Comment on "DFT+U search for the energy minimum among eight collinear and noncollinear magnetic structures of GdB_4"

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M. N. Huda and L. Kleinman [Phys. Rev. B **78**, 094424 (2008)] suggested that the magnetic structure reported for GdB_4 from spherical neutron polarimetry [J. A. Blanco, P. J. Brown, A. Stunault, K. Katsumata, F. Iga, and S. Michimura, Phys. Rev. B **73**, 212411 (2006)], a noncollinear antiferromagnetic arrangement of the Gd^{3+}, whose magnetic space group is $P4/m'b'm'$ may not be the right one. Instead of that proposed from neutron scattering, they concluded based on DFT+U calculations that the ground state corresponds to a collinear arrangement. This conclusion that the DFT+U method leads to a completely magnetic arrangement from that found experimentally demonstrates the limitation of this method when used to predict magnetic structures.

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GdB_4 has a tetragonal crystal structure at room temperature, but below T_N=42 K it orders antiferromagnetically with the magnetic moments within the *ab* plane.[1–3] The first attempt to determine the magnetic structure was made using resonant x-ray scattering azimuthal angle scans at the Gd L_3 edge, where the existence of phase quadrature between the magnetic and charge amplitudes was interpreted as due to the presence of a collinear arrangement of the Gd^{3+} magnetic moments.[4] However, at the L_3 edge one is tuning electronic transfers from a $2p$ shell to an unoccupied $5p$ one and not directly to the $4f$ shell responsible for the magnetic moments on Gd^{3+} ions. In fact, our interpretation[5–7] of the x-ray data was also compatible with several noncollinear arrangements which fitted the experimental data just as well as the collinear one reported in Ref. 4. Moreover, the existence of a collinear arrangement implies a structural phase transition for which up to now there is no evidence. Nevertheless, one can always ascribe a collinear arrangement to a very tiny orthorhombic distortion below the experimental resolution. It is for this reason that we made a spherical neutron polarimetry experiment on a single crystal in order to determine the magnetic arrangement of GdB_4 unambiguously.[8]

In a recent theoretical paper Huda and Kleinman[9] suggested that the canted tetragonal structure determined from this experiment may not be correct. The comments made in the introduction to their paper[9] demonstrate a complete lack of understanding of the technique of neutron polarimetry and the information it provides. This information depends neither on precise values of the lattice constants (the cell parameters quoted in the *A. Sample preparation section* in Ref. 8, $a=b=7.1316(2)$ Å and $c=4.0505(3)$ Å, were those used as starting set for the orientation of the single crystal at room temperature) nor on fine details of the low-temperature crystal structure. The conclusion that the structure must retain tetragonal symmetry rests rather on the absence of the depolarisation effects which would necessarily arise from the presence of domains with orthogonal spin directions which must be formed in any tetragonal to orthorhombic transition. The noncollinear tetragonal structure is further confirmed by the good agreement between the observed and calculated values of the components of the polarization matrices which could not be obtained with any collinear model. One may note that the expression for the magnetic interaction vector given below, on which the scattered polarization depends, does not involve the cell parameters directly at all.[10] The magnetic interaction vector $\mathbf{M}_\perp=\mathbf{k}\times\mathbf{F_M}(\mathbf{k})\times\mathbf{k}$, where $\mathbf{F_M}$ is the magnetic structure factor being the kth Fourier component of the magnetization distribution and is given by

$$\mathbf{F_M(hkl)} \propto \sum_{j=1}^{4} \mathbf{M_j} \exp(i\mathbf{r_j} \cdot \mathbf{k}), \tag{1}$$

$$= \sum_{j=1}^{4} \mathbf{M_j} \exp[2\pi i(hx_j + ky_j + lz_j)], \tag{2}$$

where the coordinates $\mathbf{r_j}=\mathbf{x_j a}+\mathbf{y_j b}+\mathbf{z_j c}$ for the four Gd moments ($\mathbf{M_j}$) are given in Ref. 8 and the Miller indices (hkl) define the scattering vector $\mathbf{k}=\mathbf{ha}^*+\mathbf{kb}^*+\mathbf{lc}^*$.

The fact that the DFT+U method[9] predicts a completely different magnetic arrangement from that found experimentally[8] is interesting. It should encourage other groups to study the extent to which different theoretical techniques are able to predict magnetic structure.

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