Supporting Information

Strain control of giant magnetic anisotropy in metallic perovskite $SrCoO_{3-\delta}$ thin films

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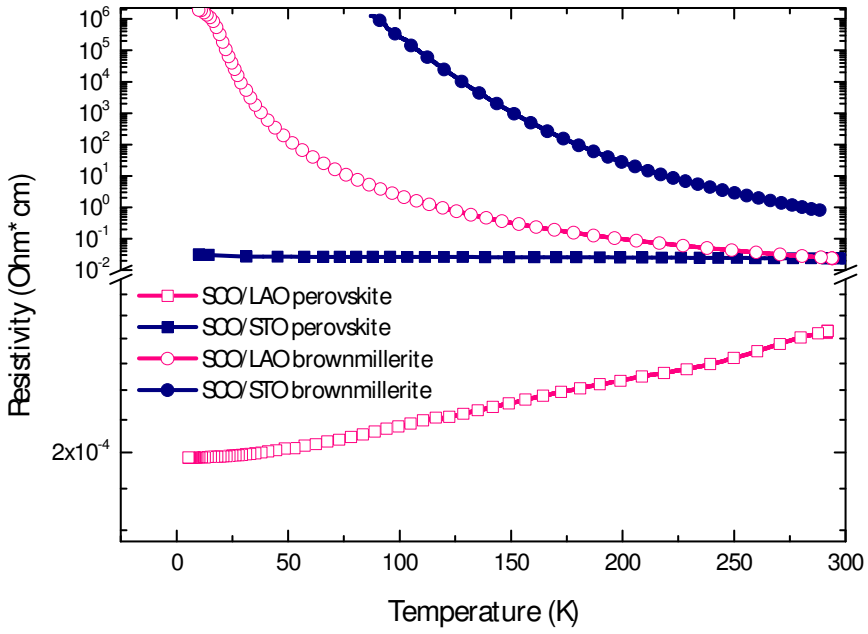

Figure S1. Temperature dependent resistivity of perovskite and brownmillerite SCO thin films on LAO and STO substrates. The resistance of the sample was measured by a Keysight 2587A high resistance meter. The perovskite structure SCO thin films are metallic below RT.

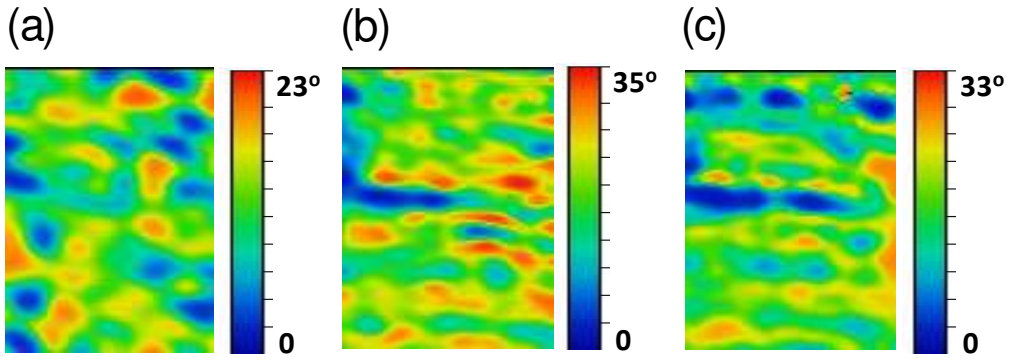

Figure S2. Evolution of magnetic domains in SCO/LAO after being exposed to high field. (a) measured at 0.1 T without exposure, (b) at -0.1 T after 9 T exposure and (c) at 0.1 T after -9 T exposure.

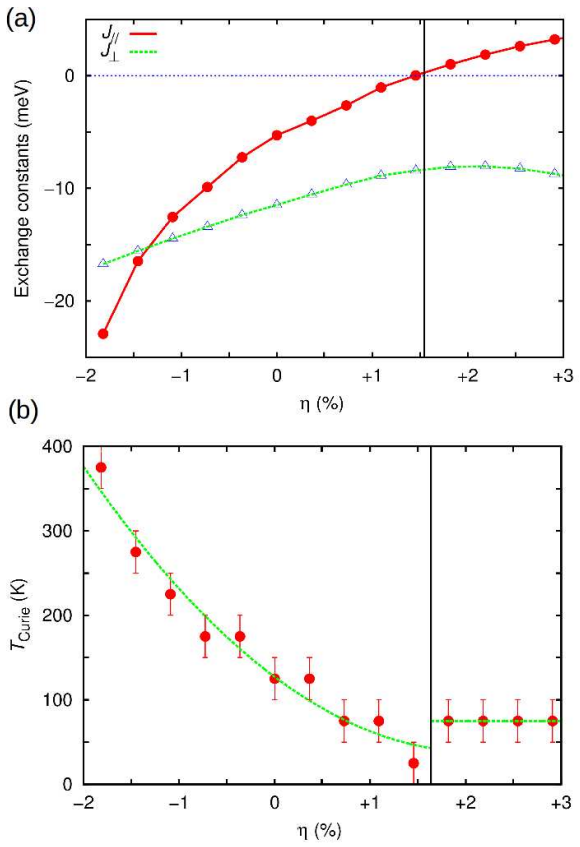

Figure S3. DFT-PBE and MC Heisenberg spin model results obtained in SCO thin films. (a) Value of the out-of-plane and in-plane exchange constants obtained from DFT-PBE calculations and expressed as a function of epitaxial strain. (b) Critical Curie temperature obtained from MC Heisenberg spin model simulations and expressed as a function of epitaxial strain. The vertical solid lines in (a) and (b) indicate the occurrence of a ferromagnetic→antiferromagnetic phase transition at zero temperature.

We calculated the zero-temperature energy of stoichiometric SCO thin films over a wide epitaxial strain interval by also using the PBE variant of the generalized gradient approximation to density functional theory (DFT). As noted in the main text, we found that the magnetic interactions estimated in SCO thin films depend significantly on the employed exchange-correlation energy functional. Those differences are easily appreciated by comparing the results shown in the Fig. S3 (obtained with the PBE functional) to the

analogous results reported in Fig. 3 of the main text (obtained with the PBEsol functional). In particular, the critical Curie temperatures, T_{Curie}, obtained with the PBE functional are systematically lower than those obtained with the PBEsol functional. In both PBE and PBEsol cases, however, the crucial strain-induced effect on the magnetic interactions is to reduce the value of the corresponding out-of-plane and in-plane exchange constants, and the magnetic interactions out-of-plane are more intense than in-plane. At null and non-zero epitaxial strain conditions the PBEsol functional provides a better agreement with respect to the experimentally measured T_{Curie}'s, hence in this context the PBEsol functional appears to be more reliable than the PBE functional. We note that a similarly noticeable dependence of the computed magnetic interactions on the selected exchange-correlation energy functional has been reported recently for the oxide perovskite $BiCoO_3$[1]; such a drawback, therefore, appears to be a common feature in the description of cobalt-based oxide pervoskites with standard DFT methods.

References

(1) Cazorla, C.; Dieguez, O.; Iniguez, J. Multiple Structural Transitions Driven by Spin-phonon Couplings in a Perovskite Oxide. *Sci. Adv.* **2017,** *3*, e1700288