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Role of competing magnetic interactions and anisotropies in determining the ground states in magnetoelectric/multiferroic honeycomb $M_4Ta_2O_9$ ($M = Mn, Co, Fe$ and Ni)

In magnetoelectrics (ME) magnetic ordering and electric polarization (EP) coexist. The ME effect is larger, if in these materials EP is of spin origin and the coupling is strong. This coupling can be utilized in applications such as MRAMs. Although technically important, the coupling mechanism is complicated. Two types of materials fulfill this requirement: (i) Type II multiferroics (ii) Materials which lack spontaneous EP in the ground state and it is induced by an external magnetic field. The magnetic order parameter should break both time reversal and inversion symmetries [1-4]. $M_4A_2O_9$ ($M = Mn, Co, Fe$ and Ni and $A = Nb, Ta$) are a rare family of materials where depending on M , either (i) or (ii) are induced below the magnetic ordering [5-8]. The structure is built from an alternative stacking of two different honeycombs leading to a competition between various magnetic interactions, anisotropy and dimensionality. In order to elucidate the emergence of (i) or (ii) in $M_4A_2O_9$, a comprehension of these competing interactions is essential, which in turn also necessitates the investigation of its electronic structure. In this work, we combined neutron powder diffraction, inelastic neutron scattering and theoretical methods including density functional theory to determine the magnetic structures, magnetic excitations and electronic structure of $M_4Ta_2O_9$. These investigations led to a discovery of a variety of fundamental spin systems (easy-axis vs easy-plane) and excitations (gapped vs ungapped) in this family of materials, which exhibit a variety of ground states [9].

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