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Hendrik Meer, Stephan Wust, Christin Schmitt, Paul Herrgen, Felix Fuhrmann, Steffen Hirtle, Beatrice Bednarz, Adithya Rajan, Rafael Ramos, Miguel Angel Niño, Michael Foerster, Florian Kronast, Armin Kleibert, Baerbel Rethfeld, Eiji Saitoh, Benjamin Stadtmüller, Martin Aeschlimann, Mathias Kläui

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Laser-Induced Creation of Antiferromagnetic 180-Degree Domains in NiO/Pt Bilayers

*Hendrik Meer, Stephan Wust, Christin Schmitt, Paul Herrgen, Felix Fuhrmann, Steffen Hirtle, Beatrice Bednarz, Adithya Rajan, Rafael Ramos, Miguel Angel Niño, Michael Foerster, Florian Kronast, Armin Kleibert, Baerbel Rethfeld, Eiji Saitoh, Benjamin Stadtmüller, Martin Aeschlimann, and Mathias Kläui**

The antiferromagnetic order in heterostructures of NiO/Pt thin films can be modified by optical pulses. After the irradiation with laser light, the optically induced creation of antiferromagnetic domains can be observed by imaging the created domain structure utilizing the X-ray magnetic linear dichroism effect. The effect of different laser polarizations on the domain formation can be studied and used to identify a polarization-independent creation of 180° domain walls and domains with 180° different Néel vector orientation. By varying the irradiation parameters, the switching mechanism can be determined to be thermally induced. This study demonstrates experimentally the possibility to optically create antiferromagnetic domains, an important step towards future functionalization of all optical switching mechanisms in antiferromagnets.

1. Introduction

To overcome the limitations of ferromagnetic systems (FMs) it is a key step to transition to antiferromagnetic systems (AFMs) in future spintronic devices. Due to their net magnetic moment, FMs are limited in their bit packing density and stability against external fields compared to antiferromagnets.[1] Another key advantage of antiferromagnets is their potential for ultrafast applications due to their inherent dynamics with resonant frequencies in the THz range.[2] Recently, the electrical switching of antiferromagnets has been intensely investigated.[3–5]

H. Meer, C. Schmitt, F. Fuhrmann, B. Bednarz, A. Rajan, B. Stadtmüller, M. Kläui
Institute of Physics
Johannes Gutenberg-University Mainz
55099 Mainz, Germany
E-mail: mathias.klaeui@magnetism.ch

S. Wust, P. Herrgen, S. Hirtle, B. Rethfeld, B. Stadtmüller, M. Aeschlimann
Department of Physics and Research Center OPTIMAS
Technische Universität Kaiserslautern
67663 Kaiserslautern, Germany

R. Ramos
Centro de Investigación en Química Biolóxica e Materiais Moleculares (CIQUS)
Departamento de Química-Física
Universidade de Santiago de Compostela
15782 Santiago de Compostela, Spain

R. Ramos, E. Saitoh
WPI-Advanced Institute for Materials Research
Tohoku University
Sendai 980-8577, Japan

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/adfm.202213536.

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M. A. Niño, M. Foerster
ALBA Synchrotron Light Facility
Carrer de la Llum 2-26
Cerdanyola del Vallés
08290 Barcelona, Spain

F. Kronast
Helmholtz-Zentrum Berlin für Materialien und Energie
Albert-Einstein-Strasse 15, 12489 Berlin, Germany

A. Kleibert
Swiss Light Source
Paul Scherrer Institut
PSI 5232 Villigen, Switzerland

E. Saitoh
Department of Applied Physics
The University of Tokyo
Tokyo 113-8656, Japan

E. Saitoh
Center for Spintronics Research Network
Tohoku University
Sendai 980-8577, Japan

E. Saitoh
Advanced Science Research Center
Japan Atomic Energy Agency
Tokai 319-1195, Japan

M. Kläui
Graduate School of Excellence Materials Science in Mainz
55099 Mainz, Germany

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However, to achieve switching on an ultrashort timescale, we must transition from electrical to optical control of the antiferromagnetic order.

In ferri- and ferromagnetic material systems, fs-laser-induced all-optical switching (AOS) has been intensively studied.[6–9] Thermally induced switching has been observed in ferrimagnetic GdFeCo alloys[6,10] and all-optical helicity-dependent switching based on the inverse Faraday effect has been observed in a wide range of ferri- and ferromagnetic materials.[11] For antiferromagnetic materials, studies on the all-optical switching have focused on the excitation of magnon modes.[12–15] There are experimental reports of large-scale optical switching of antiferromagnetic order in the tilted antiferromagnet $TbMnO_3$.[16] However, the underlying mechanism relies on the electric polarization and cannot be easily transferred to other AFM systems. Theoretical studies predict possible optically induced switching in antiferromagnetic NiO,[17,18] NiO/FM bilayers[19] and other collinear antiferromagnets.[20] Recently, first experimental evidence for light-induced manipulation of antiferromagnetic domains in NiO crystals has been reported.[21] NiO is a prototypical collinear insulating antiferromagnetic system exhibiting promising features for future potential spintronic devices: current-induced switching of the antiferromagnetic order,[5,22] electrical readout,[23] antiferromagnetic shape anisotropy,[24] and ultrafast spin dynamics in the THz range.[2,25,26] The magnetic order of NiO thin films can be temporarily modulated by irradiation with ultrafast laser pulses,[27] and several studies have reported helicity-dependent excitation of coherent magnons in NiO.[13,28] NiO exhibits a strong magnetoelastic coupling[29] and thus the above-mentioned optical manipulation of antiferromagnetic domains[21] has been attributed to a particular phononic mechanism.[30] While several mechanisms have theoretically proposed optically induced switching of NiO, there are no experimental reports on the light-induced domain switching of the antiferromagnetic order in NiO thin films.

Here, we investigate the domain structure of NiO/Pt bilayers using X-ray photoemission electron microscopy (XPEEM) with magnetic linear dichroism as the contrast mechanism, that were irradiated by circularly and linearly polarized laser-light. We observe optically induced changes of the domain structure. In contrast to the often considered switching between different Néel vector axes, we observe the creation of 180° domains and domain walls, independent of the laser polarization. Variation of the irradiation parameters allows us to identify a thermal origin of the optically induced antiferromagnetic order. We demonstrate the possibility to optically create antiferromagnetic domains in a prototypical antiferromagnetic system.

2. Results

We prepared 10 nm thick NiO(001) thin films that are epitaxially grown on MgO(001) substrates by reactive magnetron sputtering. The films are additionally capped with a 2 nm thick platinum layer to allow for imaging with X-ray magnetic linear dichroism photoemission electron microscopy (XMLD-PEEM). We have previously characterized and investigated similarly grown NiO(001) thin films and observed that the strain from the substrate mismatch leads to preferential out-of-plane alignment

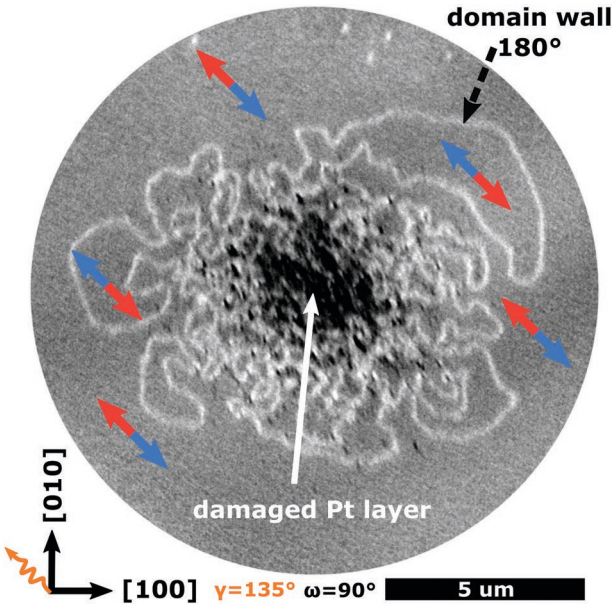

Figure 1. XMLD-PEEM image of the AFM domain structure, of an area that was illuminated for 2 s with circular right polarized laser light with a pulse length of 6.7 ps and a fluence of 12 ± 2 mJ cm^{-2}. The incoming X-ray is at an angle of 135° with respect to the [100] axis and the X-ray polarization is vertical to the sample plane with a 16° incidence angle. The blue arrows indicate the direction of the in-plane projection of the Néel vector, the different colors correspond to the different sublattices.

of the Néel vector in these thin films, stabilizing only one type of Spin-domains (S-domain).[31–34] Therefore, only four different Twin-domains (T-domains) are present in our films, each accompanied by a strong rhombohedral distortion. Similar to our previous studies on 10 nm NiO thin films,[34] we observe a domain structure in our field of view which predominantly consists of one T-domain. To study the effect of laser-irradiation on NiO/Pt bilayers, we use an ultrafast amplified laser system with a central wavelength of 800 nm and with a pulse repetition rate of 1 kHz. We irradiated our samples ex-situ with pulse trains of different pulse duration, pulse fluences, illumination time, and polarization. We imaged the antiferromagnetic domain structure of the laser irradiated regions using energy dependent XMLD-PEEM at the double peak of the Ni L_2 edge.[35] **Figure 1** shows the antiferromagnetic domain structure of a region irradiated with circularly right-polarized laser light. Several narrow domain walls (bright lines) can be observed around a central area where the platinum layer was structurally changed by the accumulated heat (dark spot). The structural changes in the capping layer are visible due to the high surface sensitivity of XPEEM and overlay with the magnetic XMLD contrast of the NiO layer in the center. The XMLD contrast depends on the orientation of the incoming electric field and the orientation of the Néel vector. We varied the polarization of the incoming X-rays (ω = 90°, 112.5°, 135°, 157.5°, 0°) and the azimuthal angle (γ = 90°, 120°, 135°, 150°, 165°, 180°) of the sample with respect to the incoming beam. For all combinations of ω and γ we observe no difference in contrast between the domains inside and outside the domain walls. The absence of contrast changes indicates that the projection of the Néel vector in these domains onto the X-ray polarization is identical for all angles

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of ω and γ and points along the same directions.[36] Thus, the domain wall between them can be identified as a 180° domain wall between two domains with 180° different orientation of the Néel vector.[37] The orientation of the Néel vector is identical on both sides of the wall, but the spins in the antiferromagnetically coupled sublattices are interchanged, as indicated by the differently colored arrows in Figure 1. Thus, the domains inside and outside the domain walls are in the same T-domain, are accompanied by the same distortion, and their S-domains have the same out of plane components. However, their sublattices are interchanged, making them 180° differently oriented domains. Note that the contrast of all the domains indicated by the blue and red arrows is identical as sub-lattice orientations that are 180° reversed yield the same XMLD contrast.

For different combinations of ω and γ we observe a variation and inversion of the contrast between the domain wall and the surrounding domains (see Supporting Information), confirming the magnetic origin of the contrast. The contrast of the domain wall appears uniform, as previously reported in our recent study on domain walls between T-domains in NiO thin films.[38] We can estimate the width of the 180° walls to be around 100 ± 30 nm (see Supporting Information). By irradiation with a laser, we are able to observe the creation of 180° domain walls (bright lines in Figure 1), which indicate the creation of domains with 180° different orientation of the Néel vector. The creation of 180° domains is independent of the polarization of the laser and can also be achieved with linearly polarized laser light (see **Figure 2**a).

To verify the antiferromagnetic nature of the laser-induced domains, we annealed our film in vacuum for 10 min without magnetic field above the Néel temperature, at 550 K. This has changed the domain structure and we can observe a disappearance of the 180° domain walls in Figure 2b. In Figure 2c, we imagined the area from Figure 1 after annealing and we can also observe that most 180° domain walls have disappeared. We can observe one 180° domain wall going across the laser irradiated area, which changes contrast depending on beam polarization (see Supporting Information).

As discussed above, the laser-induced 180° domains in Figure 2b have disappeared. However, the domains near the structural defect were not significantly altered by the heating. In NiO, strain or changes in the surface anisotropy that are introduced by patterning or defects can lead to the preferential stabilization of domains.[24] The optically induced 180° domains can be manipulated. Thus, the laser-induced domains are not generated by irreversible ablation-induced defects, which introduce strain or change the surface anisotropy, but originate from a fast heating and cooling of the antiferromagnetic NiO system. The laser-induced domain creation is dominated by thermal processes, as we can observe polarization independent creation, which occurs close to the threshold for structural changes. Only the accumulated heating and threshold for structural changes differ slightly between different laser polarizations.[39] For linearly polarized light under the same conditions as in Figure 2a, but with a lower fluence of 12 ± 2 mJ cm^{-2}, we could not observe a visible structural change or the creation of 180° domain walls, indicating that the accumulated heat was not sufficient to allow a reorientation of the spins. We can estimate the laser-induced temperature increase during irradiation by considering the

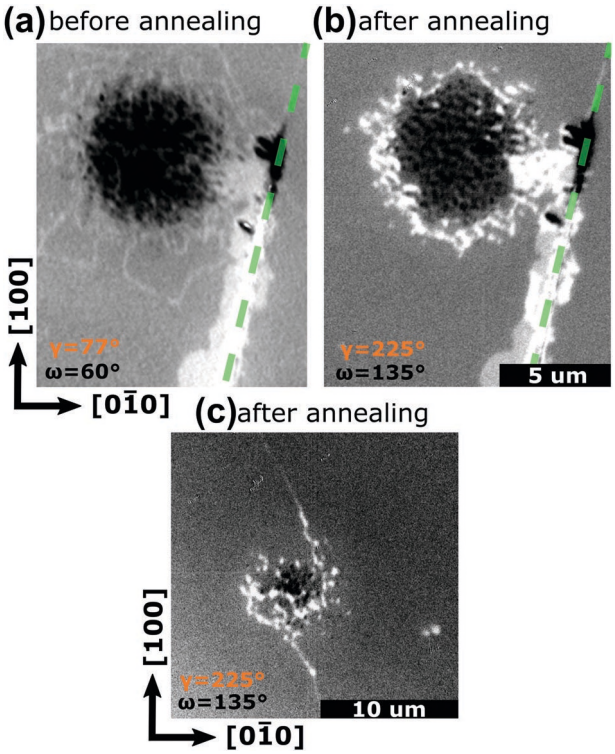

Figure 2. a) XMLD-PEEM image of an area that was illuminated for 2 s with linearly polarized laser light with a pulse length of 6.7 ps, 2000 pulses and a fluence of 16 ± 2 mJ cm^{-2}. b) Domain structure of the same laser-irradiated area after annealing above the Néel temperature. The green line indicates a structural defect at which domains are stabilized. c) XMLD-PEEM image of the area depicted in Figure 1 after annealing.

melting temperature of Pt as a lower boundary for the temperature increase near the ablated area. The temperature of the NiO near ablated areas can be estimated to be around 520 K, which is slightly below the bulk NiO Néel temperature, but above the reduced Néel temperature in our thin film ($T_{N,\,thin}$ = 400–460 K, see Supporting Information). Thus, the laser-induced heating near the threshold for structural changes can be assumed to be sufficient to allow a reconfiguration of the domain structure. In the case that the laser-induced domain creation is of thermal origin, it can be achieved without structurally affecting the Pt layer.

To create domains without damaging the Pt layer, we next investigate different laser pulse parameters. We patterned grids of markers onto the sample and irradiated the sample in the center of the grids. This allows us to identify the laser irradiated region in the XPEEM even if there is no structural changes to the Pt layer. We could observe the creation of domains without discernible structural changes to the Pt layer using 45 fs pulses and irradiation with 500 pulses over 0.5 s. The characteristic formation of 180° domain walls could be observed, as shown in **Figure 3**a,b, for irradiation with linearly polarized light. The threshold fluence for the formation of 180° domain walls under these irradiation conditions is found to be 7.2 ± 0.9 mJ cm^{-2}. Thus, we can optically create antiferromagnetic 180° domains and domain walls without damaging the NiO or the platinum capping layer.

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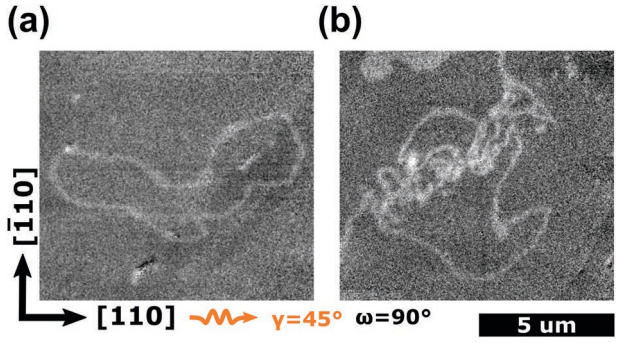

Figure 3. XMLD-PEEM image of two areas that were illuminated for 0.5 s with linearly polarized laser light with a pulse length of 45 fs and a fluence of a) 8 ± 1 mJ cm^{-2} or b) 7.2 ± 0.9 mJ cm^{-2} .

3. Discussion

We observe the optically induced formation of 180° domain walls in NiO, indicating the creation of 180° domains. We observe polarization-independent creation of the antiferromagnetic domains. The underlying mechanism is not based on a polarization dependent effect but on heat-induced domain formation, since the nucleation of the 180° domains is independent of the polarization of the exciting light.

Irradiation with the laser light heats up the NiO and its spin system. The lattice system around the heated area is still strained, exerting strain on the system that is heated up. During cooling of the irradiated area, this strain leads to preferential alignment of the spins into the previous twin domain after irradiation. The heated disordered spins settle into one of the two energetically degenerate states, with spins in the different sublattices pointing in opposite directions. Thus, 180° domains and 180° domain walls are formed. The situation is different when the whole sample is annealed, as shown in Figure 2b. As the creation of 180° domain walls requires additional energy, it is energetically unfavourable. In the slow cooling process of the annealing, (3.0 ± 0.2 K min^{-1}, see Supporting Information), the domain wall formation is avoided and their size is minimized. These results are consistent with observations on bulk NiO: it was observed that a fast cooling through the Néel temperature (opening the furnace door) can lead to a multidomain state, while annealing with slow cooling (5 K min^{-1}) leads to crystals with only few domains.[40] The size of the optically induced domains is not necessarily linear with fluence, as can be seen in Figure 3a,b, but depends strongly on the local anisotropy landscape of the irradiated area. However, by tuning the timescale of the laser heating pulse, one can potentially switch reversibly between different magnetic states.

The observed width of the antiferromagnetic 180° domain wall of 100 ± 30 nm is comparable to previous observations of 140-192 nm wide 180° degree walls in bulk NiO.[37] The determination of the Néel vector orientation inside the domain wall is challenging, due to the single-domain background in our angle dependent study. However, the higher XMLD intensity of the domain wall for out of plane polarization ($\omega = 90°$) under all investigated azimuthal angles ($\gamma = 77° - 180°$) indicates a larger out of plane component of the Néel vector inside the domain wall compared to the background domains.

Previous experiments based on current-induced switching have allowed for the controlled switching between different T-domains.[34] The laser-induced domain creation offers the exciting possibility to additionally create and study 180° domain walls, which have recently been reported to be rather narrow and even atomically sharp in antiferromagnets.[41] Further, domain walls themselves can play a crucial role in the magnon coupling of the NiO modes.[42,43] By controlled irradiation of a device, one could artificially introduce additional domain walls into the system, decreasing the size of the individual domains. In this way, laser-induced domain creation can be used to locally lower the potential switching energy for a device or to manipulate magnon transport inside a device.

While all-optical creation of 180° domains itself offers exciting opportunities, we believe that the demonstration of optically induced creation of large domains in NiO is an important step for the investigation of all-optical switching in AFMs. The pulse duration is found to be a crucial parameter in this process, since the underlying mechanism is based on the heating and the subsequent cooling of the system. Therefore, there is potential to push the writing speed to faster regimes by optimizing this optical parameter, while simultaneously tailoring the material parameters of the sample. Additionally, the variation of irradiation parameters could provide a tool for the realization of reversible optical switching. Different heat cycles could be used to switch between multi- and mono-domain states, similar to phase-change memory.[44]

It should be emphasized that our observation of a heat-induced optical domain formation does not imply the absence of other optical switching mechanisms in antiferromagnets or NiO. In the experiment presented here, we have observed 180° domain creation inside one T-domain, due to the preferential stabilization of only one S-domain, in our NiO (001) thin films.[34] In other systems with other domain configurations the optically induced domain formation might not be as restrained. To investigate the all optical switching in antiferromagnets one has to therefore carefully choose the material platform, a promising candidate might be NiO(111). In NiO (111) are three different S-domains and a total of six different magnetic states per T-domain present. Thus, NiO (111) could be the ideal platform to explore the theoretical proposed optical switching mechanisms, as the spins could switch between multiple S-domains.[19] We believe that the mechanism found here can also be achieved in other antiferromagnetic systems with strong magnetoelastic coupling, such as CoO or Hematite. The optically induced creation of 180° domains and domain walls thus provides an additional handle to manipulate antiferromagnetic domain structures.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

antiferromagnets, domains, insulators, spintronics, switching

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