Phononic manipulation of antiferromagnetic domains in NiO

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Abstract
The interaction between the crystal lattice and spins appears to be a very efficient mechanism for non-thermal manipulation of magnetic order. A particularly intriguing question is whether lattice vibrations can also be used to control or even switch domains in collinear antiferromagnets, where all other mechanisms fail. To answer this question, here we study the magnetic effects induced by tunable and narrow-band infrared optical pulses in single crystals of antiferromagnetic nickel oxide. Our measurements reveal that a resonant excitation in the vicinity of the frequency of longitudinal optical phonons leads to the displacement of antiferromagnetic domains in NiO, potentially revealing a promising method to manipulate antiferromagnetic order.

1. Introduction

The question of how to efficiently and quickly manipulate the antiferromagnetic order parameter represents one of the great challenges in condensed matter physics [1]. The net magnetic moment associated with ferromagnets allows their magnetic ordering to be easily manipulated by modest magnetic fields [2], giving rise to a vast set of technologies ranging from the compass and electrical engines to magnetic-based logic and hard-disk-drives [3, 4]. In contrast, while antiferromagnets (AFMs) represent the overwhelming majority of magnetically-ordered materials [5], their intrinsic lack of net magnetic moment leads to the intense search for alternative means to leverage their magnetic ordering [6, 7]. If successful, such identification of suitable levers will allow us to design and construct AFM-based technologies that would far supersede the speed, scalability and robustness of their ferromagnetic counterparts [8–10].

Recently, electric currents were shown to facilitate the manipulation and switching of magnetic domains in AFMs. For example, in CuMnAs and Mn$_2$Au, it is possible to switch the antiferromagnetic ordering at room temperature using the staggered action associated with electric currents [11–13]. Moreover, the domains in CuMnAs can also be switched using current-induced domain-wall motion [14]. Detailed studies of current-assisted magnetization switching on NiO/Pt bilayers using optical birefringence and the spin-Seebeck effect were published lately [15, 16], in which the combination of thermal excitation with a thermo-magnetoelastic mechanism was found to be responsible for the observed current-induced switching. However, while electrical control over the AFM order is technologically appealing, it is not a universal approach, being limited to a few AFMs with specific crystallographic and magnetic order.

Alternatively, the possibility of ultrafast all-optical magnetic switching in NiO has already been proposed and discussed theoretically [17, 18]. While this has not yet been demonstrated experimentally in AFMs, this mechanism does show its potential in exciting coherent magnons [19]. On the other hand, all-optical switching of magnetization already offers the fastest and most energy-efficient route for magnetic recording. Non-thermal coherent switching of magnetic bits, for example, can be achieved in co-doped iron-garnet within 20 ps and with a projected dissipation of only 22 aJ via the selective excitation of electronic transitions at resonance [20].

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Resonances in general offer the most energy-efficient and fastest means to affect change in condensed-matter systems. It is therefore straightforward to envision that non-linear phononics [21], in which infrared (IR)-active phonons are strongly driven at resonance and consequently modify material properties via coupled otherwise-inaccessible modes, may actually hold the ultimate key to unlocking the efficient and rapid switching of ordering in AFMs. Already, studies of non-linear phononics have revealed its capability to introduce complex structural transformations in materials, leading to striking phenomena such as the transient stimulation of insulator-to-metal phase-transitions in correlated oxides [22], the enhancement of superconducting properties [23] and the triggering of ultrafast AFM-to-ferromagnet phase-transitions [24].

In addition to driving the aforementioned short-lived phenomena, it is also possible for the resonant excitation of longitudinal optical (LO) phonons to permanently switch magnetic ordering in a ferrimagnetic iron-garnet [25]. Can the same mechanism be used to switch antiferromagnetic ordering as well? Strain, which can be induced via a nonlinear phononic mechanism, represents one of the most promising stimuli for manipulating the order parameter of AFMs on ultrafast timescales [26], as indicated by very recent studies of ultrafast phonon-induced AFM-to-ferromagnet phase transition [24]. It was also theoretically suggested to play important role in a magnetic-switching scenario in NiO [27].

In this article, we experimentally explore whether optical phonons can switch antiferromagnetic ordering. Our experiments consist of illuminating single-crystal samples of NiO with narrow-band IR optical pulses across the spectral range of 12.5–30 μm (800–330 cm⁻¹). We show that a resonant excitation at the frequency of LO phonon mode leads to the displacement of the antiferromagnetic domains, while the strongly-absorbing transverse optical (TO) mode produces no discernible change. Our results reveal that optical phonons are indeed capable of manipulating antiferromagnetic domains in NiO, and open important questions about the underlying mechanism.

2. Antiferromagnetic NiO samples

Our material of choice for this investigation is the classical collinear type-II AFM NiO. In the paramagnetic phase above the Néel temperature \( T_N = 523 \) K [28], NiO possesses cubic structure of sodium chloride type (point group \( m\overline{3}m \)). At temperatures below \( T_N \), the exchange interaction couples spins ferromagnetically within the 111 planes, and antiferromagnetically between the neighboring planes [29]. Because of magnetic ordering, the crystal structure acquires a rhombohedral distortion along the ⟨111⟩ direction. The crystallographic point group symmetry reduces to \( \overline{3}m \) and results in a sizeable optical birefringence of \( \Delta n = 0.003 \) [30], with the optical axis coinciding with that of the distortion. The birefringence allows one to see antiferromagnetic domains thus formed along the four equivalent ⟨111⟩ directions \([{111}], [{1\overline{1}T}], [{1\overline{1}T}1], [{\overline{1}T}1]) \) (the so-called T-domains) [31].

Within each of the T-domains, there are moreover three equivalent ⟨1\overline{1}T⟩ directions that are perpendicular to that of the rhombohedral distortion [32]. This allows therefore the existence of three other domains, usually referred to as S-domains. We therefore count a total of twelve possible domain states in NiO. The formation of such S-domains give rise to another magnetostriuctive distortion, resulting in a reduced crystallographic point group symmetry \( 2/m \) that defines the point group symmetry of the magnetic lattice [33, 34]. The distortion and associated optical linear birefringence originating from the T-domains is, however, two orders of magnitude larger than that derived from the S-domains [35], and so the S-domains practically have negligible influence on the polarization of the propagating light. Therefore, our observations below mostly concern with T-domains.

In our experiments, we used single crystals of NiO (111) and NiO (100), both with dimensions \((5 \times 5 \times 0.1) \) mm³, commercially produced by Surfacenet GmbH and MaTecK GmbH respectively using float zone crystal growth [36]. Both sides of the NiO (111) and NiO (100) samples were polished with roughness nominally less than 0.01 μm and 0.10 μm respectively, and the orientation is specified with an accuracy ±0.1° and ±0.3° respectively.

In figure 1, photographs of the domain structure of the two NiO samples are shown. The images were obtained using a Leica DM IRM microscope with a ×30 objective lens and a Sony IMX273 CMOS sensor. The alternating regions of high contrast manifesting in the form of lines correspond to antiferromagnetic domain walls. The estimated average width of the domain wall is 890 ± 80 nm. The length of the domain walls varies substantially across the sample, depending most likely on the crystalline quality. The domains tend to be clustered in groups of 5–10, with the groups orienting differently with respect to neighboring domains at 90° for NiO (100) and 120° for NiO (111). This is in clear agreement with the samples’ crystalline structure.

It is worth noting that, apart from having less roughness and better orientation accuracy, the NiO (111) sample features much larger (up to 3–4 μm) and more pronounced defects compared to NiO (100). The
defects are represented as black spots on figure 1 and are visible across the whole thickness of the samples. These defects seem to be inclusions of alien material; they appear to be non-magnetic and do not affect the domain pattern. However, the presence of a higher amount of defects of larger size in the NiO (111) sample lowers the efficiency of birefringence-related contrast and hence results in less pronounced visibility of magnetic domains. This leads to a poorer response being observed for this sample. In the NiO (100) sample, however, the smaller number and size of defects promote sharper contrast as per each domain as well as for domain structures across an image.

3. Experimental setup

To provide resonant excitation of the phonon modes in NiO, we sourced IR optical pulses from the FELIX (Free Electron Laser for Infrared eXperiments) facility in Nijmegen, The Netherlands. The IR beam with photon energy ranging between 99 meV and 42 meV (wavelength 12.5 μm–30 μm) was focused to a spot of diameter ≈200 μm on the surface of the NiO sample. The IR radiation consist of bursts (‘macropulses’) at a repetition-rate of 10 Hz and with duration 8 μs. Each macropulse (with energy ranging between 2 mJ and 4 mJ) contains micropulses at a repetition rate of 25 MHz. The duration of each micropulse is defined by the cavity detuning of the free-electron laser, and have been shown to be Fourier-transform limited [37]. Their bandwidth is experimentally tunable in the range of 0.5%–2.0%. A fast optical shutter was used to expose the samples to a single macropulse, i.e. a burst of ≈200 micropulses. To assess the spectral dependence of the observed effects, we tuned the photon energy to different spectral lines while keeping the same beam fluence.

To visualize the antiferromagnetic domains in the NiO samples, we illuminate the samples with linearly-polarized light, of central wavelength 520 nm, delivered by a MatchBox CW laser. The light transmitted through the sample is collected by either a ×10 or ×100 objective lens, filtered using an analyzer and projected on to a Kiralux 5.0 MP CMOS Compact Scientific Camera. The antiferromagnetic domain walls are thus detected via linear birefringence in the same manner as in figure 1. The average power of the defocused CW laser was ≈10 mW. To detect the changes of the AFM domains induced by the IR macropulse, the camera was externally triggered to take an image before and after optical illumination.

4. Resonant effect on NiO domains

Upon tuning the IR wavelength across the whole phononic spectral region, we indeed observe transient effects of the FELIX pulse on the domains in both NiO samples. In figure 2, we present typical images showing the change of the AFM domain structure in NiO (100) induced by the IR light, obtained with ×100 magnification and taken about 100 μs after the optical exposure. The domain states before and after irradiance are shown in rows (a) and (b) respectively, with the difference shown in row (c).

4.1. Data treatment

The ‘difference’ row in figure 2(c) clearly reveal a pronounced displacement of AFM domains when the IR pulse has a central wavelength of 13.5 μm and 17 μm, which is otherwise minimized at other optical wavelengths. The former wavelengths may actually be directly referred to as 2nd-order transverse (2TO)
Figure 2. Transient changes of the antiferromagnetic domain structure in NiO (100) induced by irradiation with a single IR macropulse at different wavelength, as indicated in the figure. Images in rows (a) and (b) were obtained before and after IR illumination respectively, with row (c) showing the difference. All images are of size (12 × 12) μm².

phonon and LO phonon lines in NiO crystals respectively, as measured by Raman spectroscopy [38]. To facilitate comparison between the phonon modes and our results, we reproduce in figure 3(a) the Raman spectrum characteristic of NiO measured by Aytan et al in reference [38]. The IR-active TO phonon at the wavelength 25.8 μm was identified by IR ellipsometry performed by Willett-Gies et al in reference [39].

To quantitatively evaluate and compare the effect of the different radiation, we employ the analysis technique 'structural similarity index measure' (SSIM). SSIM is a metric that measures the difference between two similar images [40] and is defined as:

\[
SSIM(x, y) = \left[ l(x, y) \right]^\alpha \cdot \left[ c(x, y) \right]^\beta \cdot \left[ s(x, y) \right]^\gamma,
\]

(1)

where \( \alpha > 0, \beta > 0, \gamma > 0 \) denote the relative importance of each of the metrics. The comparison between the two images is performed on the basis of 3 features: \( l \)—luminance, \( c \)—contrast and \( s \)—structure. It is important to note that SSIM was developed specifically to correlate better to human perception. As in specific cases, its expression is identical to Weber’s Law of luminance sensitivity of the human eye [41]. Even though the expressions state high correlations, they are often not very mathematically tractable, and in particular are rarely metrics in the strict mathematical sense. However, SSIM finds use in such applications as automated quality assurance [42], development of optimal JPEG 2000 encoders [43] and in multiple applications for machine vision [44, 45]. For a detailed and more in-depth understanding of SSIM, we refer to the original paper [40] introducing this measure. Here, we merely employ the algorithm as a tool for qualitative analysis.

Instead of applying the above metrics across the whole image at once, it is often better to apply the metrics in \( M \) small sections of the image and taking the mean overall. This method is often referred to as the ‘mean structural similarity index measure’ (MSSIM) and is calculated as follows:

\[
MSSIM(X, Y) = \frac{1}{M} \sum_{j=1}^{M} SSIM(x_j, y_j).
\]

(2)

MSSIM was averaged for a set of five independent measurements for both samples. Each set of measurements was performed using two different magnifications, \( \times 10 \) and \( \times 100 \). This was done to observe both large-scale changes in domains structure (with \( \times 10 \) magnification) as well as changes in individual domains (with \( \times 100 \) magnification). Using objectives with different magnification, however, requires adjusting both the power of the CW laser and the analyzer’s orientation in order to optimize magnetic contrast. This readjustment causes the images obtained with \( \times 10 \) magnification to have different luminance and contrast compared to those obtained with \( \times 100 \) magnification. The advantage of having a wider field of view with \( \times 10 \) magnification compared to using \( \times 100 \) magnification also results in the imaging showing slightly different structures. These factors all affect the processing of the images. After processing the images, we extract the MSSIM value that quantifies the change between the images taken before and after optical exposure, with an MSSIM value equal to 1 or 0 corresponding to minimal or maximal change respectively.

4.2. Results

Our results for NiO (100) are presented in figure 3(b). The images obtained with \( \times 10 \) magnification (green curve) reveal an effect of 15% in the magnitude of MSSIM, in the form of a broad dip lying between the 2TO and LO phonon lines, and no noticeable change in the vicinity of the TO phonon. There are two
Figure 3. (a) Normalized Raman spectrum of NiO taken from reference [38], with the frequencies of longitudinal (LO), transverse (TO) and 2TO optical phonons indicated. (b) Spectral dependence of MSSIM for NiO (100), calculated by image analysis using MSSIM. Green and orange points were obtained using ×10 and ×100 magnification respectively. (c) Spectral dependence of MSSIM for NiO (111), with blue and red points being obtained using ×10 and ×100 magnification respectively. In panels (b) and (c), the error bars show the standard deviation of the averaged data, and the solid lines are guides-to-the-eye.

potential reasons for the merging of the LO and 2TO phonon lines in to one broad dip in the case of ×10 magnification. The first relates to the spectral broadening of the IR pulse, with the bandwidth measured to be ≈0.25 μm. The second involved the calculation of MSSIM, which becomes less efficient for a larger field of view due to the poorer spatial resolution. We overcome the latter problem upon studying the images obtained with ×100 magnification (orange curve). With better spatial resolution, the broad dip previously identified actually consists of two dips correlating very well with the LO phonon (efficiency of 70%) and reasonably well with the 2TO phonon. This clearly shows that the direct optical excitation of the LO phonon results in a strong change in the AFM domains of NiO (100).

Our results for NiO (111) are shown in figure 3(c). Due to the lower sample quality, as discussed earlier, the data obtained using ×10 magnification (blue curve) may be excluded from the analysis since MSSIM only fluctuates in the range of 0.5% and shows no resonant features. With better spatial resolution afforded by ×100 magnification (red curve), however, a stronger effect of 10% is observed. Similar to the case of ×10 magnification for NiO (100), we observe a clear dip across the spectral range spanning the 2TO and LO phonon modes, albeit with a slight shift towards the LO phonon line.

To clarify the origin of the error bars in figures 3(b) and (c) we performed noise variance estimation $\sigma^2$ for the same datasets. The calculation is performed using the algorithm developed in [46]. The $\sigma^2$ was also averaged across the five independent measurements. We find that $\sigma^2 \approx 0.02$ for both magnifications of NiO (111), and $\sigma^2 \approx 0.09$ and 0.18 for ×10 and ×100 magnification of NiO (100) respectively. From these results, one may conclude that the error bars in figures 3(b) and (c) are mostly defined by raw data noise.
5. Discussion

Interestingly, for both NiO samples with orientations (100) and (111), we clearly observe no discernible effect on the AFM domains when the optical pulse drives at resonance the IR-active TO phonon. It is at this particular frequency, however, that substantial effects would be expected according to the existing understanding of non-linear phononics [21, 47], since the IR pulse can couple directly to this mode and thus anharmonically displace the equilibrium of the crystalline potential. In contrast, we find that exciting the sample at the frequency of the LO phonon leads to a substantial change of the AFM domains, with an a possible effect also manifesting at the frequency of the 2TO phonon mode. Note that while the TO phonon could be directly observed in IR absorption experiments performed at normal incidence, the LO and 2TO phonons were observed in Raman scattering measurements only. From this argument, our discovery that the strongest effect comes from the excitation of the LO phonon is rather inexplicable.

The influence of the LO phonon was noticed in the phononic switching of magnetization in iron-garnets [25]—in that case, however, the influence of the LO phonon was rather related to the symmetry of the crystal lattice, which is evidently not the case for NiO.

The following ideas and notes should be put forward to achieve at least a plausible hypothesis of the phenomenon: firstly, despite the apparent simplicity of NiO’s rock-salt structure, any model of phonon modes in NiO must overcome several complications [38]. The LO and TO modes are not Raman-active in rock-salt crystals but the weak rhombohedral distortion induced by the AFM order, and the presence of defects may make them apparent in Raman measurements. Moreover, the potential for irreversibly damaging NiO samples at high temperatures (close to $T_N$) inhibits studies of lattice anharmonicities. Secondly, LO phonon modes in ionic crystals are coupled to the electrostatic fields, and thus interact with their environment stronger than the TO ones. This has been revealed by studies of the electron–phonon (Fröhlich) interaction in semiconductor crystals and nanodots [48–50]. The question of how the IR light seemingly drives the LO phonon mode, however, remains open.

6. Conclusions

We have experimentally studied, using linear birefringence in combination with polarizing microscopy, how narrow-band IR optical pulses manipulate antiferromagnetic domains in single-crystal NiO samples of orientation (100) and (111). Our results show that the domains are most strongly affected when the optical pulses match in frequency the characteristic frequencies of the LO and 2TO phonon modes in NiO. In contrast, minimum effect is found when we excite the IR-active LO phonon. Our results unambiguously reveal that antiferromagnetic domains in collinear AFMs can be manipulated via phonon-based processes, and will undoubtedly stimulate further investigation of the underlying mechanism.

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Data availability statement

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

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(2022) 023009 P Stremoukhov et al