 0\ q_i)$ direction is evident, as is the complex form of the response in the basal plane $(q_h\ 0\ 1/2)$,

together with the subsidiary maximum at the position $\mathbf{Q}^* = (1/2 \ 0 \ 1/2)$, first reported by Metoki *et al.*²³ For temperatures at and below T_N the intensity at the \mathbf{Q}^* position has an energy gap, i.e. no quasielastic term, and exhibits no observable change as the temperature is lowered through T_{sc} (shown further below). The maximum intensity at \mathbf{Q}^* appears at an energy transfer of ~ 3 meV, and this is further illustrated in **Fig. 13**, where the data, taken at 1.5 K with $k_f = 2.662 \text{ \AA}^{-1}$ extend to 6 meV transfer. This figure also illustrates the strong dispersion of the spin wave in the $(0 \ 0 \ q_l)$ direction and the broad extent of the scattering around \mathbf{Q}^* in the $(q_h \ 0 \ 1/2)$ direction. The response around \mathbf{Q}^* is highlighted in the individual q -scans at different energy transfers (using different spectrometers) of **Fig. 14**, where, as remarked above, at low energy transfer (bottom scan in Fig. 14) there is no observable (quasielastic) response. The spatial extent of magnetisation correlations centred around \mathbf{Q}^* are estimated from this figure, over the broad range of energy transfer, $0.6 < E < 2.4$ meV, to be approximately isotropic and in the range of $\sim 20 \text{ \AA}$.

Figure 15 emphasizes the difference in response at 0.2 K between \mathbf{Q}_0 and \mathbf{Q}^* in the superconducting state: at \mathbf{Q}^* there is no observable change in response on entering the superconducting state in sharp contrast with the strong time correlations around \mathbf{Q}_0 which are also seen in the mappings of Fig. 2 for \mathbf{Q}_0 at $k_f = 1.15 \text{ \AA}^{-1}$ and Fig. 12 for both \mathbf{Q}_0 and \mathbf{Q}^* at $k_f = 1.3 \text{ \AA}^{-1}$. As Fig. 15 indicates, the response at \mathbf{Q}^* is little influenced by the quantum correlations induced by the coherence of the superfluid state. The absence of an active role of the superconducting phase coherence for excitations of high energy c.f. T_{sc} is not unexpected⁹ and has already been noted for the 1.5 meV response at \mathbf{Q}_0 . On short time scales the dynamic correlations are expected to average out the slow phase-field coherence associated with the pairing potential correlations of the condensate. Furthermore, the absence of an emerging condensate response at \mathbf{Q}^* below T_{sc} underscores both the stability of the antiferromagnetic correlations in the phase coherent state and the axial gap symmetry along c^* [30-36].

Previous work found the cross section for modes propagating in the basal plane to be poorly defined in momentum and energy transfer at all temperatures below T_N .¹⁸⁻²³ The broad response at \mathbf{Q}^* , which has a typical energy scale of 35 K, is shown in the contour plots of **Fig. 16**, which give the scattered intensity across the zone for three temperatures, 2.5, 12 and 20 K. A similar build up of intensity for wave vectors around $|\mathbf{Q}^*|$ is noted in the report of Krimmel *et al.*¹⁹ by the time of flight technique on polycrystalline material where an enhanced response at 25 K occurs for $|\mathbf{Q}| \sim 1 \text{ \AA}^{-1}$ with a typical energy ~ 2.2 meV and width

0.75 meV (*fwhm*). There is no equivalent enhancement around \mathbf{Q}_0 at these elevated temperatures in agreement with the finding of Fig. 7. **Fig. 17**, left hand panel, extends the observations from 20 K, as in the mappings of Fig. 16, to considerably higher temperatures in the paramagnetic regime at respectively higher energy transfer. Under these conditions, above T_N , quasielastic scattering of similar intensity extending to at least 8 meV is present at both \mathbf{Q}_0 and \mathbf{Q}^* . As shown here at 2 K and in the mappings of Fig. 16, only at and below ~ 20 K does the signal at \mathbf{Q}^* become inelastic with a maximum at about 3 meV and a range of ~ 8 meV. However, as the right hand panel in Fig. 17 and Fig. 15 show, at and below T_N for the smallest energy transfer measured, ~ 0.2 meV, the situation reverses. Here the enhancement occurs at \mathbf{Q}_0 with no long time correlations building up at \mathbf{Q}^* even in the neighbourhood of T_N . This lack of a low energy response in the vicinity of \mathbf{Q}^* may be contrasted with the incommensurate ordering in the isoelectronic compound UNi_2Al_3 close to \mathbf{Q}^* at $(1/2 \pm 0.11 \ 0 \ 1/2)$ [Ref. 39].

In summary, at \mathbf{Q}^* and low temperature, there is an inelastic response broad both in energy and wave vector transfer which becomes quasielastic for temperatures above 20 K. However, there is no quasielastic feature at any temperature in the normal antiferromagnetic phase or low energy inelastic response analogous to that seen around \mathbf{Q}_0 well below T_{sc} .

3. DISCUSSION

An overview of the inelastic neutron scattering data collected at ILL, JAERI and PSI on the low temperature magnetic response in UPd_2Al_3 has been given primarily in the form of extensive maps in energy and momentum space. These maps encompass the two major symmetry directions of the reciprocal lattice, parallel and perpendicular to the hexagonal axis over a wide range of temperatures extending from 150 mK to well above T_N . The dynamical response of UPd_2Al_3 is strongly structured both in momentum and energy with key information on the nature of the cross section contained in the polarisation, form factor, temperature and field dependence of the signal close to the magnetic zone centre $\mathbf{Q}_0 = (0 \ 0 \ 1/2)$.

In the normal antiferromagnetic phase there are *two* distinct features in the energy spectrum in the vicinity of the antiferromagnetic wave vector as illustrated in Fig. 2. First, the quasielastic component, which concentrates around \mathbf{Q}_0 as the temperature is lowered through T_N and remains as a major feature of the excitation spectrum down to T_{sc} . Second, the

dispersive spin wave like¹⁸ (exciton) mode which persists essentially unchanged in form from $T_N/2$ to the lowest temperatures. The uniform scaling of both features in different Brillouin zones, Fig. 11, suggests they may be associated with the quasiparticle states that give rise to the heavy-fermion like properties of this material.

On approaching T_{sc} from above we observe another important feature associated with the quasielastic scattering around \mathbf{Q}_0 . Its correlation length (as measured along the [001] axis) increases on descending below T_N until, at temperatures just above T_{sc} , the correlation length of this scattering is about 100 Å, i.e. close to the estimated pairing coherence length of the condensate¹, Figs. 6, 7. It may be conjectured that, at temperatures well below T_{sc} the large entropy removal could be associated with the reduction of this response and its replacement by a gapped condensate contribution. However, experiments below T_{sc} at 0.4 K and \mathbf{Q}_0 for fields up to 4 Tesla²², reveal the inelastic pole associated with the superconducting condensate to be progressively quenched in amplitude at fixed frequency even by modest fields ($B \leq B_{c2}/2$), Fig. 10. This appears at variance with its behaviour on heating in zero field, where up to $T_{sc}/2$ it is stable in amplitude and form. These apparent differences, of fundamental importance, are under current investigation. In the similar temperature regime, the stable spin wave like mode, which has been identified with the existence of a van-Hove type singularity at ~ 1.4 meV in tunnelling experiments performed with injection parallel to the hexagonal axis³², has been invoked to provide a pairing mechanism in some models of the superconducting state.³¹⁻³⁴

In addition to this rich energy structured response in the vicinity of \mathbf{Q}_0 , there is a secondary maximum at the wave vector \mathbf{Q}^* , which persists from 150 mK in the antiferromagnetic-superconducting state to above T_N in the paramagnetic phase as illustrated in Figs. 12-17. The detailed implication on thermodynamic properties of having such multiple wave vector maxima remains unclear, although there has been a proposal that the nominally ordered state in UPd₂Al₃ remains dynamic in nature on account of the \mathbf{Q}^* mode. In this respect, the contrasting q -space local response at \mathbf{Q}_0 and the q -space spread form at \mathbf{Q}^* , identified in the present studies, may have a more general bearing on the existence of an antiferromagnetic-superconducting ground state.²⁴

The task of understanding a coherent antiferromagnetic-superconducting ground state remains a major challenge in condensed matter physics. In the interim, we hope the rich and robust nature of the data on UPd₂Al₃ presented here will stimulate further experiments and discoveries of other model systems. In Part II we complement these studies with a critical

appraisal of the assumptions, scope and limits inherent in analyses of inelastic neutron scattering data and the modelling of the magnetic response function.

4. ACKNOWLEDGEMENTS

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Figure Captions

Fig 1: (colour online) The crystallographic and magnetic structure of UPd₂Al₃. The large circles represent the positions of uranium ions with the bold arrows marking the relative directions of the magnetic moments. The smaller red circles in the same planes represent the positions of the palladium ions whilst the smallest blue circles, in the intercalating plane, represent the aluminium ions.

Fig 2: (colour online) Contour plots of the normalised intensity (counts/monitor) at four temperatures (as marked) as a function of $\mathbf{q} = (0 \ 0 \ q)$ and neutron-energy loss with final wave vector $k_f = 1.15 \text{ \AA}^{-1}$ (energy resolution: $fwhm \sim 0.09 \text{ meV}$). In this and succeeding contour maps the dots indicate the data collection points used in construction of the map. The smearing parallel to the abscissa is the result of graphical interpolation between the constant- q (i.e. parallel to ordinate) scans. On the energy transfer scale red and blue arrows, respectively, mark the energies corresponding to the antiferromagnetic, T_N , and the superconducting, T_{sc} , transitions with the sample temperature, T indicated by black arrows. The colour scheme, designed to highlight the behaviour around T_{sc} , leads to a saturation at the lowest energy transfers for $T > 7 \text{ K}$ (for details see Figs. 4 and 5 on semi-logarithmic scales). The cross section at the smallest energy transfers is inaccessible due to incoherent elastic scattering, and, at $\mathbf{Q}_0 = (0 \ 0 \ 1/2)$, due to the developing antiferromagnetic Bragg peak for $T < T_N$. Attention is drawn to the reduction in q -space extent of the scattering at low energy on lowering the temperature, and the appearance of a gap in the low energy response in the superconducting phase with the growth of two maxima in the spectral response below T_{sc} as brought out in detail with the semi-logarithmic plots of Figs. 4 and 5. Data taken on IN14 [26].

Fig. 3: (colour online) Contour plot of the intensity at \mathbf{Q}_0 as a function of temperature and energy transfer with $k_f = 1.15 \text{ \AA}^{-1}$. Marked on the plot are the energies of the characteristic temperatures T_{sc} and T_N and the line $E = k_B T$ to indicate the approximate division between thermal and quantum induced fluctuations. Three features are evident: (i) The scaling of the quasielastic scattering $\sim k_B T$ for $T_{sc} < T < T_N$ (red, triangular-shaped area), (ii) the growth of an inelastic (spin wave or exciton) response with an intensity maximum at increasing energies on lowering temperature, reaching an energy of $\sim 1.5 \text{ meV}$ at low temperatures and showing no apparent change at T_{sc} (green arc) and (iii) the strong inelastic response in the superconducting phase below T_{sc} (diffuse orange area). Data taken on IN14 [26].

Fig 4: The temperature dependence of the inelastic response at \mathbf{Q}_0 . Left hand panel for $T < T_{sc}$ and right hand panel for $T > T_{sc}$. Note the alternating logarithmic intensity scales displaced by $1/2$ decade as indicated on left and right hand ordinates; open symbols refer to left hand and closed ones to right hand scales, respectively. For $T \ll T_{sc}$ two clear inelastic features are resolved and indicated by vertical arrows. The low energy response collapses above 1.5 K into the elastic peak and no clear statement on its nature (i.e. quasielastic or inelastic) may be made. The upper, inelastic peak maintains an essentially fixed energy and intensity for all $T < T_{sc}$. Above T_{sc} , the low energy response appears as a quasielastic pole with the higher energy spin wave like feature collapsing smoothly to be indistinguishable from the quasielastic response above 8 K . The solid line is smooth

fit to 1.75 K data (top left hand panel) and this line has been scaled by the Bose factor (with a constant background subtracted) and overlaid over the 0.15 K and 2.5 K data. At 0.15 K this procedure clearly fails to reproduce the data. The horizontal bar indicates the instrumental resolution. Data taken on IN14 with $k_f = 1.15 \text{ \AA}^{-1}$.

Fig. 5: The dispersion of the inelastic response along the c^* axis. Left hand panel at 0.15 K and right hand panel at 1.8 K, i.e. close to T_{sc} . Note logarithmic vertical scale and the zero level of successive scans are displaced by 1 decade for clarity. Both inelastic features disperse significantly for $T \ll T_{sc}$. Whilst the low energy response changes to a quasielastic form on passing to higher temperatures the high energy response is largely unaffected in either intensity or dispersion. Data taken on IN14 with $k_f = 1.15 \text{ \AA}^{-1}$.

Fig. 6: Neutron intensities as a function of q_l along the c^* axis taken at constant energy transfer of 0.4 meV in both the normal and superconducting states. The widths correspond to a correlation length in real space of $\sim 100 \text{ \AA}$. At both 0.45 K and 2 K the response is confined to the immediate vicinity of \mathbf{Q}_0 . Data taken at JAERI with incident wave vector fixed at $k_i = 1.5 \text{ \AA}^{-1}$ with a corresponding energy resolution of $\sim 0.2 \text{ meV}$ (*fwhm*).

Fig. 7: Comparison of the scattering in the hexagonal plane ($q_h \ 0 \ 1/2$) (left) and along the hexagonal c^* axis ($0 \ 0 \ q_l$) (right) at different temperatures in the normal state. Note logarithmic vertical scale and different steps in reciprocal space (a reciprocal lattice unit corresponds to $a^* = 1.355 \text{ \AA}^{-1}$ and $c^* = 1.500 \text{ \AA}^{-1}$ along the two axes). Only for temperatures $\sim T_N$ does the quasielastic response become more isotropic and extend significantly out into the zone in both the basal plane and along the hexagonal axis. Data taken on IN14 with $k_f = 1.15 \text{ \AA}^{-1}$.

Fig. 8: Plot of intensity maxima from scans with $T \sim 2 \text{ K}$ presented as a dispersion relation. The closed symbols indicate data from scans in which a single maximum is observed. Open symbols indicate regions where two features are observed in energy scans. Note that for $T < T_{sc}$ the quasielastic response is replaced by a low lying excitation, but without any effect on the inelastic feature at 1.5 meV. Away from \mathbf{Q}_0 , the grey area of $\pm 0.7 \text{ meV}$ indicates the region over which the intensity has at least 50% of its peak value. The dashed line corresponds with a stiffness of $14.6 \text{ meV} \cdot \text{\AA}$ in the c^* direction (left hand panel) and $10.5 \text{ meV} \cdot \text{\AA}$ in the basal plane (right hand panel). The abscissa are scaled to accommodate the different a and c axis lattice parameters.

Fig. 9: The lower plots show the transverse (solid points) and longitudinal (open points) response at \mathbf{Q}_0 for $T = 0.15 \text{ K}$ and $T = 10 \text{ K}$ obtained by polarisation analysis as explained in the text. The inelastic response is polarised transverse to the magnetic moment vector both above and below T_{sc} . The shape is similar to the response obtained without polarisation analysis shown in the upper panels.

Fig. 10: (colour online) Overview of intensity at \mathbf{Q}_0 for $T = 0.4 \text{ K}$ measured as a function of applied magnetic field up to 4 Tesla. The lower panel is a contour plot made from the five scans shown in the upper part of the figure. The upper critical field $B_{c2} = 3.6 \text{ T}$ is indicated by the blue arrow. For $B \ll B_{c2}$ two clear inelastic features are resolved and indicated by vertical arrows. Data taken at JAERI with $k_f = 1.5 \text{ \AA}^{-1}$ [13,14].

Fig. 11: Dispersion of both the low energy and high energy inelastic modes at $T = 0.15$ K in the superconducting state. Left hand panel for $\mathbf{Q} = (0\ 0\ q_i)$ and right hand panel $\mathbf{Q} = (1\ 0\ q_i)$. The solid line shown in the $(1\ 0\ 0.50)$ scan is the fit to the scan at $(0\ 0\ 0.50)$ reduced by the factor 2.2 (with a constant background subtracted), which is expected from the uranium form factor (squared) difference at these two wave vectors. Data taken on IN14 with $k_f = 1.15\ \text{\AA}^{-1}$.

Fig. 12: (colour online) Contour map at 0.15 K showing the response at relatively low energy transfer across the Brillouin zone. The magnetic zone centres (\mathbf{Q}_0) are $(0\ 0\ 1/2)$ and $(1\ 0\ 1/2)$. In the $(0\ 0\ q_i)$ direction (left hand panel) the response is centred around \mathbf{Q}_0 , whereas it is more complex in the $(q_h\ 0\ 1/2)$ direction. This figure shows the secondary maximum in the inelastic response at the position $\mathbf{Q}^* = (1/2\ 0\ 1/2)$. The abscissa are scaled to accommodate the different a and c axis lattice parameters. The colour scheme, designed to highlight the behaviour around \mathbf{Q}^* , leads to a saturation close to \mathbf{Q}_0 (for details see Fig. 14 on semi-logarithmic scales). The cross section at the smallest energy transfers is inaccessible due to incoherent elastic scattering, and, at $\mathbf{Q}_0 = (0\ 0\ 1/2)$, due to the antiferromagnetic Bragg peak. Data taken on IN14 with $k_f = 1.3\ \text{\AA}^{-1}$.

Fig. 13: (colour online): Contour plots of the intensity across the zone as determined with $k_f = 2.662\ \text{\AA}^{-1}$ (where the resolution is ~ 1 meV (*fwhm*)). Note the inelastic dispersive feature emanating from the $\mathbf{Q}_0 = (0\ 0\ 1/2)$ position, and the response at $\mathbf{Q}^* = (1/2\ 0\ 1/2)$, which is centered at about 3 meV and extends to ~ 6 meV. Data taken on IN8.

Fig. 14: Constant E -scans across the zones taken on IN8 with $k_f = 2.662\ \text{\AA}^{-1}$ (top panel) and IN14 with $k_f = 1.3\ \text{\AA}^{-1}$ (lower panel). Note (i) the dispersive feature starting at \mathbf{Q}_0 shifted further out in the zone at higher energy transfer and (ii) that at low energy transfer there is no observable response at \mathbf{Q}^* . Since, in this temperature range, only the spectral features around \mathbf{Q}_0 are strongly temperature dependent, the different temperatures are not crucial in discussing features away from \mathbf{Q}_0 . Note logarithmic vertical scale and that the zero level of successive scans are displaced for clarity.

Fig. 15: Constant q -scans at \mathbf{Q}_0 and \mathbf{Q}^* taken below T_{sc} showing the absence of any features in the response at \mathbf{Q}^* up to an energy transfer of 2 meV. Data taken on IN14 with $k_f = 1.15\ \text{\AA}^{-1}$.

Fig. 16: (colour online) Inelastic response across the zone from $(1/2\ 0\ 1/2)$ to $(1\ 0\ 1/2)$ at three temperatures. Note the response (green island) at \mathbf{Q}^* persists with approximately constant intensity both in zero point and thermal excitation from low temperature to well above T_N . The intensity recorded at $(1\ 0\ 1/2)$ marks an equivalent \mathbf{Q}_0 position. Data taken at PSI with $k_f = 1.5\ \text{\AA}^{-1}$.

Fig. 17: Left hand panels: constant q -scans at \mathbf{Q}_0 and \mathbf{Q}^* at different temperatures. Above T_N , the response persists to at least 8 meV in energy transfer. Quasielastic scattering is present at both positions for $T_N < T < 80$ K but only persists below T_N at \mathbf{Q}_0 , see right hand panel and Fig. 15. Note the maximum at finite energy transfer in both cases below T_N . The data below 1 meV have been suppressed since they fall within the (elastic)

resolution window of the spectrometer. Data taken on IN8 with $k_f = 2.662 \text{ \AA}^{-1}$. Right hand panel: Temperature dependence of the low energy response at the two positions \mathbf{Q}_0 and \mathbf{Q}^* taken at 0.2 meV energy transfer. Note that the response is strongest at T_N (marked by arrow) at \mathbf{Q}_0 with negligible temperature dependence at \mathbf{Q}^* . Data taken at PSI with $k_f = 1.15 \text{ \AA}^{-1}$.

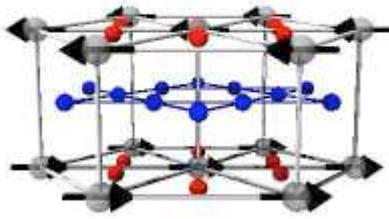


Fig. 1

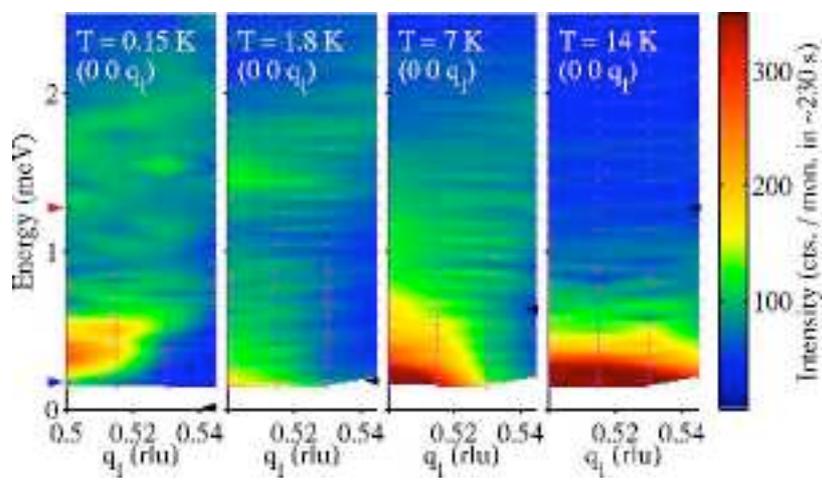


Fig. 2

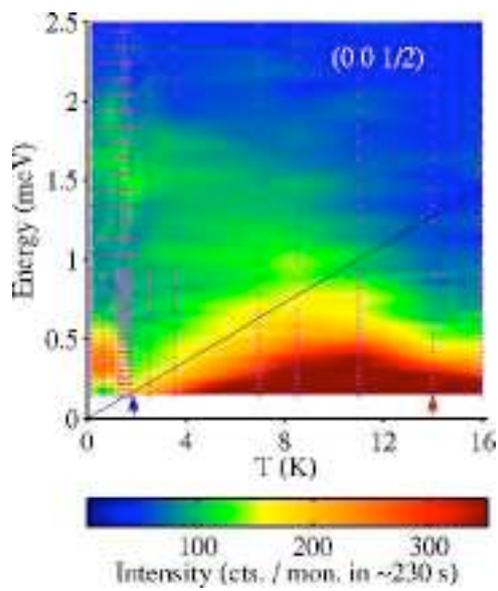


Fig. 3

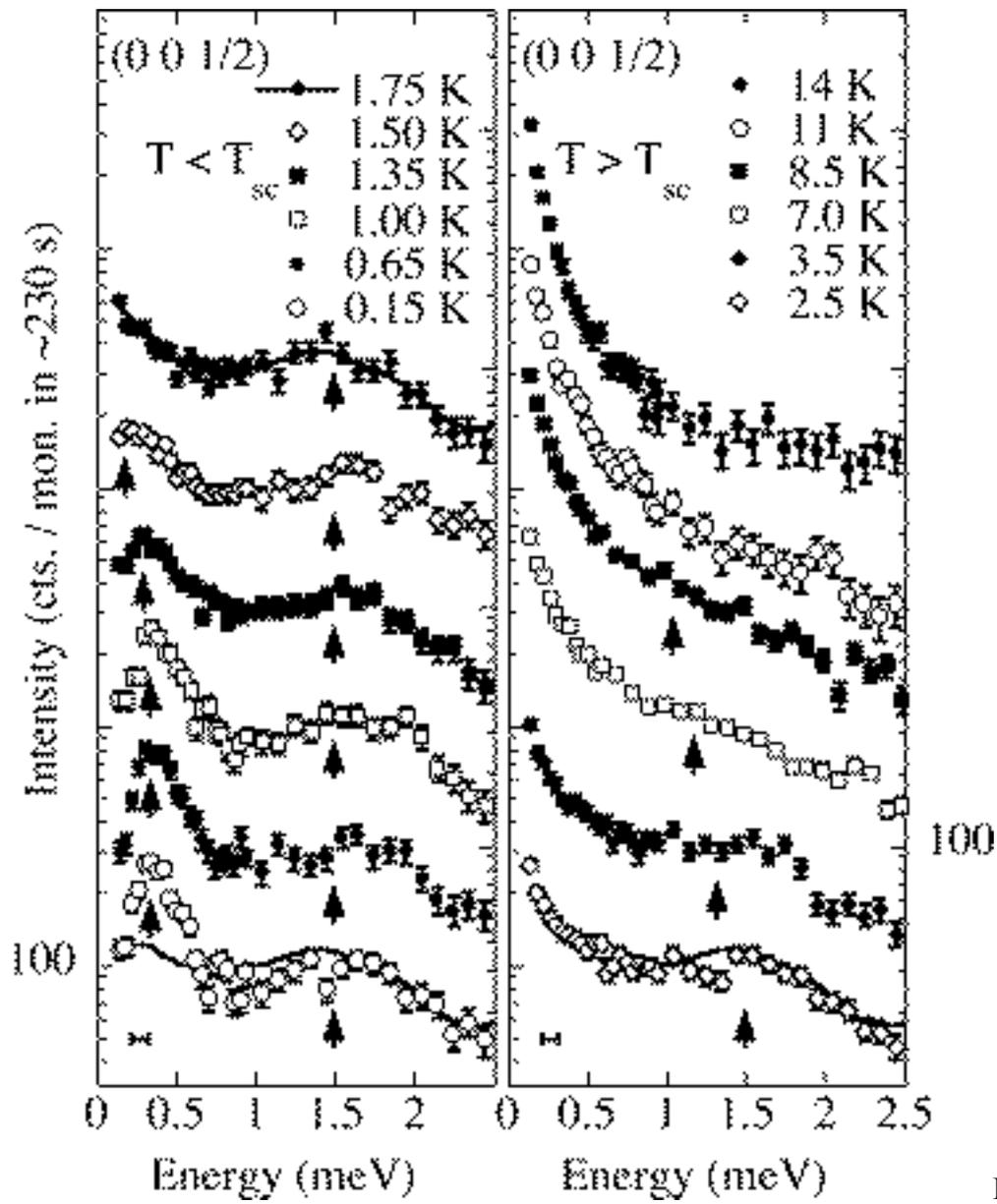


Fig. 4

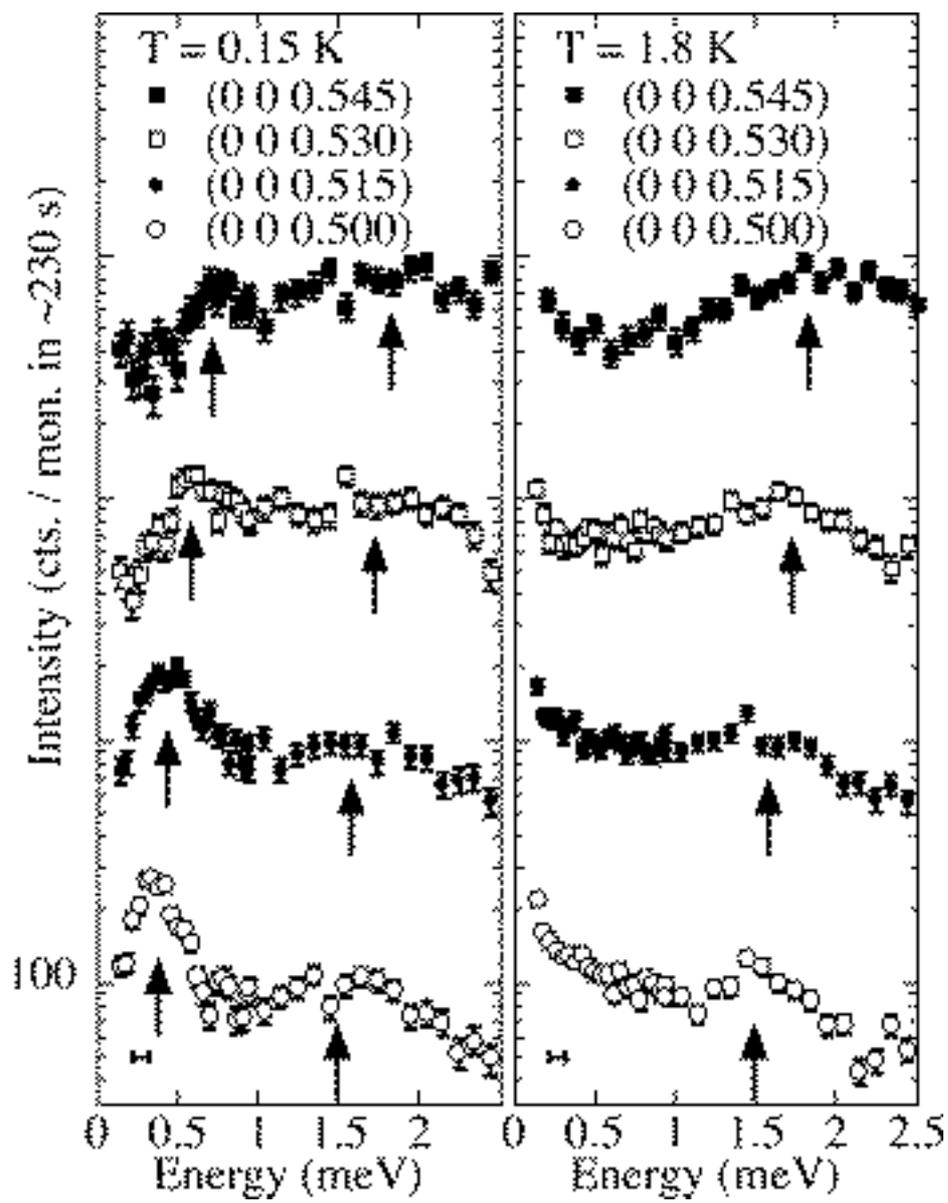


Fig. 5

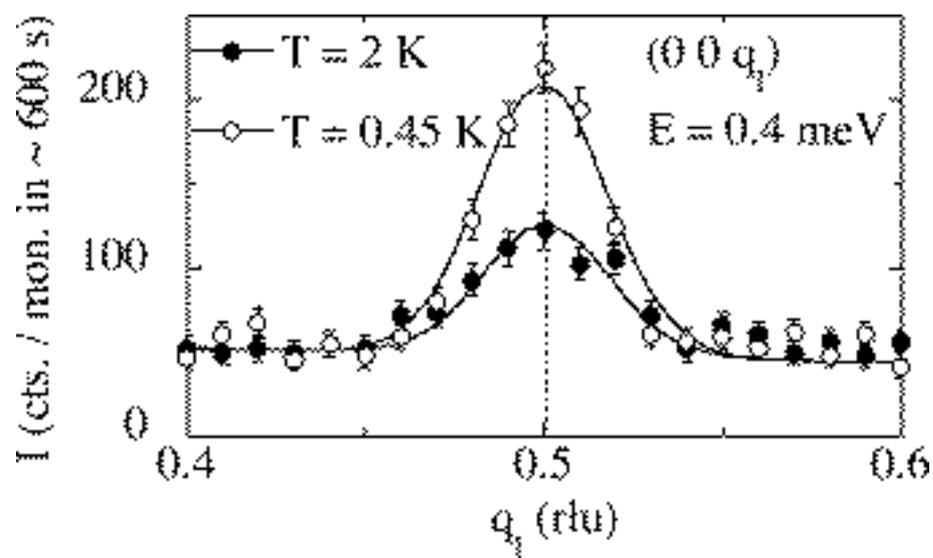


Fig. 6

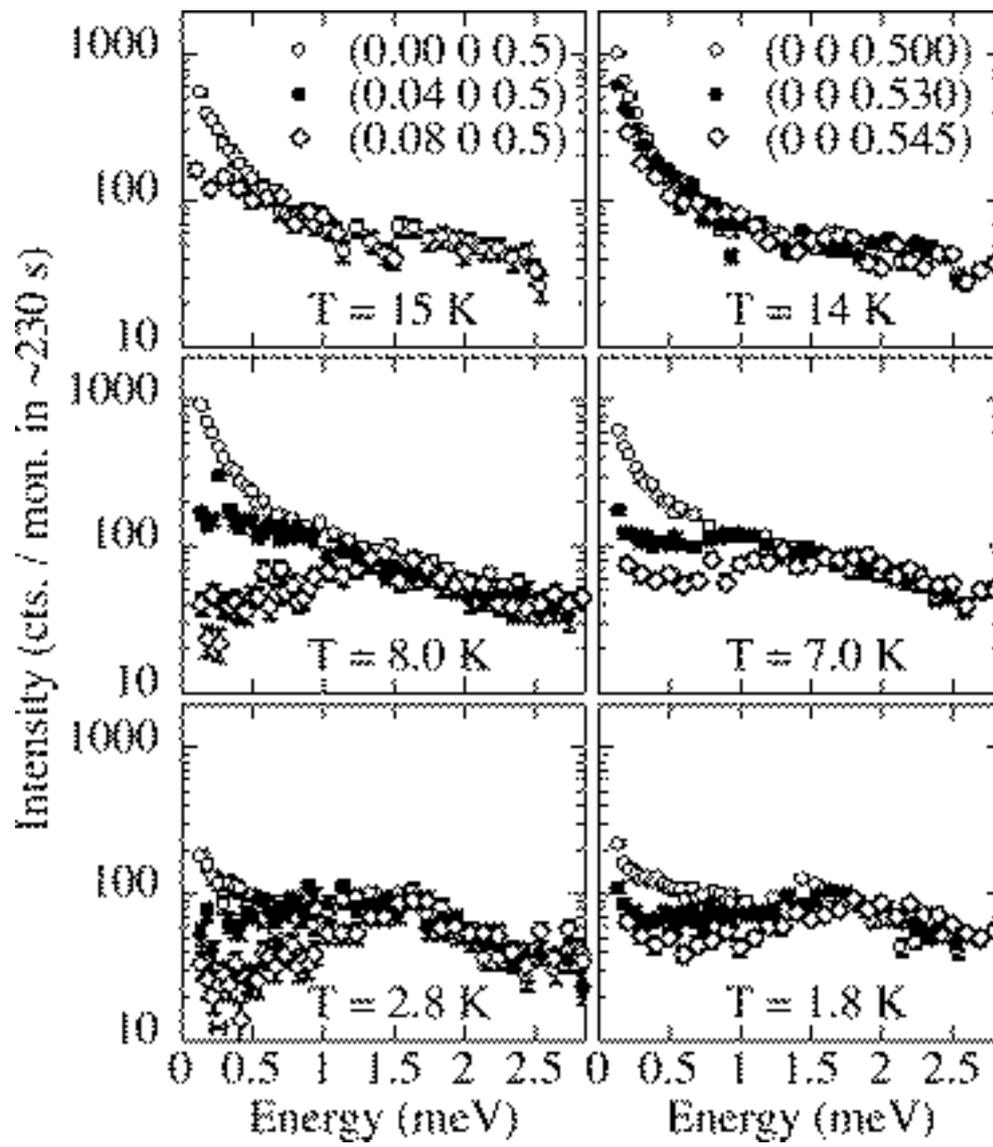


Fig. 7

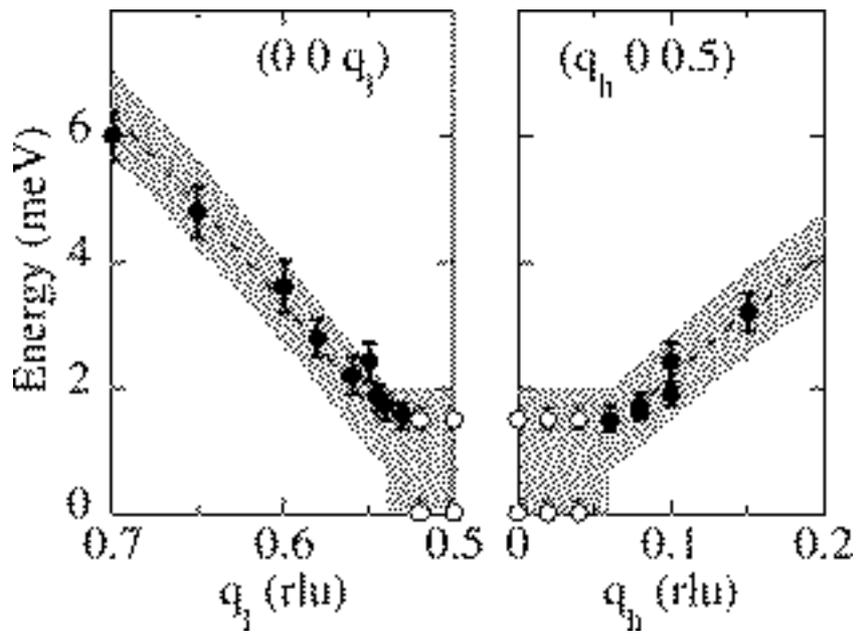


Fig. 8

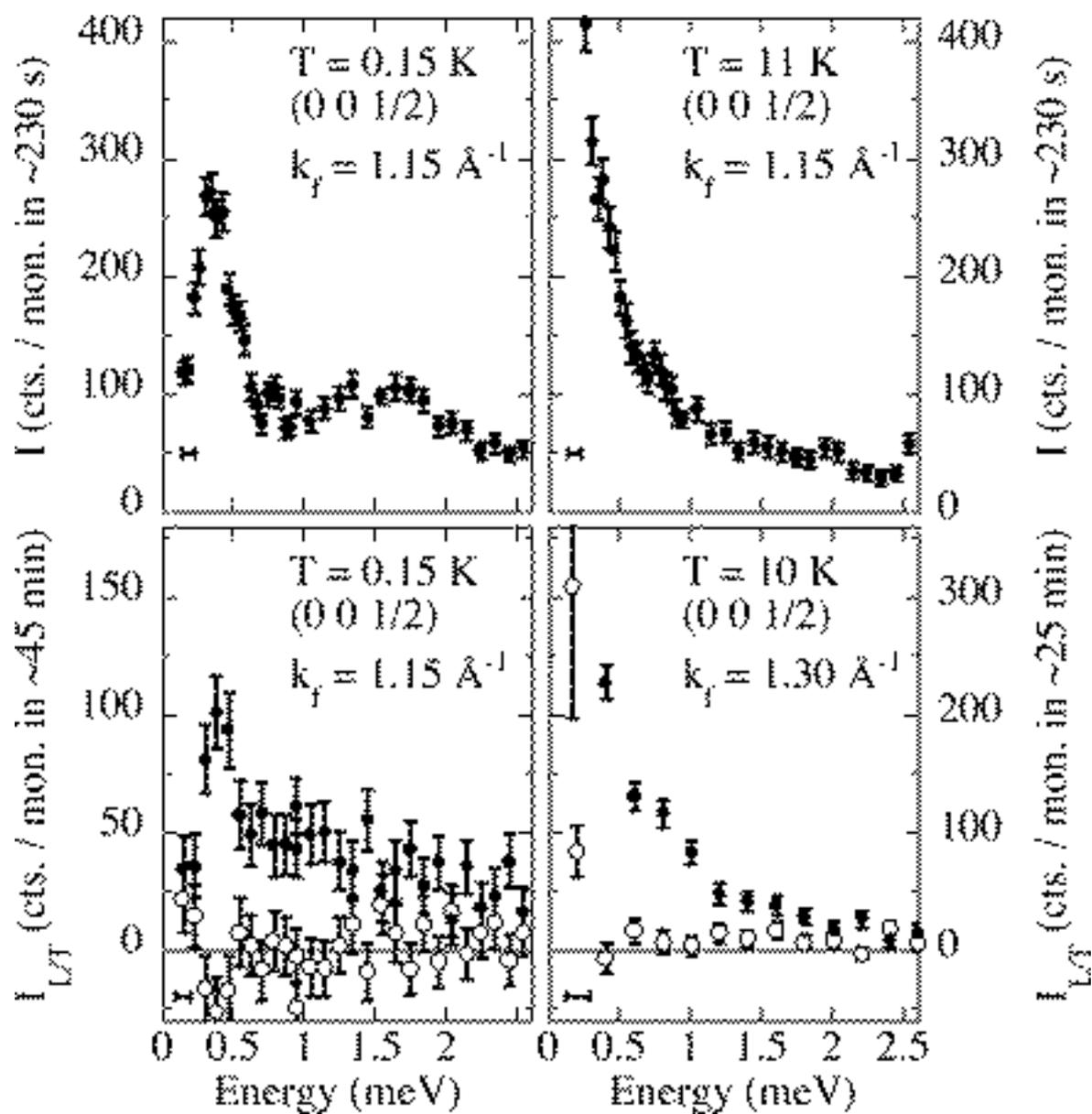


Fig. 9

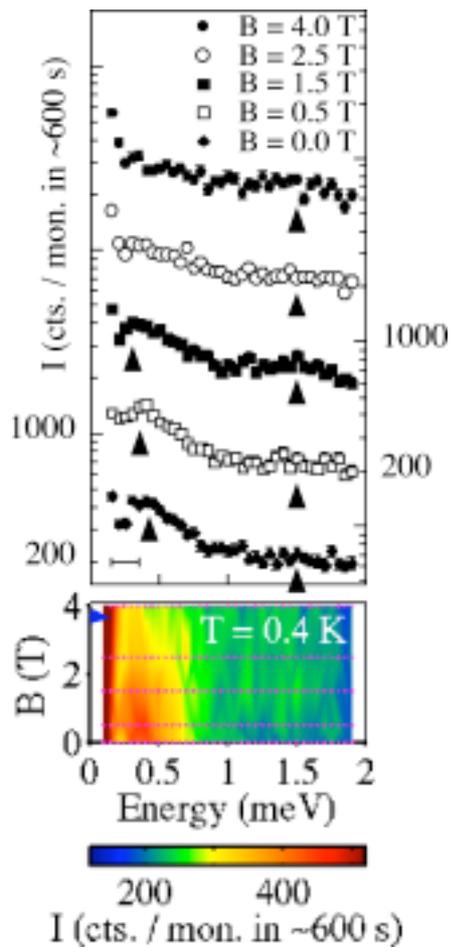


Fig. 10

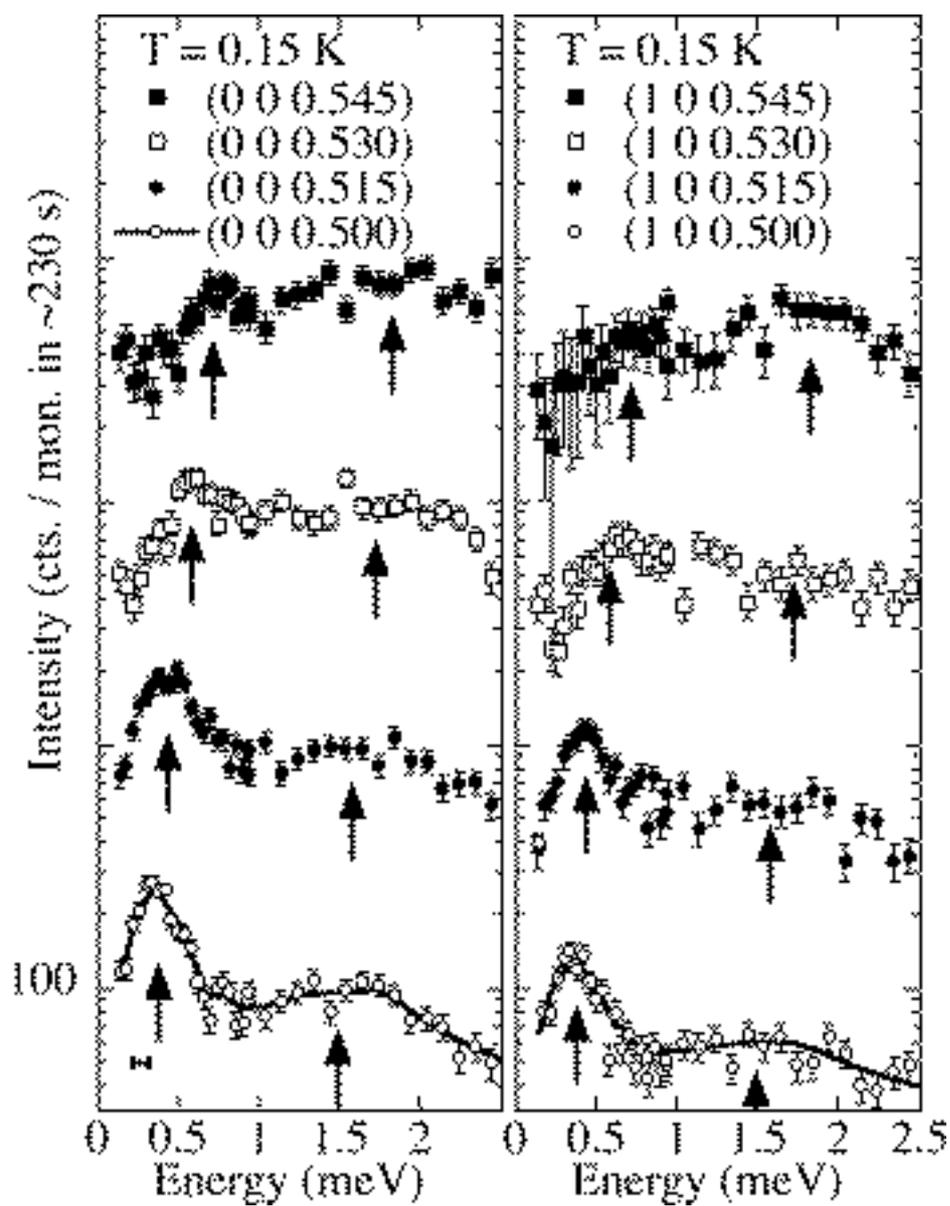


Fig. 11

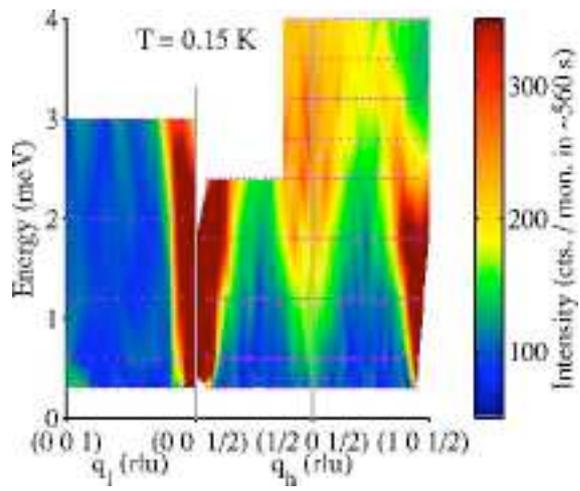


Fig. 12

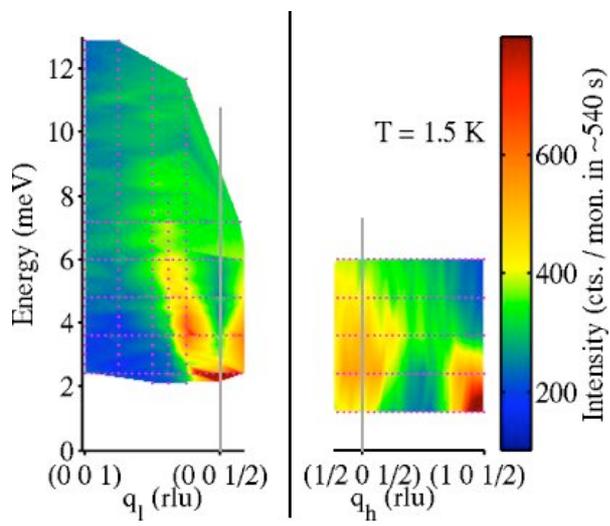


Fig. 13

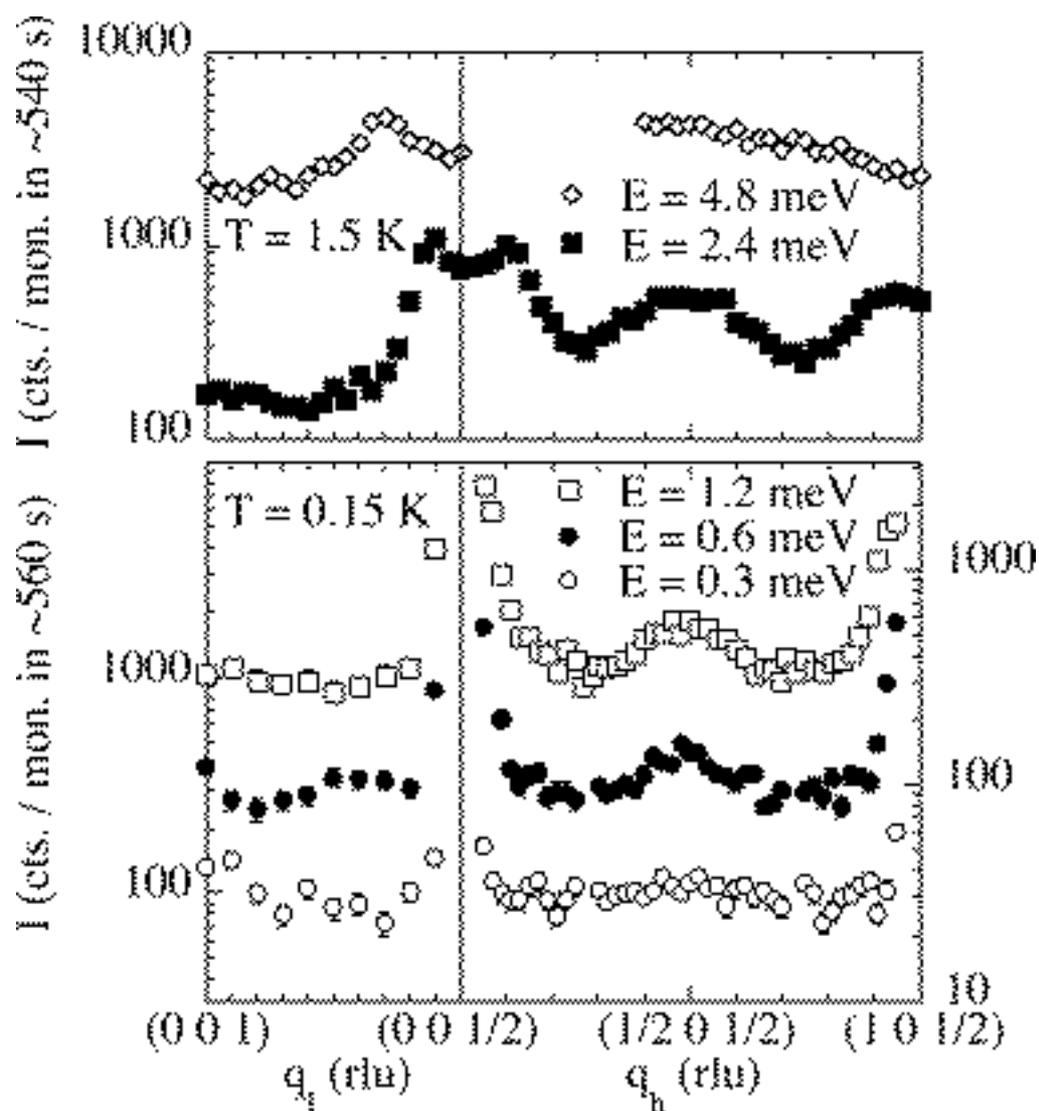


Fig. 14

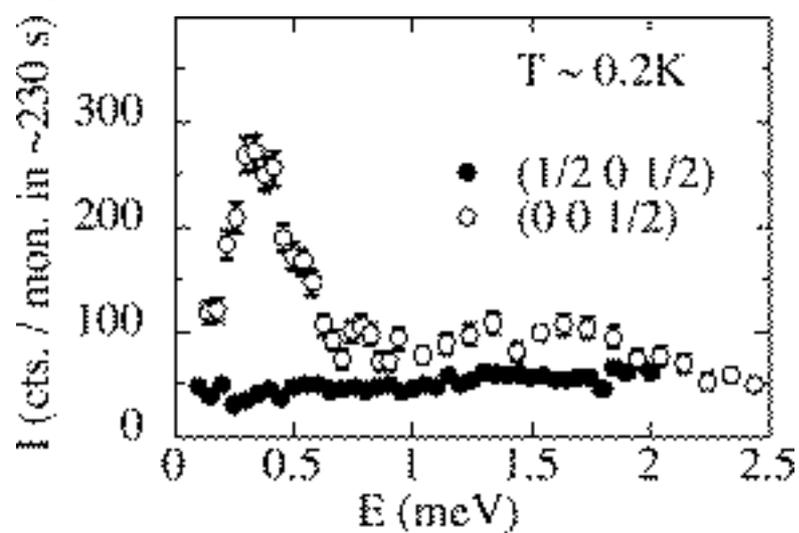


Fig. 15

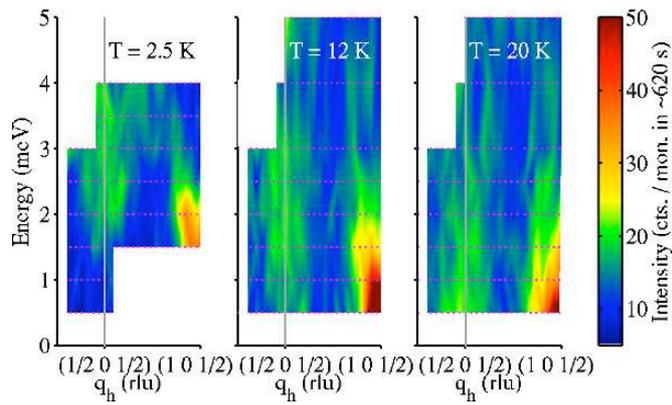


Fig. 16

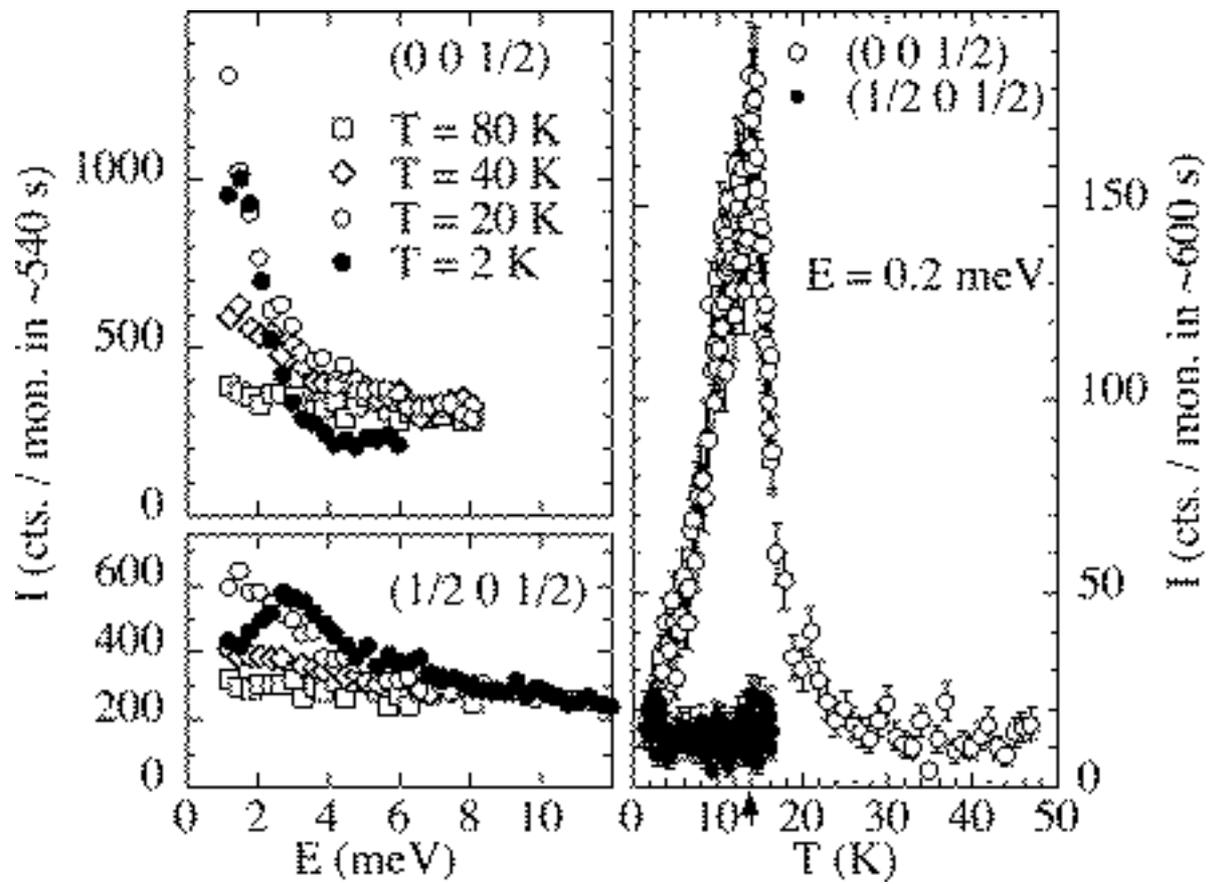


Fig. 17