Additional information concerning experimental details:

Fine tuning of ferromagnet/antiferromagnet interface magnetic anisotropy for field-free switching of antiferromagnetic spins

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Preparation and structural characterization of the samples

The Fe(110) films were grown by Molecular Beam Epitaxy (MBE) on atomically clean W(110) single crystal at room temperature and then annealed at 675 K. We made use of shutter shadowing the sample during the growth of Fe in order to prepare macroscopic Fe steps with the thickness $d_{Fe} \in \{50, 85, 90, 95, 100, 105, 150, 200\}$ Å. Next, on such prepared Fe(110) samples, NiO overlayers with the thickness d_{NiO} = 40 Å were grown at room temperature by reactive deposition of Ni in partial oxygen pressure 1×10^{-6} Torr. NiO overlayer copies the macroscopic structure of underlying Fe steps. Additionally series of samples was prepared with the width of the macroscopic steps ranging from 0.2 mm up to 4 mm. XMLD studies show that magnetic properties of NiO/Fe bilayers are totally independent on the width of these macroscopic terraces prepared by MBE shutter. After each processing step, the structure of the surface was in-situ monitored using low-energy electron diffraction (LEED).

Crystallographic orientation of NiO(111) in respect to Fe(110)

The exemplary Low Energy Electron Diffraction (LEED) pattern of the uncovered Fe(110) surface is shown in Fig. 1a of this supplementary material. Sharp diffraction spots indicate a smooth unreconstructed (110) surface. Within the studied 50 – 200 Å thickness range of Fe we find the in-plane lattice spacing a_{001} along the Fe[001] direction a_{001}=2.88±0.02 Å, which corresponds to an almost fully relaxed Fe(110)/W film when compared to 2.86 Å value for bulk iron. The LEED pattern from the surface of the NiO-covered Fe(110)/W(110) sample shown in Fig. 1b indicates a hexagonal NiO(111) surface structure independently on the thickness of the underlying Fe film. Corresponding ball models of (c) Fe(110) and (d) NiO(111)/ Fe(110) surfaces are also shown in Fig.1. From the LEED patterns we conclude that the Fe[1-10] and Fe[001] in-plane directions are parallel to NiO[-211] and NiO[01-1] directions within NiO(111) plane, respectively.

Magnetic characterization of the samples

We followed the magnetic properties of the NiO/Fe(110) bilayers by means of X-ray Magnetic Linear and Circular Dichroism (XMLD and XMCD) techniques complemented by both *in situ* and *ex situ* longitudinal magneto-optic Kerr effect (MOKE).

Most of the spectroscopic data presented in the body of the article was obtained in a total electron yield (TEY) mode in the XAS end-station of the Polish synchrotron SOLARIS [1]. Both XMLD and XMCD measurements were performed in two geometries, namely with circular and linear polarization of incoming X-rays parallel either to Fe[1-10] or Fe[001] in-plane direction. In order to ensure the required in-plane sensitivity to orientation of AFM magnetic moments, temperature dependent XMLD measurements were

performed at normal incidence geometry. Since data sets corresponding to both measurements geometries are fully consistent with each other, we restrict ourselves to present only the results of the first mentioned geometry, in which large XMCD value is expected when $\mathbf{M}_{Fe}$ is parallel to Fe[1-10] direction and in case of XMLD studies electric field is oriented along NiO[-211] || Fe[1-10] direction, as shown in the inset of the Fig. 2d, in the body of the article.

Micro-spectroscopic and spectro-microscopic studies presented in Fig.1 of the article were performed in the spectroscopic photoemission and low energy electron microscope (SPELEEM) which is the end-station of the Nanospectroscopy beamline in Elettra synchrotron (Trieste, Italy) [13]. In the SPELEEM setup the X-rays were incident on the sample at 16° grazing angle from the surface. Therefore, only one of the two linear polarization states was within the sample plane giving sensitivity to the change in the in-plane spin orientation of NiO. The XMLD-PEEM images of the boundary area shown in Fig. 1b, f were obtained by subtracting the two PEEM images (and normalizing to their sum) acquired at 868.9 and 870.2 eV photon energy, which correspond to the two absorption peaks in the Ni L_2 edge visible in the XMLD spectra in Fig. 1 d. The XMCD imaging with circular X-ray polarization was mostly sensitive to in-plane magnetization along the beam direction. The XMCD algebra resulted in "dark" contrast for beam propagation parallel to the magnetization direction, and "bright" contrast for the antiparallel configuration.

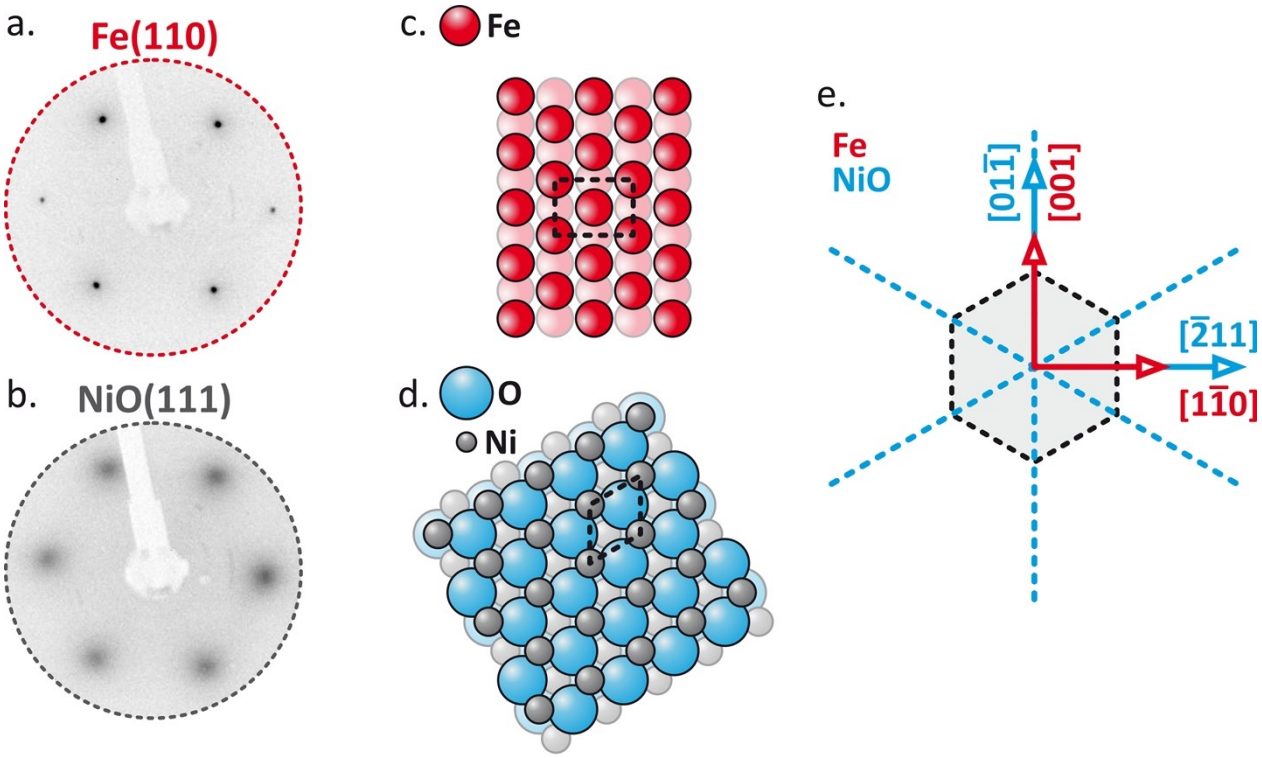

Fig.1 LEED patterns of (a) uncovered Fe(110) and (b) NiO(111)/Fe(110) surfaces. Corresponding ball models of (c) Fe(110) and (d) NiO(111)/Fe(110) surfaces. The relative Fe and NiO in-plane directions as concluded from LEED analysis are sketched in (e).

[1] The PEEM/XAS beamline: https://synchrotron.uj.edu.pl/en_GB/linie-badawcze/peem-xas