

Controlling magnetic sublattices in heterometallic ludwigite  $\text{Fe}_{3-x}\text{Mn}_x\text{BO}_5$ 

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Ludwigite oxyborates  $\text{M}_2\text{M}'\text{BO}_5$ , where M and M' are divalent and trivalent 3d metal ions, respectively, have an intriguing orthorhombic (SG : Pbam) crystal structure made of interconnected low dimensional units in the form of three-leg (3LL) ladders, 3LL1 and 3LL2 [1] (Figure 1). 3LL1 is made of three edge-sharing octahedra, while 3LL2 is made of three corner-sharing octahedra, in the ab plane. These three-octahedra units, or triads, share edges along c to form square ladder sublattices.

The existence of two crystallographically distinct sublattices is actually not anodyne : in vonsenite  $\text{Fe}_3\text{BO}_5$ , Mössbauer and X-ray diffraction studies at room temperature have evidenced that  $\text{Fe}^{3+}$  species occupy preferentially 3LL1, while 3LL2 is occupied by  $\text{Fe}^{2+}$  [2]. In addition, a charge ordering transition has also been observed at  $\text{TCO} = 283 \text{ K}$  [1], [4], resulting from the ordering on 3LL1 of the extra itinerant electron within each  $\text{Fe}^{3+}$  triad. This also impacts the magnetic properties : according to neutron diffraction results, 3LL1 and 3LL2 magnetically order, but independently, at  $\text{TN}_1 = 112 \text{ K}$  and  $\text{TN}_2 = 70 \text{ K}$ , with different propagation vectors,  $k_1 = (0 \ 0 \ 1/2)$  for 3LL1, and  $k_2 = (0 \ 0 \ 0)$  for 3LL2.

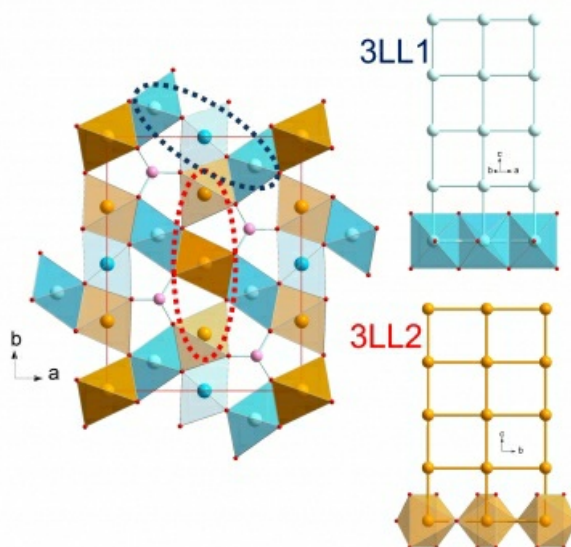
To investigate the origin of this behavior, the isostructural system  $\text{Fe}_{3-x}\text{Mn}_x\text{BO}_5$  has been studied by electron microscopy and low temperature neutron powder diffraction, combined with physical properties measurements. A decrease of both  $\text{TN}$ 's, more pronounced for  $\text{TN}_2$ , along with a reduced ordered moment, are observed with increasing x, up to  $x = 1$ . Interestingly, results show that Mn is substituted preferentially on 3LL2 : for  $x = 1$ , in addition to the disappearance of charge ordering features, only short-range magnetic ordering is observed on 3LL2, leading to superparamagnetic ac susceptibility response [3]. Relaxor type ferroelectric properties are observed up to  $x = 1$  at low temperature. Surprisingly, for  $x = 1.5$ , 3D long-range ordering below  $\text{TN} = 100 \text{ K}$  is observed, which couples both 3LLs within a collinear  $k = 0 \ 0 \ 0$  structure, without any sign of magnetic disorder.

The existence of two magnetic sublattices, whose composition and coupling can be tuned through preferential substitution, make of this ludwigite system a promising one to study the effect of controlled magnetic disorder in transition metal compounds.

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