Reply to "Contradicting Reports on Magnetic Properties of Layered Molecule-Based Material $N(n\text{-}C_3H_7)_4[Fe^{II}Fe^{III}(C_2O_4)_3]$"

Bhattacharjee's comment highlights the experimental difficulty in studying the fascinating magnetic properties of the family of mixed valence Fe(II, III) oxalate salts, namely, their sensitivity to small changes of structure and composition varying the synthetic procedure. These changes are of two kinds.

First, the extent of interlayer disorder changes with the preparative conditions. We demonstrated the presence of stacking faults in the title compound and conjectured that they could influence the bulk magnetic properties.[1] In that regard, the earliest samples studied showed Néel type Q ferrimagnetism while samples prepared by a modified procedure, giving better long-range structural order, were type N.[2,3] For the related salt containing butyl-triphenylphosphonium cation we showed that the magnetic order of the product changed from Q to N simply by adding the cation in aqueous or methanol solution.[4]

The second potential influence on the bulk magnetic order is chemical stoichiometry, for example, an excess of Fe(III) creating vacancies on the Fe(II) sublattice. That was investigated in the tetra-butylammonium salt, which behaves as type N,[2,3] by systematic doping of Zn(II) to simulate Fe(II) vacancies. A changeover from N to Q was indeed observed with increasing Zn(II) content.[5] Finally, by elemental analysis of the propyl-triphenylphosphonium salt, a clear correlation was demonstrated between increasing Fe deficiency and a change from N to Q.[4]

In conclusion, the unusual magnetic properties of the $AFe^{II}Fe^{III}(C_2O_4)_3$ ferrimagnets are strongly influenced by structural disorder and nonstoichiometry. Great care is needed to ensure that the properties being studied are intrinsic.

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