Large coupled magnetoresponses in EuNbO2N

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Supporting Information for:

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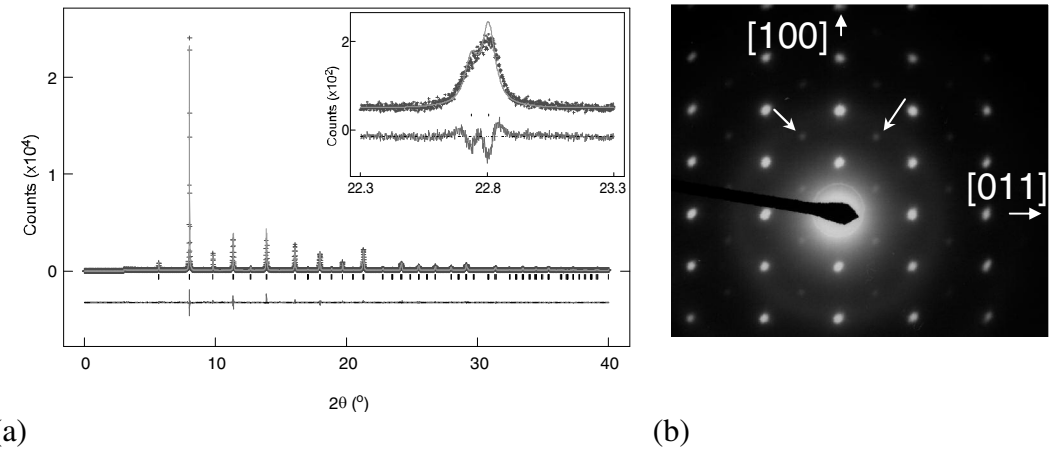
Experimental Details

Samples of $EuTaO_2N$ and $EuNbO_2N$ were prepared by ammonolysis at 950 ^oC of $EuTaO_4$ and $EuNbO_4$ respectively. The oxide precursors were prepared by solid state reaction in air between Eu_2O_3 (Aldrich, 99.99 %) and Ta_2O_5 or Nb_2O_5 (Aldrich, 99.99 %). Eu_2O_3 was previously treated at 900 ^oC for 12 hours. The binary oxides were mixed in the stoichiometric proportions, pelletized and treated at 1000 ^oC for 12 hours, then retreated at 1400 ^oC for 12 hours with intermediate regrinding. These two cycles were performed three times, until no impurities were detected in the X-ray diffraction patterns. $EuTaO_4$ and $EuNbO_4$ were nitrided by two15 hours treatments in flowing NH_3 (Carburos Metálicos, 99.9%) at a rate of 200 cm^3/min with subsequent quenching to room temperature in the ammonia atmosphere. The second treatment was done on a pellet of 7 mm diameter pressed at 3 tons for performing the electrical measurements.

N contents were determined by combustion analysis using a Carlo Erba instrument. Synchrotron powder X-ray diffraction data were collected at 290 K on instrument ID31 at the ESRF, Grenoble from capillary samples in the angular range $3 \leq 2\theta \leq 40^o$ with a wavelength of 0.3983 Å and electron diffraction patterns were obtained in a JEOL 1210 electron microscope operating at 120 kV (see Figure).

Magnetic measurements were performed between 2 and 300 K in fields of 0.0025 and 1 T using a Quantum Design SQUID magnetometer. M(H) curves were measured up to 5 T for temperatures between 2 K and 20 K. The dielectric constant and resistivity were measured using an Agilent 4284A LCR meter and an Agilent 3458A multimeter, respectively. The magnetocapacitance was measured at several frequencies and we show the values measured at 100 kHz. The contacts were made using silver epoxy. The temperature and magnetic field were controlled through a Physical Properties Measurement System PPMS (Quantum Design), using a home-built insert and wiring.



(a) (b)

Supporting Figure. Diffraction data for $EuNbO_2N$. (a) The fitted synchrotron X-ray diffraction profile. The expanded peak in the inset is split by the tetragonal lattice distortion. The data are fitted using a centrosymmetric P4/mmm model. An acentric P4mm refinement model that would be consistent with a ferroelectric distortion gave no significant improvement and was unstable. (b) Electron diffraction pattern along the [0-11] zone axis with additional spots (arrowed) that reveal a further $\sqrt{2}$ x $\sqrt{2}$ x 2 body-centred superstructure.