



HAL
open science

Accessing d-d Excitations Around Misfit Dislocations in Strongly Correlated NiO Thin Films Using High Energy-Resolution EELS

Matthieu Bugnet, Khalil El Hajraoui, Adam Kerrigan, Vlado K Lazarov, Guillaume Radtke, Quentin M Ramasse, Demie Kepatsoglou

► **To cite this version:**

Matthieu Bugnet, Khalil El Hajraoui, Adam Kerrigan, Vlado K Lazarov, Guillaume Radtke, et al.. Accessing d-d Excitations Around Misfit Dislocations in Strongly Correlated NiO Thin Films Using High Energy-Resolution EELS. *Microscopy and Microanalysis*, Jul 2024, Cleveland, United States. hal-04801592

HAL Id: hal-04801592

<https://hal.science/hal-04801592v1>

Submitted on 28 Nov 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Meeting-report

Accessing *d-d* Excitations Around Misfit Dislocations in Strongly Correlated NiO Thin Films Using High Energy-Resolution EELS

Matthieu Bugnet^{1,2,3,*}, Khalil El Hajraoui^{1,4}, Adam Kerrigan⁵, Vlado K. Lazarov^{4,5}, Guillaume Radtke⁶, Quentin M. Ramasse^{1,2,7}, and Demie Kepatsoglou^{1,4}

¹SuperSTEM Laboratory, SciTech Daresbury Campus, United Kingdom

²School of Chemical and Process Engineering, University of Leeds, United Kingdom

³Centre National de la Recherche Scientifique, Mateis Laboratory, University of Lyon, France

⁴School of Physics, Engineering and Technology, University of York, United Kingdom

⁵York-JEOL Nanocentre, University of York, United Kingdom

⁶Sorbonne Université, Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, France

⁷School of Physics and Astronomy, University of Leeds, United Kingdom

*Corresponding author: mbugnet@superstem.org

Quantum materials are in the center of academic research due to their potential on resolving the ever-increasing demand in energy-efficient and high-speed microelectronic devices. In that respect, transition metal (TM) oxides, for which the partially filled *d* shell of the TM element leads to a wide range of unusual properties, are of particular interest. Their properties could in principle be tuned with the controlled interplay of several degrees of freedom like lattice, orbital, charge, and spin. Among the plethora of candidate compounds under investigation, nickel oxide (NiO) has become a major focus since the experimental evidence of excitonic excitations involving Ni *d* electrons [1, 2].

The nature of the non-dipolar *d-d* excitations has previously been studied in single crystal NiO using two different theoretical bases to interpret nonresonant inelastic X-ray scattering experiments, which are appropriate to provide a spatially-average response with high momentum resolution [1, 2]. Electron energy-loss spectroscopy (EELS) in an electron microscope is a unique tool allowing access to such excitations at high spatial resolution, down to the scale of atomic columns in crystals. The technique was previously employed to demonstrate its viability to detect *d-d* excitations in NiO at the nanoscale [3]. Nevertheless, *d-d* excitations have thus far been probed only in pristine NiO, and it is of interest to understand their evolution in imperfect crystals, and specifically at defects.

To reach this goal, we use high energy-resolution (monochromated) EELS coupled to aberration-corrected scanning transmission electron microscopy (STEM) in a Nion HERMES instrument, equipped with a Nion monochromator and aberration-corrector, a high-stability Nion Iris EEL spectrometer fitted with a Dectris Ela direct electron detector. NiO thin films were grown by pulsed laser deposition and molecular beam epitaxy on Yttrium stabilized zirconia substrates and cross-section lamellae were prepared by focused ion beam milling in a Hitachi Ethos NX5000. Due to the lattice mismatch, the films present a high density of dislocations close to the interface, which induces a strong and localized structural distortion of the NiO lattice (Fig. 1a).

In this work, we combine monochromated low-loss and core-loss EELS analysis at atomic resolution to study the effect of the structural distortion around dislocations on the electronic structure of NiO. We also investigate the non-dispersive nature of the *d-d* excitations by mapping the electronic band-structure using a slit as the spectrometer entrance aperture. Here, the Ni-L₂₃ fine structures are only weakly sensitive to the distorted environment, while the O-K near-edge fine structures are not as sharp as in pristine areas and do not contain the ferromagnetic signature previously reported [4] (Fig. 1b). On the other hand, spectral variations of the *d-d* excitations can be detected around the dislocations as compared to the bulk (Fig. 1c), which suggests a complementary perspective to core-loss EELS to explore the complex interplay between the atomic and electronic structure of NiO. This is of particular interest given the intrinsic complexity to reproduce both O-K and Ni-L₂₃ edges in distorted environments from single-electron or multielectronic calculations, contrary to interband excitations like *d-d* transitions. Based on these results, the link between structural distortion and low energy excitations will be discussed. This investigation brings light on the behavior of excitonic excitations around defects in a prototypical quantum material like NiO [5].

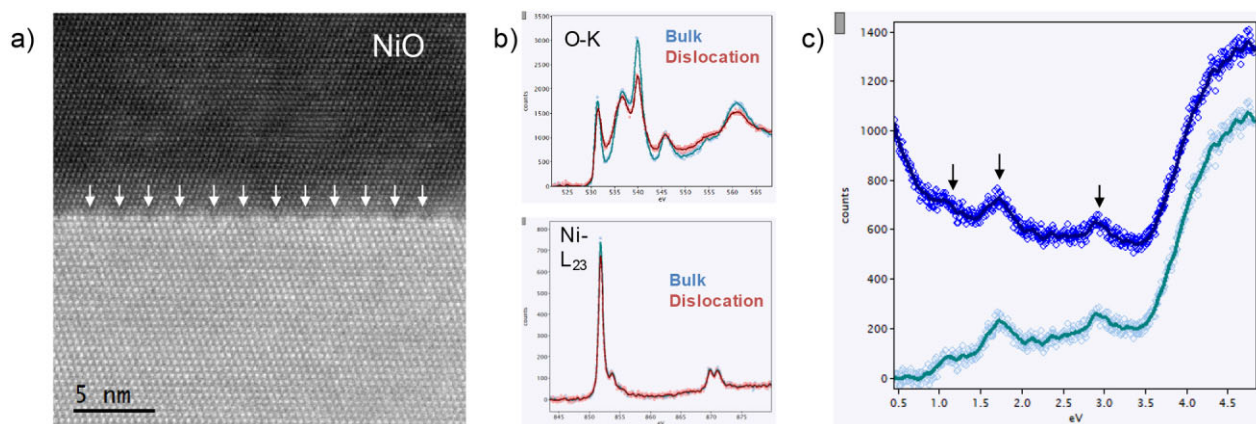


Fig. 1. a) STEM-HAADF images of the NiO thin film illustrating the periodic array of dislocations, indicated by the white arrows. b) O-K and Ni-L₂₃ near-edges fine structures in the bulk of the film and in the vicinity of the dislocations. c) Evidence of d-d excitations (black arrows) in the bulk of the NiO film with and without background extracted.

References

1. M. W. Haverkort *et al.*, *Physical Review Letters* **99** (2007), p. 257401. <https://doi.org/10.1103/PhysRevLett.99.257401>
2. B. C. Larson *et al.*, *Physical Review Letters*, **99** (2007), p. 026401. <https://doi.org/10.1103/PhysRevLett.99.026401>
3. A. Gloter *et al.*, *Ultramicroscopy* **109** (2009), p. 1333-1337. <https://doi.org/10.1016/j.ultramic.2009.06.005>
4. I. Sugiyama *et al.*, *Nature Nanotechnology* **8** (2013), p. 266-270. <https://doi.org/10.1038/nnano.2013.45>
5. M. Bugnet acknowledges access to SuperSTEM, the EPSRC national facility for advanced electron microscopy (through grants EP/V036432/1 and EP/W021080/1), and support from a visiting associate professorship from the School of Chemical and Process Engineering at the University of Leeds.