INTRODUCTION

Electric field control of magnetism, also known as converse magneto-electric (ME) coupling, in ferromagnetic (FM)/ferroelectric (FE) composites is of considerable interest because of the potential for its development as next-generation memory storage and sensing technologies (1, 2). Of particular interest for use as the FE layer are relaxor-ferroelectrics, such as $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ (PMN-PT), which show a large piezoelectric response for compositions near a morphotropic phase boundary ($x \sim 35\%$ for PMN-PT) (3). By coupling the relaxor-ferroelectric with an FM containing large magnetostriction, converse ME coupling is achieved through transfer of the voltage-induced strain from the FE layer into the FM layer that can result in strain-mediated control of in-plane magnetic anisotropy (4–6), tunneling magnetoresistance (7), FM resonance (8), and conductivity (9).

For many strain–mediated ME coupling applications, including rotation of the in-plane magnetization of a coupled FM, anisotropic in-plane strains are required. Therefore, the abovementioned studies (4–7, 9) used (011)-oriented bulk single crystals of PMN-PT that develop large anisotropic in-plane strains under an electric field. Fig. 1A shows the crystal geometry for the (011) orientation. The $x = 30\%$ composition of PMN-PT is rhombohedral (R) with spontaneous polarization along the (111) directions (10). These polarization directions can be grouped as rhombohedral up ($R_{UP}$), rhombohedral in-plane ($R_{IP}$), and rhombohedral down ($R_{DOWN}$) (4). In addition, applying a large electric field can stabilize a polarization parallel to the applied field direction [011], giving the crystal an orthorhombic (O) up ($O_{UP}$), purple symmetry (11). Each of these polarization groups results in average strained unit cells projected into the (011) plane as shown in Fig. 1B, with the unstrained cubic cell (dashed lines) as a reference. This uses equal-weight averaging of each polarization vector present in the group. The normal strains associated with each polarization group may be calculated using the PMN-PT piezoelectric rotation with 3 V PMN-PT bias, much less than the bulk PMN-PT ~100-V requirement. Scanning transmission electron microscopy and phase-field simulations clarify the membrane response. These results provide a crucial step toward understanding the microstructural behavior of PMN-PT thin films for use in piezo-driven ME heterostructures.
RESULTS

Fabrication of (011)-oriented membrane heterostructures

Previously, we demonstrated the fabrication of (001)-oriented PMN-PT films on Si (25) and fabrication of membranes via etching of a Si substrate (33). Because of difficulties associated with growth of epitaxial PMN-PT on (011) Si, that method was incompatible for this study. Therefore, we instead use a (011)-oriented SrTiO3 (STO) substrate with a water-soluble Sr3Al2O6 (SAO) sacrificial layer to create the (011) membrane (34). Fabrication details can be found in Methods, and key steps are highlighted in Fig. 2. First, epitaxial SAO and a capping STO layer are grown on top of (011)-oriented STO substrates by pulsed laser deposition (PLD), followed by sputtering of epitaxial SrRuO3 (SRO) and PMN-PT layers (Fig. 2A). After depositing a Pt electrode, the entire heterostructure is then attached topside down onto a polydimethylsiloxane (PDMS) and glass platform followed by H2O etching of the SAO layer to release the films from the STO substrate (Fig. 2B). After removing the STO buffer layer, deposition and patterning of the FM Ni layer into 160-μm-diameter circular patterns, as well as deposition and patterning of protective polymer SU-8 and Au-lifted electrode top layers, result in the final membrane heterostructure shown in Fig. 2C. A scanning electron microscope (SEM) image of the final heterostructure is shown in Fig. 2D. To confirm that the PMN-PT retained its high-quality single-crystalline structure, x-ray diffraction was performed before and after substrate removal (fig. S1A). The PMN-PT out-of-plane lattice parameter exhibits no change upon removal of the substrate (fig. S1B), and the full width at half maximum of the (011) PMN-PT peak rocking curve remains the same as well (fig. S1C).

Symmetry-enabled rotation of Ni in-plane anisotropy

Strain-induced changes of magnetic anisotropy in the Ni overlayers were measured by longitudinal magneto-optic Kerr effect (MOKE) hysteresis loops as a function of PMN-PT bias electric fields. When the applied magnetic field is swept along a magnetic easy axis (EA), a square hysteresis loop results from the magnetization reorienting between parallel and antiparallel to the applied field. A magnetic field applied along a hard axis (HA) continuously rotates the magnetization away from the EA, resulting in a linear hysteresis loop that saturates at full rotation. Therefore, with the field applied along the [011]pc y direction, we can observe a 90° rotation of magnetic anisotropy as the MOKE loop transitions from an EA to HA upon application of the electrical bias, as seen in Fig. 3A.

Because of Ni’s negative magnetostriction, it will align its EA along the more compressive direction in the presence of anisotropic strain. At 0 kV/cm bias, the as-grown Ni has a weak EA anisotropy along the y direction. Application of a large electric bias (positive or negative) results in HA MOKE loops (Fig. 3A, green curves), meaning that the strain is more compressive along the [100]pc x direction. To confirm that the EA anisotropy is now along the x direction,
MOKE loops were measured with the magnetic field parallel to the [100] pc direction, rotated 90° in-plane from Fig. 3A (fig. S2), and EA MOKE loops were observed at high fields. This matches the expected strain behavior associated with driving the film toward O UP symmetry (Fig. 1). When the electric bias is removed, however, the Ni returns to the as-grown state regardless of bias history, indicating that the strain is relaxed upon removal of the bias. Application of –30 kV/cm bias results in an EA MOKE loop with a higher coercive field (Fig. 3A, purple curve). The reinforcement of the EA along the y direction indicates a reversal in the strain anisotropy from the high-field case, i.e., more compressive along the [01 1 ] y direction, and fig. S2 confirms that the MOKE loop around the FE imprint shows a HA along the x direction. Therefore, we observe a 90° rotation of the Ni in-plane anisotropy over the range of −30 to 30 kV/cm applied bias, corresponding to a 3-V bias across the thickness of our 500-nm PMN-PT membranes. Overall, the MOKE hysteresis behavior is symmetric about –30 kV/cm, which we will show to be due to the FE imprint (discussed in the next section). Similar experiments were performed on a 500-nm clamped PMN-30PT thin film still attached to its STO substrate (fig. S3). Even up to an applied bias of ±400 kV/cm (±20 V), there is no change in the MOKE loop hysteresis. This demonstrates the importance of removing mechanical clamping by the substrate, without which the large anisotropic in-plane strains cannot be achieved.

**Strain behavior and FE properties of PMN-PT/Ni membranes**

To understand the strain behavior inferred from the MOKE hysteresis, we plotted the calculated magnetic anisotropy energy density ($K_U$) determined from the saturation field of HA loops and the associated differential strain ($\varepsilon_{xx} - \varepsilon_{yy}$) using the known magnetostriction...
of Ni in Fig. 3B. Polarization ($P$) versus electric field hysteresis loops (PE loops) are in Fig. 3C, and permittivity versus electric field are in Fig. 3D. Note S2 details the calculation of $K_U$ and $\varepsilon_{xx} - \varepsilon_{yy}$. The PE loops in Fig. 3C show an FE imprint of approximately $-30$ kV/cm, which we believe to be due to the asymmetric electrode configuration of SRO (top) and Pt (bottom) (35). This results in the zero bias polarization of the PMN-PT film to be in a partially polarized state of $\sim 15$ $\mu$C/cm$^2$ pointing toward the SRO electrode. The MOKE hysteresis loops (Fig. 3A), as well as the calculated strain (Fig. 3B) and permittivity (Fig. 3D), show similar symmetric behavior about the FE imprint.

In Fig. 3 (B to D), guidelines have been added that separate the membrane behaviors into three regions: a low-field region near the FE imprint and high-field regions away from the imprint. In the low-field region, we observe that the strain remains relatively constant (Fig. 3B), while the polarization is switching between negative (down) and positive (up) (Fig. 3C). As will be shown in the next section, the film at 0 kV/cm exhibits a mixture of both in-plane rhombohedral (R$_{IP}$) and out-of-plane rhombohedral (R$_{OP}$) domains. Within the low-field region, the film maintains this mixed R state, while the polarization switches between positive and negative, resulting in only minor changes to the differential strain. When the polarization begins to saturate in the high-field region, the strain exhibits the largest changes with applied bias as observed in Fig. 3B as well as the MOKE hysteresis in Fig. 3A. The high-field strain behavior arises from monoclinic distortions as the spontaneous polarizations of R$_{OP}$ domains rotate toward the O direction, as demonstrated later in the “Phase-field simulations” section.

Another interesting feature of the PMN-PT membranes is that the PE loops show a slim-loop hysteresis with low remnant polarization. Similar PE behavior has been reported in other studies of PMN-PT thin films with similar composition (15, 16, 25, 36) and resembles the PE hysteresis of a canonical relaxor becoming nonergodic, such as PMN (PMN-xPT with $x = 0\%$) around 250 K (37). This supports the claim that the morphotropic phase boundary may be shifted to a higher PT content in PMN-PT thin films (36), suggesting that changing the composition of the film may increase the hysteresis of the membrane. The hysteresis also decreases as we approach the DC limit as seen by the quasi-static PE loop at 0.1 Hz (see Methods). Because all MOKE measurements must be performed under DC bias, the reduced hysteresis of the quasi-static loop demonstrates that the nonpermanent strain behavior observed in MOKE is closely related to the nonpermanent polarization behavior of the PMN-PT film.

In the study by Wu et al. (4) using bulk (011) PMN-PT with a Ni overlayer, as well as the study of nanosized Ni ellipses on bulk (011) PMN-PT by Buzzi et al. (6), the primary mechanism for anisotropy rotation in the Ni was anisotropic in-plane strain generated by permanent 71°/109° switching between R$_{IP}$ and R$_{UP}$ polarization states. A primarily R$_{IP}$ state would be expected, where the overall polarization in the film approaches 0 $\mu$C/cm$^2$. This would occur near the FE imprint ($-30$ kV/cm) as seen in Fig. 3C. As we will show in the next section, at 0 kV/cm, where the polarization is $\sim 15$ $\mu$C/cm$^2$, we have a mixture of R$_{IP}$ and R$_{OP}$ domains. Therefore, transitioning from the mixed state at 0 kV/cm to a fully R$_{IP}$ state at $-30$ kV/cm would result in $\varepsilon_{xx} - \varepsilon_{yy} < 0$ (Fig. 1) and should result in a HA MOKE loop. However, Fig. 3A shows an EA MOKE loop at the imprint field instead. Therefore, we do not observe permanent switching to a fully R$_{IP}$ state as observed in bulk (011) PMN-PT.

**Phase-field simulations of PMN-PT membrane**

Phase-field simulations were performed to understand the strain behavior of the PMN-PT membrane. Details of the simulation can be found in Methods. The initial domain configuration consists of a mixture of R$_{IP}$ and R$_{OP}$ as seen in the spontaneous polarization diagram in Fig. 5A, as well as the [011] ($z$ direction) stereographic projection of the spontaneous polarizations in Fig. 5B. The evolution of the spontaneous polarization distribution with electric field is shown for 10 kV/cm (Fig. 5, C and D), 20 kV/cm (Fig. 5, E and F), and 100 kV/cm (Fig. 5, G and H). The simulation is summarized by plotting the average polarization for the $x$, $y$, and $z$ directions (Fig. 5I), as well as the average in-plane strain (Fig. 5J). The average strain was calculated by averaging the strain contribution of individual spontaneous polarization elements multiplied by the electrostriction tensor described in note S1. In the simulation, 0 kV/cm corresponds to no electric bias across the PMN-PT membrane, including any built-in bias from an FE imprint. Therefore, the starting point of the simulation corresponds to the expected structure around the FE imprint of the experimental PMN-PT membrane ($-30$ kV/cm). In addition, histograms of the absolute angle between spontaneous polarizations and the O$_{UP}$ [011] direction are shown in Fig. S5.
Guidelines are added to Fig. 5 (I and J) to separate low-field behavior from high-field behavior. As seen in Fig. 5I, the polarization increases rapidly in the low-field region as the mixed R\(\text{IP}\), R\(\text{DOWN}\), and R\(\text{UP}\) state switches to primarily R\(\text{UP}\) by 20 kV/cm (Fig. 5, C to F, and fig. S5, B and C). Within this region, we observe two behaviors of the spontaneous polarizations within the PMN-PT: (i) polarization switching from R\(\text{DOWN}/R\text{IP}\) to R\(\text{UP}\), as seen in the decrease in polarizations near the R\(\text{IP}\) regions of the stereographic projection, and (ii) polarization rotation from R\(\text{UP}\) to O\(\text{UP}\), as indicated by the region between the two R\(\text{UP}\) polarizations being populated in the stereographic projections. The increase in R\(\text{UP}\) polarizations and polarization rotation toward O\(\text{UP}\) (indicated by the shift in the peak toward a lower angle) are also visible in fig. S5. While the switching between R\(\text{DOWN}\) and R\(\text{UP}\) does not result in a change of in-plane strain, the switching between R\(\text{IP}\) to R\(\text{UP}\) results in \(\varepsilon_{xx} - \varepsilon_{yy} > 0\) (Fig. 1C). On the other hand, the polarization rotation between R\(\text{UP}\) and O\(\text{UP}\) results in \(\varepsilon_{xx} - \varepsilon_{yy} < 0\). Therefore, the low-field region experiences a large increase in average polarization in the z direction, while the competing strain behaviors act to keep the in-plane strain relatively constant (Fig. 5, I and J).

In the high-field region, the average polarization is nearly saturated but continues to increase as the polarization continues to rotate from R\(\text{UP}\) toward the O\(\text{UP}\) state. Nearly all of the polarizations have been switched to R\(\text{UP}\), meaning that the polarization rotation toward O\(\text{UP}\) dominates the strain behavior resulting in a large decrease in differential in-plane strain (Fig. 5J). By 100 kV/cm, all of the polarizations lie in the region between R\(\text{UP}\) and O\(\text{UP}\) (Fig. 5, G and H, and fig. S5D), corresponding to a large monoclinic distortion away from R\(\text{UP}\). The simulation results qualitatively agree with the experimental strain and polarizations measured in the PMN-PT/Ni membrane (Fig. 3), showing relatively constant in-plane strain during polarization switching and large changes of in-plane strain at higher fields. Note that the strains calculated from the experimental MOKE loops (Fig. 3B) exhibit a horizontal and vertical shift relative to the calculated strains from simulation (Fig. 5J), although, qualitatively, the two curves are similar. The vertical shift arises from the as-grown Ni being weakly anisotropic with the EA along the y direction at 0 kV/cm, while the horizontal shift comes from the FE imprint of the PMN-PT membranes being approximately ~30 kV/cm.

**DISCUSSION**

We have provided demonstration of the low-voltage strain-mediated ME effect in an all-thin-film heterostructure that only relies on the large anisotropic strains inherent to (011) PMN-PT thin films by completely removing them from their substrate.
The PMN-PT/Ni membranes achieve a robust, piezo-driven, 90° rotation of the in-plane magnetic anisotropy of the Ni overlayer under application of only a few volts of bias across the thickness of the PMN-PT membrane. This is roughly two orders of magnitude less voltage than demonstrations using bulk single crystals of PMN-PT that require application of >100 V (4). The ME coupling is achieved by driving the PMN-PT polarization toward orthorhombic symmetry under the applied bias, resulting in strain anisotropy controlled by the in-plane crystal symmetry of the PMN-PT film. STEM measurements show that the zero-field domain structure of the PMN-PT membrane consists of a mixture of both R$_{IP}$ and R$_{OP}$ domains, as well as additional regions with B-site cation displacements along directions between the two R states. Phase-field simulations confirm that the in-plane differential strain does not change in the low-field region near the FE imprint because of the switching between R$_{IP}$ and R$_{OP}$ domains competing with polarization rotation toward O$_{Up}$. However, at higher fields, polarization rotation toward O$_{Up}$ dominates and once again results in a giant piezoelectric effect.

Demonstrations using bulk PMN-PT show permanent switching between in-plane and out-of-plane R polarization states and, consequently, between distinct strain states. The permanent switching behavior of bulk PMN-PT is typically a trait that is deemed desirable for applications such as memory storage, but the nonpermanent strains in our PMN-PT membranes may still be able to provide the 180 magnetization switching needed for memory devices (40, 41). Understanding the differences between bulk and membrane PMN-PT response to external stimuli is key to their use in future technologies. Such differences may arise owing to various effects, such as smaller domain sizes in membrane versus bulk, reduced chemical ordering in PMN-PT membranes due to differences in material processing parameters (e.g., growth temperature), as well as different electrical/mechanical boundary conditions in membranes arising from higher defect concentration, or enhanced role of the interface (42). Studying the membrane PMN-PT electrical bias response using in situ STEM or synchrotron x-ray diffraction, as well as studying compositions other than the $x = 30\% (1 - x)$PMN-$x$PT used here, could provide key information for future applications. Our work provides key insight into the microstructural behavior of PMN-PT thin-film membranes and demonstrates how they can be used in ME coupling devices. In addition, coupling the PMN-PT membrane with a variety of other materials, such as complex oxides, two-dimensional materials, and III-V semiconductors, can lead to the discovery of previously unknown piezo-driven phenomena.

**METHODS**

**Membrane fabrication**

Twenty nanometers of epitaxial SAO was grown on top of (011) STO substrates via PLD. The SAO was grown at a substrate temperature of 780°C and $p(O_2) = 1 \times 10^{-6}$ torr using a laser fluence of 1 J/cm$^2$ on the polycrystalline SAO target. A 20-nm capping layer of STO, preventing possible cation diffusion at the interface (43), was grown at 750°C and $p(O_2) = 1 \times 10^{-6}$ torr using a laser fluence of 2 J/cm$^2$ on the single-crystal STO target. One hundred nanometers of SRO was grown by radio frequency (RF) magnetron sputtering at a power of 100 W in 200 mtorr of Ar:O$_2$ (12:8) with a substrate temperature of

![Fig. 5. Phase-field simulations of the (011) PMN-PT membrane. Spontaneous polarization and [011] stereographic projection of the PMN-PT membrane at (A and B) 0 kV/cm, (C and D) 10 kV/cm, (E and F) 20 kV/cm, and (G and H) 100 kV/cm. The legend for the coloring of spontaneous polarization is included in (A). (I) Average polarization in the x, y, and z directions versus applied field. (J) Field dependence of the average anisotropic in-plane strain $\varepsilon_{xx} - \varepsilon_{yy}$. In (I) and (J), guidelines have been added to separate the low-field and high-field regions.](image-url)
600°C. Five hundred nanometers of 70% Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–30% PbTiO\textsubscript{3} (PMN-30PT) was grown by RF magnetron sputtering at a power of 100 W in 500 mtorr of Ar:O\textsubscript{2} (17:3) with a substrate temperature of 625°C. One hundred nanometers of Pt was deposited at room temperature by DC magnetron sputtering. The edges of the heterostructure were ground slightly to remove any sidewall deposition of SRO, PMN-PT, or Pt that would prevent H\textsubscript{2}O from etching the SAO later on.

PDMS with a weight ratio of 10:1 (base:cross-linking agent) was spin-coated onto a 10 mm by 10 mm glass substrate at 5000 rpm for 10 s, resulting in a PDMS thickness of ~30 μm. Before the PDMS is cured, the PMN-PT film heterostructure is placed into the PDMS Pt-side down. The entire sample is placed in vacuum for a minimum of 5 hours to remove any bubbles between the PDMS and Pt layers, followed by curing of the PDMS on a hot plate at 100°C for 1 hour. Placing the film into the uncured PDMS is crucial, as it allows the PDMS to mold to the surface of the film, ensuring that the PMN-PT membrane is as flat as possible after the substrate is removed. One consequence of curing the PDMS after the film is attached is that it will mold to the sides of the substrate. Once cured, removing the PDMS from the sides of the substrate with a razor is necessary so that the H\textsubscript{2}O can reach the SAO.

The sample is placed in a beaker of water to etch the sacrificial SAO layer. This process can take anywhere from 24 to 72 hours. Heating the water to 70° to 80°C was found to speed up the etching in some cases. Etching progress was monitored by visual inspection under a microscope, and once found to be completed, the substrate was removed using tweezers. Dipping in isopropyl alcohol was used to displace water and reduce surface tension between the substrate and film in some instances.

With the substrate removed, the exposed surface of the membrane now consists of the STO layer that was used to cap the SAO layer. Ion milling was used to remove the STO layer and expose the SRO film. A 35-nm Ni film was deposited by DC sputtering at room temperature on top of the SRO to act as the FM layer for our FE/FM composite. Photolithography and wet etching were used to pattern the SRO/Ni into 160-μm-diameter disks. SU-8 photoresist was spin-coated and patterned by photolithography to create a protection layer that left the Ni/SRO disks exposed while covering the PMN-PT. Thirty nanometers of Au was deposited by DC sputtering at room temperature, followed by photolithography and patterning to create Au electrodes partially overlapping the Ni/SRO disks, and partially on top of the SU-8. This allowed for electrical contact to be made with probe tips or wire bonding to the Au on the SU-8 layer without risk of damaging the fragile membrane heterostructure.

**Scanning transmission electron microscopy**

Two kinds of cross-sectional samples having [100] and [01T] pseudo-cubic projections were prepared using a dual-beam focused ion beam system (Helios G3, FEI) to determine the FE domain structure. We used a Ga ion beam at 30 kV to make a thin specimen and using a STEM (JEM-ARM200F, JEOL, Japan) at 200 kV equipped with a fifth-order probe corrector (ASCOR, CEOS GmbH, Germany) at the Materials Imaging and Analysis Center of Pohang University of Science and Technology (POSTECH) in South Korea. The optimum size of the electron probe for STEM observation was ~78 pm. The collection semiangles of the high-angle annular dark-field (HAADF) detector were adjusted from 68 to 280 mrad to collect scattered electrons in a large angle for clear Z-sensitive images. HAADF-STEM images were acquired using SmartAlign (HREM Research Inc., Japan), which conducted the multistack of images and aligned them using rigid registration to correct the sample drift and scan distortions. The obtained raw images were processed using a bandpass difference filter with a local window to reduce background noise (Filters Pro, HREM Research Inc., Japan).

**Phase-field simulations**

The phase-field method is used to simulate the effect of applied bias on the polarization distribution in (011) PMN-30PT freestanding membranes. In the phase-field model, the polarization is selected as the order parameter to describe the domain structures, and its spatial and temporal evolutions are controlled by the time-dependent Ginzburg-Landau equation (44, 45): \( \frac{\partial P}{\partial t} = -L(\Delta P/\delta P) \), with \( L \) being the kinetic coefficient related to the domain-wall mobility and \( F \) being the total free energy, which includes the bulk chemical energy, polarization gradient energy, electric energy, and elastic energy (44). The parameters for the bulk chemical energy are from literature (46). The freestanding membrane is represented by a grid of 128×128×(N\text{bottom-air} + N\text{membrane} + N\text{top-air}) Å with \( \Delta x = \Delta z = 1 \) nm, \( N\text{bottom-air} = 2 \), \( N\text{membrane} = 20 \), and \( N\text{top-air} = 2 \). Periodic boundary conditions are assumed in the in-plane directions. To consider the (011) orientation, we define the simulation coordinate system \((x, y, z)\) to be \( x/\langle100\rangle, y/\langle01T\rangle, \) and \( z/\langle011\rangle, \) and the tensor coefficients are rotated by the corresponding rotation matrix.

Stress-free boundary conditions are assumed for both the top and bottom surfaces for solving the elastic equilibrium equation of the membrane. This is achieved by considering an inhomogeneous system including two layers of vacuum at both the top and bottom surfaces of the membrane. The elastic equilibrium equation of such an inhomogeneous system is solved using the spectral iterative perturbation method (47). To incorporate the applied electric field, a superposition method is used to solve the electrostatic equilibrium equation with a uniform preset voltage bias at the top surface while the bottom surface is grounded (48).

**MOKE measurements**

The PMN-PT membrane was mounted between the poles of an electromagnet and a polarized red HeNe (632 nm) laser was reflected off of the sample surface at approximately 45° from normal incidence. The beam was focused to an approximately 10-μm spot near the center of the 160-μm Ni discs using an achromatic lens. The reflected beam’s polarization was analyzed with a differential detection scheme. A polarizing beam splitter directed the \( s \) and \( p \) components of the reflected beam onto two channels of a Thorlabs PDB210A differential photodetector and a half-wave plate before the beam splitter was used to balance the inputs to the detector. A fitting procedure was
used to extract $H_{sat}$ from HA MOKE loops, resulting in the values plotted in Fig. 4D. The loop was normalized so that the Kerr rotation at magnetic saturation is ±1. Data with normalized Kerr rotation values between 0.85 and −0.85 were fit to a line. The magnetic field values where the fitted line intersects with +1 and −1 normalized Kerr rotation were respectively taken to be $H_{sat}^+$ and $H_{sat}^-$, with $H_{sat}$ as the average.

FE measurements

Permittivity measurements were performed by slowly (0.5 Hz) sweeping a bias electric field across a device while also applying an AC waveform [3.5 kV/cm root mean square (rms) at 4 kHz], detecting the resulting 4-kHz AC current with a lock-in amplifier. High-frequency PE loops were obtained by integrating the current flowing to the device while sweeping the electric field at 30 kHz. The observed frequency dependence of the PE loop required determining the PE response at very low frequency to compare with MOKE measurements, where magnetic hysteresis data were acquired with static electric field. A 0.1-Hz equivalent polarization at an electric field $E_0$ was determined by letting the polarization decay from its initial high-frequency sweep value for 10 s at fixed electric field $E_0$ and then saturating with a high-frequency sweep and integrating the resulting current to determine the polarization change. Both the initializing sweep and saturating sweep were complete PE loops with the same electric field extent, but phase-shifted to begin and end at $E_0$.

SUPPLEMENTARY MATERIALS

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