Comment on "Magnetic field effects on neutron diffraction in the antiferromagnetic phase of UPt$_3$"

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Moreno and Sauls have recently tried to reanalyze earlier neutron-scattering studies of the antiferromagnetic order in UPt$_3$ with a magnetic field applied in the basal plane. In their calculation of the magnetic Bragg-peak intensities, they perform an average over different magnetic structures belonging to distinct symmetry representations. This is incorrect. In addition, they have mistaken the magnetic field direction in one of the experiments, hence invalidating their conclusions concerning the experimental results.

Neutron elastic-scattering measurements of the magnetic order in UPt$_3$ have shown that a high magnetic field applied in the basal plane or along the hexagonal c axis has virtually no effect on the size of the magnetic moment, the Néel temperature, or on the magnetic structure.[1,2] Within the precision of these measurements, no change of the domain populations[1,2] or of the moment direction in the basal plane[1] was observed. Recently, Moreno and Sauls[3] (MS) have tried to reanalyze the two experiments in Refs. 1 and 2 under the assumption that the pinning energy of the domain walls is larger than the in-plane anisotropy. Unfortunately, the actual analysis of MS is incorrect for reasons that will be discussed below.

UPt$_3$ orders antiferromagnetically below $T_N = 6$ K with a propagation vector $\mathbf{k} = (0.5,0,0)$ and the Fourier component of the moment $\mathbf{m}_k$ parallel to $\mathbf{k}$. For a sample without strain and in zero magnetic field, neutron scattering cannot distinguish between a single-k structure with three K domains and multi-k structures with or without domains. Recent neutron-scattering measurements under uniaxial pressure[4] indicate that the magnetic structure is single k, and we will only discuss this case in this Comment. We restrict ourselves also to magnetic fields in the basal plane (Ref. 2 also treated $H \| c$), as this is the only case analyzed by MS. Since the temperature dependence of the moment is smooth without any jump at the transition temperature and there is no evidence of any hysteresis or latent heat, we assume that the transition is second order. We also assume that the moment is static and that the crystal structure in the paramagnetic phase is hexagonal with space group $P6_3/mmc$ (D_{6h}^4), although a lower trigonal symmetry was recently reported.[5] These assumptions were also made by MS.

Group-theory analysis[6–8] indicates that for a propagation vector $\mathbf{k} = (0.5,0,0)$ the magnetic representation that describes a magnetic moment at the U position ($2c$) can be decomposed into six irreducible representations (IRs) of order one. A ferromagnetic alignment of the moments within the unit cell (shown by neutron-scattering measurements) is compatible with only three of these, namely, Γ_2, Γ_4, and Γ_6.[9] Application of the Landau theory for a second-order phase transition provides an important simplification to the analysis of the resulting magnetic structure because it requires that only one IR become critical. Consequently, we can limit the *symmetry-allowed* magnetic structures to those defined by a single IR. As each of these IRs has only one basis vector associated with it in the present case, we find immediately that the moments are fixed along specific crystallographic directions. The corresponding moment directions (assuming a single-k structure) are parallel to $\mathbf{k}$, perpendicular to $\mathbf{k}$ in the basal plane, and parallel to the c axis, respectively. Since the $\mathbf{Q} = (0.5,0,0)$ magnetic Bragg reflection is absent in neutron-scattering measurements, the antiferromagnetic phase is described by Γ_2 with the moment parallel to the propagation vector. In this case, there are no S domains,[10] i.e., there is only one possible orientation of the moment $\mathbf{m}$ with respect $\mathbf{k}$. There are, however, three K domains, corresponding to the three equivalent orientations of $\mathbf{k}$ in the basal plane: $\mathbf{k}_1 = (0.5,0,0)$, $\mathbf{k}_2 = (0,0.5,0)$, and $\mathbf{k}_3 = (0.5,-0.5,0)$. For unstrained (annealed) samples, the three K domains have equal populations, as seen from the intensity in neutron-scattering measurements.[11] This is the standard picture of the magnetic order in UPt$_3$ as observed by neutron and x-ray scattering.

When a magnetic field is applied within the basal plane, the K domain with moments perpendicular to the applied field is favored over the other K domains. For a sufficiently strong field, one would expect a repopulation of the different K domains. Within current precision, this has not been observed by neutron-scattering measurements.[1,2] However, recent measurements under uniaxial pressure suggest a domain repopulation.[4]

Moreno and Sauls assume in their work that there are three "domains" for a given K domain, as illustrated in their Fig. 1. However, the magnetic structures shown in Figs. 1(b) and 1(c) are not domains of the structure in Fig. 1(a). Rather, they are $2S$ domains of a different magnetic structure. While Fig. 1(a), which corresponds to the actual magnetic structure of UPt$_3$, is described by the basis vectors associated with Γ_2, the structure shown in Figs. 1(b) and 1(c) corresponds to a mixing of those associated with Γ_2 and Γ_4. Hence the structure presented as Fig. 1(a) has a different symmetry than that shown in Figs. 1(b) and 1(c). Since ordering under Γ_2 and Γ_4 involves more than one IR, the magnetic structure shown in Figs. 1(b) and 1(c) must involve a first-order transition, in contrast to the structure of Fig. 1(a) which is compatible with a second-order phase transition. Although it appears as if the magnetic structures shown in Fig. 1 have the same energy, they have different symmetries. Hence, one would expect that either the structure in Fig. 1(a) or the structure in Figs. 1(b) and 1(c) is established. The absence of the $\mathbf{Q}=(0.5,0,0)$ magnetic Bragg reflection in neutron-scattering data shows unambigiously that the structure shown in Fig. 1(a) is established. It is also the only one of the structures shown in Fig. 1 that is compatible with a second-order phase transition (provided that the nonmagnetic space group is $P6_3/mmc$). Even if the magnetic phase transition were first order so that the restrictions of Landau theory no longer apply, the structure of Fig. 1(a) has still a different symmetry from that in Figs. 1(b) and 1(c).

In their actual analysis of the experimental data, MS evaluate the ratio r of the magnetic Bragg-peak intensities in field and in zero field, given by Eq. (4) in Ref. 3. However, they average Eq. (4) over the two *different structures* shown in Fig. 1. This is clearly wrong. Since the moment is parallel to the propagation vector [see Fig. 1(a)], there are no S domains,[12] and there should be no averaging. If the moment were not parallel to $\mathbf{k}$, but still in the basal plane forming an angle α with respect to the propagation vector (which would require a first-order transition), the two S domains corresponding to $+\alpha$ and $-\alpha$ [these are illustrated in Figs. 1(b) and 1(c) for the case of $\alpha=60°$] should be averaged. The incorrect averaging over different magnetic structures in MS invalidates their analysis of both Refs. 1 and 2. In particular, the results given in Eqs. (5)–(8) are all incorrect.

A second problem is that MS's analysis of the work by van Dijk *et al.*[2] assumes wrongly that the applied field was along the particular a axis that was at 30° with respect to the observed moment. However, van Dijk *et al.* clearly stated that they measured the $\mathbf{Q}=(0.5,0,1)$ magnetic Bragg peak using a vertical-field magnet, which means that the magnetic field was applied perpendicular to the horizontal scattering plane, and hence at 90° with respect to the moments of the studied Bragg peak. The field was thus along the a axis that in the notation of Ref. 2 can be labeled (0, 1, 0) in real space or $(-1,2,0)$ in reciprocal space. In this geometry, there is no reason for the moment in the $\mathbf{k}=(0.5,0,0)$ domain to rotate, so no intensity change is expected. However, if the other K domains would be depopulated and instead contribute to the $\mathbf{k}=(0.5,0,0)$ domain, the intensity at $\mathbf{Q}=(0.5,0,1)$ would increase by a factor of 3 (for a full domain repopulation) as stated by van Dijk *et al.*

In the experiment by Lussier *et al.*,[1] the lower magnitude of the applied field allowed the use of a horizontal field magnet, which gives a much larger choice of geometries. Their analysis, which is correct, shows that for a field of 3.2 T in the basal plane there is no domain repopulation, not even for a field-cooled sample where the pinning energy is irrelevant, as only the most favored domain will form on cooling through the Néel temperature. Notably, they also did not observe any moment rotation.

MS also suggest that it is not known whether the Fourier component of the magnetic moment is parallel to the propagation vector in zero field. However, Hayden *et al.*[11] showed beyond any doubt that $\mathbf{m}_k$ is parallel to $\mathbf{k}$. For the same sample, they first showed that the K domains are equally populated, by measuring at three different Bragg peaks, each corresponding to a different $\mathbf{k}$ vector. Next, they showed that a magnetic Bragg peak with $\mathbf{Q}\|\mathbf{k}$ in one of these domains has zero intensity, which proves unambiguously that $\mathbf{m}_k$ is parallel to $\mathbf{k}$. The same result has been found by other groups, including ours.

In summary, although the scenario with a large pinning energy and the discussion of the symmetry-breaking properties of a triple-k structure are interesting, the actual analysis by Moreno and Sauls of the field dependence of the magnetic Bragg-peak intensities in UPt$_3$ is incorrect, as they perform an average over different magnetic structures that are symmetry inequivalent. There is no experimental evidence that the moment rotates away from the propagation vector when a magnetic field is applied in the basal plane. Also, such a rotation is not compatible with the symmetry properties of a second-order phase transition.

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[1] B. Lussier, L. Taillefer, W.J.L. Buyers, T.E. Mason, and T. Petersen, Phys. Rev. B **54**, 6873 (1996).

[2] N.H. van Dijk, B. Fåk, L.P. Regnault, A. Huxley, and M.T. Fernández-Díaz, Phys. Rev. B **58**, 3186 (1998).

[3] J. Moreno and J.A. Sauls, Phys. Rev. B **63**, 024419 (2001).

[4] N.H. van Dijk, P. Rodière, F. Yakhou, M.T. Fernández-Díaz, B. Fåk, A. Huxley, and J. Flouquet, Phys. Rev. B **63**, 104426 (2001).

[5] D.A. Walko, J.-I. Hong, T.V. Chandrasekhar Rao, Z. Wawrzak, D.N. Seidman, W.P. Halperin, and M.J. Bedzyk, Phys. Rev. B **63**, 054522 (2001).

[6] E.F. Bertaut, J. Appl. Phys. **33**, 1138 (1962); E.F. Bertaut, Acta Crystallogr., Sect. A: Cryst. Phys., Diffr., Theor. Gen. Crystallogr. **24**, 217 (1968); E.F. Bertaut, J. Phys. Colloq. **C1**, 462 (1971); E.F. Bertaut, J. Magn. Magn. Mater. **24**, 267 (1981).

[7] Yu. A. Izyumov, V.E. Naish, and R.P. Ozerov, *Neutron Diffraction of Magnetic Materials* (Consultants Bureau, New York, 1991).

[8] A.S. Wills, Physica B **276-278**, 680 (2000). Program available from ftp://ftp.ill.fr/pub/dif/sarah/

[9] The labeling of nonzero IRs follows the scheme used by O.V. Kovalev, *Representations of the Crystallographic Space Groups*, 2nd ed. (Gordon and Breach Science Publishers, Yverdon, Switzerland, 1993).

[10] S domains, also termed orientational domains, differ only in the choice of the equivalent local axes. They are necessarily of the same symmetry and so may be described by the same irreducible representation.

[11] S.M. Hayden, L. Taillefer, C. Vettier, and J. Flouquet, Phys. Rev. B **46**, 8675 (1992).

[12] There are also two so-called 180° domains, which differ only by the phase of the stacking sequence (i.e., $+-+-$ vs $-+-+$), but since they are indistinguishable, we do not count them here.