Large interfacial magnetostriction in \((\text{Co/Ni})_4/\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3–\text{PbTiO}_3\) multiferroic heterostructures

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ABSTRACT: The magnetoelastic behavior of multiferroic heterostructures –coupling of magnetic anisotropy or domain dynamics to structural deformations – has been intensively studied for developing materials for energy-efficient, spin-based applications. Here we report on a large, interface-dominated magnetostriction in \((\text{Co/Ni})_4/\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3–\text{PbTiO}_3\) multiferroic heterostructures. Ferromagnetic resonance spectroscopy under voltage-induced strains enabled estimation of the saturation magnetostriction as a function of Ni thickness. The volume and the interface components to the saturation magnetostriction are \((6.6 \pm 0.9) \times 10^{-6}\) and \((-2.2 \pm 0.2) \times 10^{-14} \text{ m, respectively. Similar to perpendicular magnetic anisotropy in Co/Ni, the large, negative magnetostriction originates from the Co/Ni interfaces. This interfacial functionality delivers an effect over 300% larger than the bulk contribution, and can enable low energy, nanoelectronic devices that combine the tunable magnetic and magnetostrictive properties of Co/Ni multilayers with the ferroelectric properties of Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3–\text{PbTiO}_3.\)

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1. Introduction

Manipulation of the magnetization and magnetic anisotropy of ultrathin magnetic films by electric-field induced strain in artificial multiferroic heterostructures has generated much interest as a pathway toward potentially more energy efficient control of magnetic states compared with current-generated Oersted fields or spin-transfer torques.\textsuperscript{1-5} Previous studies have shown that strains induced by electric fields applied to a piezoelectric substrate material like Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}\textsubscript{1-x-PbTiO\textsubscript{3}}x (PMN-PT) or Pb(Zr,Ti)O\textsubscript{3} (PZT) may significantly alter the preferred magnetic anisotropy direction in an overlaid magnetic thin film.\textsuperscript{6-14} This approach has been demonstrated for shifting the ferromagnetic resonance response of thin ferromagnetic films\textsuperscript{6,15-17} and for the generation and detection of spin waves.\textsuperscript{18-21} In combination with the voltage-controlled magnetic anisotropy effect, there are efforts to develop a charge- and strain-controlled magnetic memory device, promising lower energy write operations than current-controlled memories.\textsuperscript{22-24} Recently, implementations of artificial multiferroic heterostructures have been proposed for clocking in spin logic gates permitting both Boolean and non-Boolean circuits.\textsuperscript{25-29}

Multiferroic heterostructures in which the ferromagnetic element exhibits a perpendicular magnetic anisotropy (PMA) carry significance for potential applications. Due to their improved scaling compared to planar magnetized materials, PMA materials are used in spin-transfer-torque magnetic random access memory devices,\textsuperscript{30-32} as well as magnetic recording media.\textsuperscript{33} Heterostructures combining a piezoelectric layer with PMA CoFeB,\textsuperscript{7} Co/Pt,\textsuperscript{2,10,34} Co/Pd\textsuperscript{4} and Cu/Ni\textsuperscript{3} and Co/Ni,\textsuperscript{35} have shown potential for applications through reductions in the switching energy; enhancement of the domain wall propagation speed; and 90- and 180-degree field-free switching. A recent study\textsuperscript{10} has shown that the electric field response in multiferroic (Co/Pt)-PMN-PT has distinct interface- and bulk contributions to the magnetic anisotropy energy, demonstrating the importance of the overlaid magnetic film thickness on electric-field control of magnetism in PMA multilayered thin films.

Quantifying the strain-mediated effects on PMA in Co/Ni multilayers is significant for a broad application space, from magneto-optic storage and all-perpendicular spin-valve memory devices to magnetic tunnel junction magnetic random access memories, spin-orbitronic and voltage-controlled magnetic memories.\textsuperscript{36-42} Unlike previously studied magnetoelastic compounds Ni/Cu or Co/Pt, each constituent layer in Co/Ni is ferromagnetic and consequently should carry a
distinct contribution to the total magnetic response to strain, defined by an effective
magnetostriction constant. Moreover, for technological applications, Co/Ni exhibits low intrinsic
damping\textsuperscript{43} and high spin polarization\textsuperscript{44} compared to other PMA multilayers, enabling potentially
faster dynamics and higher sensitivity read-out for Co/Ni integrated into a magnetic tunnel
junction.\textsuperscript{45} Although the lower damping may cause an undesirable enhancement in relaxational
time,\textsuperscript{46} clear benefits are afforded by the lower write energy for prospective toggle-mode
magnetostrictive memory devices and the longer sustained precession cycles for strain-mediated
spin-wave devices.\textsuperscript{47-48} A recent study indicated that a Ni-rich multilayer [Co(0.15 nm)/Ni(0.6
nm)]\textsubscript{16} exhibited a reduction in the magnetic coercive field and a reduction in the out-of-plane
magnetic remanence under out-of-plane strains.\textsuperscript{35} In this report, electric-field-biased, stripline
ferromagnetic resonance measurements of \((\text{Co/Ni})_4/\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-\text{PbTiO}_3\) multiferroic
heterostructures enables the direct and unambiguous measurement of changes in the PMA due to
electric-field induced changes in the magnetoelastic anisotropy energy. Owing to the tunable
magnetic anisotropy in Co/Ni multilayers, it would be possible to select a film stack with
sufficiently modest PMA for strain-tuning of the magnetization direction.

We demonstrate that Co/Ni multilayers exhibit a tuneable magnetostriction that can be
enhanced by reducing the Co and Ni sublayer thicknesses. This is in contrast to an earlier
prediction of a dominant volume magnetostriction in these multilayers estimated by taking a
weighed sum of the positive magnetoelastic modulus of hcp Co (29 MJ/m\textsuperscript{3}) with the smaller
negative modulus of fcc Ni (-10 MJ/m\textsuperscript{3}).\textsuperscript{49} Taking into account the total thickness of each Co/Ni
multilayered sample, we find that the saturation magnetostriction increases in inverse proportion
to the total thickness, whereby the interfacial component of the magnetostriction exceeds the
volume component by more than 300\%. Our results demonstrate that the interfaces are
fundamental to understanding the magnetoelastic anisotropy in Co/Ni multilayers. The ability to
tune the magnetostriction through engineering the Co/Ni interfaces carries technological promise
for electric-field-controlled composite multiferroic devices with perpendicular magnetic
anisotropy.

2. Results and Discussion

2.1. Composition and structure of Co/Ni multilayers

The multilayers of Ta (3.0 nm)/Pt(2.0 nm)/[Co(0.6 nm)/Ni (\(t_{\text{Ni}}\))]\textsubscript{x4}/Co(0.6 nm)/Pt(2.0
nm)/Ta(3.0 nm) were grown on top of [001]-cut PMN-PT substrates by magnetron sputtering
(see Methods section). The bilayer Ta (3.0 nm)/Pt (2.0 nm) complex grown directly on top of the PMN-PT was chosen to promote the growth of polycrystalline fcc (111) Co/Ni and with adequate thickness to effectively screen accumulated surface charge on the ferroelectric PMN-PT crystal. Where previous studies have indicated a ferroelectric field effect on the magnetic anisotropy comparable to the magnetoelastic response due to strain, these Co/Ni multilayers samples are deliberately isolated from the charged PMN-PT surface. Only the Ni thickness is varied, while keeping the thicknesses of the other layers fixed. In order to apply a bias voltage through the 0.5 mm-thick PMN-PT substrate, a back electrode of Ti(2 nm)/Ta(20 nm) was grown on the back side of all substrates by electron beam evaporation. In this way, electric fields are generated in the PMN-PT substrate by applying a voltage between the top and bottom Ta electrodes.

The PMA strength of Co/Ni multilayers strongly depends on fcc (111) stacking of the Co and Ni layers and smooth interfaces. For sufficiently thin Co layers, the atomic planes can grow in a fcc (111) stacking order, in which the cobalt layers may continue the stacking order of the platinum or nickel on which they were deposited. Figure 1 shows the XRD spectra for samples with nominal Ni thicknesses \( t_{\text{Ni}} = 0.45 \) nm and \( t_{\text{Ni}} = 0.9 \) nm. The Bragg scattering peak associated with the [Co/Ni](111) reflection (2\( \theta = 43.8^\circ \)) can be seen on the shoulder of the PMN-PT(002) reflection (44.7\(^\circ\)). Also present are the Pt(111) (39.6\(^\circ\)) and Pt(200) (46\(^\circ\)) reflections. Notably absent from these scans are the Co(200) and Ni(200) peaks in the vicinity of 2\( \theta = 50^\circ \), thereby indicating partial fcc(111) texturing in these films deposited on PMN-PT.

To further elaborate the microstructure of the [Co/Ni] multilayers on PMN-PT, we conducted a transmission electron microscopy (TEM) study on a [Co(0.6 nm)/Ni(0.9 nm)]\(_{x4}\) sample, shown in Figure 2. In this cross-section TEM micrograph, the scale bar was calibrated using the lattice parameter of the PMN-PT, which was obtained by tilting the substrate to its [010] zone axis. The amorphous Ta adhesion layer is visible just above the top surface of the PMN-PT, followed by a symmetric Pt/[Co/Ni]/Pt trilayer in which the interfaces between the [Co/Ni] complex and the Pt seed and cap layers can be distinguished. The microstructure of the Co/Ni multilayers is textured and extends from seed to cap, with polycrystalline fcc (111) grains suggested by Fast Fourier Transforms of the micrograph in these regions in Figure 2(b). The observed microstructure is similar to results observed in Co/Ni multilayers grown on thermally oxidized Si, demonstrating the suitability of PMN-PT substrates for growth of Co/Ni layers.
multilayers.\textsuperscript{40, 45} Intensity peaks in the FFT can be found at 0.23 nm for Pt and 0.21 nm for [Co/Ni], which agree with the lattice spacing for (111)-oriented face centered cubic growth of the Pt-seeded [Co/Ni] multilayers extracted from the XRD spectra shown in Figure 1. The presence of stacking faults and dislocations common to polycrystalline, sputtered films are also evident.

\section*{2.2. Magnetostriction in Co/Ni multilayers determined by broadband ferromagnetic resonance spectroscopy}

\subsection*{2.2.1. Modification of the ferromagnetic resonance condition in Co/Ni multilayers in response to variable strain}

The sign and strength of the magnetostriction coefficient is estimated from strain-mediated changes to the magnetic anisotropy field measured using a stripline ferromagnetic resonance (FMR) spectrometer, modified to enable \textit{in situ} strains in response to moderate electrical biases to the piezoelectric substrate, which can shift the ferromagnetic resonance field in response to modulation of the magnetoelastic anisotropy. In the case of Co/Ni samples, the multilayered thin film is shown to have a (111)-oriented polycrystalline structure, for which the magnetic free energy density, describing the Zeeman energy, demagnetizing energy, uniaxial-magnetocrystalline and magnetoelastic anisotropies, respectively, is expressed as follows:

\begin{equation}
E = -\mu_0 M_S H \cos (\theta - \theta_H) + \frac{1}{2} \mu_0 M_S^2 \cos^2 \theta + K_2 \sin^2 \theta + K_{\text{elastic}} \sin^2 \theta.
\end{equation}

Here, \( \mu_0 \) is the vacuum permeability, \( H \) is an applied external field, \( M_S \) is the saturation magnetization, \( K_2 \) is the first-order magnetocrystalline anisotropy energy, \( K_{\text{elastic}} \) is the first-order uniaxial magnetoelastic anisotropy, \( \theta_H \) and \( \theta \) are respectively the polar angles of the applied field and the magnetization relative to the sample surface normal. Neglecting hysteretic effects in the ferroelectric crystal, the out-of-plane tensile strain due to the \( d_{33} \) inverse piezoelectric response of the PMN-PT(001) crystal is accompanied by biaxial compression in the plane (e.g. \( d_{31}=d_{32}=-2\nu d_{33}/(1-\nu) \)), where \( \nu \) is the Poisson ratio for PMN-PT. The biaxial symmetry of the planar strain transferred completely to the overlaid thin film multilayers reduces the magnetoelastic anisotropy energy term to a simple expression:

\begin{equation}
K_{\text{elastic}} = B_2 (\varepsilon_\parallel - \varepsilon_\perp),
\end{equation}

for which \( \varepsilon_\parallel (\varepsilon_\perp) \) is the biaxial planar (out-of-plane) strain in Co/Ni multilayers, and \( B_2 = -3c_{44}\lambda_S^{111} \) is the second magnetoelastic modulus, related to the saturation magnetostriction.
along (111) \( \lambda_{\parallel}^{\perp} \) and the elastic constant \( c_{44} \). Biaxial compression in the thin film generates out-of-plane expansion, in proportion to the Poisson ratio of the multilayered film. Consequently, for positive (negative) \( \lambda_{\parallel}^{\perp} \), voltages generating longitudinal expansion of the PMN-PT generate in-plane compression in the Co/Ni multilayers and make the out-of-plane orientation more (less) energetically favorable by enhancing (reducing) the total uniaxial anisotropy energy.

Through the Kittel equations for in-plane (\( \parallel \)) applied fields, the magnetic free energy components factor into the ferromagnetic resonance field versus frequency dispersion in Equation (3):

\[
f = \frac{g \mu_B \mu_0}{2 \pi \hbar} \sqrt{H_{\text{res}} \parallel (H_{\text{res}} \parallel + M_{\text{eff}})},
\]

for which \( f \) is the ferromagnetic resonance frequency, \( \hbar \) is the reduced Planck constant, \( \mu_B \) is the Bohr magneton, \( g \) is the spectroscopic \( g \)-factor, and an effective magnetization \( M_{\text{eff}} \) is defined for convenience as follows:

\[
M_{\text{eff}} = M_S - 2 \frac{K_2 + K_{\text{elastic}}}{M_S}.
\]

For negative \( M_{\text{eff}} \), the equilibrium magnetization configuration lies normal to the plane, and for positive \( M_{\text{eff}} \), the magnetization can lie within the plane. At a given frequency, incremental increases (decreases) in the magnetoelastic anisotropy strength \( \Delta K_{\text{elastic}} \) due to strain will decrease (increase) the effective magnetization \( \Delta M_{\text{eff}} = -2 \Delta K_{\text{elastic}} / M_S \) and shift the in-plane resonance field to higher (lower) field magnitudes.

2.2.2. **Broadband ferromagnetic resonance spectroscopy with in-situ tunable strains**

The magnetostriction in Co/Ni multilayers is evaluated by the change in magnetic anisotropy in response to electric-field induced strains in the PMN-PT substrates. Figure 3 illustrates the approach to estimating the change in magnetic anisotropy. In Figure 3(a), the multiferroic heterostructure is shown with a thick Ta (20 nm) back electrode and a thinner (5 nm) protective capping electrode of Pt(2 nm)/Ta(3 nm) over the Co/Ni multilayers. Voltages applied to the Ta electrodes across the (001)-cut surface of the PMN-PT induce an out-of-plane strain \( \varepsilon_{\parallel}^{\text{PMN}} > 0 \) due to the inverse piezoelectric effect along the electric biasing axis. The electromechanical coupling factor associated with this expansion was measured using a linear motion transducer and is \( d_{33} = (1650 \pm 50) \text{ pm V}^{-1} \), as shown in the strain-electric field curve.
in Figure 3(b). Whereas an established approach for electric-field-biased ferromagnetic resonance measurements has been to utilize a cavity to couple microwave magnetic fields into an electrically biased, multiferroic heterostructure,\textsuperscript{2, 6-7, 10, 53} the approach discussed herein and depicted in Figure 3(c) uses the common ground plane of a grounded coplanar waveguide (GCPWG) board and a back electrode to simultaneously generate static electric fields in the PMN-PT and generate microwave magnetic fields in the Co/Ni multilayers with the potential to apply arbitrary microwave frequencies. At a fixed microwave frequency, cycling the electric field through the PMN-PT ferroelectric hysteresis loop (Figure 3(b)) generates shifts in the ferromagnetic resonance field due to strain, which can be seen in Figure 3(d) for a Co(0.6)/Ni(0.9) multilayers at 18 GHz microwave excitation frequency. This change in resonance field is related to strain-mediated changes in the magnetic anisotropy through the inverse magnetostriction effect.

Figure 4 presents the protocol for estimating the strain-mediated changes to the magnetic anisotropy energy in the Co/Ni multilayers sample with a 0.9 nm thick Ni layer. First, a series of in-plane FMR spectra are collected under zero electric field bias, as shown in Figure 4(a). Equation (3) is used to fit the frequency versus FMR field dispersion in Figure 4(b), from which we estimate the $g$-factor ($g = 2.18 \pm 0.01$) and the effective magnetization ($\mu_0 M_{\text{eff}} = 0.478 \text{T} \pm 0.004 \text{T}$) under zero bias electric field. Uncertainty in our best-fit parameters herein comprises the standard uncertainty from a least-squares fit to Equation (3). Then, at a fixed microwave frequency (18 GHz), FMR spectra are collected under increasing- and decreasing electric bias. This frequency (18 GHz) was chosen because it showed the cleanest FMR absorption lineshape, although other frequencies were tested and showed a similar characteristic. The resonance field versus PMN-PT electric field shown in Figure 4(c) exhibits electric hysteresis, with a coercive field approximately ($E_c \cong (0.12 \pm 0.02) \text{MV m}^{-1}$), consistent with strain-electric field measurements conducted on the same sample. We use the data in Figure 4(c) to invert Equation (3) and solve for $M_{\text{eff}}$ as a function of electric field, as shown in Figure 4(d). To neglect changes in the anisotropy due to inhomogeneous planar strains taking place during ferroelastic domain switching in the PMN-PT substrate, our estimations of the magnetostriction focus on the observed change $\Delta M_{\text{eff}}$ while decreasing the applied voltage from the saturating electric field down to zero, as depicted by the overlaid line on Figure 4(d).\textsuperscript{53-54} For this [Co(0.6 nm)/Ni(0.9 nm)] sample, the estimated change is $\mu_0 \Delta M_{\text{eff}} = (0.013 \text{T} \pm 0.002 \text{T})$. 

7
The electrically-biased ferromagnetic resonance spectroscopy shown in Figure 4 was carried out on the full series of five [Co(0.6 nm)/Ni(tNi)] multilayers. For each sample, the $\mu_0\Delta M_{\text{eff}}$ value was estimated as detailed by the previous paragraph. Supporting Information Figures S1-S4 present the individual measurements of the 0.15 nm, 0.3 nm, 0.45 nm and 0.6 nm thick Ni sublayer samples for estimating $\mu_0\Delta M_{\text{eff}}$. The saturation magnetization for each sample was also measured using vibrating sample magnetometry (Supporting Information Figure S9).

The change in magnetic anisotropy energy is estimated from $\frac{1}{2}\mu_0 M_S \Delta M_{\text{eff}}$. The PMN-PT strain $\varepsilon_{\perp}^{\text{PMN}}$ at the two electric field values used to estimate $\Delta M_{\text{eff}}$ is approximately $(\Delta \varepsilon_{\perp}^{\text{PMN}} \approx 0.08 \% \pm 0.01 \%)$. Consistent with previous studies,\textsuperscript{12-13,54-56} we will assume negligible strain relaxation at the Co/Ni multilayers thicknesses explored here, and that the biaxial compression in the PMN-PT substrate is transferred homogeneously through the thickness of the overlaid Co/Ni film. By setting this value to the predicted change in magnetoelastic anisotropy due to inverse magnetostriction from Equation (3), we solve for $\lambda_{s111}^\text{vol}$, the saturation magnetostriction.

### 2.3. Influence of Ni thickness on saturation magnetostriction in Co/Ni multilayers

Figure 5 shows the estimated saturation magnetostriction values for the series of Co/Ni multilayers plotted against the inverse of the total thickness of the [Co(0.6 nm)/Ni(tNi)]$_n$/Co(0.6 nm) multilayered films. Immediately evident is the negative sign for the magnetostriction value for the entire series of multilayers. Therefore, out-of-plane tensile strain in Co/Ni multilayers tends to reduce the PMA. The largest estimated magnetostriction value is estimated for the [Co (0.6 nm)/Ni (0.15 nm)] multilayers ($\lambda_{s111}^\text{vol} = (−51 \pm 3) \cdot 10^{-6}$), taking into account the uncertainty in measured quantities for the strain, the effective demagnetization change $\mu_0\Delta M_{\text{eff}} = (.016 \ T \pm .002 \ T)$ and the saturation magnetization ($M_S \approx 1200 \pm 20 \ \text{kA m}^{-1}$).

For comparison, the magnetostriction of fcc (111) Ni is shown as a horizontal line on Figure 5.

Upon inspection of the data in Figure 5, there is an apparent linear relationship between the inverse thickness and the saturation magnetostriction. Using a phenomenological form for the magnetostriction that includes a volume and an interfacial component, we can fit the magnetostriction data with the following model:

$$\lambda_{s111}^\text{vol}(t) = \lambda_{s}^\text{vol} + \frac{(2n + 1)}{t} \lambda_{s}^\text{int},$$

where $\lambda_{s}^\text{vol}$ and $\lambda_{s}^\text{int}$ are the volume and the interface components of the saturation magnetostriction, respectively, $n$ represents the number of Co/Ni bilayer repeats and $t$ represents...
the aggregate Co/Ni multilayers thickness. Where increases in the Ni content appear to reduce the magnitude of the magnetostriction of Co/Ni multilayers below the Ni bulk value, an underlying thickness dependence to the magnetostriction appears to be more significant here, suggesting that the interfacial component is dominant. This result is consistent with recent indications that magnetostriction can be significantly enhanced by interfacial engineering.56-57

Magnetostriction originates from spin-orbit coupling between magnetization and the lattice, similarly relevant to the development of magnetocrystalline anisotropies.58 There are several possible origins of a large surface or interface magnetostriction in ultrathin layers, including crystal field effects,59 bilayer intermixing and roughness at the interfaces,60-61 and dipolar effects.62 As Co/Ni multilayers show a significant interface-induced orbital moment asymmetry underlying the perpendicular magnetic anisotropy energy,63 also reproduced here in Supporting Information Figure S10, we anticipate a similarly large interface contribution to the magnetostriction. Indeed, the best-fit line of Equation 5 to the data in Figure 5 estimates that the volume \(\lambda_{S}^{\text{vol}}\) and the interface \(\lambda_{S}^{\text{int}}\) components to the saturation magnetostriction are \((6.6 \pm 0.9) \times 10^{-6}\) and \((-2.2 \pm 0.2) \times 10^{-14}\) m, respectively. While this relationship between saturation magnetostriction and thickness would imply that the volume magnetostriction is positive - in contrast to established values for bulk Co and Ni specimens - it must be stressed that this is an empirical model that fits data for a heterostructured material over a limited thickness range. On the other hand, this empirical relation may be useful for thin film heterostructures, in which the sub-10 nm behavior is most relevant. This large interfacial magnetostriction component approaches four times larger than the volume contribution across our presented thickness range, indicating the significance of the interfaces for generating a large magnetoelastic response.

Recent work has demonstrated the significance of elastic effects on the electronic states in Co/Pt multilayers, modifying the magnetic anisotropy through modifications to the 3d-5p hybridization.10 Although Co/Ni multilayers are composed entirely of 3d transition metals, orbital state occupation \((d_{xy} \text{ and } d_{x^2-y^2})\) is expected to play a significant role in the perpendicular magnetic anisotropy.36 Future \textit{ab initio} calculations could shed light on the role of orbital states on strain-mediated changes to the anisotropy.

3. Conclusion
This work addresses the magnetostriction in Co/Ni multilayers grown on piezoelectric PMN-PT and PZT substrates. In conjunction with the tunable magnetization and interfacial perpendicular anisotropy explored previously in Co/Ni multilayers, the saturation magnetostriction also varies strongly with Co-to-Ni content, for which the most Co-rich multilayers exhibit the largest value for $\lambda^{111}_s = (-51 \pm 3) \times 10^{-6}$ as measured along the out-of-plane direction. In contrast to earlier predictions, Co/Ni multilayers exhibit negative magnetostriction for both Co-rich and Ni-rich heterostructures, which suggests that fcc-Co complements the negative magnetostriction already evident in fcc-Ni in Co/Ni multilayers. Accounting for the total thickness of each Co/Ni multilayered sample, we find that the saturation magnetostriction increases in inverse proportion to the total thickness, whereby the interfacial component of the magnetostriction exceeds the volume component by more than 300%. This strongly suggests that the negative magnetostriction in Co/Ni multilayers originates at the interfaces or originates from a material property associated with the interfaces, an effect that has been previously forecast, but required the in-situ biased stripline ferromagnetic resonance measurements for direct and unambiguous evaluation of the magnetoelastic anisotropy energy. This approach isolates the interfacial magnetoelastic effect from similarly present effects in Co/Ni multilayers carrying $1/t$ dependence such as the Néel-type interface anisotropy.49, 63

The moderate inverse magnetostriction observed in Co/Ni multilayers also reveals the potential for multiferroic composites in which electric fields that tune a piezoelectric layer can switch the magnetization orientation of Co/Ni. Although the saturation magnetostriction is moderately lower than in Co/Pt, the comparatively low Gilbert damping ($\sim 0.02$, see Supporting Information Figure S11) and tunable demagnetization field could compensate for this to enable fast, energy-efficient magnetization control for electric-field controlled devices.47, 64-65 This has potential applications in voltage-controlled magnetic memory and logic devices, whereby the stability of in-plane magnetization and PMA can be toggled with an electric field applied to a piezostrictive underlayer, and in spin-torque nano-oscillators, in which strain mediated changes in the magnetic anisotropy can be used to modify the oscillator’s operating frequency.

4. Methods

Magnetic multilayered films were obtained by direct current magnetron sputtering on (001)-cut single crystal PMN-PT substrates with dimensions of $4 \times 4 \times 0.5 \text{ mm}^3 (L \times W \times H)$ temperature in a multi-target deposition chamber with a base pressure below $7 \times 10^{-6} \text{ Pa } (5 \times$
$10^{-8}$ Torr). Following a 1 minute clean under low energy Ar$^+$ ion bombardment (500 eV) to remove organic contamination, multilayered films of varying thicknesses were grown with identical seed layers, starting with a 3 nm Ta adhesion layer followed by 2 nm Pt to encourage (111) face-centered-cubic growth of the subsequent film. Five Co/Ni multilayered films were synthesized on PMN-PT: [Co(0.6 nm)/Ni(tNi))$_{x4}$/Co(0.6), in which the Ni thickness was varied between 0.15 nm and 0.9 nm. The films all had identical capping layers, starting with a 2 nm Pt layer with a protective 3 nm Ta capping electrode. Sputtering was carried out at fixed power (40 W) and Ar pressure (0.27 Pa), resulting in sputtering rates of 1.2 nm min$^{-1}$ (Ta), 2.2 nm min$^{-1}$ (Pt), 0.7 nm min$^{-1}$ (Co) and 0.8 nm min$^{-1}$ (Ni), monitored by in-situ quartz crystal oscillators.

Prior to growth of multilayered magnetic film, a back electrode was deposited on the opposite surface of the substrate to apply electric fields. Using an electron beam evaporator with a base pressure of approximately $3 \times 10^{-4}$ Pa, a Ti(2 nm)/Ta(20 nm) bilayer was evaporated at a rate of approximately 6 nm min$^{-1}$. Prior to evaporation, the substrates were cleaned using an O$_2$ plasma clean at 150 W and under elevated temperature (50 ºC).

Samples were characterized by x-ray diffraction (XRD) theta-two theta measurements (D8 DISCOVER, Bruker AXS) to identify the phase and texture of the Co/Ni multilayers. XRD patterns were taken with the Bruker AXS system equipped with a crossed-wire area (two-dimensional) detector.$^{66}$ In the measurement, the scattering intensity was measured over a range of $35^\circ \leq 2\theta \leq 55^\circ$ with the detector held fixed. The resulting two-dimensional spectra were integrated over the $\chi$ (transverse tilt) range of $68^\circ < \chi < 85^\circ$ and $95^\circ < \chi < 112^\circ$ to provide a single graph of integrated intensity versus $\theta$. By avoiding the region around $\chi = 90^\circ$, the intense reflection from the (002) PMN-PT substrate peak is minimized to better examine the weaker intensity peaks from the deposited film. XRD spectra were collected for each sample at ambient temperature using Cu K$\alpha$ radiation for 300 min scan durations.

Cross-section transmission electron microscopy images were obtained in high-resolution mode (bright-field, parallel illumination, no aperture) using a 300 kV instrument. Initially, we aligned the PMN-PT [001] zone axis with the microscope optic axis. Subsequent image calibration was provided by the PMN-PT lattice parameter.

Ferroelectric properties of the PMN-PT substrates were measured with a home-built Sawyer-Tower system with a linear variable displacement transducer. A Trek 10 kV amplifier
was used to provide electric field and a lock-in amplifier (Stanford Research Systems SR830) was used to record the displacement.

The static and magnetic properties of the Co/Ni multilayers were characterized by vibrating sample magnetometry (VSM, MicroSense) and ferromagnetic resonance spectroscopy (FMR). Magnetization versus applied magnetic field measurements were measured using VSM to determine saturation magnetization and to identify easy and hard axes of magnetization. FMR spectroscopy was conducted on a grounded coplanar waveguide (GCPWG), implementing a 50 GHz zero-bias diode detector (Krytar) and a 50 GHz radio frequency current source (Agilent HP86530, Keysight). Using a fixed current amplitude ($I_{pk} = 0.4$ mA) and varied frequencies, the magnetic field from a dc electromagnet was swept between 0.003 T and 1.2 T. A pair of secondary coils embedded in the dc electromagnet provides a low frequency alternating field ($f = 377$ Hz, $\mu_0 H_{pk} = 1.0$ mT) for lock-in detection of the differential power absorbed by the GCPWG and sample as the dc magnetic field is swept. The applied dc and ac fields were monitored with a Hall probe sensor. In-situ electric biasing was achieved by applying a voltage between 20 V and 400 V on the back electrode of the substrate referenced to GCPWG ground plane (also common to the Co/Ni multilayers film surface of the ferroelectric substrate).

5. Supporting Information

Derivation of the saturation magnetostriction from electric-field induced changes to the ferromagnetic resonance field in [Co/Ni]//PMN-PT heterostructures, supporting data for the effective demagnetization versus applied electric field for four additional Co/Ni multilayers samples, estimation of the perpendicular magnetic anisotropy versus Ni thickness from frequency versus ferromagnetic resonance field spectra, magnetization versus applied magnetic field for Co/Ni multilayers samples and Gilbert damping of Co/Ni multilayers on PMN-PT versus Ni thickness.

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Figure 1. X-ray diffraction of $[\text{Co}(0.6 \text{ nm})/\text{Ni}]$ multilayered samples on PMN-PT with two different Ni thicknesses: (a) 0.9 nm and (b) 0.45 nm.
**Figure 2.** (a) TEM image of the microstructure of a [Co(0.6 nm)/Ni(0.9 nm)]₄₄ multilayers sample on PMN-PT, with included fast fourier transforms in regions associated with Co/Ni multilayers (b) and the PMN-PT substrate (c).
Figure 3. (a) Schematic of (Co/Ni)/PMN-PT multiferroic heterostructure. (b) Out-of-Plane (⊥) Strain versus Electric field curve for [001]-cut PMN-PT substrate before poling (black curve) and after poling (red curve) as measured by a Linear Voltage Displacement Transducer. (c) Depiction of broadband ferromagnetic resonance spectroscopy with *in-situ* electric biasing of multiferroic heterostructure in (a) for which the top (3 nm thick) Ta layer is facing the grounded coplanar waveguide (CPW) and the bottom (20 nm thick) Ta layer is facing upward and connected to high voltage supply. The applied static field is in the plane and perpendicular to the *rf* field from the CPW. (d) Example ferromagnetic resonance (FMR) spectra at 18 GHz for a [Co(0.6)/Ni(0.9)]₄ multilayers sample on PMN-PT showing electric field modification of FMR field.
Figure 4. (a) Schematic representation of broadband ferromagnetic resonance spectroscopy with *in-situ* electric biasing of a piezoelectric substrate. Series with different colors represent different excitation frequencies. (b) Frequency versus resonance field spectra for $[\text{Co}(0.6)/\text{Ni}(0.9)]_{x4}$ multilayers sample on PMN-PT with zero electric field. (c) Resonance field versus electric field generated in PMN-PT sample in (b) at 18 GHz. To account for hysteresis in ferroelectric PMN-PT, increasing (decreasing) electric field hysteresis branches were designated using red (blue) arrows and markers. (d) Effective demagnetizing field versus PMN-PT electric field, estimated from results of (b) and (c). Error bars in (c) and (d) reflect the measured field uncertainty and standard uncertainty from a least-squares fit to a Lorentzian model for the ferromagnetic resonance field. The maximum anisotropy change is indicated by the line with the two-pointed arrows.
Figure 5. Estimated Saturation Magnetostriction (open red circles) along (111) versus total inverse thickness for \([\text{Co}(0.6 \text{ nm})/\text{Ni}(t_{\text{Ni}} \text{ nm})]_4/\text{Co}(0.6 \text{ nm})\) from electrically-biased, broadband ferromagnetic resonance spectra with linear fit (solid red line) to the data and comparison to the bulk saturation magnetostriction (dashed blue line) of Ni(111). Error bars reflect the one-sigma standard error of the estimated magnetostriction, derived from uncertainty estimates for the strain, the change in the effective demagnetization and the saturation magnetization.