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

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We report time-of-flight neutron spectroscopy and neutron and x-ray diffraction studies of the 5d double perovskite magnets, Ba\textsubscript{2}M\textsubscript{2}OsO\textsubscript{6} (M = Y, Zn, Mg, Ca)\textsuperscript{[1,2]}. These materials host antiferromagnetically coupled 5d\textsuperscript{2} (Os\textsuperscript{6+}) or 5d\textsuperscript{3} (Os\textsuperscript{5+}) ions decorating a face-centered cubic (fcc) lattice (Fig. 1a) and remain cubic down to the lowest temperatures. In 5d\textsuperscript{2} osmate compounds the large spin-orbit coupling splits the t\textsubscript{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\textsuperscript{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.


\begin{figure}[h]
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\includegraphics[width=\textwidth]{figure1}
\caption{(a) Ba\textsubscript{2}M\textsubscript{2}OsO\textsubscript{6} (with M = Y, Mg, Ca and Zn) osmates double perovskites crystallize in a perfectly cubic Fm\textoverline{3}m structure. (b) Inelastic neutron scattering measurements show evidence for a gap \Delta \sim 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.}
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