Interplay between charge transfer and magnetic transition in the quadruple perovskite $(YMn_3)Mn_4O_{12}$

Marine Verseils[1], Francesco Mezzadri[2], Davide Delmonte[2], Benoit Baptiste[1], Yannick Klein[1], Laurent Chapon[3*], Edmondo Gilioli[2] and Andrea Gauzzi[1]

[1]*IMPMC-UMR 7590, UPMC, Paris, France*

[2]*IMEM-CNR, Parma, Italy*

[3]*ILL, Grenoble, France (* Present address: Diamond, Oxford, UK)*

The quadruple perovskitesystem*A*$Mn_3Mn_4O_{12}$(QP-Mn) (A=Na, Ca, La, Bi), has attracted a great deal of interest because of theunique charge, spin and orbital orderings arising from two distinct magnetic A'- and B-sublattices and from the absence of oxygen defects. Most notable are a full charge order [1, 2] and multiferroicity [3,4,5,6].The stability conditions of these orderings depend on two distinct B-O-B and A'-O-B superexchange paths, characterized by an unusuallylarge tilt of the BO6 octahedra with Mn-O-Mn bondangles ~136°, thusleading to competing AFM or FM exchange interactions not explained by the GKA rules.

In order to investigate the role of the bond geometry on the stability of competing magnetic structures in the system, we have successfully synthesized under high pressure and studied the structural and magnetic properties of $(YMn_3)Mn_4O_{12}$, where the small Y^{3+} ion is expected to enhance the magnetic exchange interaction with respect to the isovalent compound $(LaMn_3)Mn_4O_{12}$. According to this expectation, we have observed the highest AFM ordering temperature for the B-sites, $T_{N,B}$=108K. On the other hand, we have found four unexpected features, not reported before in other *A*$Mn_3Mn_4O_{12}$ compounds:(1) a second-order structural phase transition at 200 K that seems to be a precursor of the AFM ordering of the B-sites at low temperature; (2) the absence of any magnetic ordering of the A' sublattice, which has systematically been found in other *A*$Mn_3Mn_4O_{12}$ compounds in the 20-60 K range; (3) on the other hand, susceptibility measurements at low field unveil an anomaly suggesting the existence of a latent ordering; (4) at low-temperature the specific heat follows an unusual quadratic - instead of cubic - dependence, which suggests that excitations other than acoustic phonons dominate the entropy.

The above anomalies call for further studies, e.g. RIXS, that may probe the electronic excitation spectrum, possibly dominated by the proximity of competing magnetic orderings.

References:

[1]A. Prodi *et al.,* Nature materials, **3**, 48-52 (2004)[4] R.D Johnson *et al.,* Physical Letters B, **108**, 6 (2012)

[2] A. Prodi *et al.,* Physical review B, **90**, 180101 (2014) [5] X.Z. Lu *et al.,*Physical review Letters, **108**, 18 (2012)

[3] G.Zhang *et al.,* Physical review B, **84**, 17 (2011)[6] F. Mezzadri *et al.,* Physical review B, **79**, 10 (2009)