

Switching of magnetic domains in a noncollinear antiferromagnet at the nanoscale

Atul Pandey ^{1,2} Prajwal Rigvedi ¹ Edouard Lesne ³ Jitil Deka ¹ Jiho Yoon ¹ Wolfgang Hoppe ² Chris Koerner ²
Banabir Pal ¹ James M. Taylor ² Stuart S. P. Parkin ¹ and Georg Woltersdorf ^{1,2,*}

¹Max Planck Institute of Microstructure Physics, Weinberg 2, 06120 Halle, Germany

²Institute of Physics, Martin Luther University Halle-Wittenberg, Von-Danckelmann-Platz 3, 06120 Halle, Germany

³Max-Planck-Institute for Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany



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Antiferromagnets that display very small stray magnetic fields are ideal for spintronic applications. Of particular interest are noncollinear, chiral antiferromagnets of the type Mn_3X ($\text{X} = \text{Sn}, \text{Ge}$), which display a large magnetotransport response that is correlated with their antiferromagnetic ordering. The ability to read out and manipulate this ordering is crucial for their integration into spintronic devices. These materials exhibit a tiny unbalanced magnetic moment such that a large external magnetic field can, in principle, be used to set the material into a single antiferromagnetic domain. However, in thin films of Mn_3Sn , we find that such fields induce only a partial magnetic ordering. By detecting two orthogonal in-plane components of the magnetic order vector, we can demonstrate switching along multiple easy axes in Mn_3Sn and find that the nonswitchable fraction has a unidirectional anisotropy. Studying the switching at the nanoscale allows us to correlate the pinning behavior to crystal grain boundaries in the Mn_3Sn nanowire structures.

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Antiferromagnetic materials are promising candidates for high-performance spintronic devices due to their fast dynamics, stability against external perturbations, and ability to miniaturize components due to the lack of stray fields [1–4]. Of particular interest are antiferromagnetic materials with topologically protected bandstructures [5,6]. These include noncollinear antiferromagnets (NCAFs). Here, triangular spin textures with an inherent chirality [7,8] lead to the emergence of Weyl points in their bandstructure [9,10]. These, in turn, act as a strong internal effective magnetic field [11,12]. The Berry curvature associated with such Weyl points leads NCAFs to exhibit magnetotransport and magneto-optical effects analogous to conventional ferromagnetic materials. These include the anomalous Hall effect [13], anomalous Nernst effect (ANE) [14], magneto-optical Kerr effect (MOKE) [15–17], and x-ray magnetic circular dichroism [18,19].

The behavior of the magnetic order in response to external stimuli, for example, magnetic fields or spin torques, can be understood in terms of a magnetic octupole order parameter (MOOP) [19–21]. This is defined by a vector given by the sense of rotation of the chiral spin texture within a kagome plane. The advantage of NCAFs is that magnetic octupole-driven electrical or optical readout signals are much larger than those obtained from collinear antiferromagnetic materials [13,14,22]. Being able to switch the orientation of the

MOOP in NCAFs and read its state electrically or optically brings such devices closer to realization. However, to properly understand the switching mechanisms at play, it is important to study the dynamics and magnetic domain structures in NCAFs. Specifically, it is required to image their antiferromagnetic domains, which correspond to regions of triangular spin texture with the same chiral sense of rotation but oriented along one of three symmetry related directions in the hexagonal plane. Because these domains are typically very small [15,23,24], imaging must be performed with nanometer resolution. Previously, scanning ANE measurements have been performed to observe magnetic field-induced switching of the MOOP vector in NCAFs at micrometer scales [25,26]. This is achieved by using a focused laser beam to create a localized temperature gradient ∇T . This gradient generates an electrical ANE response $\mathbf{E}_{\text{ANE}}$ transverse to both ∇T and the MOOP vector.

We perform spatially resolved ANE measurements to study the spatial distribution of the MOOP in an NCAF thin film of Mn_3Sn . By detecting two orthogonal components of $\mathbf{E}_{\text{ANE}}$, we visualize switching of the MOOP into arbitrary in-plane directions of Mn_3Sn on a submicron length scale. Since the spatial resolution is not sufficient to fully understand the inhomogeneous switching behavior, we have extended the method, to map the magnetic domains with nanoscale spatial resolution. We achieve this by focusing a laser beam onto the apex of an atomic force microscope (AFM) tip to create a heat gradient on the nanometer scale, allowing us to correlate the inhomogeneous switching behavior of the NCAF to its grain structure.

Here, we investigate a 60 nm thick Mn_3Sn film grown using magnetron sputtering on an MgO (111) substrate [27]. X-ray diffraction 2θ scan reveals c -axis orientation of the film [27], which leads to a negligible OOP component of the

*Contact author: georg.woltersdorf@physik.uni-halle.de

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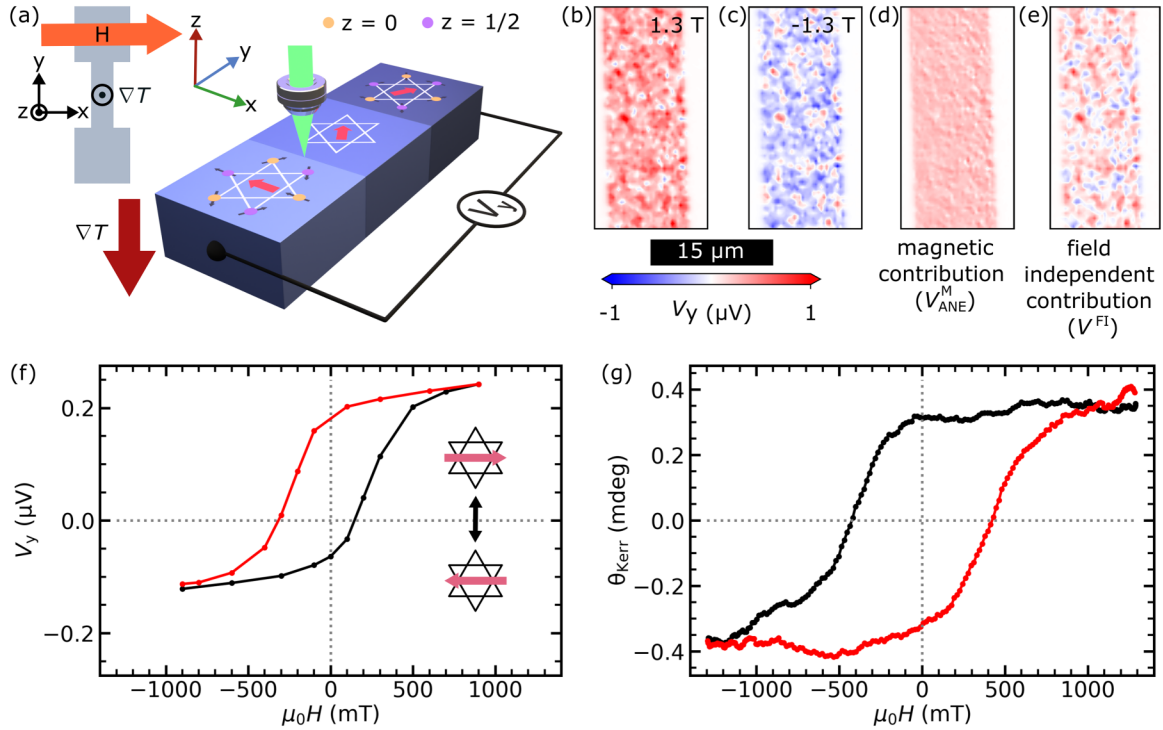


FIG. 1. (a) Schematic showing the device structure in the upper left and an illustration of the ANE microscope on the right. The laser beam with a power of 1 mW and a wavelength of 532 nm is focused on to the sample using an objective lens with a numerical aperture of 0.7 to create an out-of-plane (OOP) gradient ∇T (vertical red arrow). This generates an ANE voltage transverse to the temperature gradient and the MOOP vector (indicated by arrows on the slab). A multidomain state in the wire is indicated by three different orientations of the MOOP vector, and the Mn₃Sn spin structures for two of these domains are shown with Mn atoms at $z = 0$ and $z = 1/2$ positions in the unit cell. (b), (c) ANE microscope image of a Mn₃Sn wire structure that has been field cooled in ± 1.3 T, respectively. (d) V_y in panel (c) is subtracted pixel by pixel from that in panel (b) and divided by 2 to obtain the map of V_{ANE}^M . (e) Map of V^{FI} obtained by averaging V_y in panels (b) and (c) pixel by pixel. (f) ANE-based hysteresis loop measured at 125 °C. The inset schematically indicates the effect of the magnetic field on the magnetic order parameter. (g) Hysteresis at 125 °C obtained by longitudinal MOKE. As discussed in the Supplement Material [27], the nonswitchable background is already subtracted here.

MOOP vector ($M_z \approx 0$). The film is patterned into a wire of width $w = 10 \mu m$, as shown schematically in Fig. 1(a). In the first step, the wire structure is illuminated with a focused laser beam to study the switching behavior at the microscale. This creates a temperature gradient ∇T . The resulting ANE-induced electrical response is given by

$$\mathbf{E}_{ANE} = \mu_0 S_{ANE} \mathbf{M} \times \nabla T, \quad (1)$$

where S_{ANE} is the ANE coefficient, $\mathbf{M} = M_x \hat{x} + M_y \hat{y}$ is the in-plane (IP) polarized MOOP unit vector, and μ_0 is the permeability constant. The IP component of $\mathbf{E}_{ANE}$ is given by the following:

$$\mathbf{E}_{ANE}^{IP} = \mu_0 S_{ANE} M_y \frac{\partial}{\partial z} T \hat{x} - \mu_0 S_{ANE} M_x \frac{\partial}{\partial z} T \hat{y}. \quad (2)$$

We measure the y component of $\mathbf{E}_{ANE}^{IP}$ ($E_{ANE}^y = -\mu_0 S_{ANE} M_x \frac{\partial}{\partial z} T$) by detecting the voltage in a wire oriented along the y direction V_y . The spatially resolved measurement of V_y allows us to create a map of the x component M_x .

First, we consider the case where the laser beam is focused using a microscope objective. We switch the MOOP vector from $+\hat{x}$ to $-\hat{x}$, as shown schematically in the inset of Fig. 1(f), by applying a magnetic field ($\mu_0 H$) along the positive or negative x directions at an elevated temperature (125 °C). At this temperature, the coercivity is reduced, so that

a ± 1.3 T magnetic field is sufficient to saturate the MOOP. The effect of temperature and the applied magnetic field on the switching behavior is shown in the Supplemental Material [27]. The Mn₃Sn device is then cooled to room temperature in the presence of the magnetic field, where spatially resolved ANE measurements are performed (thus avoiding temperature-induced drift). The spatial distribution of V_y is shown in Figs. 1(b) and 1(c). When the device is cooled in the presence of 1.3 or -1.3 T, the majority of regions show positive or negative signs of the measured voltage, respectively. This sign change corresponds to the reversal of the ANE signal because of the field-induced switching of the MOOP vector.

In general, V_y may have a contribution that can be switched with the magnetic octupole moment and nonswitchable contributions related to the Seebeck effect caused by inhomogeneities. Indeed, in our experiments we find small areas where the signal polarity is insensitive to the field. We attribute these to domain pinning or grain boundaries. Field-induced switching is favored by coupling the net moment with the magnetic field, and magnetocrystalline magnetic anisotropy hinders it. Regions with a strong anisotropy or a negligible net moment, that cannot be switched by a 1.3 T field, would form pinned magnetic domains. Domain pinning results in a field-independent ANE voltage (V_{ANE}^{FI}), while

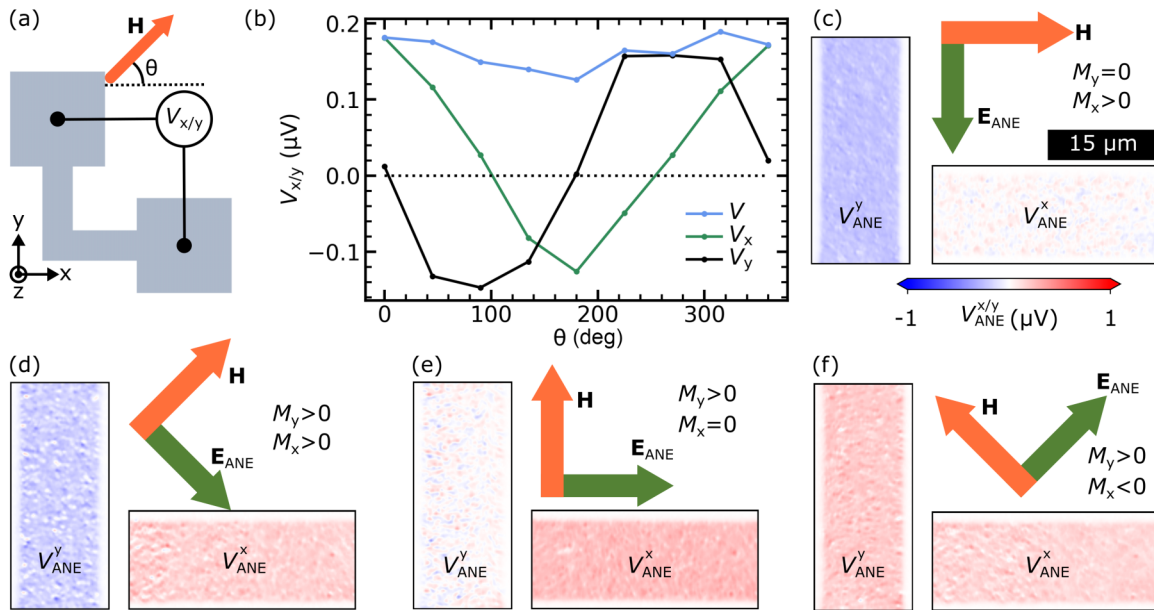


FIG. 2. (a) Schematic of the device for angle-dependent field measurements. $V_{x/y}$ indicates the voltage picked up by the lock-in amplifier while the horizontal/vertical arm of the device is illuminated with the laser beam. (b) Average voltage signal measured in the horizontal (V_x) and vertical arms (V_y) of the device against the in-plane field angle θ . $\theta = 90^\circ$ corresponds to the positive y direction while $\theta = 0^\circ$ corresponds to the positive x direction. (c)–(f) ANE microscopy domain images for different in-plane field angles, as indicated by the arrows: (c) $\theta = 0^\circ$, (d) $\theta = 45^\circ$, (e) $\theta = 90^\circ$, and (f) $\theta = 135^\circ$. The left panels show scans of a $15\text{ }\mu\text{m} \times 30\text{ }\mu\text{m}$ area in the vertical arm, while the right panels show scans of a $30\text{ }\mu\text{m} \times 15\text{ }\mu\text{m}$ area in the horizontal arm.

grain boundaries result in a nonmagnetic Seebeck effect (SE) voltage (V_{SE}^{NM}) [28], as shown systematically in the Supplemental Material [27]. The magnetic field-dependent ANE signal (V_{ANE}^M) can be separated by subtracting two images where the MOOP is field polarized in opposite directions [Fig. 1(d)]. Similarly, by adding these images, we obtain a spatial map of field-independent contributions. The resulting map is shown in Fig. 1(e) and reveals that there is a significant bipolar field-independent contribution ($V^{FI} = V_{ANE}^{FI} + V_{SE}^{NM}$) that is present throughout the wire structure. The magnetic field dependence of the MOOP reversal is further studied by recording the hysteresis loop from using the ANE signal. Figure 1(f) shows the average V_y measured over a $10\text{ }\mu\text{m} \times 15\text{ }\mu\text{m}$ area as a function of the applied magnetic field at a temperature of 125°C [27]. This demonstrates the field-induced switching of the magnetic order and is validated by comparing it with the hysteresis loop obtained using longitudinal MOKE [Fig. 1(g)]. We note, that the coercivity obtained with the ANE measurement in $150\text{ }\mu\text{m}^2$ region of the wire structure is smaller than that from the MOKE measurement of the film under a $\approx 500\text{ }\mu\text{m}$ wide laser focal spot. This might result from the inhomogeneity of the domain pinning. Furthermore, the high power density of the focused laser beam in the ANE measurement will increase the temperature of the Mn_3Sn wire by approximately 4 K. This estimate of temperature increase for the laser beam with a power of 1 mW is based on a COMSOL simulation, that shows about 19 K increase for a laser beam with a power of 5 mW [29]. The coercivity has a strong temperature dependence [27], where we find an increase of approximately 13 mT/K. Therefore, an even small laser heating-induced temperature increase of 4 K can reduce

the coercivity by up to 50 mT in the ANE-based hysteresis loop. We observe an offset in V_y in the ANE-based hysteresis loop. This results from the field-independent contributions, either through the nonmagnetic Seebeck effect V_{SE}^{NM} or as a consequence of V_{ANE}^{FI} due to partial field-induced switching. In the latter case, an offset will arise if the population of pinned domains with an MOOP vector along $+\hat{x}$ is higher relative to that along $-\hat{x}$.

To verify the presence of partial field-induced switching, we investigate the angular dependence of the measured signal. For this set of measurements, a device with two orthogonal arms was fabricated, as schematically shown in Fig. 2(a). The voltages measured in the horizontal (V_x) and vertical (V_y) arms allow us to deduce M_y and M_x , respectively [see Eq. (2)]. This configuration permits the measurement of both, V_x and V_y , using a single lock-in channel, as depicted in the schematic. When the vertical arm is illuminated, only the y component of the electrical response (V_y) is detected, whereas illumination of the horizontal arm provides V_x . Magnetic order is changed by applying a 1.3 T in-plane magnetic field along different angles θ at an elevated temperature of 125°C . Subsequently, we image the magnetic domains after field cooling to room temperature. Average values of V_x (V_y), which are directly proportional to the two components of the MOOP M_y (M_x), are plotted against θ in Fig. 2(b). A 90° phase shift between V_x and V_y confirms that they are related to the two orthogonal components of $\mathbf{M}$. Both show a 2π periodic behavior with θ , indicating that the MOOP vector switches along multiple directions. We show spatially resolved measurements of V_{ANE}^M in the horizontal (V_{ANE}^x) and the vertical (V_{ANE}^y) arms, that allow us to visualize two orthogonal components of the MOOP

vector. This is done by subtracting the nonmagnetic background from the spatially resolved measurements of V_x and V_y [27]. The nonmagnetic contributions in both the arms are mapped following the same procedure as in Fig. 1(f). When the magnetic field is applied along the positive x direction, the MOOP orients with the field, resulting in an electrical $\mathbf{E}_{\text{ANE}}$ response along $-\hat{y}$ direction [Fig. 2(c)]. This is indicated by a mostly negative value of V_{ANE}^y that is recorded in the vertical arm and a negligible value of V_{ANE}^x recorded in the horizontal arm. When the field is applied at an angle of 45° with respect to the x axis, we observe the electrical $\mathbf{E}_{\text{ANE}}$ response in the majority of the region being oriented at an angle of -45° with respect to the x axis, as revealed by the predominately blue (red) contrast in the vertical (horizontal) arms [Fig. 2(d)]. Applying the field along the $\hat{y}$ direction results in positive V_{ANE}^x and negligible V_{ANE}^y [Fig. 2(e)], while applying the field at 45° from the y axis results in positive V_{ANE}^x and V_{ANE}^y [Fig. 2(f)]. Furthermore, the absolute value of V_{ANE}^x (V_{ANE}^y) is highest when the field is applied along $\hat{y}$ ($\hat{x}$) direction. It is smaller by a factor of $\approx 1/\sqrt{2}$ when the field is applied at 45° with respect to $\hat{x}$ or $\hat{y}$ directions. Thus, we observe rotation of the MOOP vector following the angle of an applied IP magnetic field; we show this further by rotating the field in smaller steps of 20° [27]. These images demonstrate that the spatially averaged MOOP can be aligned along arbitrary in-plane directions by a magnetic field. As shown by x-ray diffraction pole scans of our arbitrary Mn_3Sn sample are hexagonal and well oriented and epitaxial growth [27]. Thus, one may expect them to show three magnetic easy axes along the $(\bar{2}110)$ directions [7,30,31]. Applying a magnetic field in the plane of the film should therefore orient the MOOP along the closest of the possible six easy directions.

However, if this field would achieve complete switching, the magnitude $|\mathbf{M}| = \sqrt{M_x^2 + M_y^2}$, and thus, the voltage $V \propto |\mathbf{M}|$ calculated by $V = \sqrt{V_x^2 + V_y^2}$ would be independent of θ . Instead, we observe a minimum in V , and therefore in the magnitude of $\mathbf{M}$, at $\theta = 180^\circ$ [Fig. 2(b)]. This indicates that the MOOP only partially aligns with the magnetic field and that this alignment is most favorable along the positive x direction.

In these images, we also observe the presence of domain structures smaller than the optical resolution of ≈ 700 nm of the ANE microscope [29]. To properly understand this switching process, we need to image magnetic domains at the nanoscale. This is achieved by applying a laser beam to the apex of a vibrating AFM tip. Here, the tip-sample interaction results in an enhanced optical near-field underneath the tip, creating a temperature gradient with a lateral spread on the nanometer length scale. The resulting ANE voltage is demodulated at the second harmonic of the tip frequency ($V_y^{2\omega}$), which corresponds purely to the voltage generated by the near-field-induced temperature gradient. We have previously demonstrated that this technique can image magnetic textures in ferromagnets with a spatial resolution of 75 nm [29]. Here, we apply the same technique to image magnetic domains in a c -axis-oriented 60 nm thick Mn_3Sn film patterned into a $300 \text{ nm} \times 10 \mu\text{m}$ nanowire.

By following the same measurement procedure used in Figs. 1(b)–1(e), we image the Mn_3Sn nanowires after positive

[Fig. 3(b)] and negative [Fig. 3(c)] field cooling. Subtracting and adding these images yields the magnetic [Fig. 3(d)] and field-independent parts [Fig. 3(e)] of V_y , respectively. The simultaneously measured AFM topography [Fig. 3(f)], provides insight into the structure and morphology of the thin film device. This allows us to correlate ANE-based magnetic domain images with the grains of the film at nanoscale.

We perform a higher-resolution measurement on a region shown by the dotted square in Fig. 3(f). The antiferromagnetic domain distribution after positive [Fig. 3(g)] and negative [Fig. 3(h)] field cooling is now imaged with a resolution of better than 100 nm.

Figs. 3(i) and 3(d) show that, at this length scale the magnetic contribution V_{ANE}^M is spatially nonuniform. This is caused by chiral domains with a sense of rotation. Some of which are pinned and cannot be switched by the applied field, supporting our previous observation of pinned domains affecting the MOOP switching process in Mn_3Sn . These pinned domains, in turn, show a higher magnitude of $V_{\text{ANE}}^{\text{FI}}$, as shown in Fig. 3(j). By comparing these images with the topographic map of the nanowire, as displayed in Fig. 3(k), we observe that the regions with large V^{FI} are correlated with positions of structural grains. We conclude, that the domains are pinned in place by crystalline grain boundaries in the thin film.

Our observations are consistent with the switching behavior of collinear antiferromagnets observed using high-resolution photoemission electron microscopy. Here, current-induced spin-orbit torque in CuMnAs [32] and Mn_2Au [33], as well as applied magnetic fields of up to 70 T in Mn_2Au [34], only induce partial switching.

Another recent study of magnetic domain images in Mn_3Sn nanowires by near-field-based ANE microscopy [28] also demonstrates magnetic field-dependent and field-independent contributions to the measured voltage signal. The field-independent contribution was attributed to a Seebeck effect-induced voltage at the grain boundaries. Our analysis of domain images together with AFM-based topography is consistent with this prior work. Furthermore, some regions were observed to show no magnetic contribution ($V_{\text{ANE}}^M = 0$). These regions were ascribed to an OOP polarization of the MOOP. However, this is inconsistent with our recent work on ANE imaging of OOP magnetized nanowires [29], where we show that the OOP magnetization results in zero ANE signal only if the magnetization underneath the heat gradient spot is uniform. Therefore, a nonuniform magnetization under the spot, for example, a domain wall or the edges of the nanowire, will result in a nonzero ANE signal due to the magnetization gradient [29,35]. For these reasons, we ascribe the regions without field-dependent signals V_{ANE}^M in Mn_3Sn to be a result of pinned domains.

In conclusion, we have imaged chiral antiferromagnetic domains in Mn_3Sn nanowires using near-field-based scanning ANE microscopy with nanoscale spatial resolution. By simultaneous observation of two orthogonal components of $\mathbf{E}_{\text{ANE}}$, we can track the orientation of the MOOP vector and demonstrate that the MOOP vector can be oriented along the in-plane magnetic field directions. Our nanoscale images reveal that

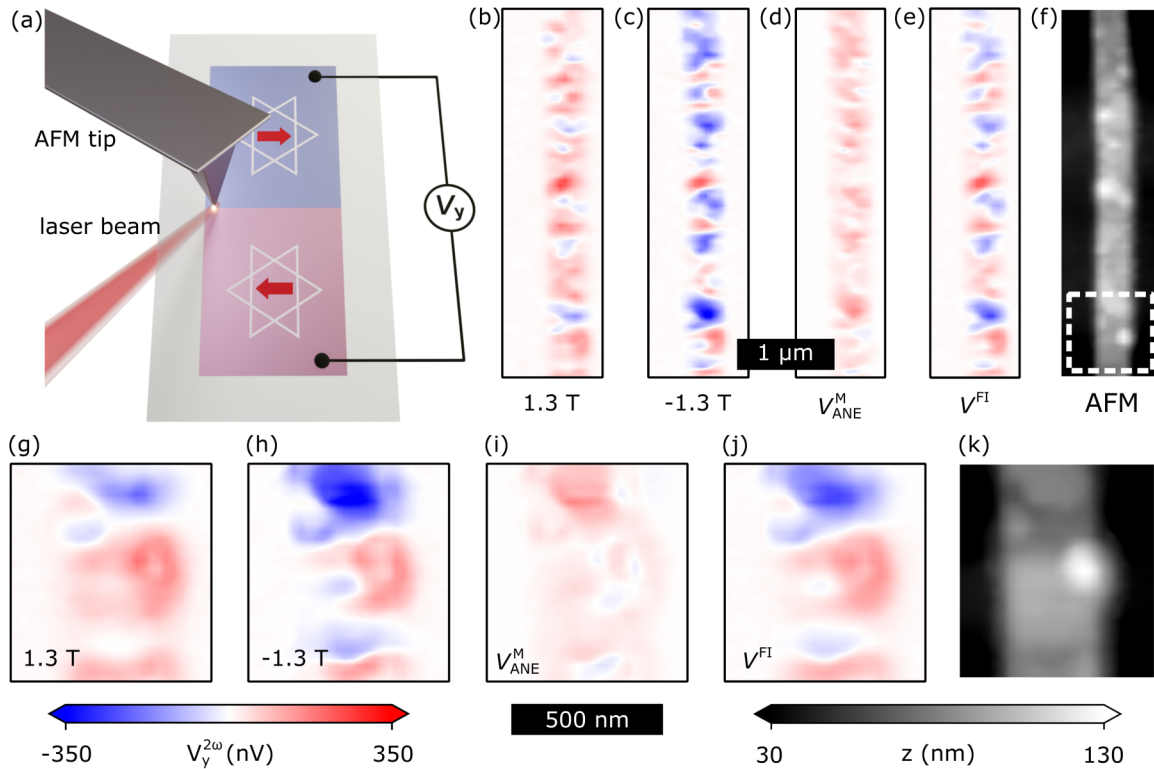


FIG. 3. (a) Schematic illustration of the near-field ANE microscope. (b)–(e) Scanning measurements performed at room temperature in a Mn_3Sn nanowire. The step size is 50 nm. (b), (c) ANE images after ± 1.3 T field cooling, respectively. (d) Magnetic part of near-field measured voltage $V_y^{2\omega}$. (e) Field-independent part of $V_y^{2\omega}$. (f) AFM topography measured simultaneously with the ANE measurements in panel (c); the topography reveals the granular structure of the film. (g)–(k) Scanning measurements performed in a smaller region, shown by the dotted square in panel (f), with a smaller 20 nm step size. The images correspond to the same experimental conditions as panels (b)–(f).

only a partial field-induced switching occurs, indicating domain pinning in regions corresponding to crystal grains. The large crystal grains are a consequence of the high-temperature annealing process that is required for chemical ordering of the Mn_3Sn compound. It is well established that switching is complete and occurs at much smaller fields in bulk Mn_3Sn crystals [15]. Our work strongly indicates pinning due to grain boundaries in the Mn_3Sn layer. Therefore, we believe that the switching behavior can be drastically improved if a more coherent epitaxial growth can be established reducing

grain boundaries and film roughness in thin Mn_3Sn layers, for instance, using alternative methods, such as templated growth [36] or molecular beam epitaxy with suitable buffer layers.

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Data availability. All primary data that support our findings of this study, as well as the codes used for the analysis, are available at Zenodo [37].

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