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# Polarized-neutron-scattering study of the spin-wave excitations in the 3-k ordered phase of uranium antimonide

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**Abstract**
The anisotropy of magnetic fluctuations propagating along the [1 1 0] direction in the ordered phase of uranium antimonide has been studied using polarized inelastic neutron scattering. The observed polarization behavior of the spin waves is a natural consequence of the longitudinal 3-**k** magnetic structure; together with recent results on the 3-**k**-transverse uranium dioxide, these findings establish this technique as an important tool to study complex magnetic arrangements. Selected details of the magnon excitation spectra of USb have also been reinvestigated, indicating the need to revise the currently accepted theoretical picture for this material.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Neutron scattering has a well-deserved reputation as the technique of choice to investigate magnetic phenomena in condensed matter [1]. In particular, spherical neutron-spin polarimetry (SNP) applied to neutron diffraction has been extensively used to determine complex magnetic structures [2, 3]. This technique has, however, less frequently been applied to inelastic neutron scattering experiments, perhaps understandably due to the unavoidable flux intensity reduction associated with beam polarization. One successful example is our recent work on uranium dioxide ($UO_2$) [4], which exhibits a transverse 3-**k** magnetic moment arrangement below the Néel temperature $T_N = 30.8$ K. The multi-**k** magnetic structure of cubic compounds in the ordered phase can be described by expressing the three components of the magnetic moment at the lattice site $j$ (with position vector $\mathbf{R}_j$) as

$$\langle S_j^\alpha \rangle = \mathrm{e}^{\mathrm{i}\mathbf{k}_0^\alpha \cdot (\mathbf{R}_j - \mathbf{R}_0)} \langle S_0^\alpha \rangle \quad (1)$$

where $\alpha = x, y, z$. Two possible arrangements (labeled by A and B in figure 1) are present for the transverse 3-**k** structure of $UO_2$:

$$\mathbf{k}_0^x = 2\pi \mathbf{u}_y/a; \qquad \mathbf{k}_0^y = 2\pi \mathbf{u}_z/a; \qquad \mathbf{k}_0^z = 2\pi \mathbf{u}_x/a, \quad (2)$$

or

$$\mathbf{k}_0^x = 2\pi \mathbf{u}_z/a; \qquad \mathbf{k}_0^y = 2\pi \mathbf{u}_x/a; \qquad \mathbf{k}_0^z = 2\pi \mathbf{u}_y/a, \quad (3)$$

$\mathbf{u}_\alpha$ being the unitary vector along the $\alpha$ direction. We showed that the polarization of the spin-wave branches measured in the ordered phase is not only perfectly consistent with RPA calculations, but more importantly is a natural consequence of the 3-**k** structure. Despite the fact that multi-**k** structures have been a common theme of actinide physics for at least the last 20 years, it is impossible to distinguish with elastic scattering experiments between a true 3-**k** configuration and the superposition of 1-**k** modulations in different zones [5]. One method that has been used is to cool the material through $T_N$ with an applied field or stress in the hopes of changing the domain populations [6]. This has been successful in some cases, but one always must be aware that the applied field or stress might be below some threshold value to cause a domain

 

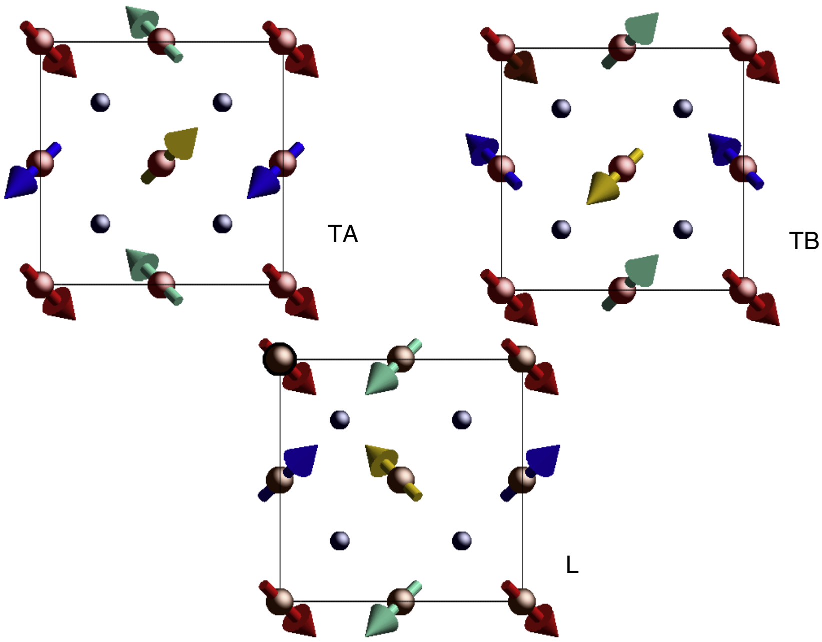


**Figure 1.** Schematic representation of type-I, 3-**k** magnetic structures in a fcc lattice, as projected down a cube axis. TA and TB indicate the moment arrangement in the two possible domains of the transverse structure, while L refers to the single-domain longitudinal structure. The magnetic moments (indicated by arrows) point along different ⟨1 1 1⟩ directions of the cubic cell for the four independent uranium sublattices of the magnetic unit cell.

imbalance. Thus, in general, a more robust method should be found.

With this in mind, we have designed and performed a polarized inelastic neutron scattering experiment on uranium antimonide, which is an fcc cubic compound exhibiting a type-I 3-**k** structure below $T_N = 213$ K with propagation direction $\mathbf{Q}_{AF} = [0\,0\,1]$ like $UO_2$, but longitudinal rather than transverse (figure 1), i.e. with $\mathbf{k}_0^\alpha = 2\pi\mathbf{u}_\alpha/a$, $\alpha = x, y, z$. The spin-wave behavior, as studied by *unpolarized* inelastic neutron scattering, was actually crucial in the past to determine the correct magnetic structure in USb [7], but the assessment of suitable general rules could make the contribution of polarized neutron inelastic scattering very important in this framework. Our results confirm that the unusual polarization properties of spin-wave branches are a natural consequence of the magnetic structure. Furthermore, we have put this knowledge to use in analyzing some details of the magnon excitation spectra which would not have been evident without polarized neutrons.

## 2. Results and discussion

The experiment was performed at ILL on a 7 g USb single crystal, aligned in the (1 0 0)–(0 1 0) scattering plane, on the IN22 three-axis spectrometer equipped with an SNP option (CRYOPAD-III). The temperature was $T = 50$ K throughout the experiment, well below $T_N$. The final wavevector $k_f$ was fixed at 4.1 Å$^{-1}$ during the whole experiment, so that the same configuration could be used to measure both the acoustic and the optic spin-wave branches along all directions.

Only the spin-flip channel was monitored, in order to exclude vibrational contributions; if the initial and final beam polarization **P** are chosen parallel to **Q**, all magnetic fluctuation components perpendicular to **Q** appear in this channel [10]. If, on the contrary, **P** is perpendicular to **Q**, the spin-flip channel contains only the components perpendicular to both **Q** and **P**. In the following we use the common reference frame in which the $x$ axis is defined parallel to the momentum transfer **Q**, the $y$ axis is perpendicular to $x$ and contained in the scattering plane, and the $z$ axis is perpendicular to the scattering plane, with $\mathbf{u}_x \times \mathbf{u}_y = \mathbf{u}_z$.

The spin-wave dispersion in the high-symmetry directions has been experimentally studied by unpolarized inelastic neutron scattering in [8]. Actually, not all the considered directions are worth reexamining with polarized neutrons: in fact, when $\mathbf{Q} \parallel [0\ 0\ 1]$ the two perpendicular directions are equivalent in cubic symmetry, while for $\mathbf{Q} \parallel [1\ 1\ 1]$ the two spin-wave branches are degenerate and the polarization properties are therefore isotropic. For the $\mathbf{Q} \parallel [1\,1\,0]$ direction, however, the situation was extremely informative in the case of $UO_2$. Figures 3 and 4 of [4] show that the acoustic spin-wave peak is absent in the spin-wave channel with $\mathbf{P} \parallel [0\,0\,1]$, while the optic branches distribute their intensity along both components. This is the expected behavior for a transverse 3-**k** dipolar order. In the case of the longitudinal structure of USb, it is predicted [9] that the acoustic branch should have the opposite polarization with respect to $UO_2$ (i.e. should contain fluctuations only along the [0 0 1] direction and therefore only appear in our $\mathbf{P} \parallel y$ spin-flip channel), and that the optical branch should be fully transverse.

The former observation, limited to the acoustic mode and only at the $X$ point, could be made even without polarized neutrons by Lander and Stirling [11] by pointing out that no magnetic scattering is found at the (0 0 1) point. Our

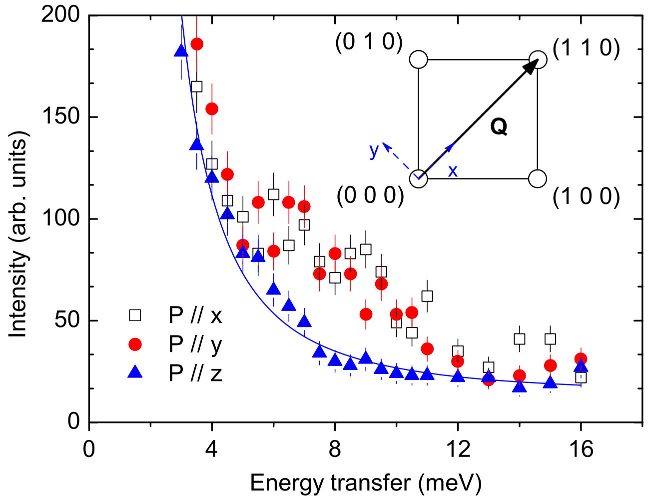


**Figure 2.** Spin-flip energy scans with $\mathbf{Q} = [1\ 1\ 0]$ and initial and final polarization along $x$ (open squares), $y$ (filled circles) and $z$ (triangles). The solid line is a guide to the eye. In the inset, a schematic representation of the scattering plane is given.

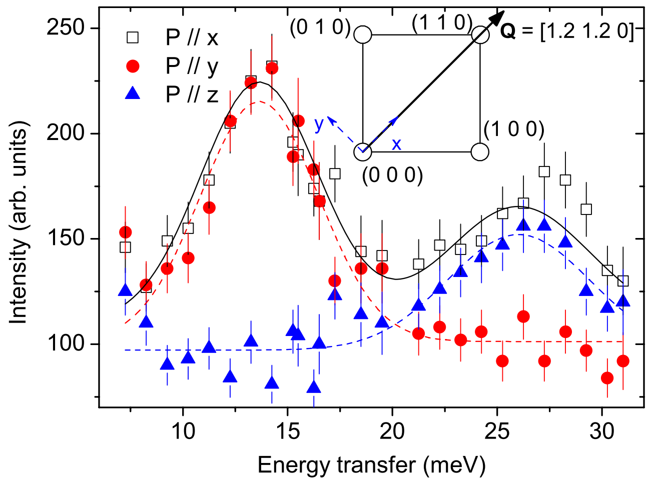


**Figure 3.** Spin-flip energy scans with $\mathbf{Q} = (1.2\ 1.2\ 0)$ and initial and final polarization along $x$ (open squares), $y$ (filled circles) and $z$ (triangles). The dashed lines are fits to the latter two data sets, each curve being the sum of a single Gaussian and a flat background; the solid line is the sum of the above two Gaussians plus a flat, roughly polarization-independent, background. In the inset, a schematic representation of the scattering plane is given.

experiment with polarized neutrons confirms this observation (figure 2), but also proves that the same is true in the whole Brillouin zone along the [1 1 0] direction, and that also the peculiar polarization of the optical mode is confirmed. The fact that the observed acoustic peak at (1 1 0) is large and asymmetric can be attributed (apart to the fact that fixing $k_f = 4.1$ Å$^{-1}$ is not the best choice when focusing on the low-energy excitations) to magnon–phonon interaction, since a strong phonon branch is also present at 6 meV at this point. With the experimental possibilities of SNP we were able to look for a signature of this mixed term by measuring the off-diagonal components of the neutron scattering functions, i.e. terms where the neutron polarization is turned by $\pi/2$, but no clear results were obtained.

Figure 3 shows spin-flip energy scans at the (1.2 1.2 0) point with $\mathbf{P}$ along $x$, $y$, and $z$: the latter two data sets were fitted by a single Gaussian peak, and the sum of both peaks correctly reproduce the former set after accounting for the presence of a (roughly polarization-independent) flat background. This observation can be easily explained as follows. In the inset of figure 3 we show the scattering plane with the $\mathbf{Q}$ vector along the [1 1 0] direction. The relevant zone centers for the nuclear structure are (1 1 1) and (1 1 $\bar{1}$), which are out of the scattering plane. Thus the magnetic propagation vectors of $\mathbf{q}_1 = [0\ 0\ 1]$ and $\mathbf{q}_2 = [0\ 0\ \bar{1}]$ are along the $z$ axis and result in the magnetic Bragg peak (1 1 0). If we consider longitudinal fluctuations, which we believe make up the acoustic modes, then these are defined by $m \parallel z$, i.e. out of the scattering plane. For (normal) transverse fluctuations they must lie in the scattering plane, and we can conveniently take them to have components parallel to $\mathbf{Q}$, (i.e. $m \parallel x$) and perpendicular to $\mathbf{Q}$ (i.e. $m \parallel y$), and, of course, no component parallel to $z$.

Now the key aspects of the polarization analysis technique are: ($a$) that we are sensitive, by virtue of the neutron interaction, to components only perpendicular to $\mathbf{Q}$, and ($b$) that if we look at the spin-flip signal the components must be perpendicular to $\mathbf{P}$. Consider first a longitudinal acoustic signal at 13 meV in figure 3. This will appear in channels with $\mathbf{P} \parallel x$ and $\mathbf{P} \parallel y$, but not in $\mathbf{P} \parallel z$ as the latter is excluded by point ($b$) above. Moreover, we should see the full strength in both the first two configurations. This is exactly what is seen in figure 3.

Now consider the optic mode at 26 meV, which we believe is only transverse. For $\mathbf{P} \parallel x$ and $\mathbf{P} \parallel z$ the $m \parallel y$ component is observed, but the $m \parallel x$ component can never be observed because of point ($a$) above. However, for $\mathbf{P} \parallel y$ the $m \parallel y$ component is forbidden in the spin-flip channel because of point ($b$) above, so the signal should be zero. This is exactly what is shown in figure 3 for the optic mode. Moreover, the intensity is smaller because only one half of the spin-wave fluctuations are observed in each channel.

The so-called *flipping ratio*, defined as $f = S_y/(S_y + S_z)$ ($S_\alpha$ being the spin-flip magnetic signal for $\mathbf{P} \parallel \alpha$) is a quantitative parameter defining the polarization of an excitation. We measured this quantity directly by counting at single energy and momentum transfer points for several hours: in particular at $\mathbf{Q} = [1.2\ 1.2\ 0]$ we chose the energy transfer values of 13 meV (corresponding to the peak of the acoustic magnon branch), 20 meV (a background position) and 27 meV (optic branch). Counts with polarization along $x$ were also recorded in order to account for the polarization-independent background. In terms of the measured intensities $I_\alpha$, the flipping ratio then becomes

$$f = \frac{I_x - I_z}{2I_x - I_y - I_z}. \quad (4)$$

The results given in figure 4 essentially agree with the theoretical values of 1 for the acoustic branch and 0 for the optical branch, while confirming the expected isotropic behavior in background positions.

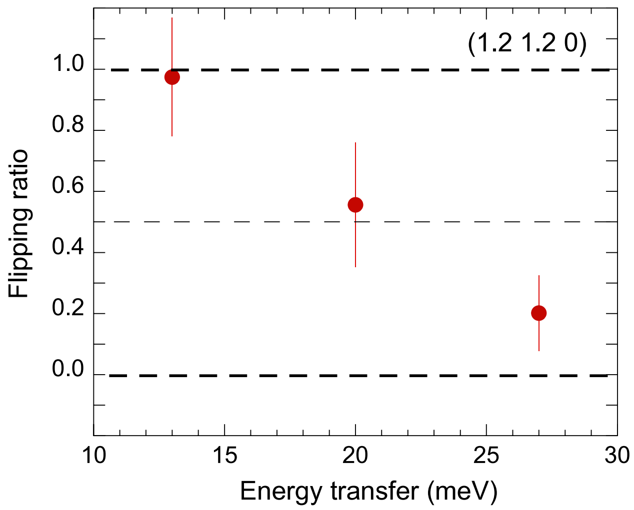


**Figure 4.** Flipping ratio (see text for details) measured at $\mathbf{Q} = [1.2\ 1.2\ 0]$ for three different values of the energy transfer: 13 meV (corresponding to the peak of the acoustic magnon branch), 20 meV (a background position) and 27 meV (optic branch). See figure 3 for a constant-$Q$ scan across these energies. The dashed lines indicate the theoretical values of 0, 0.5 and 1 expected for the optic, background and acoustic positions respectively.

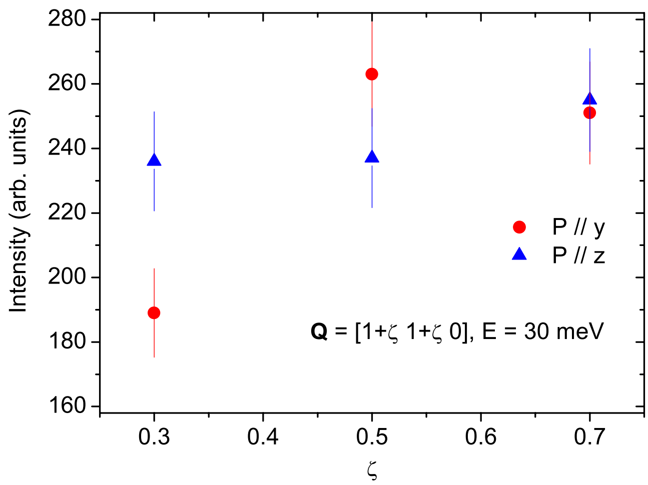


**Figure 5.** Spin-flip neutron counts with energy transfer fixed at 30 meV, different values of the momentum transfer along [1 1 0], and initial and final polarization along $y$ (filled circles) and $z$ (triangles).

Having established this point, we can now go further and use polarized neutrons to look at some subtler details of the excitation spectra. One of the points worth reinvestigating is evident in figure 18 of [8]. At the $X$ point the energy difference between the acoustic and optic branches is maximal, whereas at $\mathbf{Q} = (1.5\ 1.5\ 0)$ the two branches are degenerate from symmetry considerations. However, going from the latter to the $\Gamma$ point, a further splitting is predicted by the calculations which was not experimentally observed. Although the authors indicate an error bar smaller than the calculated splitting, knowing the properties of the two branches in a polarized neutron experiment we can check this easily. We measured the spin-flip neutron counts at three values of $\mathbf{Q} = (1 + \zeta\ \ 1 + \zeta\ \ 0)$ ($\zeta = 0.3, 0.5, 0.7$) at an energy transfer of 30 meV (figure 5), with polarization along $y$ and $z$; the acoustic branch appears only in the former and the optical branch only in the latter direction. At $\zeta = 0.3$ the $z$ counts are significantly larger because the optic peak tail reaches 30 meV in this configuration. At $\zeta = 0.5$ the scattering is isotropic as expected, since the branches are merged; but this remains true also at $\zeta = 0.7$, where according to the calculations one of the two branches should peak near 30 meV. This is an indication that the exchange and crystal-field parameters obtained from the original fitting in [8] should be revised to reduce this splitting considerably.

## 3. Conclusions

The polarization of the spin-wave modes in the ordered state of USb has been studied by three-dimensional polarization analysis techniques. No evidence for any cross terms in the $3 \times 3$ polarization matrix was found; the results in this paper could therefore equally well be obtained by regarding the diagonal elements of the matrix, i.e. by using a Helmholtz arrangement that can change the incident polarization of the neutrons to the three perpendicular directions. However, in practice, the use of CRYOPAD often simplifies the experimental set-up. The measured polarizations of acoustic and optical branches are consistent with those expected for the *longitudinal* 3-**k** static ordered structure, which emerge in a natural way from the model and are largely independent of the coupling parameters chosen. The result of this study, and the earlier one on $UO_2$ [4], establish that the method of analyzing the polarization of the spin-wave spectra is a unique method of distinguishing whether the static magnetic structure is of the 3-**k** type. Furthermore, the type of 3-**k** order, either transverse or longitudinal, may also be confirmed.

Such spin-wave polarization measurements were also used to test the theoretical model suggested for USb. One consequence of this model is that the two branches should be well separated at $\zeta = 0.7$, but this is not confirmed experimentally. In addition, the crystal-field states predicted by the model have not been observed [12], suggesting that a refinement of the parameters proposed in [8] might be needed.

## Acknowledgment

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