Evolution of half-metallic ferromagnetism in (111)-oriented manganite superlattices

F. Cossu,[1] H.-S. Kim,[1] I. di Marco,[2,3] J. do Nascimento,[4] and V. Lazarov[4]

[1] *Department of Physics and Institute of Quantum Convergence and Technology, Kangwon National University – Chuncheon, 24341, Korea*

[2] *Asia Pacific Center for Theoretical Physics – Pohang, 37673, Korea*

[3] *Institute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University, Grudziadzka 5, 87-100, Toruń, Poland*

[4] *Department of Physics, University of York, Heslington, York, YO10 5DD, United Kingdom*

Oxide heterostructures can host exotic phenomena – such as flat bands [1], magnetic anisotropy [2], exchange bias [3] and spin-glass [4,5], only latent in bulk – and interesting phase competition in thin films and superlattices, because of symmetry breaking and quantum confinement. Research in these topics can reveal the potential of oxides for future applications. The (001)-oriented superlattice of two anti-ferromagnetic insulators $LaMnO_3$ and $SrMnO_3$ is found to be a half-metallic ferromagnet with short periodicity and an antiferromagnetic insulator with 2 or more unit cells [6,7,8]. Our *ab-initio* work predicts a (111)-oriented $LaMnO_3|SrMnO_3$ superlattice is a half-metallic ferromagnet in spite of its large thickness due to strain and charge transfer across the interface [9]. Magnetism and half-metallicity are bulk-like phenomena rather than due to local interfacial effects [9] – unlike in (001)-oriented superlattices [6,7,8]. We compare the two ground state space groups of bulk $LaMnO_3$ ($R\bar{3}c$) and bulk $La_{2/3}Sr_{1/3}MnO_3$ ($Pnma$), finding that their competition is tuned by in-plane strain and superlattice thickness. The $R\bar{3}c$ supports breathing distortions coupled to charge/spin oscillations, similarly to Hund's metals and high-T_C superconductors. The space group competition plays also a role in the Mn magnetic coupling, as $Pnma$ promotes A-type antiferromagnetism. Computed within the Heisenberg formalism via the magnetic force theorem, it stays strongly ferromagnetic in the La region but progressively shifts towards antiferromagnetic in the Sr region as thickness grows, suggesting mixed magnetic orders. Finally, we estimate the Curie temperature via atomistic spin dynamics.

References:

[1] Rüegg et al., Phys. Rev. B **85**, 245131 (2012).
[2] Asaba et al., Phys. Rev. B **98**, 121105 (2018).
[3] Gibert et al., Nat. Mater. **11**, 195 (2012).
[4] Huangfu et al., Phys. Rev. B **102**, 054423 (2020).
[5] Ding et al., Adv. Mater. Interfaces **3**, 1500676 (2016).
[6] Bhattacharya et al., Phys. Rev. Lett. **100**, 257203 (2008).
[7] Pardo et al., Appl. Phys. Lett. **104**, 081602 (2014).
[8] Nanda and Satpathy, Phys. Rev. B **79**, 054428 (2009).
[9] Cossu et al. npj Comput. Mater. **8**, 77 (2022).

This research is funded by the National Research Foundation (NRF) of Korea (Grants No. 2022R1I1A1A01071974)