

Magnetic order induced symmetry-breaking in the coupled honeycomb system $\text{Fe}_4(\text{Nb,Ta})_2\text{O}_9$

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Magnetoelectric (ME) multiferroics are compounds which exhibit simultaneous magnetic ordering (MO) and electric polarization (EP) which are coupled. This coupling is utilized in applications such as MRAMs, sensors and capacitors. The ME effect is significant, if the emerging EP is of spin origin and the coupling is strong. However the emergence of EP and the coupling mechanism between these two orders are not completely understood and subject to extensive ongoing research. Three types of materials fulfill this requirement: (i) Type II multiferroics such as orthorhombic RMnO_3 (R=rare earth), where spontaneous EP is induced by the MO, (ii) Polar magnets such as $\text{M}_2\text{Mo}_3\text{O}_8$ (M=Fe and Mn), which also exhibit MO induced EP, but has a polar paramagnetic phase and (iii) Magnetoelectric materials such as Cr_2O_3 , where EP is induced by an external magnetic field below MO [1-6]. $\text{M}_4\text{A}_2\text{O}_9$ (M=Fe, Co, Mn and A=Nb,Ta) are a family of materials where depending on M, either (i) or (iii) emerge below MO. The unit cell contains two crystallographically distinct magnetic sites M, which form edge-shared coplanar and corner-shared buckled honeycombs which are connected along c. The interaction between the honeycombs enables a competition between the interlayer and the intralayer exchange interactions and anisotropies which results in ground states with various spin orderings [7-8]. Recently $\text{Fe}_4\text{Ta}_2\text{O}_9$, in contrast to the magnetoelectric Co and Mn counterparts, was reported to exhibit symmetry-breaking and EP below MO from dielectric property measurements [9-10]. Elucidation of the emergence of EP in $\text{Fe}_4\text{Ta}_2\text{O}_9$ necessitates a comprehension of its crystal, magnetic and electronic structure. In this work, we combined neutron powder diffraction, inelastic neutron scattering, bulk property measurements and theoretical methods to determine the crystal/magnetic structures, magnetic excitations and electronic structure of $\text{Fe}_4\text{Ta}_2\text{O}_9$ and compared it with the related compound $\text{Fe}_4\text{Nb}_2\text{O}_9$. $\text{Fe}_4\text{Ta}_2\text{O}_9$ exhibits the unique feature of the emergence of both the symmetry-breaking multiferroic and magnetoelectric phases below MO, whereas $\text{Fe}_4\text{Nb}_2\text{O}_9$ only exhibits magnetoelectric phases below MO. This enables $\text{Fe}_4(\text{Nb,Ta})_2\text{O}_9$ to be an ideal system to investigate the transition between the multiferroic and magnetoelectric phases. We elucidate these unique phenomena utilizing symmetry arguments in terms of spin and orbital degrees of freedom.

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