Design Strategy and Prediction of New Room Temperature Magnetoelectric Multiferroics

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Perovskite ABO_3 oxides display many promising functional properties including ferroelectricity and magnetism. An ongoing challenge has been to design and synthesize materials in which ferroelectric and magnetic orderings not only coexist in a single phase above room temperature, but also are nontrivially coupled. Such a cross coupling between these different ferroic orderings could lead to many novel applications, in particular, involving electrical control of magnetism that is required in case of MERAMs or magnetic capacitors.

Taking advantage of a newly developed mechanism to induce ferroelectricity - "*hybrid improper ferroelectricity*"[1-4] and supported by first-principles density functional theory (DFT) calculations, we will discuss and outline strategies where electric field control of magnetism can be possible in systems consisting of ABO_3 building blocks. First, we take the example of ABO_3 / $A'BO_3$ superlattices, which may be amenable to advanced oxide thin film growth techniques, and second, bulk $AA'BB'O_6$ double perovskites which can be realized by synthetic chemical route.

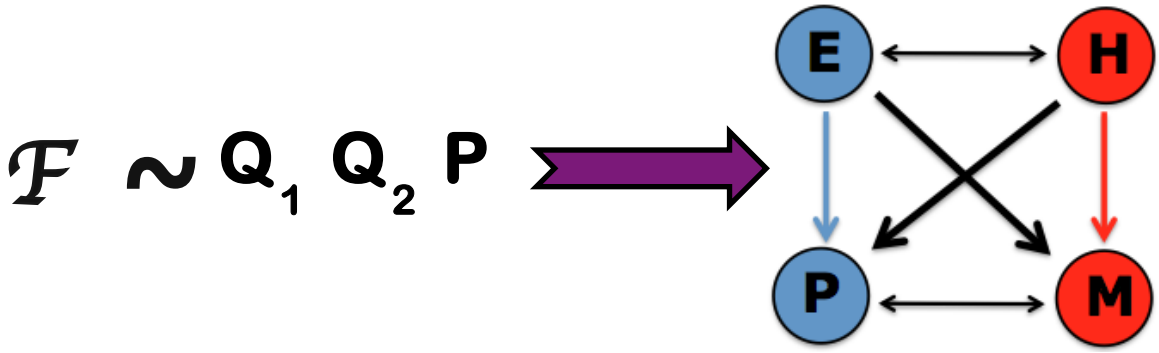

Figure: Hybrid improper ferroelectricity: route to create new room temperature magnetoelectric multiferroics, here, switchable polarization P is driven by two primary order parameters Q_1 and Q_2. Individually, Q_1 and Q_2 can drive weak magnetization as well as linear magnetoelectric coupling, which is promising for electric field control of magnetism.

ABO_3 / $A'BO_3$ Superlattice: We show that in $(LnFeO_3)_m$ / $(Ln'FeO_3)_n$ digital superlattices (Ln, Ln' = lanthanide or Y, and both *m*, *n* are odd integers) constructed from *Pnma* perovskites, BO_6 octahedral rotations induce a spontaneous electrical polarization. Furthermore, this rotation pattern is shown to induce linear magnetoelectricity and weak-ferromagnetism, much like the recently

discussed '327' manganite Ruddlesden-Popper [2]. In these ferrite superlattices, however, it is clear that both the ferroelectric and magnetic ordering temperatures should occur above room temperature. We discuss how the Ln/Ln' cation radius mismatch controls the magnitudes of the induced polarization and magnetization, as well as the barrier to switch the polarization. Finally, we suggest that these systems represent a practice way to realize a linear magnetoelectric effect at room temperature. To help in identifying a successful design strategy we have also addressed the governing factors that determine the strength of the linear magnetoelectric coupling.

AA'BB'O_6 Bulk Double Perovksites: Ordered double perovskite materials with general chemical formula $A_2BB'O_6$ (where A is an alkaline-earth or rare earth ion, BB' are transition-metal ions) exhibit rare combination of ferromagnetic/ferrimagnetic-insulating properties above or near room temperature. Here, the high magnetic T_c is due the complex interplay between B and B' transitions metal cations arranged in rock salt manner. Three types of AA' orderings are possible in $AA'BB'O_6$, namely, layer, rock salt and columnar. Symmetry arguments show that layer and rock salt orderings can have spontaneous polarization induced due to *hybrid improper* mechanism. Thus, this family of materials can be promising for investigation of electric field control of magnetism. We take up the case of Sr_2CrOsO_6, which is a ferrimagnetic insulating double perovskite with $T_c \sim$ 725K [5], the highest known T_c for a magnetic insulator with appreciable uncompensated magnetic moment among the oxide family [6]. In $AA'CrOsO_6$ systems, we proposed a chemical route to stabilize the layered AA' phase. We have also identified new multiferroic materials in layer $AA'CrOsO_6$ with low switching barriers and with strong magnetoelectric coupling, expected above room temperature. Our study, which includes an extensive choice of A and A' cations should form the platform for checking feasibility of synthesizing them and verifying our proposal.

We believe that the general conception and design strategy for room temperature electrical control of magnetism by considering ABO_3 perovskite oxides as building blocks will motivate further experimental and theoretical investigation in this direction.

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