Reply to "Comment on 'Magnetic field effects on neutron diffraction in the antiferromagnetic phase of UPt_3' "

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Fåk, van Dijk, and Wills (FDW) question our analysis of elastic neutron-scattering experiments in the antiferromagnetic phase of UPt_3. They state that our analysis is incorrect because we average over magnetic structures that are disallowed by symmetry. We disagree with FDW and reply to their criticism below. FDW also point out that we have mistaken the magnetic-field direction in the experiment reported in N. H. van Dijk, B. Fåk, L. P. Regnault, A. Huxley, and M-T. Fernández-Díaz, Phys. Rev. B **58**, 3186 (1998). We correct this error and note that our previous conclusion is also valid for the correct field orientation.

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We disagree with the claim of Fåk *et al.*[1] that our analysis of elastic neutron-scattering experiments in the antiferro magnetic (AFM) phase of UPt_3 is incorrect because we average over magnetic structures that belong to different irreducible representations of the crystallographic space group. Classification of magnetic structures and magnetic phase transitions on the basis of irreducible representations of the space group and time inversion neglects the fundamental role that exchange interactions play in magnetic phase transitions.[2–5] Exchange interactions are invariant under continuous rotations of all the moments, and typically dominate the anisotropy energies that couple the atomic moments to the lattice. Classification of magnetic structures based on the exchange group accounts for the wide variety of magnetic structures that are observed in magnetic materials. The Shubnikov classification, which does not take into account the higher symmetry of the exchange interactions, disallows some of these structures.[5]

Thus, for a magnetic instability driven by exchange interactions the primary irreducible representation is based on the combined group of continuous rotations in spin space, the crystallographic space group, and time reversal, G_{ex}. The irreducible representations of the exchange group combine several irreducible representations of the space group.[6] Thus, not only are magnetic structures corresponding to irreducible representations of the space group allowed, but on the contrary, structures that are a combination of irreducible representations of the space group, but belong to one exchange representation, are also possible magnetic structures. Many examples of magnetic structures with these types of "mixed space-group representations"[7] are described in the literature.[2,8]

In most materials the magnetically ordered phase is defined by one irreducible representation of the space group due to the anisotropy energies which resolve (at least partially) orientational degeneracies within the exchange representation.[9] However, since the anisotropy terms are relatively weak, the energy splitting of differently oriented magnetic states are small. Thus, magnetic domain structures, and particularly their response to magnetic fields, should be analyzed using the nearly degenerate states within the full exchange multiplet. We believe this is the correct approach to understanding the magnetism and to analyzing the possible magnetic structures in the heavy fermion compound UPt_3.

In our analysis, we considered a general model for UPt_3 compatible with the available data.[10] We selected one irreducible representation of G_{ex} that is consistent with elastic neutron-scattering data in zero field. If we neglect the spin-lattice couplings then only the relative orientations of the atomic moments in the magnetic unit cell are fixed by the primary irreducible representation. Anisotropy energies are also included to resolve, or partially resolve, the degeneracies of the exchange representation.

Neutron scattering and x-ray experiments in UPt_3 show AFM order with propagation vector $\vec{q}_1=\vec{a}_1^*/2$.[11] The magnetic U ions occupy two symmetry-equivalent positions in the unit cell. The magnetic representation has six dimensions (three times the number of magnetic ions). Until very recently, the crystal structure of UPt_3 was thought to be hexagonal with space group D_{6h}^4. However, a recent x-ray-diffraction experiment revealed a lower trigonal symmetry with space group D_{3d}^3.[12] In either case, the magnetic representation can be decomposed into six one-dimensional representations. Three of these correspond to ferromagnetic (FM) alignment of the ions in the unit cell; the other three representations correspond to AFM alignments. The alignment of the magnetization or sublattice magnetization may be along the $\hat{x}$, $\hat{y}$, or $\hat{z}$ axes. However, these six structures are connected with only two exchange representations corresponding to FM or AFM alignment in the unit cell. Table I shows the irreducible representations and basis functions of

TABLE I. Irreducible representations and basis functions of the space groups D_{6h}^4 and D_{3d}^3 grouped by exchange multiplets. FM and AFM refer to ferromagnetic or antiferromagnetic alignment of the two U ions on the unit cell. We use the notation of Kovalev in Ref. 13.

	FM	AFM		FM	AFM
	$\tau_2:\hat{x}$	$\tau_7:\hat{x}$		$\tau_2:\hat{x}$	$\tau_3:\hat{x}$
D_{6h}^4	$\tau_4:\hat{y}$	$\tau_5:\hat{y}$	D_{3d}^3	$\tau_4:\hat{y}$	$\tau_1:\hat{y}$
	$\tau_6:\hat{z}$	$\tau_3:\hat{z}$		$\tau_2':\hat{z}$	$\tau_3':\hat{z}$

the crystallographic space groups D_{6h}^4 and D_{3d}^3 grouped by their corresponding exchange multiplets.

Our study is based on a free-energy functional [Eq. (9) of Ref. 10] which includes the exchange, anisotropy, and Zeeman energies. First, a uniaxial anisotropy term [not shown in Eq. (9) of Ref. 10] restricts the order parameter to the basal plane. In addition, the in-plane (hexagonal) anisotropy energy favors alignment of the moments along any of the three directions perpendicular to the hexagonal lattice vectors. Note that the form of the anisotropy energy is the same for either D_{6h}^4 and D_{3d}^3 symmetry groups. The effect of a magnetic field on the AFM order is included through the Zeeman coupling to the atomic moments, which in general mixes different nearly degenerate representations of the space group within the exchange multiplet.[14] The competition between the anisotropy energy and the Zeeman coupling induces hexagonal modulations of the upper critical field as a function of the orientation of the field in the basal plane at the transition to the superconducting phase.[15,16] The in-plane anisotropy energy is small, since a large in-plane anisotropy energy would produce an orthorhombic modulation of the upper critical field, which is not observed. Higher-order anisotropy terms[17] which might resolve the remaining degeneracy and thus favor alignment of the moments along the propagation vector of the magnetic order would be extremely small. Therefore, the three structures shown in Fig. 1 of Ref. 10 are degenerate, or quasidegenerate, and should be considered in the analysis of the magnetic structure and neutron scattering in the presence of an in-plane magnetic field.

From the magnetic peak intensities reported by Hayden *et al.*[18] we estimate that the intensity at $\vec{q}_1=[1/2,0,0]$ is at most 12% of the intensity at $\vec{Q}_1=[1/2,1,0]$. From this ratio we estimate that between 4% and 8% of the sample displays magnetic moments nonparallel to their propagation vector $\vec{q}_1$ (in this case). As Fåk *et al.* discuss in their comment, it is expected that a macroscopic single crystal should show equally populated magnetic domains when only the configurations with moments laying parallel to the propagation vector of the magnetic order are present. An indication of unequally populated domains is shown in Fig. 1 of Hayden *et al.*; the magnetic Bragg peaks with propagation vectors $\vec{Q}_3=[-1,3/2,0]$ and $\vec{Q}_2=[-3/2,1/2,0]$ show only 50% and 60% of the intensity of the peak at $\vec{Q}_1=[1/2,1,0]$, respectively. Hayden *et al.*[18] do not display the intensities of $\vec{q}_2=[0,1/2,0]$ and $\vec{q}_3=[1/2,-1/2,0]$, so it is not clear whether they are larger or smaller than the intensity at $\vec{q}_1=[1/2,0,0]$. If they are smaller than the intensity at $\vec{q}_1=[1/2,0,0]$ it would prove that only a small fraction of the sample display magnetic order with moments non parallel to their propagation vector. Conversely, if the intensities at $\vec{q}_2=[0,1/2,0]$ and $\vec{q}_3=[1/2,-1/2,0]$ are larger than the intensity at $\vec{q}_1=[1/2,0,0]$, this could mean that a larger proportion of the sample displays moments nonparallel to the propagation vector.

Thus, in our analysis we consider the possibility of degenerate, or nearly degenerate, magnetic structures by making an average over different distributions of domains. We also presented results and predictions for the single magnetic structure with the magnetization parallel to the propagation vector. The authors of the comment seem to have overlooked this prediction, which if we had confined our analysis to a single representation of the space group, as Fåk *et al.* advocate, would be the only relevant structure.

We did mistake the magnetic-field direction in the experiment reported in Ref. 19. In the correct geometry of that experiment the field was along the reciprocal-lattice direction $[-1,2,0]$. The ratios reported in Eq. (5) of Ref. 10, and in the paragraph that follows that equation, should be modified as follows. When only domain "1" is populated we have

$$r=1. \tag{1}$$

For a crystal with equally populated magnetic domains, the correct ratio between the scattering rate at high field and zero field is

$$r=\frac{1-[0.441\cos(\theta_H+\pi/2)]^2}{\langle 1-[0.441\cos(\theta)]^2\rangle}=0.89. \tag{2}$$

Our previous conclusion, stated for the incorrect field orientation, is unchanged for the correct field orientation; it is not possible based on existing data to conclude whether or not the U moments rotate with the field, because of the small change in intensity that is expected for this Bragg peak and the large error bars that are reported for the intensity.

We also concluded that, in order to understand UPt_3 magnetism in the presence of magnetic field or under pressure, systematic, zero-field measurements of the intensity of a number of magnetic peaks in the same single crystal, such as those reported in Ref. 20, need to be carried out. Furthermore, our hypothesis that intrinsic stacking faults pin the AFM domain walls in the *ab* plane and fix the spatial distribution of domains with different propagation vectors has been recently reinforced. For uniaxial pressures applied to the basal plane a significant increase in the magnetic intensity has been reported[21] in contrast with the relatively small change in a magnetic field.[19,22] Pinning by intrinsic stacking faults may help explain this difference, since the applied magnetic field leaves the distribution of regions with different propagation vectors unaltered. However, uniaxial pressure likely disturbs the configuration of stacking faults leading to a stronger effect on the magnetic structure.

In conclusion, our analysis of the neutron-scattering data is based on a sound theoretical model for possible magnetic structures in UPt_3, which is more general than would be allowed based on a single irreducible representation of the space group. The relative importance of exchange interactions leads naturally to mixed irreducible representations of the crystal space group, which are relevant because they are energetically allowed. Our approach also includes and made predictions [Eq. (1) and Eq. (8) in Ref. 10] for the magnetic configuration with magnetic moments parallel to the propagation vector of the magnetic order. As we concluded in Ref. 10 the experimental data is roughly consistent with this configuration or with one where two unequally populated domains are present.

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