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Multipolar and Néel magnetic orders induced by strong spin-orbit coupling in *5d* frustrated cubic double perovskites

E. Kermarrec[a*], D. D. Maharaj[b], G. Sala[b,c], M. B. Stone[c], C. Ritter[d], F. Fauth[e], C. A. Marjerrison[f], J. E. Greedan[f,g], A. Paramekanti[h] and B. D. Gaulin[b,f]

a. Université Paris Saclay, CNRS, Laboratoire de Physique des Solides, 91405 Orsay, France
b. Department of Physics and Astronomy, McMaster University, Hamilton, Ontario L8S 4M1, Canada
c. Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
d. Institut Laue-Langevin, Boîte Postale 156, 38042 Grenoble Cedex, France
e. CELLS-ALBA Synchrotron, Carrer de la Llum 2-26, 08290 Cerdanyola del Vallès, Barcelona, Spain
f. Brockhouse Institute for Materials Research, McMaster Univ., Hamilton, Ontario L8S 4M1, Canada
g. Department of Chemistry and Chemical Biology, McMaster University, Ontario L8S 4M1, Canada
h. Department of Physics, Univ. of Toronto, 60 St. George Street, Toronto, Ontario M5S 1A7, Canada

* email : edwin.kermarrec@universite-paris-saclay.fr

We report time-of-flight neutron spectroscopy and neutron and x-ray diffraction studies of the *5d* double perovskite magnets, Ba_2MOsO_6 (*M* = Y, Zn, Mg, Ca) [1,2]. These materials host antiferromagnetically coupled $5d^2$ (Os^{6+}) or $5d^3$ (Os^{5+}) ions decorating a face-centered cubic (fcc) lattice (Fig. 1a) and remain cubic down to the lowest temperatures. In $5d^2$ osmate compounds the large spin-orbit coupling splits the t_{2g} electronic levels into effective *j*=1/2 and *j*=3/2 levels, while the latter is further splitted by the crystal field interaction (Fig. 1c). This leads to a non-Kramers doublet ground state separated by a gap to the excited triplet, as shown by our neutron spectroscopy measurements (Fig. 1b). These $5d^2$ compounds exhibit thermodynamic anomalies consistent with a single-phase transition at a temperature *T**, and a gapped magnetic excitation spectrum with spectral weight concentrated at wave vectors typical of type-I antiferromagnetic orders. However, while muon spin resonance experiments show clear evidence for time-reversal symmetry breaking below *T**, we observe no corresponding magnetic Bragg scattering signal. These results are shown to be consistent with ferro- octupolar symmetry breaking below *T**, and will be discussed in the context of other *5d* double perovskite magnets and theories of exotic orders driven by multipolar interactions.

[1] E. Kermarrec et al., Phys. Rev. B **91**, 075133 (2015); D. D. Maharaj et al., Phys. Rev. Lett. **124**, 087206 (2020).
[2] A. Paramekanti, D. D. Maharaj, and B. D. Gaulin, Phys. Rev. B **101**, 054439 (2020).

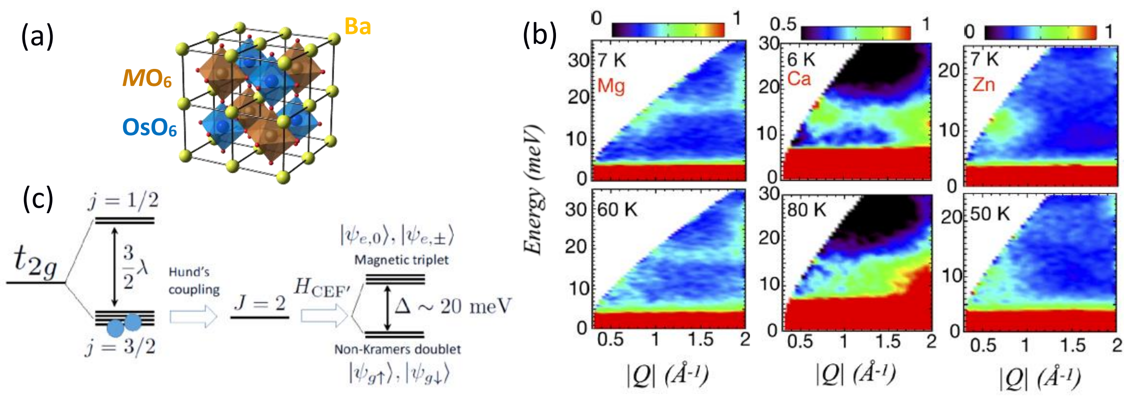

Figure 1 : (a) Ba_2MOsO_6 (with *M* = Y, Mg, Ca and Zn) osmates double perovskites crystallize in a perfectly cubic $Fm\bar{3}m$ structure. (b) Inelastic neutron scattering measurements show evidence for a gap Δ ~ 10-20 meV below *T**, which is the result of a combination of strong spin-orbit-coupling and crystal field interaction (c), that leads to a non-Kramers ground state doublet with vanishing matrix elements for the dipole operators *J*, precluding dipolar order, and promoting the emergence of multipolar orders.