Room-temperature multiferroic superlattices

L. Bellaiche[1], Xiang Ming Chen[2], Jorge Íñiguez[3],
Wei Ren[4], Yurong Yang[1] and Hongjian Zhao[1,2]

[1]*Physics Department & Institute for Nanoscience and Engineering, University of Arkansas, Fayetteville, Arkansas 72701, USA*
[2]*Laboratory of Dielectric Materials, Department of Materials Science and Engineering, Zhejiang University, Hangzhou 310027, China*
[3]*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, 08193 Bellaterra, Spain*
[4]*Department of Physics, Shanghai University, 99 Shangda Road, Shanghai 200444, China*

Discovering multiferroic materials having an electrical polarization and being strongly ferromagnetic near room-temperature is a long-time-sought quest, due to the promise of novel spintronic devices. Strikingly, it appears that these two desired features are rather difficult to simultaneously co-exist. Using first-principle and Monte Carlo calculations, we demonstrate that two kinds of multiferroic superlattices do possess these two desired features [1,2].

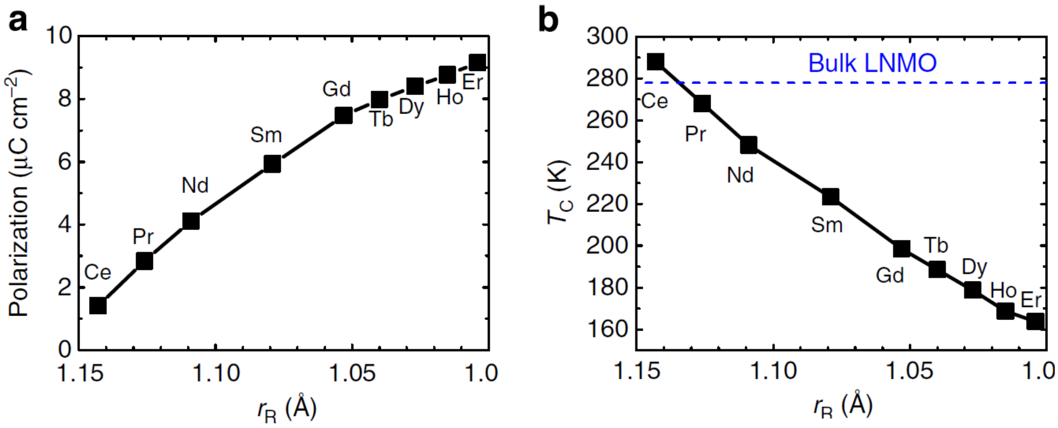

Figure 1. (a) Predicted electrical polarization and (b) magnetic Curie temperature of La_2NiMnO_6/ R_2NiMnO_6 superlattice as a function of the rare-earth ionic radius.

The first kind of superlattice is made by La_2NiMnO_6/R_2NiMnO_6, where R is a rare-earth ion. As shown in Fig. 1a, its electrical polarization increases when decreasing the rare-earth ionic radius. For instance, this polarization is about 9.2 $\mu C/cm^2$ at small temperature when R=Er (which is the rare-earth ion having the largest ionic radius). The origin of this polarization is the novel energy term $-K\omega_R\omega_M u$, proposed in Ref. [3], where K is a constant, ω_R and ω_M characterize the in-plane antiphase and out-of-plane in-phase tiltings, respectively, and u is the displacement of the La or R ions with respect to their ideal positions. In both La_2NiMnO_6 and R_2NiMnO_6 bulks, the u of any two neighboring LaO or RO layers cancel each other, therefore making the whole polarization vanishing. On the other hand, in the La_2NiMnO_6/R_2NiMnO_6 superlattices, the u of neighboring LaO and RO layers, while still opposite in direction, are different in magnitude (because their K parameters are different). As a result, the polarization is finite in these superlattices, which naturally explains the so-called hybrid improper ferroelectricity [4-6]. Regarding the magnetic order of these superlattices, the ground state is ferromagnetic with a magnetization being around 2.4 μ_B per 5 atoms (since both La_2NiMnO_6 and R_2NiMnO_6 bulks are ferromagnetic). As shown in Fig. 1b, the La_2NiMnO_6/Ce_2NiMnO_6 superlattice exhibits a magnetic Curie temperature of 290K, that is very close to room temperature [1], and this magnetic Curie temperature decreases when increasing the rare-earth ionic radius.

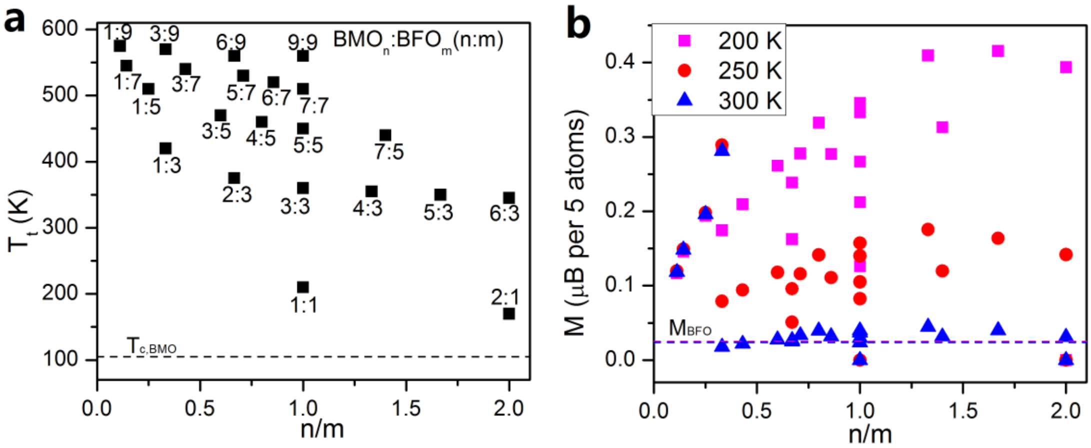

Figure 2. (a): Computed magnetic ordering temperatures for different superlattices studied by Monte Carlo simulations. The results are ordered according to the ratio of *n/m* for each $(BMO)_n/(BFO)_m$ superlattice. (b) Computed magnetization at three different temperatures for these superlattices. The horizontal dashed lines in Panels (a) and (b) show the Curie temperature of pure BMO and the bulk Magnetization of BFO thin film, respectively, for comparison.

The second discovered type of multiferroic materials possessing a large polarization and a strong ferromagnetic order at room-temperature is made of superlattices combining a high Neel temperature anti-ferromagnet and a lower Curie temperature ferromagnet insulator, e.g., $BiMnO_3/BiFeO_3$ (BMO/BFO) superlattices. Indeed, our first-principle calculations show that BMO_n/BFO_m can have an electrical polarization as large as 90 $\mu C/cm^2$ (similar to $BiFeO_3$ bulk) and are also ferromagnetic. The magnetic properties of these superlattices are strongly dependent on the thickness of BMO and BFO (*n* and *m*), as shown in Fig. 2. In particular, the magnetization can reach large values of 0.3 μ_B per five atoms at room temperature in some cases (e.g., BMO_1/BFO_3), which is larger than that of BFO films by at least one order of magnitude. Thus, our works strongly suggest that the most promising commercial application of multiferroics (magneto-electric RAM) is indeed possible [2].

These works are supported by the ONR Grants N00014-11-1-0384 and N00014-08-1-0915, Department of Energy, Office of Basic Energy Sciences, under contract ER-46612, and ARO grant W911NF-12-1-0085. Some computations were also made possible thanks to the ONR grant N00014-07-1-0825 (DURIP), a Challenge grant from the Department of Defense, and the MRI grant 0722625 from NSF.

References:

[1] Hong Jian Zhao, Wei Ren, Yurong Yang, Jorge Iniguez, Xiang Ming Chen and L. Bellaiche, Nature Communications 5, 4021 (2014).
[2] Yurong Yang, L. Bellaiche and Jorge Iniguez, submitted.
[3] L. Bellaiche and Jorge Iniguez, Phys. Rev. B 88, 014104 (2013).
[4] N.A. Benedek and C.J. Fennie, Phys. Rev. Lett. 106, 107204 (2011).
[5] J.M. Rondinelli and C.J. Fennie, Adv. Mater 24, 1961 (2012).
[6] A. Mulder, N A. Benedek, J. M. Rondinelli, and C. J. Fennie, Adv. Funct. Mater. 23, 4810 (2013).