Title: Impact of Magnetic Ion Substitution on the Crystal Structure of Multiferroic Aurivillius Phases

Abstract:

Multiferroic materials, characterised by coexisting ferroelectric and ferromagnetic/ferrimagnetic polarisation states, hold promise for advancing memory technologies beyond conventional limits. Achieving technologically-competitive devices requires enhancing the magnetic properties of multiferroics, through strongly interacting magnetic moments with long-range spin ordering. This seminar will focus on the development of a rare multiferroic system, the Aurivillius phase $Bi_6Ti_xFe_yMn_zO_{18}$ (B6TFMO), which uniquely exhibits ferroelectricity, ferrimagnetism, and magnetoelectric switching at room temperature.[1,2]

Aurivillius phases are a family of layered ferroelectrics with the general formula $Bi_2O_2(A_{m-1}B_mO_{3m+1})$, where '*m*' denotes the number of perovskite-type layers interleaved between $[Bi_2O_2]^{2+}$ interface layers. To optimise the magnetic response of B6TFMO for next-generation data storage applications, it is essential to understand how magnetic ion substitution influences its crystal structure. Atomic-resolution energy dispersive X-ray analysis reveals that cation partitioning in B6TFMO arises from elastic strain and electrostatic energy contributions, which depend on distance from the $[Bi_2O_2]^{2+}$ fluorite-type layers.[3] This partitioning causes manganese ions to preferentially locate within the centre perovskite layers, resulting in up to a 90% increase in potential ferromagnetic spin alignments within these layers, compared to a scenario with random distribution of magnetic cations across all five available B-sites. Density functional theory studies confirm that the presence and specific distribution of manganese are key to establishing long-range ferrimagnetic order within B6TFMO.[4] Additionally, integrated differential phase contrast imaging shows that significant octahedral tilting is necessary to position magnetic ions close enough to facilitate this long-range order.[5]

Motivated by an observed enhancement in magnetisation and ferrimagnetic Curie temperature with higher manganese content, we investigated the solubility limits of magnetic ion inclusion in B6TFMO. Our findings demonstrate that transitioning to higher-*m* Aurivillius phases allows for greater incorporation of magnetic ions while maintaining in-plane ferroelectric properties.[6] These studies offer valuable insights into the structural constraints of magnetic ion substitution in multiferroics, informing the design of future materials that integrate magnetic and ferroelectric properties for data storage applications.

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