

Antiferromagnetic insulatronics: spintronics in insulating 3d metal oxides with antiferromagnetic coupling

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Antiferromagnetic transition metal oxides are an established and widely studied materials system in the context of spin-based electronics, commonly used as passive elements in exchange bias-based memory devices. Currently, major interest has resurged due to the recent observation of long-distance spin transport, current-induced switching, and THz emission. As a result, insulating transition metal oxides are now considered to be attractive candidates for active elements in novel spintronic devices. Here, we discuss some of the most promising materials systems and highlight recent advances in reading and writing antiferromagnetic ordering. This article aims to provide an overview of the current research and potential future directions in the field of antiferromagnetic insulatronics.

I. INTRODUCTION

In spin-based electronics, writing, storing, and reading information relies on the electron's spin rather than its charge. Spintronic devices are commonly implemented in ferromagnets¹. Despite major advances, real devices utilizing conventional ferromagnetic metals and spin-polarised charge currents have several drawbacks: parasitic magnetic stray fields, intrinsically low characteristic dynamic frequencies, large magnetic damping, and ohmic losses. These limit the device density, integration, and operation speed, as well as increase the power consumption.

AFMs have moved to the forefront of condensed matter physics and especially spintronics, due to their unique and favorable properties, which have recently started to be exploited²⁻⁶. In particular, the zero net magnetic moment makes AFMs insensitive to external stray fields, thus enhancing their stability. Furthermore, the absence of stray fields implies that there is no dipolar coupling between different areas in an AFM. If used for storage, this could lead to more than a 100-fold increase in the potential storage density⁷. In collinear AFMs, 180° reversal of the magnetic ordering is not easily detectable. Therefore, in contrast to ferromagnets, where a logic "1" and a logic "0" are commonly encoded by 180° reversal of the magnetization, information is stored along multiple directions in AFMs. Beyond the static properties, the exchange enhancement of the dynamics⁸ leads to eigenmode frequencies that are orders of magnitude higher compared to ferromagnets. The ultrafast dynamics holds promise for antiferromagnetic devices with THz operation speed⁹⁻¹¹.

The three key strategies for developing novel devices for the next-generation information and communication technologies are thus to i) eliminate stray fields to increase the density, ii) integrate low-damping insulators

to decrease the power consumption and increase the efficiency and, iii) employ materials with ultrafast dynamics to increase the operation speed.

Theoretically, it was predicted that pure spin currents can be generated, transported, and used in antiferromagnetic insulators for writing, reading, and transporting spin information to enable such new devices^{2,3,12}. The experimental verification for electrical detection and control of the antiferromagnetic order¹³ has recently propelled AFMs into the limelight and paves the path towards utilizing AFMs as active components in spintronic devices. Ferromagnets currently still dominate the field of spintronics, but insulating antiferromagnets (AFMs) have been shown to exhibit properties that have the potential to revolutionize the industry by enabling ultrafast, low-power spin-based electronic devices in the future¹⁴.

Insulating magnetic oxides have been of particular interest due to their tunable magnetic ordering, magnetic properties and their chemical stability¹⁵⁻¹⁷. While ferro- and ferrimagnetic oxide systems have been widely studied, insulating antiferromagnetic oxides have recently also gained significant interest. Most conspicuous are 3d metal (Fe, Ni, Co, Cr, etc.) oxides that commonly exhibit antiferromagnetic order with well-defined spin structures and can be grown in high quality both as bulk crystals and thin films. This review presents an overview of insulating antiferromagnetic 3d oxide materials and discusses recent developments in detecting and manipulating the antiferromagnetic state based on a variety of mechanisms ranging from static to ultrafast phenomena.

II. IMAGING METHODS TO REVEAL THE MAGNETIC ORDERING IN ANTIFERROMAGNETS

A key challenge for realizing antiferromagnetic devices is the readout of the antiferromagnetic state. The absence of net magnetic moments and stray fields makes it more challenging to detect the antiferromagnetic order compared to their ferromagnetic counterparts. However,

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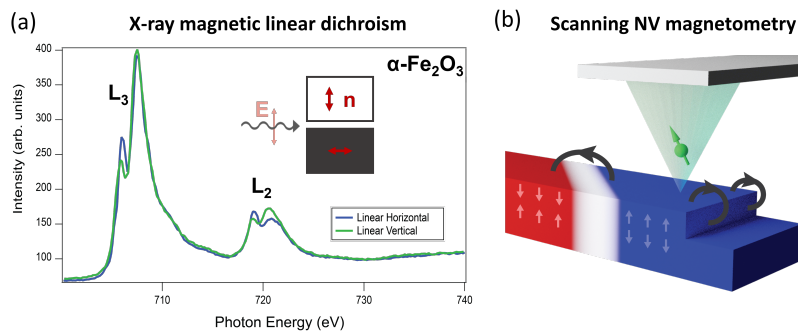


FIG. 1. (a) X-ray absorption spectra for linear horizontal (blue) and vertical (green) polarization at the Fe $L_{3,2}$ edges for $\alpha\text{-Fe}_2\text{O}_3$. Inset shows the bright (dark) contrast for parallel (perpendicular) alignment of the Néel vector (red) and the X-ray polarization. (b) Illustration of imaging the stray field of AFMs at domain walls and topographic features using scanning NV magnetometry.

88 recent developments in experimental magnetic imaging¹²⁰
 89 techniques have led to easier access to the antiferromag-
 90 netic domain structure with increased spatial and tem-
 91 poral resolution^{10,18}.

92 A. Birefringence imaging

93 The first imaging of antiferromagnetic domains uti-
 94 lized birefringence imaging to visualize antiferromag-
 95 netic domains in the insulating collinear antiferromagnet
 96 NiO ^{19,20}. Birefringence imaging is based on the polar-
 97 ization rotation of reflected or transmitted light from an
 98 antiferromagnetic material. Many magneto-optical ef-
 99 fects that are linearly dependent on the magnetization
 100 \mathbf{M} of a material (such as the Kerr or Faraday effect)
 101 can be used to investigate materials with a net magne-
 102 tization $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$, but are not suitable for com-
 103 pensated AFMs because the opposite sub-lattice magne-
 104 tizations $\mathbf{M}_1 = -\mathbf{M}_2$ cancel each other out. However,
 105 there are also second-order effects that depend quadrat-
 106 ically on the Néel vector $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$ and do not van-
 107 ish even for compensated AFMs^{10,21}. While differences
 108 in the birefringence of antiferromagnetic domains can
 109 originate directly from second order magneto-optical ef-
 110 fects (Cotton-Mouton or Voigt Effect)^{22,23}, birefringence
 111 of AFMs with a strong magnetoelastic coupling is often
 112 dominated by strain-induced birefringence²⁴⁻²⁶. Birefrin-
 113 gence imaging using a polarizing microscope is a power-
 114 ful and easily accessible tool to investigate the antiferro-
 115 magnetic domain structure of bulk crystals^{20,27} and thin
 116 films^{28,29}. Moreover, it can be readily combined with ad-
 117 ditional techniques such as current-induced switching ex-
 118 periments³⁰⁻³², application of magnetic fields³³ or pump-
 119 probe techniques²².

B. X-ray magnetic dichroism

122 Element specificity, sensitivity to chemical sites, and
 123 variable depth sensitivity make polarized X-ray absorp-
 124 tion spectroscopy a powerful tool³⁴. While X-ray mag-
 125 netic circular dichroism (XMCD) vanishes for fully com-
 126 pensated magnetic moments, antiferromagnetic order
 127 can be studied by measuring the X-ray magnetic linear
 128 dichroism (XMLD)³⁵⁻³⁸. The XMLD signal is given by
 129 the difference in absorption of linearly polarized X-rays
 130 with polarization parallel and perpendicular to the Néel
 131 order (see Fig. 1a). In addition to XMLD, crystal fields
 132 can also induce linear dichroism, which has to be care-
 133 fully disentangled from the magnetic contribution³⁹.

134 Today, XMLD detected by photoemission electron mi-
 135 croscopy (PEEM) is one of the most widely used tech-
 136 niques to image antiferromagnetic domain structures.
 137 Vector maps reconstructed from angle-dependent X-ray
 138 imaging reveal the orientation of the Néel order^{40,41}.
 139 In contrast to PEEM, scanning transmission X-ray mi-
 140 croscopy (STXM) allows for measuring XMLD in sizable
 141 magnetic fields. The necessity to deposit the material of
 142 interest on a membrane can be circumvented by detecting
 the XMLD in the total electron yield (TEY)^{42,43}.

C. Nitrogen-vacancy center magnetometry

143 Nitrogen-vacancy (NV) centers in diamond have been
 144 shown to be highly sensitive and non-perturbative probes
 145 for sensing the stray field of magnetic materials, oper-
 146 ating over a wide range of temperatures and magnetic
 147 fields, and a dynamic range spanning from direct current
 148 to gigahertz⁴⁴⁻⁴⁷. While AFMs do not produce global
 149 stray fields, small magnetic stray fields can arise locally
 150 due to uncompensated magnetic moments at the sur-
 151 face, topographic features, or domain walls (see Fig. 1b).
 152 Wide-field NV microscopes use a camera to image a dense
 153 layer of NV centers close to the surface of a diamond
 154 crystal adjacent to the magnetic layer and are, hence,

156 diffraction limited. The spatial resolution of scanning
 157 NV magnetometry, which is based on scanning a single
 158 NV center at the apex of a diamond tip across the sam-
 159 ple, can reach the nanometer scale. A major challenge
 160 of NV magnetometry is the reconstruction of the mag-
 161 netization from the stray fields. To lift the ambiguity
 162 of the reconstruction, a dual approach enabling detec-
 163 tion of both, the stray field and the magnetic order, can
 164 be used⁴⁸. Recent reports have successfully probed the
 165 magnetic domain structure of 3d metal oxide AFMs us-
 166 ing NV magnetometry^{49–52} as well as the magnetic noise
 167 and spin waves via spin relaxometry^{53–56}.

168 III. 3D OXIDE MATERIALS AND THEIR SPIN 169 STRUCTURES

170 In ferromagnets, magnetic domains are defined by re-
 171 gions of uniform magnetization. In order to minimize²¹⁴
 172 the stray field, ferromagnets typically adopt a multi-
 173 domain state in absence of an external magnetic field²¹⁶
 174 depending on the interplay of a range of relevant energy²¹⁷
 175 terms such as the exchange interaction, dipolar interac-
 176 tion, and anisotropy. While complex spin textures have²¹⁹
 177 been studied widely in ferromagnets, much of the under-
 178 lying physics of their analogues in AFMs remains to be²²¹
 179 explored.²²²

180 In collinear AFMs, the order parameter, the Néel vec-
 181 tor, is given by the direction of the staggered magneti-
 182 zation of the sublattices. Given the zero net magnetic²²³
 183 moment, one might expect that magnetic fields are un-
 184 able to change the Néel vectors. However, in order to²²⁴
 185 explain the magnetic susceptibility observed in some an-
 186 tiferromagnetic materials, Néel proposed that domain²²⁶
 187 walls separating different antiferromagnetic domains can²²⁷
 188 be displaced by small magnetic fields⁵⁷. Indeed, multi-
 189 domain states have been observed in a broad range of²²⁹
 190 materials today^{20,38,58–62}. The staggered magnetic order²³⁰
 191 in collinear AFMs gives rise to a zero net stray field.²³¹
 192 Thus, based on the intuition developed for ferromagnets,²³²
 193 spontaneous formation of domains in AFMs might come²³³
 194 as a surprise. In antiferromagnetic oxides, strong mag-
 195 netoelastic interactions come into play and can lead to²³⁵
 196 the formation of domains, as discussed in more detail be-
 197 low. Antiferromagnetic domains resulting from magne-
 198 toelastic interactions predominantly form along different²³⁸
 199 (noncollinear) directions^{43,63–65}. However, antiferromag-
 200 netic domains can also form with the Néel vector along²⁴⁰
 201 the same axes in the adjacent domains. In collinear anti-
 202 ferromagnets composed of two sub-lattices the direction²⁴²
 203 of the spins of the sub-lattices can be interchanged be-
 204 tween two domains, as shown in Fig. 2. Thus, the Néel²⁴⁴
 205 vector in these two domains is pointing along the same²⁴⁵
 206 axes, but along the opposite direction, creating domains²⁴⁶
 207 with 180° different Néel vector orientation delineated by²⁴⁷
 208 a 180° domain wall.²⁴⁸

209 To understand the formation of 180° domains, we have²⁴⁹
 210 to consider additional mechanisms^{66,67}. For instance,²⁵⁰

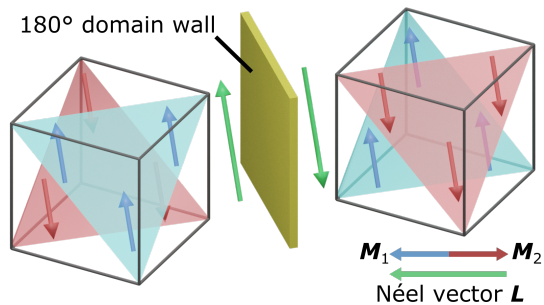


FIG. 2. Depiction of 180° domains in which the magnetization of the sub-lattices (red and blue) is interchanged and the Néel vector (green) points along the opposite direction.

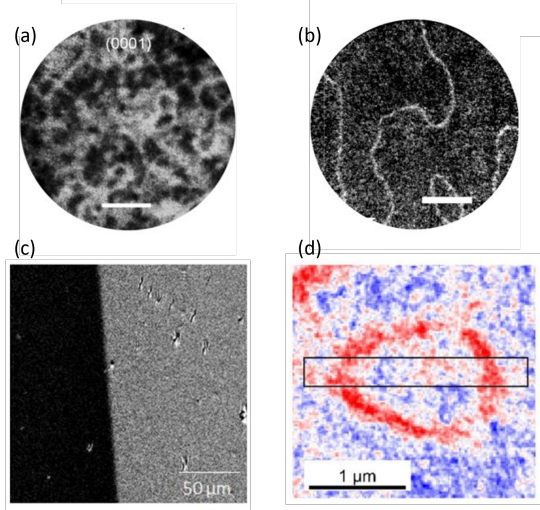
180° domains can result from defects where domains are
 nucleated during cooling from above the ordering temper-
 ature, the Néel temperature T_N ⁴⁹. Another possibility is
 the merging of two 90° domain walls when the antiferro-
 magnet is driven above the spin flop transition⁶⁷. Dur-
 ing this process, two domains that have nucleated sepa-
 rately meet and may form a 180° domain wall. In AFMs,
 the formation of domains and complex spin textures is
 mainly governed by the interplay between exchange in-
 teraction, anisotropy, and magnetoelastic coupling and
 can vary strongly between different materials systems as
 discussed below.

A. α -Fe₂O₃

A promising material candidate for antiferromagnetic
 spintronics is the well-known insulating AFM α -Fe₂O₃
 (hematite) in which recently long-distance transport of
 spin-information was observed (see section VB). α -
 Fe₂O₃ exhibits a range of exciting properties such as
 the Dzyaloshinskii-Moriya Interaction (DMI) leading to
 the Morin transition⁶⁸: at low temperatures the inter-
 play between anisotropy and DMI favors an uniaxial
 anisotropy with the Néel vector oriented along the c-axis,
 while above the Morin transition (260 K) the canting of
 the two sublattices favored by the DMI leads to a weak
 magnetic moment with the Néel vector perpendicular to
 the c-axis⁶⁹. In this canted moment state, the antiferro-
 magnetic domains can be controlled by modest magnetic
 fields, leading to large single domain states⁷⁰. The Morin
 transition and the anisotropies can be tailored by doping
 and a strong dependence on the structure and the result-
 ing direction of the easy axis has been observed^{71,72}.

To study the magnetic properties, early experiments
 on bulk α -Fe₂O₃ were carried out using neutron scat-
 tering^{73,74}. Using electrical methods based on spin Hall
 magnetoresistance (see section VA), the anisotropies,
 DMI and the spin-flop have been determined as a func-
 tion of temperature⁷⁵. To understand the spin dynam-
 ics properties, additionally, antiferromagnetic resonance
 measurements⁷⁶ and spin pumping measurements⁷⁷ were
 carried out, and a low magnetic damping was estab-

251 lished⁷⁶.



252 FIG. 3. XMLD-PEEM images of the domain structures of (a) 100 nm thick (0001) α -Fe₂O₃ at 100 K and (b) 100 nm thick (1 $\bar{1}$ 02) α -Fe₂O₃ at 100 K, the scale bar corresponds to 2 μ m. (c) Magneto-optical Kerr microscopy image of a bulk (1 $\bar{1}$ 02) bulk crystal at room temperature showing large domains. (d) A spin structure with the topology of an antiferromagnetic antiskyrmion found in (1 $\bar{1}$ 02) α -Fe₂O₃. (a-b) Reprinted with permission from Ref. 78. Copyright 2022 American Chemical Society. (d) from supplementary information of Ref. 71.

252 The spin structure in α -Fe₂O₃ can be ascertained by
 253 magnetic imaging. In bulk α -Fe₂O₃ crystals, large do-
 254 mains are observed, and macroscopically large areas of
 255 single domain can be found⁷⁹. More interesting are thin
 256 films, where the growth-strain can generate additional
 257 domain configurations. As seen in Fig. 3, very different
 258 domain structures exist. In (0001) α -Fe₂O₃ small do-
 259 mains are observed below the Morin transition (Fig. 3
 260 (a)), while in (1 $\bar{1}$ 02) films, larger domains with clear do-
 261 main walls are found (b). In the easy-plane phase above
 262 the Morin transition also multi-domain states are ob-
 263 served in thin films, while large domains can generally
 264 be stabilized in bulk crystals (Fig. 3 (c)). The transition
 265 from the easy-axis phase to the easy-plane phase that
 266 occurs in the Morin transition also leads to a transition
 267 from 180° to 60° domain walls, given the six-fold in-plane
 268 symmetry.

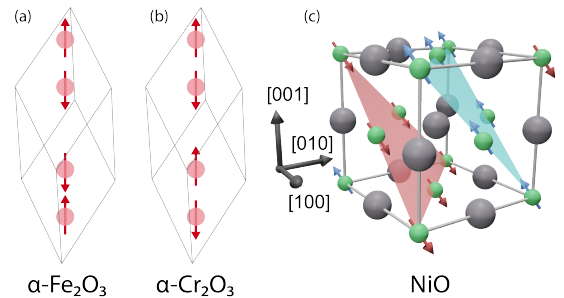
269 Finally, α -Fe₂O₃ also exhibits topologically non-trivial
 270 spin structures (Fig. 3 (d)), which is surprising since
 271 the symmetry of α -Fe₂O₃ does not allow for Lifshitz-
 272 Invariants that could stabilize a chiral magnetic struc-
 273 ture. The antiskyrmion-like structures observed in α -
 274 Fe₂O₃ are likely rather domain structures stabilized by
 275 local pinning sites^{41,71,80}.

276 B. α -Cr₂O₃

277 Similar to α -Fe₂O₃, the α -Cr₂O₃^{81,82} adopts a corun-
 278 dum structure and is an insulating collinear AFM below
 279 the Néel temperature ($T_N = 308$ K⁸³). The crucial differ-
 280 ence between the two isomorphous α -Fe₂O₃ and α -Cr₂O₃
 281 lies in the arrangement of the spins along the [111] axis as
 282 shown in Fig. 4 (a) and (b)⁸⁴. The symmetry of the mag-
 283 netic crystal also gives rise to the linear magnetoelectric
 284 effect in Cr₂O₃^{66,85,86}.

285 Due to the the easy-axis anisotropy in Cr₂O₃, there
 286 are two types of domains separated by 180° domain walls
 287 which makes the detection of the domain structure chal-
 288 lenging. The first experimental observation of the do-
 289 main structure was achieved via second-harmonic gen-
 290 eration⁸⁷. The equilibrium boundary magnetization of
 291 magnetoelectric AFMs⁸⁸ leads to the formation of sur-
 292 face magnetization domains due to uncompensated mag-
 293 netic moments at the interface. These domains have been
 294 imaged by XMCD, magnetic force microscopy, and scan-
 295 ning NV magnetometry^{50,89}, revealing the existence of
 both Bloch and Néel type domain walls⁵¹. In addition to
 the magnetic imaging technique, the antiferromagnetic
 order has also been studied electrically in Cr₂O₃/ heavy
 metal heterostructures⁹⁰⁻⁹².

Recent work has demonstrated the high potential of
 Cr₂O₃-based devices for a wide variety of further re-
 search, ranging from spin superfluidity to novel memory
 technologies^{50,93,94}.



296 FIG. 4. Spin structure of the AFMs (a) α -Fe₂O₃, (b) α -
 297 Cr₂O₃, and (c) NiO.

298 C. NiO

299 NiO was possibly the first material in which anti-
 300 ferromagnetic domains could be observed directly^{19,20}
 301 and this material has been used as an insulating
 302 spacer to observe tunnel-magneto resistance in a
 303 reproducible manner⁹⁵. The high Néel temperature of
 304 $T_N = 523$ K⁹⁶ in the bulk makes NiO an attractive
 305 candidate for investigating antiferromagnetic domain
 306 structures and antiferromagnetic spintronics. Below its
 307 Néel temperature, NiO adopts a type-II antiferromag-
 308 netic order: the spins are antiferromagnetically
 309 coupled between

the $\{111\}$ planes due to superexchange⁹⁷ and ferromagnetically coupled inside the $\{111\}$ planes⁹⁸ as shown in Fig.4 (c). The exchange striction between the antiferromagnetically coupled planes leads to a contraction between the $\{111\}$ planes and a temperature dependent^{99,100} rhombohedral distortion of the cubic crystal^{101–104}. The symmetry of the original fcc lattice allows four possible combinations of antiferromagnetically coupled $\{111\}$ planes, which are each associated with a different strain. These domains are considered to be Twin domains (T-domains)²⁰. An additional smaller anisotropy due to dipolar interactions leads to a threefold set of preferential spin orientations within the ferromagnetic planes along the $[112]$ directions, the so-called Spin domains (S-Domains)^{20,27,105,106}. If one additionally considers 180° domains, we arrive at a total of 24 possible domain configurations in the NiO bulk system.

The impact of strain on the antiferromagnetic domain structure has been studied extensively in NiO^{19,20,27,106–113}. In particular, external strain such as strain during cleaving, polishing, sample glue, or handling with tweezers can easily manipulate the antiferromagnetic domains^{27,114–117}. Furthermore, the domain structure of both, bulk and thin film NiO samples is influenced by the growth conditions such as growth temperatures^{118,119}, oxygen pressure¹²⁰, and film thickness^{36,395}. Importantly, the strain resulting from the lattice mismatch with the substrate plays a significant role for the anisotropy^{119,121}. Large domain sizes can be achieved by annealing^{19,122}. As a result, it is crucial to study the domain structure of NiO thin films by an imaging technique like XMLD-PEEM, to understand observations in current-induced switching experiments^{123,124}.

D. CoO

First neutron diffraction data has indicated a similar magnetic structure for CoO as for NiO¹⁰². However, in contrast to NiO, the orbital moment in CoO is not quenched and CoO exhibits a strong spin-orbit coupling, which affects the orientation of the spins^{102,125,126}. Investigations of the crystallographic structure of CoO revealed a tetragonal distortion of the fcc lattice below the Néel temperature (290°K) in contrast to the rhombohedral distortion in NiO or MnO^{127–129}. Neutron diffraction measurements could not provide clear information on the spin structure of CoO and different models were proposed¹³⁰. Initially, a collinear spin structure was assumed in which the spins are ferromagnetically coupled inside the $\{111\}$ planes^{102,131–133}. However, the tetragonal symmetry of the crystal led to the proposal of a noncollinear spin structure in a multi-spin-axis model^{133–135}. Further studies revealed that the tetragonal deformation can occur in a collinear spin alignment and is additionally accompanied by a monoclinic deformation^{125,136,137}. After the experimental observation of the additional monoclinic distortion, the magnetic struc-

ture of CoO is widely accepted to be collinear^{130,137–141}. However, we note that recent studies have again proposed a noncollinear structure for CoO^{142–144}. In the collinear structure, the spins of CoO are coupled ferromagnetically inside the $\{111\}$ planes and antiferromagnetically between the planes. The tetragonal distortion of CoO along one of the three cubic axes leads to the possible formation of three different twin domains, compared to four different twin domains in NiO⁶¹. Large domains in CoO crystals can be obtained by annealing the crystals above the Néel temperature^{137,139,145,146} and the application of external strain can lead to the preferential stabilization of certain domains¹³⁹. Similar to NiO, the magnetic structure of CoO depends on the growth parameters and on the substrate-induced strain^{61,147}. In thin films, compressive strain favors an in-plane alignment of the magnetic moments^{148,149} and tensile strain leads to an out-of-plane alignment^{148,149}. For CoO grown on MgO(001) the spins align in the plane of the film¹⁵⁰ and two in-plane magnetic easy axes along $[110]$ or $[\bar{1}10]$ are present^{151,152}. Birefringence imaging can be used to image the domains of thin-films^{29,33} and second-order magneto-optical effects can also be used to study the magnetization dynamics of CoO thin films¹⁵³. CoO is often considered to be similar to NiO, however, the unquenched orbital moment leads to significant differences in its crystallographic structure, magnetic properties¹⁵⁴, and rich electrical properties¹⁵⁵.

IV. ANTIFERROMAGNETIC SHAPE ANISOTROPY

In ferromagnets shape anisotropy due to stray fields has long been a key tool to tailor device properties. In AFMs this effect is absent due to the lack of a net magnetic moment. Nevertheless, it has been predicted that in AFMs with a strong magnetoelastic coupling, strains can lead to antiferromagnetic shape-anisotropy^{156,157}. First investigations of this effect were carried out on antiferromagnetic LaFeO₃ and local changes of the domain structure near the edge of patterned devices were observed^{158–161}. However, recent investigations on NiO have revealed that the patterning of devices can lead to two different effects. In devices patterned along the projection of the easy axes of NiO, local changes of the surface anisotropy lead to a preferential stabilization of certain domains in the vicinity of the patterned edge. Each domain is associated with a different deformation of the crystallographic structure due to the strong magnetoelastic coupling. Thus, long-range magnetoelastic strains which originate from the domain formation determine the equilibrium domain structure in the center of the device. The equilibrium domain structure depends here on the interplay between multiple effects such as the destressing of the domains, the coupling with the non-magnetic substrate and the energy of the domain walls¹⁶². One way to control the antiferromagnetic ground state is to vary the aspect ratio of the patterned elements, see Fig. 5¹⁶².

Therefore, antiferromagnetic shape anisotropy is a powerful tool to engineer antiferromagnetic domain structure in devices and is not limited to NiO, but can be extended to other AFMs with strong magnetoelastic coupling such as CoO or α -Fe₂O₃.

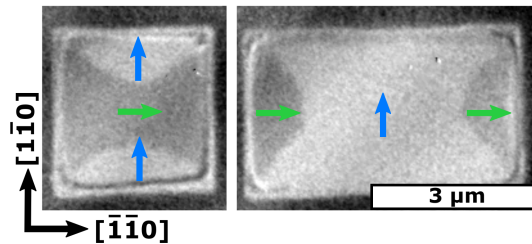


FIG. 5. XMLD-PEEM image of the domain structure in patterned NiO/Pt bilayer devices after annealing. The arrows indicate the direction of the inplane projection of the Néel vector, adapted with permission from Meer et al., Phys. Rev. B 106, 094430 (2022). Copyright 2022 American Physical Society. Appl.

V. CURRENT-INDUCED EFFECTS

A. Electrical detection

Electrical detection of the antiferromagnetic order is one of the main building blocks for realizing antiferromagnet-based memory technologies. For insulating AFMs, electrical reading of the magnetic state is commonly achieved in antiferromagnet/heavy-metal bilayer structures making use of the spin Hall magnetoresistance (SMR)^{163,164}. A charge current in a heavy-metal creates a spin current flowing toward the interfaces due to the spin Hall effect. The interfacial interaction of the spins in the heavy-metal with the magnetic insulator depends on the relative orientation of the spins and the magnetic order. The interfacial exchange of angular momentum can be detected electrically via the inverse spin Hall effect in the heavy-metal. This gives rise to the characteristic $\sin(2\theta)$ dependence of the SMR on the angle θ between the charge current direction and the magnetic order. In contrast to ferromagnets, the perpendicular alignment of the Néel vector with respect to the external magnetic field results in a negative sign of the SMR signal for AFMs^{75,165–169}. Given the simple planar device structure required for electrical detection, SMR has proven to be a powerful tool for probing magnetic properties and enables the integration of AFMs in the next-generation computing technologies.

B. Spin Transport

Many new ideas regarding the generation and transport of spin-currents in antiferromagnetic materials have

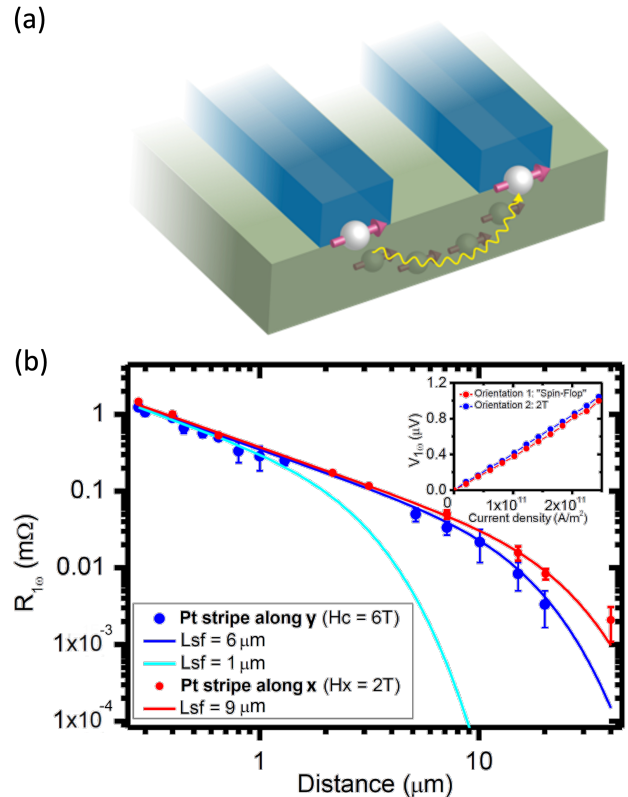


FIG. 6. (a) Schematic of non-local spin transport. Spins are injected by the spin Hall effect from a heavy metal wire (left blue injection wire) as a magnonic spin current into the antiferromagnetic insulator (gray), where they propagate to a second heavy metal wire (right blue detection wire). There, the spin current is absorbed and detected electrically by the inverse spin Hall effect. (b) Measured non-local resistance as a function of distance in a bulk α -Fe₂O₃ (1120) crystal, showing spin transport over micrometer distances. The inset shows a linear dependence of the signal on the current density with no threshold, indicating diffusive transport. (b) adapted with permission from Lebrun et al., Nature 561, 222–225 (2018). Copyright 2018 Springer Nature Limited.

been generated in the last few years, but most of them remained at the stage of predictions. Conventional experimental work has so far mainly consisted of studying AFMs in coupled AFM/FM systems^{170–173} in which the antiferromagnetic properties are deduced from a detection in the adjacent FM, as for instance realized in a magnon spin-valve device¹⁷⁴. The transport perpendicular to the plane across the layer thickness in antiferromagnetic insulators with adjacent ferro-(ferri-)magnetic layers has typically yielded only very short spin transport length scales of a few to a few tens of nm^{171,175–177}. To study the pure AFM transport without other magnetic layers involved, non-local transport (see Fig. 6 (a)) is a suitable technique. Recently, some of the longest known spin transport length scales were shown for low damping antiferromagnetic insulators like YFeO₃¹⁷⁸, Cr₂O₃⁹³ and

475 α -Fe₂O₃ in the easy axis phase (Fig. 6 (b))¹⁷⁹. While spin⁵³⁰
 476 transport is expected in the easy-axis phase due to the⁵³¹
 477 circular polarization of the magnons, lateral spin trans-⁵³²
 478 port has also been demonstrated in the easy-plane phase
 479 of α -Fe₂O₃. Fundamentally, no magnonic spin transport
 480 is expected because the magnons in easy-plane AFMs
 481 are in the simplest description linearly polarized. How-
 482 ever, pairs of linearly polarized magnons with different
 483 k-vectors but the same energy can be combined into cir-
 484 cularly polarized magnons, leading to magnons with a
 485 certain degree of circular polarization and thus transport
 486 of spin angular momentum^{76,180,181}. This means that in
 487 easy-plane and orthorhombic AFMs, one can electrically
 488 generate pairs of linearly polarized spin waves, which
 489 carry an effective circular polarization and thus spin in-
 490 formation⁷¹.

491 Finally, by linking the spin structures to the transport,
 492 it has been found that the domain structure significantly
 493 impacts the length scale of spin transport. The domain
 494 walls act as efficient spin scatterers and thus strongly
 495 governing the spin transport^{71,78}.

496 C. Electrical manipulation

497 In addition to reading, writing AFMs efficiently is an⁵³⁵
 498 actual key open goal of the field. One possible approach⁵³⁶
 499 relies on Néel spin-orbit torques, creating a staggered⁵³⁷
 500 effective field of opposite sign on each magnetic sub-⁵³⁸
 501 lattice for special crystallographic compounds, as stud-⁵³⁹
 502 ied in the particular metallic compounds CuMnAs and⁵⁴⁰
 503 Mn₂Au^{13,182}. An approach, applicable to a broader⁵⁴¹
 504 range of AFMs including the 3d oxide materials reviewed⁵⁴²
 505 here, is to use the non-staggered, antidamping-like torques⁵⁴³
 506 exerted by a spin accumulation at the interface of a heavy
 507 metal and an AFM-insulator due to a charge current in
 508 the heavy metal^{183,184}. This mechanism leads to a dis-⁵⁴⁴
 509 placement of domain walls that can potentially be very
 510 fast due to the absence of a Walker breakdown phe-⁵⁴⁵
 511 nomenon¹⁸⁴. Here, the direction of the wall motion is⁵⁴⁶
 512 governed by their sense of rotation. If there are no Lif-⁵⁴⁷
 513 shitz - invariants that break the inversion symmetry and⁵⁴⁸
 514 lead to a chiral spiralization of the AFM domain walls,⁵⁴⁹
 515 this means that different domain walls move in different⁵⁵⁰
 516 directions and no overall switching is achieved. In par-⁵⁵¹
 517 ticular, with the recent observation that domain walls⁵⁵²
 518 in NiO/Pt are not chiral¹²⁴, this mechanism would be⁵⁵³
 519 difficult to use for devices. Another mechanism is a pon-⁵⁵⁴
 520 deromotive force effect that has been predicted to yield⁵⁵⁵
 521 deterministic 90° domain wall motion¹⁸⁵.⁵⁵⁶

522 Experimentally, the possibility of switching was⁵⁵⁷
 523 demonstrated in NiO/Pt^{30,185-188} and also in het-⁵⁵⁸
 524 erostructures of CoO/Pt¹⁵¹ and α -Fe₂O₃/Pt^{189,190}. As⁵⁵⁹
 525 contradicting directions of the switching were reported,⁵⁶⁰
 526 other effects beyond spin orbit torques had to be in-⁵⁶¹
 527 voked. In particular, by comparing direct imaging and⁵⁶²
 528 transport data, it was found that some of the transport-⁵⁶³
 529 based detection suffered from artifacts due to electromi-⁵⁶⁴

gration. Resistances changed due to structural changes
 of the material, rather than switching of the magnetiza-
 tion^{30,151,191-193}.

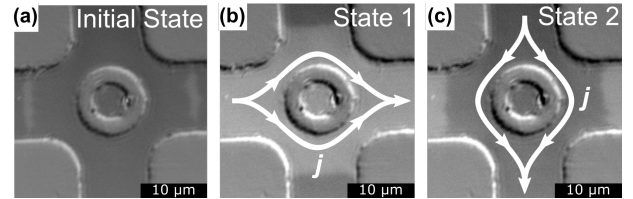


FIG. 7. (a) Birefringence image of the domain structure in a NiO/Pt bilayer device used for current-induced switching experiments before any current application. The circular area in the center of the cross is electrically isolated from the rest of the device. (b) When a current is applied to the device, the current flows around the dot. Nevertheless, after the current pulse has been applied, the domain inside the electrically isolated part is switched. (c) The application of an orthogonal pulse leads to orthogonal heat-induced strain and the dot can be switched back. Adapted with permission from Meer et al., Nano Lett. 21, 114–119 (2021). Copyright 2022 American Chemical Society.

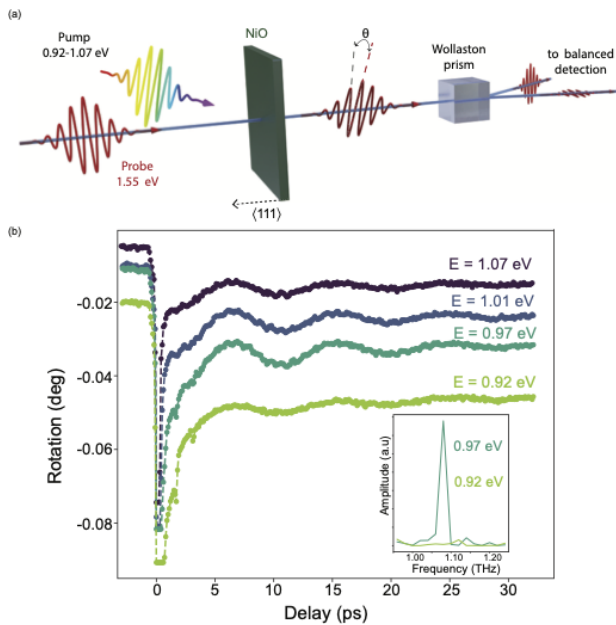
Furthermore, the magnetic switching observed by direct imaging also turned out to comprise contradicting directions of the switching, indicating that not only spin-orbit torques are involved. In a careful analysis, it was shown that localized heating leading to spatially varying strain induces switching by magneto-elastic coupling that depends strongly on the device geometry^{32,151,194}. As shown in Fig. 7, switching can even be achieved in areas where no current is flowing, indicating that the governing mechanism in NiO/Pt bilayers studied here is not due to spin-orbit torques.

VI. LIGHT-INDUCED EFFECTS

While the current-induced torques enable switching between different equilibrium antiferromagnetic states, the fast intrinsic dynamics of AFMs can be accessed effectively by optical pulses. One of the most promising applications of AFM-based devices is the optical generation of THz radiation induced by oscillations of the Néel vector.

Light-induced excitation of the magnetic dynamics can be governed by different mechanisms. Direct excitation via magneto-optical effects (inverse Faraday effect, Cotton-Mouton effect) induces coherent rotation of the Néel vectors. It occurs on short (subpicosecond) time scales and is sensitive to the polarization of the light. According to first-principle calculations¹⁹⁵ and atomistic simulations¹⁹⁶, the polarization should ideally be circular. Experimentally, optically-induced coherent magnetic dynamics and generation of THz magnons were observed in NiO¹⁹⁷⁻²⁰¹ and in Cr₂O₃^{197,198}. However, magnon dynamics were also induced by linearly polarized pulses²⁰⁰, which was attributed to the multi-domain structure of

565 NiO²⁰². A less explored pathway to light-induced genera-592
 566 tion of coherent magnons involves excitation-magnon tran-593
 567 sitions. Using this technique, the ultrafast spin dynamics594
 568 of multi-domain NiO was explored²⁰³ (see Fig. 8).



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All-optical manipulation of antiferromagnetic state was demonstrated both in single crystals^{197–199,203,204,208} and thin films with a heavy metal layer^{200,201,205,206}. In the latter case, the heavy metal enables magnon-to-light conversion via inverse spin Hall effect and re-emission of optical pulses.

Both the electrical and optical manipulation of antiferromagnetic states involve magnetic dynamics that can be excited either by spin torques or by varying the magnetic anisotropy landscape. Spin torques can be excited in insulators, by interfacial spin exchange via spin Hall effect^{75–77}, by spin-transfer torque from optically induced spin polarized photoelectrons²⁰⁹, or via the inverse Faraday effect¹⁹⁹. The first two effects are interfacial and are efficient in thin films. In 3d metal oxides with strong magnetoelastic coupling, varying the magnetic anisotropy by stress or strain is also an efficient tool for spin switching. Similarly to current-induced switching, laser-induced heating of heterostructures creates additional shear strains that set preferable orientation of the magnetic vectors and induce switching. Remarkably, in addition to quasi static switching³², such thermo-magnetoelastic effects in NiO/Pt bilayers were also observed in the THz dynamics²⁰⁶. Nevertheless, in real devices, both switching mechanisms, spin-assisted (by spin torques) and non-spin-assisted (by strain), co-exist. Proper tailoring of the sample geometry can optimize the combination of both mechanisms and, thus, the switching.

VII. OUTLOOK

Insulating 3d metal oxides are a promising class of materials for future antiferromagnetic devices, offering a variety of approaches for controlling and reading the antiferromagnetic order. A key advantage over their metallic counterparts is the observed long distance spin transport in these antiferromagnetic insulators^{178,179}. Transporting information without transporting charge carriers is a key aspect of insulating spintronics and builds the foundation for future energy-efficient spintronic devices.

Due to the pronounced magnetoelastic coupling in AFMs, the domain structure is very sensitive to strain. As a result, strain can be used to tailor device properties during the growth or by strain-induced shape anisotropy. This static approach known as “straintronics” allows for the design of energy-efficient devices^{210,211}. Dynamic manipulation of the Néel order via current-induced switching is based on two competing mechanisms, spin-orbit torque-based switching and switching due to current-induced heat and strain. Optically induced switching and detection of the antiferromagnetic order extends the study of insulating transition metal oxides into the ultrafast regime, making them an attractive candidate for THz emitters due to their resonance frequencies in the THz range¹¹.

Another area in which antiferromagnetic 3d metal ox-

ides could play a crucial role is the emerging field of “orbital bitronics”, where the orbital angular momentum is used to control the magnetic ordering²¹². Comparing 3d metal oxides with a quenched orbital moment, such as NiO, and strong orbital moment, like CoO, could be used to study the influence of the orbital moment.

In recent years, several key concepts for the control of the antiferromagnetic ordering in 3d metal oxides have been developed. Today, a major challenge is the exploitation of the combination of these fundamental phenomena in spintronic devices. Overall, the extensive research into the fundamental properties of transition metal oxides and the recent advances in the reading and writing of their antiferromagnetic ordering in thin films make them promising candidates for the exploration of novel concepts in future antiferromagnetic spintronic devices.

VIII. AUTHOR DECLARATION

A. Conflict of Interest

The authors have no conflicts to disclose.

B. Contributions

All authors contributed equally to this work.

C. Data Availability

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

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