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Anisotropic magnetic field dependence of the magnetization dynamics in UPd_2Al_3

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The magnetization dynamics of the magnetic superconductor UPd_2Al_3 has been investigated by inelastic neutron scattering in the normal and superconducting, antiferromagnetically ordered state under a magnetic field applied parallel to the hexagonal c axis. Within the available resolution, the dynamic response below 2.5 meV is insensitive to the applied field on the scale of the field dependence reported by E. Blackburn *et al.* [Phys. Rev. B **74**, 024406 (2006)] in which the field was applied in the basal plane. Our results support that the previously reported field dependent quasielastic contribution is related to the dynamics of the rotation of the magnetic moment. The changes observed in the inelastic part of the excitation spectrum are consistent with those expected from spin-wave theory. Interestingly, the anisotropic field dependence of the normal-state response may be correlated with the superconducting properties of this material.

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I. INTRODUCTION

In UPd_2Al_3, superconductivity ($T_{sc}=1.8$ K) develops inside an ordered magnetic phase ($T_N=14.3$ K) and thermodynamic measurements suggest that both phenomena are generated by the same electronic states.[1] The interplay between the magnetism and the superconductivity is still being intensively discussed, and the superconductivity in UPd_2Al_3 is clearly unconventional.[2] Anomalous inelastic neutron scattering in the superconducting state at the antiferromagnetic (AFM) zone center $\mathbf{Q}_0=(0\ 0\ 0.5)$ indicates a link between antiferromagnetism and superconductivity.[3,4]

Recent inelastic neutron scattering experiments have investigated the magnetization dynamics at the antiferromagnetic zone center of UPd_2Al_3 as a function of a magnetic field applied in the hexagonal basal plane.[5] We here report on the magnetic response when the field is applied along the hexagonal c axis. Comparing the results of both investigations provides insight into the normal-state magnetization dynamics of UPd_2Al_3, which may be correlated with the superconducting properties of this material.

UPd_2Al_3 has a hexagonal unit cell with lattice parameters $a=b=5.35$ Å and $c=4.185$ Å at ambient conditions. Below $T_N=14.3$ K, the magnetic moments on the uranium atoms align in the basal plane and stack antiferromagnetically up the c axis, resulting in a propagation vector $\mathbf{q}=[0\ 0\ 0.5]$. The magnetic moment associated with each uranium site is μ is $0.85\mu_B$.[6–8] Moment rotations in the basal plane are observed[6] when a magnetic field of $B^b=4.2$ T is applied parallel to the b axis. When the magnetic field B^c is applied perpendicular to the basal plane, it is perpendicular to both the $B=0$ T time-averaged and fluctuating magnetic moments (the latter are known by polarization analysis to be confined to the basal plane[3]) and so does not couple directly to them. In this case, no moment rotations are observed up to the maximum field measured, 35 T.[9]

Superconductivity develops inside the antiferromagnetically ordered phase for temperatures $T_{sc}<1.8$ K and below anisotropic upper critical fields of $B^b_{c2}=B^a_{c2}=3.3$ T and $B^c_{c2}=3.7$ T.[2]

II. EXPERIMENT

A crystal of 2.5 g and cylindrical shape was obtained from a melt of high purity elements by the Czochralski method.[10] It was used in our previous investigations,[5] aligned to give an $\mathbf{a}^*$-$\mathbf{c}^*$ scattering plane. We applied a magnetic field B^c up to 3.8 T parallel to the c axis using a horizontal field cryomagnet. The measurements were performed on the cold neutron three-axis spectrometer IN14 at the Institut Laue-Langevin, Grenoble, using a fixed final energy $E_f=2.74$ meV ($k_f=1.15$ Å^{-1}) and a horizontally focusing pyrolytic graphite (PG002) analyzer in conjunction with a PG002 monochromator as well as 60′ collimation and a Be filter between monochromator and sample. The energy resolution was 0.06 meV full width half maximum (FWHM) at the elastic position. We measured the inelastic response at the antiferromagnetic zone center $\mathbf{Q}_0=(0\ 0\ 0.5)$ at $T=2.5$ K and at $T=0.3$ K.

The data with B^b parallel to the b axis[5] were obtained in the same manner, but using a vertical split-coil cryomagnet providing a maximum field of $B=5$ T. Using a different cryomagnet changes the experimental conditions slightly, e.g., the absorption of cryogenic equipment and the beam divergence tolerated by the magnet opening. As we will see, the observed inelastic count rates differ by about 20% for different field directions but the same measuring time. Nevertheless, within one experimental setup, the variations are significantly smaller, e.g., less than 5% in the elastic intensity of nuclear Bragg reflections. All data, measured per monitor in the incident beam, are presented for a similar measuring time, which corresponds to about 10 min. Besides this difference in absolute count rate, the observed relative variations at zero field are close to the statistical error of an individual data point. These variations are not relevant when analyzing data taken under identical experimental conditions,

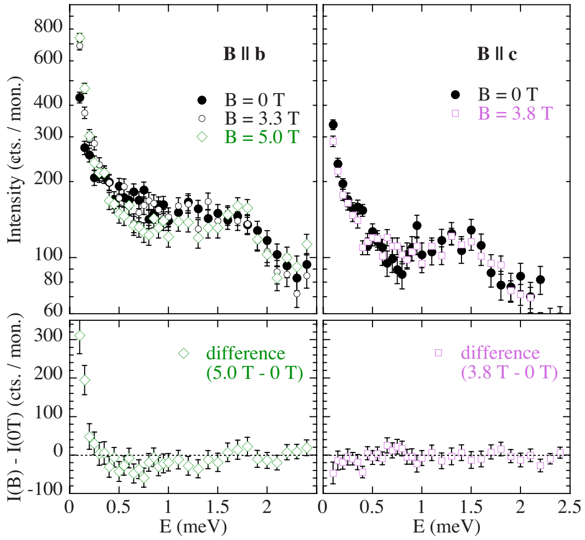

FIG. 1. (Color online) The dynamic response of $\mathrm{UPd_2Al_3}$ at $T=2.5$ K at the antiferromagnetic zone center (0 0 0.5) for energies $\Delta E \leqslant 2.5$ meV as measured by inelastic neutron scattering on IN14 with $k_f=1.15$ Å. Left-hand side panels: When the magnetic field is parallel to the b axis, the low-energy response $\Delta E \leqslant 0.3$ meV increases in field, whereas the inelastic scattering intensity is reduced between 0.5 and 1.5 meV. Data taken from Ref. 5. Right-hand side panels: When the magnetic field is parallel to the c axis, no such changes are observed.

and, for this reason, we restrict ourselves to the comparison of data obtained within one experiment, i.e., under identical experimental conditions.

III. RESULTS

Figure 1 shows the dynamic response at the antiferromagnetic zone centre $\mathbf{Q}_0=(0\ 0\ 0.5)$ up to $\Delta E=2.5$ meV at $T=2.5$ K with the magnetic field applied in two different directions, compared to the signal in zero field. As previously reported,[5] a change in the dynamic response is observed when the field is applied within the basal plane [Fig. 1, left-hand side]: The low-energy response $\Delta E \leqslant 0.3$ meV increases for both fields $B^b=3.3$ T and $B^b=5.0$ T shown in the figure, whereas the inelastic scattering intensity is reduced between 0.5 and 1.5 meV for $B^b=5$ T only. This is best visible in the difference plot in the lower part of the figure. No such changes are observed with a field of $B^c=3.8$ T applied along the c axis [Fig. 1, right-hand side].

At the magnetic Bragg reflection $\mathbf{Q}_0=(0\ 0\ 0.5)$, the elastic neutron scattering intensity is sensitive to all projections of the magnetic moment in the basal plane. The precision of the crystal alignment and the reproducibility of instrument positioning lead to variations of less than 5% in the elastic intensity of nuclear Bragg reflections. Within this precision, the elastic intensity at $\mathbf{Q}_0$ does not change for any field investigated, as expected for a time-averaged static magnetic moment confined to the basal plane. In agreement with previous diffraction work,[6] we could not observe any change in the momentum width of the elastic signal. The energy resolution (FWHM) is 0.06 meV, and investigating the energy width at a nuclear position (0 0 1) reveals a constant flat background signal for $\Delta E \geqslant 0.2$ meV. We therefore presume that the observed changes in the spectrum are not related to any static moment component. Within the resolution of the three-axis instrument IN14, the observed low-energy signal may be considered as quasielastic at all fields. In zero field, this is supported by high-resolution neutron spin-echo investigations.[11]

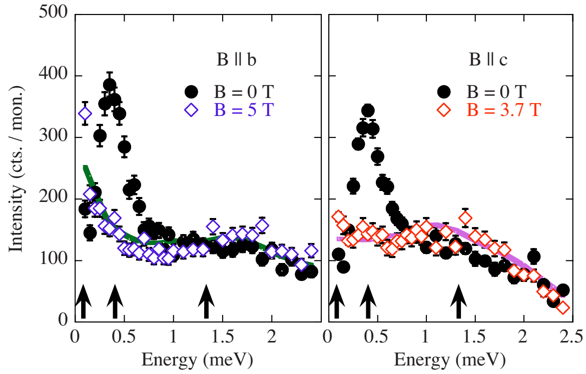

FIG. 2. (Color online) The dynamic response of $\mathrm{UPd_2Al_3}$ at $T=0.3$ K at the antiferromagnetic zone center (0 0 0.5) for energies $\Delta E \leqslant 2.5$ meV as measured by inelastic neutron scattering on IN14 with $k_f=1.15$ Å at zero field and as well as with magnetic fields $B > B_{c2}$ applied along the b axis (left-hand side, $B_{c2}^b=3.3$ T) and the c axis (right-hand side, $B_{c2}^c=3.7$ T). The solid line is a smooth fit to the 2.5 K data from Fig. 1 at similar fields, scaled by a Bose temperature factor. Three contributions marked by arrows are discussed in the text.

We continue our discussion with additional comments on the inelastic response at higher energy transfers. From our previous experiments, it is known that working at an alternative $\mathbf{Q}$ position such as (1 0 0.5) enhances the visibility of the inelastic response. Nevertheless, in this case, the elastic intensity (Bragg peak) from the static magnetic moments changes when the moment reorientation occurs, leading to an additional complication for any quantitative analysis. We therefore performed our entire experiment at the (0 0 0.5) position only.

The dynamic response at $\mathbf{Q}_0$ and low temperatures is shown in Fig. 2. With magnetic fields $B > B_{c2}$ applied, the sample does not become superconducting, and indeed for both investigated field directions, the temperature dependence of the normal-state dynamic response follows Bose scaling. As previously reported,[3] the dynamic response changes in three respects (marked by arrows in the figure) when the sample is superconducting at low temperatures and zero field: Inside the superconducting state, a strong inelastic feature exists, centered at $\Delta E \approx 0.4$ meV and marked by the middle arrow, in conjunction with the broader inelastic feature at higher energies (right arrow). Nevertheless, the latter is reduced in intensity compared to its counterpart in the normal state. The intensity at low energies, marked by the left arrow and attributed to quasielastic scattering, is suppressed when the sample is superconducting.

IV. DISCUSSION

Since the changes observed in the dynamic response in the normal state depend on the field direction, we suggest that they are related to a dynamical aspect of the moment reorientation, which takes place only for a field applied in the basal plane parallel to the b direction. For $B^b=4.2$ T, the effective bulk magnetic moments rotate by 30° from the a direction to the a^* direction.[6] The inelastic scattering data, which are sensitive to the space-time moment-moment decorrelation function, establish a change in the temporal correlations in the vicinity of the moment rotation. There is no apparent change in the momentum width of the observed quasielastic signal with field, suggesting that the spatial extent of the dynamic correlations does not change.

At 2.5 K and with the field parallel to the b axis, the inelastic scattering intensity is reduced between 0.5 and 1.5 meV once the moments are pointing along a^*, as happens above the transition at $B^b=4.2$ T.[5] Such behavior is consistent with a description of this part of the excitation spectrum as being spin-wave-like with a finite excitation energy arising from the hexagonal anisotropy of the underlying lattice.[12] The magnetic field then affects the energy of this part of the excitation spectrum only if it has a component in the direction of the time-averaged magnetic moments. At $B=0$ T, both the fluctuating and static components are confined to the basal plane.[3] Assuming this to hold for the B^c used in our experiment, no changes are anticipated, and, indeed, for the magnetic field applied parallel to c, no field dependence has been observed.

In the perspective of a link between superconductivity and magnetism, any quantitative analysis between the field dependence of the dynamic scattering and the anisotropic upper critical field of the superconductivity requires further theoretical input. It is nevertheless intriguing to note that the upper critical field $B_{c2}^b=3.3$ T is reduced with respect to $B_{c2}^c=3.7$ T, whereas the low-energy magnetic fluctuations are *enhanced* when the magnetic field B^b is applied parallel to the b direction. In this context, we also point out that our previous neutron spin-echo work established the absence of low-energy fluctuations in the superconducting state at lowest temperatures and in zero field.[11]

The observations may be rationalized within a BCS-like scenario, where superconducting properties are generally enhanced by pair-making high-frequency modes and, conversely, depressed by low-frequency pair-breaking fluctuations.[13] In such a scenario, the increased quasielastic scattering observed for a field applied parallel to the b axis acts as pair breaking, and so the upper critical field B_{c2}^b is reduced with respect to B_{c2}^c for a field applied parallel to the c axis, where no change occurs.

Moreover, the observed changes of the scattering intensity at higher energies in the range between 0.5 and 1.5 meV for fields parallel to the b axis may suggest a weakening of the coupling. The relevance of the high-frequency modes for superconductivity may be deduced from the low temperature data in Fig. 2. Based on its (zero-field) temperature dependence, this redistribution in the dynamic response has already been evoked in earlier work[14] as important for the superconducting pairing mechanism. More recent theoretical work outlines the relevance of the AFM state for the superconductivity in more detail.[4,15] The observation of reduced inelastic intensity in the normal AFM state within this energy range and reduced B_{c2}^b when a field is applied within the basal plane adds further evidence to this proposal for the superconducting pairing mechanism.

V. CONCLUSION

To summarize, we investigated the dynamic magnetic response of UPd_2Al_3 by inelastic neutron scattering with a magnetic field applied parallel to the b axis (see also Blackburn *et al.*[5]) and the c axis. By comparing these, we suggest that the anisotropic dependence of the superconducting upper critical field B_{c2} may be correlated with the magnetization dynamics involved with the rotation of the moments when a field is applied in the basal plane. This finding should be taken into account in the current scenarios for the superconducting pairing mechanism.

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