# **Supporting Information**
for
# Coupled Lattice Polarization and Ferromagnetism in Multiferroic $NiTiO_3$ Thin Films

Tamas Varga,[1*] Timothy C. Droubay,[2] Libor Kovarik,[1] Manjula I. Nandasiri,[3] Vaithiyalingam Shutthanandan,[1] Dehong Hu,[1] Bumsoo Kim,[4,5] Seokwoo Jeon,[5] Seungbum Hong,[4,5] Yulan Li,[2] and Scott A. Chambers[2]

[1]*Environmental Molecular Sciences Laboratory, Richland, WA*
[2]*Physical and Computational Sciences Directorate, Pacific Northwest National Laboratory, Richland, WA*
[3]*Imaging and Chemical Analysis Laboratory, Montana State University, Bozeman, MT*
[4]*Materials Science Division, Argonne National Laboratory, Argonne, IL*
[5]*Department of Materials Science and Engineering, KAIST, Daejeon 34141, Republic of Korea*

**Corresponding author: tamas.varga@pnnl.gov*

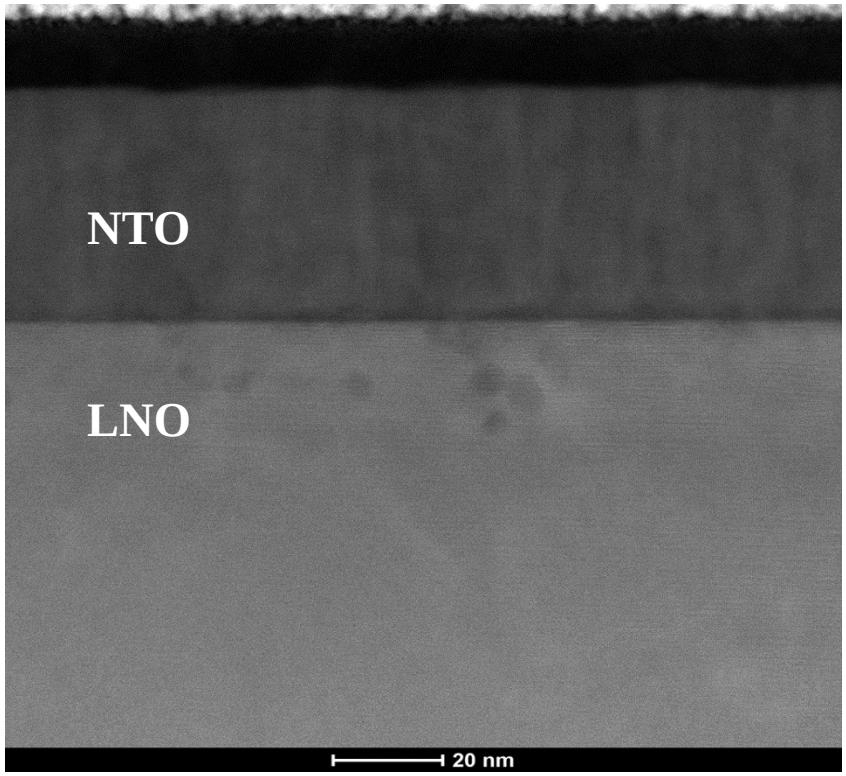



**Figure S1:** TEM image of an $NiTiO_3$/LNO film showing columnar grain growth

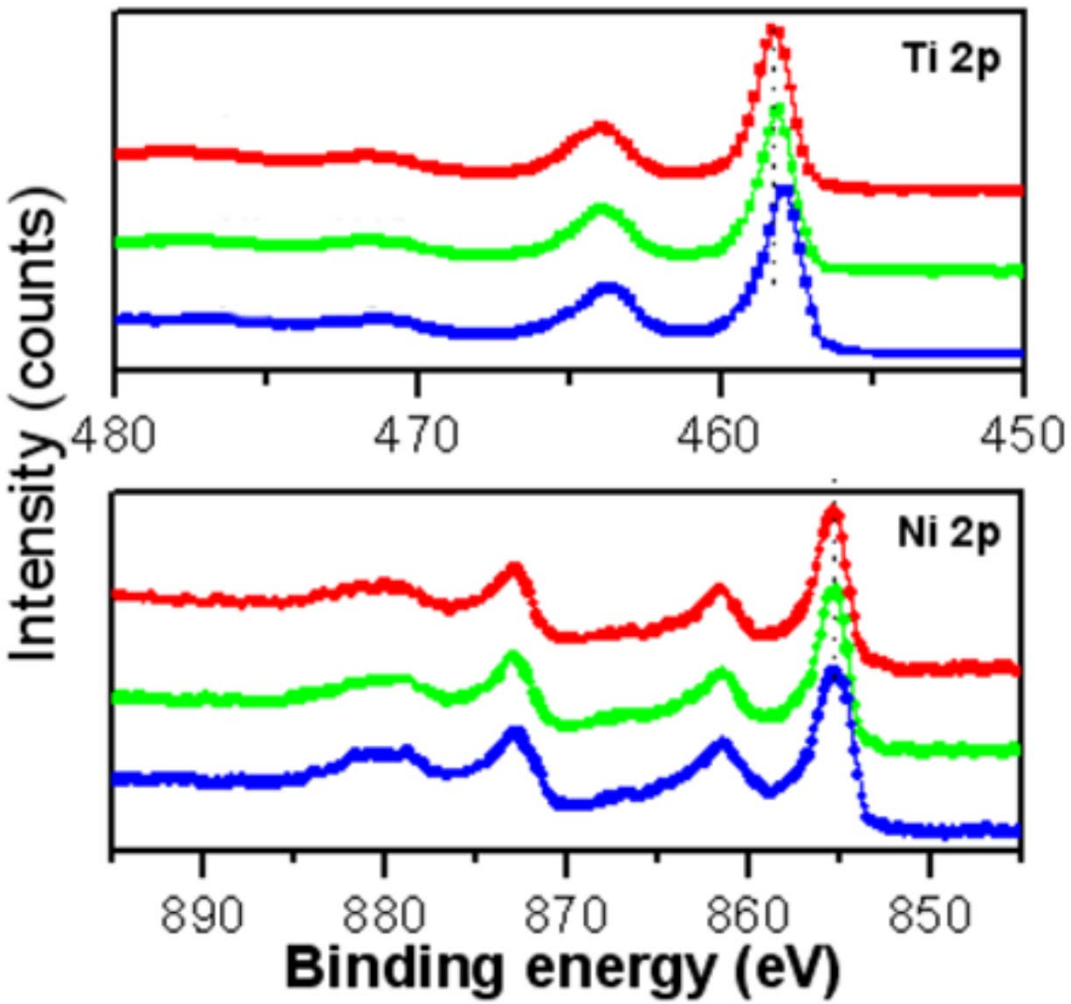
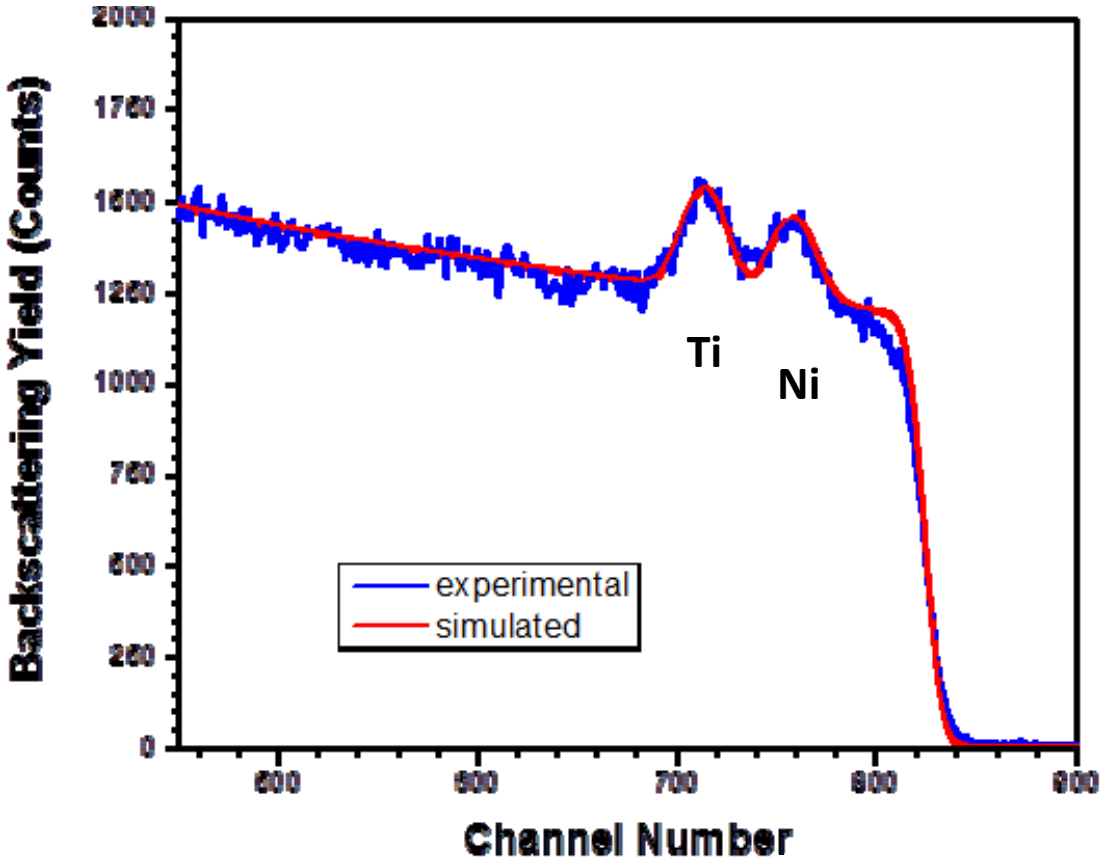



**Figure S2:** XPS for Ti and Ni in a representative $NiTiO_3$/LNO film




| Sample ID | Layer # | | Thickness | | Composition | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | (1 x $10^{15}$ at/$cm^2$) | (nm) | Ni | Ti | O | Al | O | Li | Nb |
| | | | | | (atomic %) | | | | | | |
| NTO-LNO-29 | | | | | | | | | | | |
| | 1 | | 510 | | 25 | 30 | 55 | | | | |
| | 2 | | | | | | | | 48.5 | 16.5 | 35 |

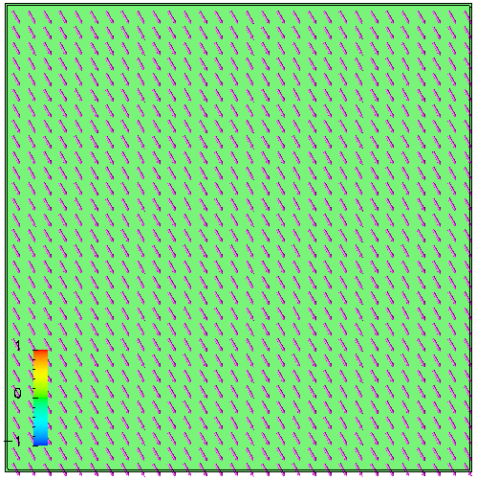
**Figure S3:** RBS analysis of two $NiTiO_3$/LNO film samples

The considered film has a rhombohedral crystal structure. One of the rhombohedral diagonal directions is parallel to the film normal and one of them is in the film plane (see Fig.3c in paper). By setting an orthogonal coordinate **x**={$x_1$,$x_2$,$x_3$} with axis $x_3$ being normal to the film plane, the magnetic anisotropic energy for a rhombohedral crystal is generally described by

$$f_{anis} = K_1\left(1-m_3^2\right)+K_2\left(1-m_3^2\right)^2$$

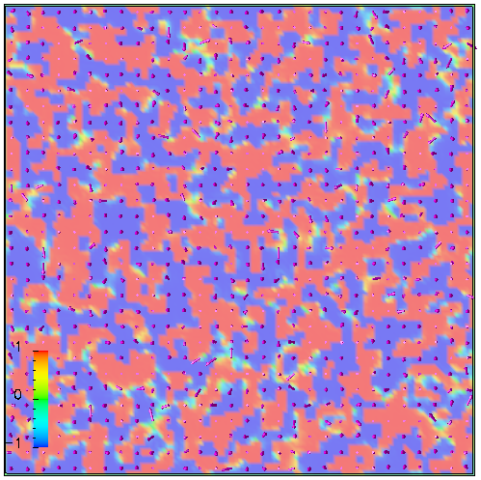
where $m_3$ is the component of the magnetization vector **m**={$m_1$,$m_2$,$m_3$} along the $x_3$-axis. When $K_1$>0 and $K_2$>-$K_1$, the magnetic easy axis is along the direction of [0,0,1]; When $K_1$<0 and $K_2$<-$K_1$/2, the magnetic easy axis is in the plane of (0,0,1), along the $[1/2,\sqrt{3}/2,0]$ direction as shown in Fig. 3c.

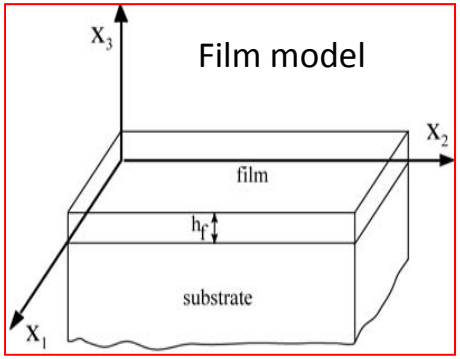
The simulation considered periodic boundary conditions along the $x_1x_2$ directions in the film plane. The obtained ferromagnetic domain structures are shown below and viewed from the top of the thin film. The arrows are the magnetization vectors. The color refers to the magnitude of the out-of-plane magnetization component ($m_3$) as indicated by the color bar.



$x_2$=[010]

$x_1$=[100]






(a) Ferromagnetic domain morphology by taking $K_1$<0 and $K_2$=0. [1/2,-√3/2, 0] is a magnetic easy axis. The obtained magnetic domain structure is a single domain. All the magnetization in the film are all along the [1/2,-√3/2, 0] direction.

(b) Ferromagnetic domain morphology by taking $K_1$>0 and $K_2$=0. [0,0,1] is a magnetic easy axis. The obtained magnetic domain structure is a multi-domain structure. The magnetization in the film is along either the direction of [0,0,1] (red) or [0,0,-1] (blue).

**Figure S4: Film orientation, magnetic easy axes and domain morphologies**

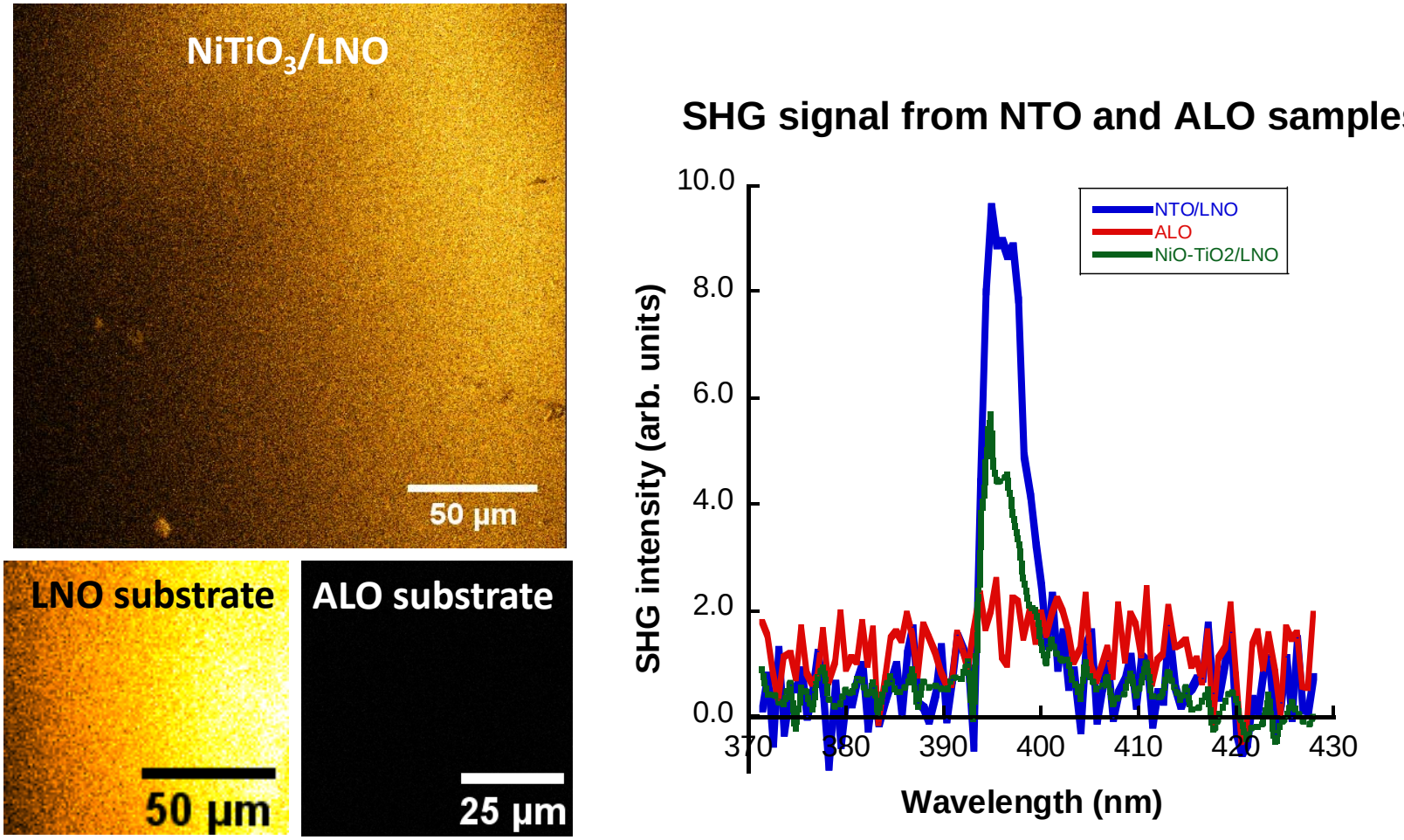



**Figure S5: Optical SHG in transition mode (confocal imaging ) and reflection mode**

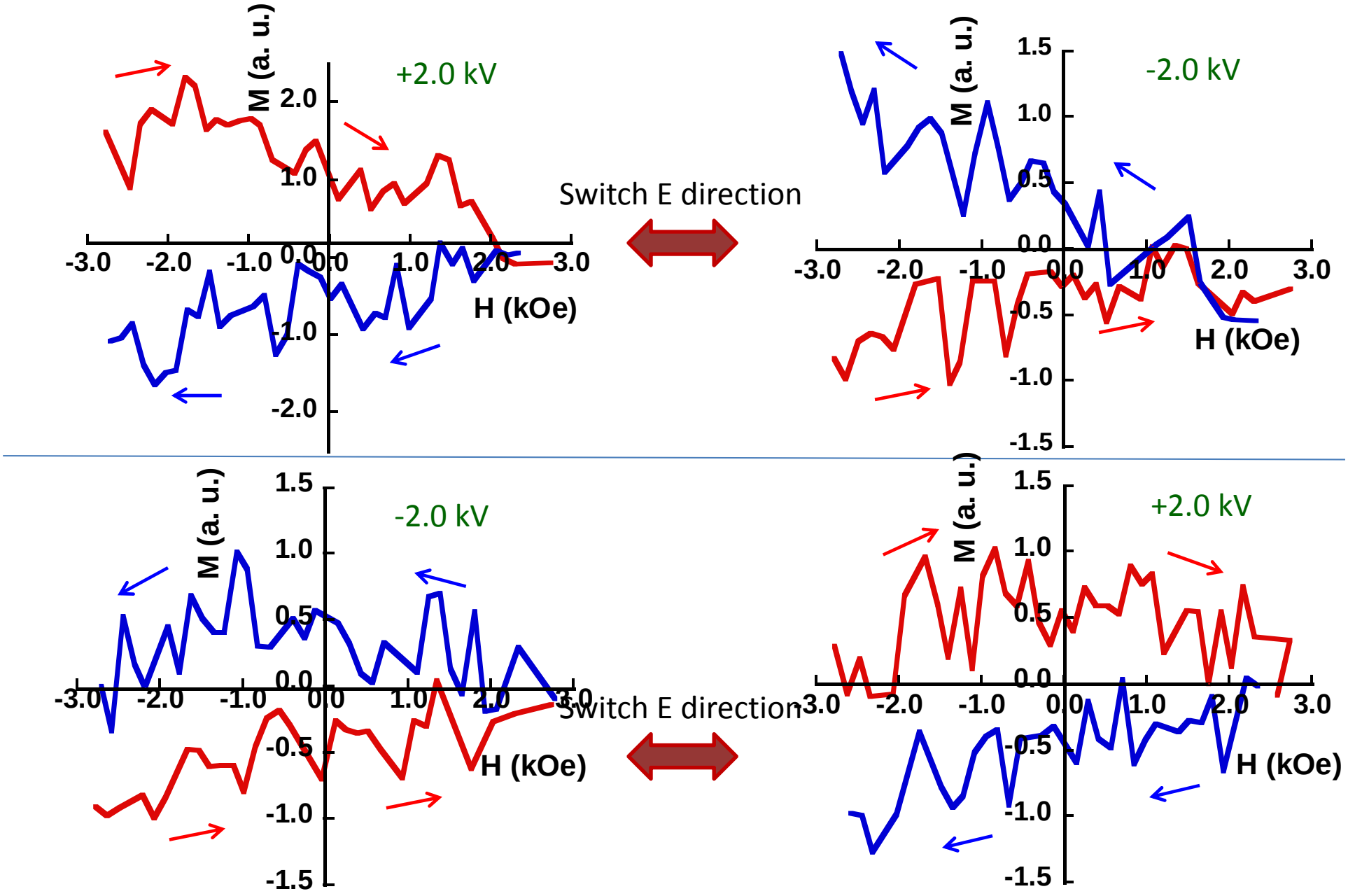



**Figure S6: Reversible coupling from magnetic hysteresis curves for sample NTO/LNO-30 at 2 kV**