Supplementary Information for

Oxygen Driven Anisotropic Transport in Ultra-thin Manganite Films

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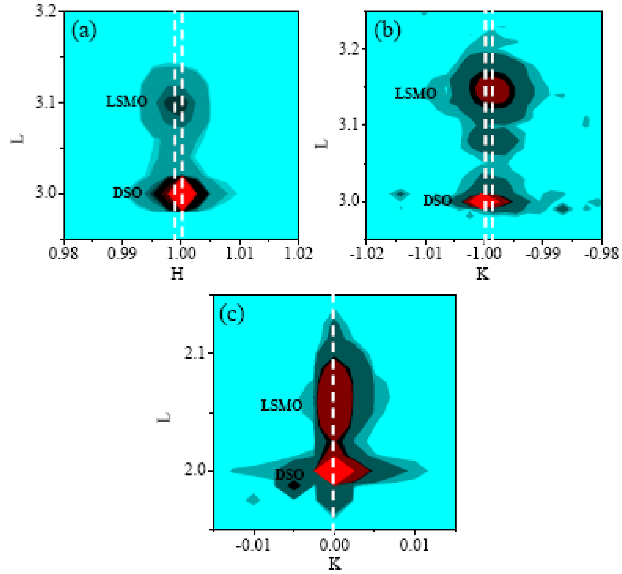
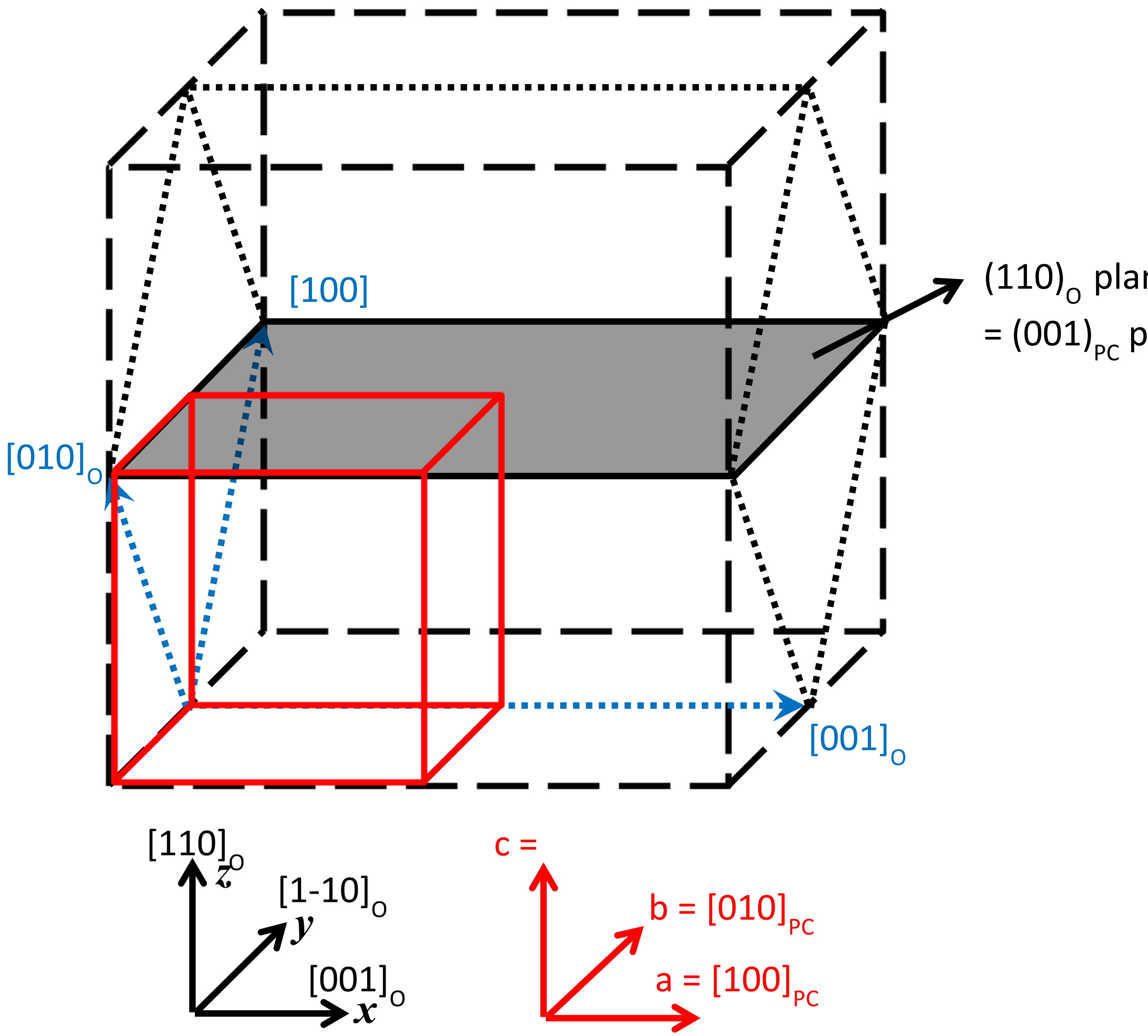
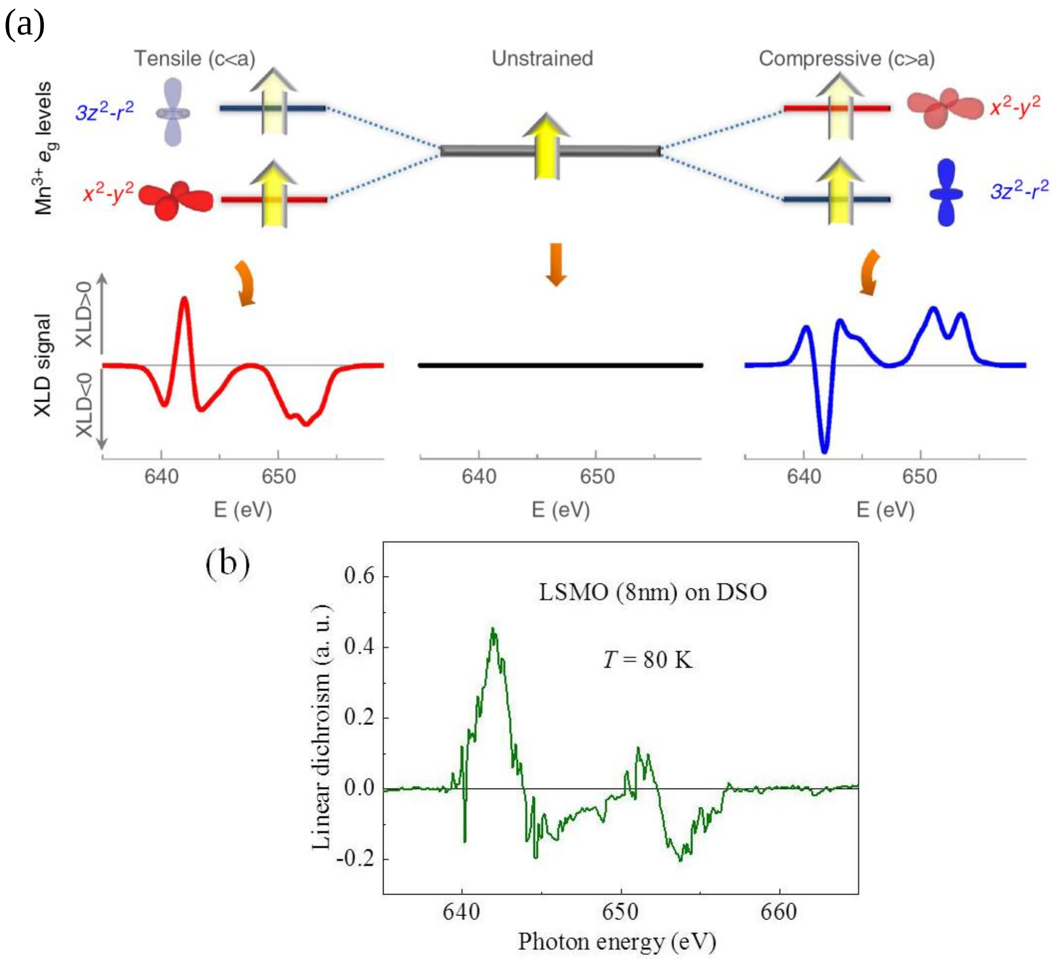


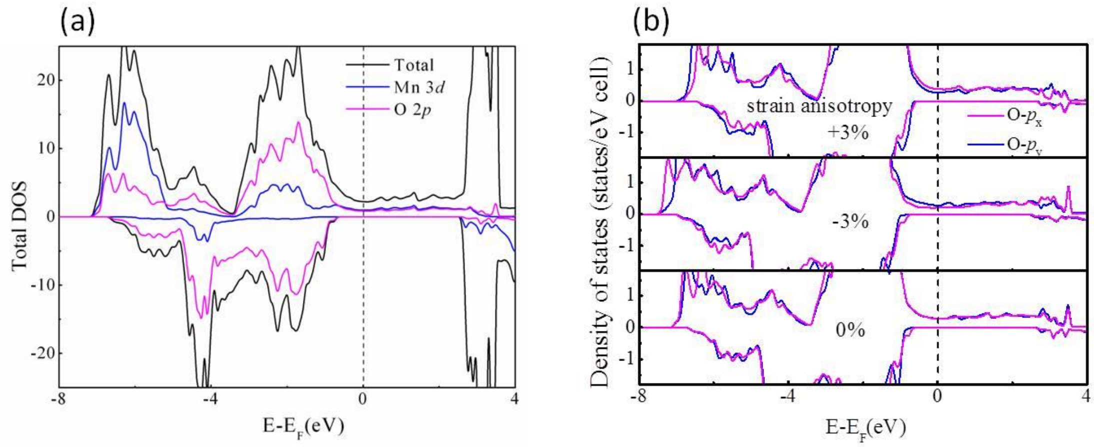
Supplementary Figure S1, Reciprocal space mapping (RSM) for the 8 nm LSMO film on DSO. (a), (b) and (c) are around the pseudocubic (103), $(0\bar{1}3)$, and (002) peaks, respectively. The RSMs are plotted in reciprocal lattice units (r.l.u.) of the DSO substrate.



Supplementary Figure S2, Definition of the *x*, *y*, *z* coordination. The black dotted lines indicate the orthorhombic unit cell of the DSO substrate. The red solid lines represent the pseudocubic unit cell of (110)-oriented DSO, which have the same *x*, *y*, *z* coordination as the pseudocubic LSMO thin films grown on top.



Supplementary Figure S3, Linear dichroism of LSMO and orbital occupancy of the LSMO on DSO. By comparing the simulated linear dichroism of LSMO with different orbital occupancy with that obtained in our ultra-thin film on DSO, it is clear that the Mn $3d_{x^2-y^2}$ orbital is preferably occupied for LSMO on DSO with large tensile strain (~1.9 %). (a) Simulated results [adapted from *Nat Commun* **3**, 1189 (2012)] and (b) our experimental result for LSMO on DSO (b).



Supplementary Figure S4, First-principles calculations for unstrained LSMO with a_0=3.876 Å. The same supercell as in the paper with FM ground state is used. (a), Density of stated (DOS) of LSMO obtained using GGA+U=6 eV. (b), DOS from O p_x (pink lines) and O p_y (blue lines) orbitals under various strain anisotropies with fixed *y* direction lattice constant (*b*=3.876 Å).

Supplementary Note 1

Structural analysis of the LSMO on DSO

Supplementary Figure S1 shows the reciprocal space mappings (RSMs) around the pseudocubic (103), (0$\bar{1}$3), and (002) peaks for the 8 nm LSMO film on DSO. For DSO, the pseudocubic lattice constants are a = 3.952 Å along $[001]_o$ and b = 3.947 Å along $[1\bar{1}0]_o$. In Supplementary Fig. S1a, H = 0.9992 and 1.0002 for the centers of LSMO and DSO peaks (0.1% difference), respectively. So we can obtain that a = 3.948 Å for the LSMO film. In Supplementary Fig. S1b, K = -1.0003 and -0.9986 for the centers of film and substrate peaks (0.17% difference), respectively. So we can obtain that b = 3.940 Å for LSMO. Thus the in-plane strain anisotropy in LSMO film is (3.948-3.940)/3.948 = 0.2 % [Note the asymmetric shape of the peaks and the accuracy of the equipment (~0.1 %)]. The results suggest that the 8 nm LSMO film is fully-strained on DSO. In Supplementary Fig. S1c, L = 2.0633 and 2.0001 for the peaks of film and substrate, respectively, which leads to c = 3.826 Å for LSMO film and the tetragonal distortion c/a = 0.968.

Supplementary Note 2

First-principles calculations of the density of states of unstrained LSMO

We have conducted first-principles calculations on unstrained LSMO using bulk lattice constant of a_0=3.876 Å. The supercell used is the same at that described in the manuscript and FM ground state is assumed. The results show that LSMO is a half metal as expected. When a tensile strain is applied along the x direction, the contribution to density of states (DOS) at Fermi level from O $2p_x$ increases, similar to that obtained in the strained LSMO films. When a compressive strain is applied, the opposite is observed as shown in Supplementary Fig. S4b.