

# Heterogeneous Magnetic Order Melting Triggered by Ultrafast Lattice Control at the LaAlO<sub>3</sub>/NdNiO<sub>3</sub> Interface

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**Abstract:** We use femtosecond Resonant Soft X-ray Diffraction to observe heterogeneous melting of antiferromagnetic order in an epitaxially grown NdNiO<sub>3</sub> film. Cross-interface coupling launches a propagating phase front following direct lattice excitation of the LaAlO<sub>3</sub> substrate.

**OCIS codes:** (320.7130) Ultrafast processes in condensed matter, including semiconductors; (160.6990) Transition-metal doped materials; (340.7480) X-rays

## 1. Introduction

Transition metal oxides offer a variety of functional electronic properties likely to have a significant impact in applications. In particular, the exceptional sensitivity of these materials to structural distortions paves new ways for the design and control of their phase state via strain engineering in artificial oxide heterostructures.

Ultrafast control of the lattice as a means to modulate various electronic phases has been demonstrated in complex oxides. Control of transport and magnetic properties in manganites [1,2] and light-induced superconductivity in cuprates [3] were demonstrated. These experiments based on the direct lattice excitation of the bulk, and the photo-excited volume has been driven into the new phase state rather homogeneously.

Recently, we have extended the same approach to complex oxide heterostructures. By triggering a lattice mode in the substrate we steer the electronic phase state in an epitaxial film deposited on its surface. This cross-interface coupling can unleash the full potential of high-speed vibrational control in a broad class of artificial materials.

The understanding of these phase transitions requires knowledge of their dynamics on the nanometer length and the femtosecond time scale. Only recently, these spatiotemporal dynamics have become accessible by monitoring the ultrafast evolution of charge, orbital and magnetic ordering via femtosecond resonant soft X-ray diffraction (RSXD) at free electron lasers [3,4-6].

## 2. Experiment

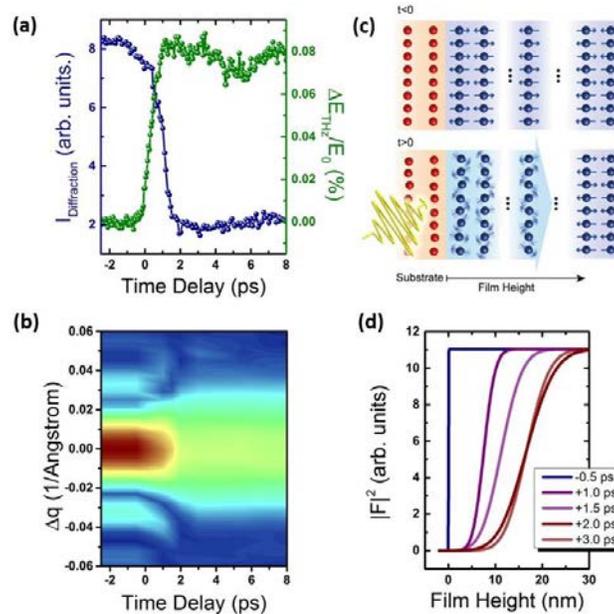
We study a compressively strained NdNiO<sub>3</sub> film, 100 unit cells thick, deposited on a pseudocubic (111) LaAlO<sub>3</sub> substrate by off-axis RF magnetron sputtering. This film transforms at about 130 K from a high-temperature metallic to a low-temperature insulating phase, accompanied by (1/4 1/4 1/4) wave vector antiferromagnetic (AFM) ordering on both, the Ni and Nd sublattices. Ultrafast THz spectroscopy has shown, that resonant excitation of an IR-active phonon mode of the LaAlO<sub>3</sub> substrates induces a long-lived five-order-of-magnitude increase of the electrical conductivity of the NdNiO<sub>3</sub> epitaxial thin film [7].

Here, we monitor the evolution of the AFM ordering on the Ni sites via femtosecond RSXD at the Linac Coherent Light Source. This free electron laser is tuned to the L<sub>3</sub> edge at 852 eV to measure the diffraction at the (1/4 1/4 1/4) wave vector. We use the identical 15- $\mu$ m wavelength 200-fs excitation pulses as in the above mentioned THz-probe experiments, thus resonantly exciting the Al-O stretch phonon mode of the substrate. The pump pulses are focused to a 4 mJ/cm<sup>2</sup> fluence onto the sample, kept at 40 K.

## 3. Results

Figure 1(a) shows the photo-induced change of the height of the AFM order related diffraction peak. The optical excitation reduces the peak intensity by about 80 % and recovers on the time scale of tens of picoseconds. The 1.6 ps response time significantly exceeds the 250 fs time resolution of the experiment, which is determined by the jitter

between the synchronized optical laser and the FEL. The concomitant increase of the THz reflectivity (green data points in Fig. 1(a)), results from the increase of the electrical conductivity in the NdNiO<sub>3</sub> epitaxial film. This insulator-metal transition was proposed to be driven by a light-induced structural distortion at the substrate-film interface [7].



**Fig. 1:** (a) Time-resolved intensity of the AFM order related  $(1/4\ 1/4\ 1/4)$  diffraction peak, measured at the Ni L<sub>3</sub> edge, following direct optical excitation of the LaAlO<sub>3</sub> substrate. The green data points show the transient THz reflectivity of the same sample under identical excitation conditions. (b) Momentum dependence of the same diffraction peak, as function of time delay. (c) Schematic illustration of this AFM order melting process. At negative time delay, the NdNiO<sub>3</sub> film is homogeneously ordered. Direct excitation of a LaAlO<sub>3</sub> substrate phonon triggers a magnetic melt front, propagating from the interface into the film. (d) Quantitative analysis of this phase front propagation in real space, carried out in the frame of kinematical diffraction theory.

Information about the real space dynamics of the magnetic ordering along the (111) sample growth direction can be extracted from Fig. 1(b), where we plot the transient momentum dependence ( $\theta$ - $2\theta$  scans) of the  $(1/4\ 1/4\ 1/4)$  diffraction peak. At negative time delay, strong magnetic correlations and sharp domain wall boundaries, given by the 30 nm film thickness, are manifested through the presence of Laue oscillations. The mid-infrared excitation forces a decrease of the peak intensity, a significant peak broadening and concomitant extinction of the Laue oscillations. In real space, these dynamics translate into a reduction of the magnetic correlation length along the thin film out-of-plane direction, together with a broadening of the domain boundaries.

Taking into account that we resonantly drive the lattice mode of the substrate, we show that the AFM order dynamics in the NdNiO<sub>3</sub> film involve the propagation of a demagnetizing phase front, travelling from the substrate interface into the thin film as sketched in Fig. 1(c). We quantitatively capture these dynamics in the frame of the kinematical diffraction theory. Figure 1(d) shows the early time scale evolution of the space-dependent AFM order parameter, represented by the square modulus of the structure factor  $|F(z)|^2$  as obtained by fitting the diffraction curves shown in Fig.1(b)) at each time delay.

The spatiotemporal evolution of the AFM order melting resembles that of a magnetic soliton. A careful analysis shows that the propagation speed exceeds the longitudinal sound velocity. We will discuss possible scenarios for the excitation of these heterogeneous dynamics, which are nucleated at the interface through the light-driven lattice distortion of the substrate.

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