Charge-Transfer Engineering at Polar Double-Perovskite/Perovskite Interfaces

De Luca G[1.7*], Spring J[1], Kaviani M[2], Jöhr S[1], Campanini M[3], Zakharova A[4], Guillemard C[5], Herrero-Martin J[5], Erni R[3], Piamonteze C[4], Rossell MD[3], Aschauer U[2], Gibert M[1,6].
*gdeluca@icmab.es
1 University of Zurich, Switzerland.
2 University of Bern, Switzerland.
3 Swiss Federal Laboratories for Materials Science and Technology, Switzerland.
4 Swiss Light Source, Switzerland.
5 ALBA Synchrotron Light Source, Spain.
6 Technical University of Wien, Austria.
7 Instituto de Ciencia de Materiales de Barcelona (ICMAB-CSIC), Spain.

In the epitaxy of complex oxides, interfacial charge-transfer phenomena are a promising pathway to engineer functionalities that are absent in the bulk, the discovery of a two-dimensional electron liquid at the interface between $LaAlO_3$ and $SrTiO_3$ (STO) being a prominent example [1]. In this context, advanced approaches to manipulate the electronic state of a buried interface are thus very intriguing.

Here, first, we demonstrate that at the interface between a polar double perovskite (La_2NiMnO_6, LNMO) and a non-polar single perovskite (STO), a charge transfer triggered by a polarity mismatch arises. LNMO is a ferromagnetic insulator with a bulk Curie temperature (T_C) around 280 K, resulting from positive superexchange between the long-range-ordered Ni^{2+} and Mn^{4+} cations. Bulk-like behavior is well reproduced in high-quality epitaxial films [2]. Nevertheless, the electron doping induced by the electronic reconstruction perturbs the magnetism of the ultrathin LNMO films via formation of Mn^{3+}, resulting in a strong reduction of T_C.

To recover the optimal electronic configuration of the buried LNMO//STO interface, we design a top-interface engineering approach through a $LaNiO_3$ (LNO) overlayer. The presence of LNO is fundamental in displacing the interfacial charge and allows to re-establish the ferromagnetism in LNMO. The LNO-induced electron migration is also obtained on polar LNMO//LSAT interfaces, suggesting that a similar method can be adapted to other polar oxide interfaces [3].

References

[1] Ohtomo A and Hwang HY. A high-mobility electron gas at the LaAlO3/SrTiO3 heterointerface, Nature 2004:427, 423
[2] De Luca G, Spring J, Bashir U, Campanini M, Totani R, Dominguez C, Zakharova A, Döbeli M, Greber T, Rossell MD, Piamonteze C and Gibert M, Ferromagnetic insulating epitaxially strained La2NiMnO6 thin films grown by sputter deposition, APL Mater. 2021:9, 081111
[3] De Luca G, Spring J, Kaviani M, Jöhr S, Campanini M, Zakharova A, Guillemard C, Herrero-Martin J, Erni R, Piamonteze C, Rossell MD, Aschauer U, Gibert M, Top-Layer Engineering Reshapes Charge Transfer at Polar Oxide Interfaces, Adv. Mater 2022:34, 2203071