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Title: Stress induced domain wall motion in FeCo based magnetic microwires for realization of energy harvesting

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Abstract

Energy harvesting is getting significant interest due to the emergence of internet-of-things (IoT). As IoT devices are required to generate power on their own, several researchers are studying ferroelectric materials for voltage generation. However, ferroelectric materials suffer from high resistance at low frequencies, which reduces the output power. Domain wall propagation in ferromagnetic materials, as induced by stress and a pick-up voltage using coils, has been investigated as an alternate form of energy harvesting. Such studies which were reported in multiferroic structures featured a voltage applied to induce the stress, which defeats the purpose of self-power generation. In this article, power generation from mechanical vibrations in purely ferromagnetic structures is shown. These results are achieved by showing that the domain walls can be moved entirely by stress in a tri-layer stack of ferromagnetic microwires. The use of flexible substrates with low Young's modulus and a trilayer magnetic stack enabled the achievement of significant magnetization rotation or domain wall motion even from ambient vibrations. Here, the rotation of magnetization or

domain wall motion is exploited to induce voltages in the pickup coils. The results shown in this paper provide an alternative way to power IoT devices.

1. Introduction

Energy harvesting gets further significance with the advent of Internet of things (IoT) devices, which need to harvest their own energy. Ferroelectric materials, which have been investigated intensively, suffer from low output power due to their high resistance^[2-6]. Hence, researchers are exploring alternative materials for application in energy harvesting devices. In the field of spintronics, the motion of domain walls in nanoscale structures has been demonstrated by using spin-polarized currents and external magnetic field^[7-13]. Domain wall movement is useful for magnetic memory applications, where the domains store information states and the information is read by moving them^[11, 14-18]. Domain wall movement can also be explored for energy harvesting by converting stress into changes in magnetization and thus into electrical voltage. In the energy harvesting investigations reported so far, domain wall motion is accomplished only in multiferroic structures, where stress was induced by providing an electrical energy to the ferroelectric layers^[19-25]. Employing significant amounts of electrical energy, however, defeats the purpose of energy harvesting. Moreover, ceramic ferroelectric layers, like PZT, require high temperature deposition or post deposition annealing (>400 °C) under severe oxidation conditions, which inevitably degrade the magnetic properties^[26]. Therefore, stress induced motion of domain walls in pure magnetic films without the use of ferroelectric layers and a supplied energy is a better approach for self-power generation. In the past, amorphous magnetic materials have been studied for domain wall dynamics under stress^[27, 28]. Pickup of voltage using a pick-up coil was used for sensing the domain wall motion^[29-31]. An analytical model for stress-induced transverse domain wall movement in ferromagnetic nanostripe has also been proposed^[32]. In this article, we report that the domain walls in magnetic microwires made of crystalline ferromagnetic films and lithography technique can

be moved by merely applying mechanical stress. The key recipes for the successful observation of such a domain wall movement are: (i) a magnetic stack of CoNi/Fe₆₅Co₃₅/CoNi with which we have controlled the microstructures (see supporting information for more details about the material stack), (ii) an induced magnetic easy axis that we set by the direction of fringing magnetic field during the deposition (see supporting information) and (iii) the use of flexible substrates with which we could enhance the stress delivered to the device from ambient sources, which is otherwise impossible with rigid substrates like Si. With a prototype device, we have shown in this article, the motion of domain walls under applied stress and have harvested energy.

2. Results and Discussion

Figure 1. Domain evolution of a magnetostrictive magnetic microwire under stress (micromagnetic simulations). a) Domains of an as-deposited magnetostrictive magnet, with an anisotropy easy axis transverse to the length of the wire. b), c) d), e), f), Motion of domain walls under applied stress. The stress introduced to the microwires was modelled by increasing the magnetic anisotropy along the length of wires. b) and c) Widening of domain walls when a stress equivalent to 10% (1b) and 20% (1c) of anisotropy is applied along the

length of wires. d) Tilting of domain walls due to rotation of magnetization resulting in reduction of domain width when the increased anisotropy is 40% of as-deposited anisotropy. e) Expansion of domains, for stress anisotropy value equivalent to 80% of the as-deposited anisotropy. f) Diminishing of domain walls when the increased anisotropy along the length is equivalent to the as-deposited anisotropy (see supporting information).

In Figure 1, we summarize the micromagnetic simulations that motivated our experiments. For this simulation, we chose the material parameters corresponding to $\text{Fe}_{65}\text{Co}_{35}$ alloy. FeCo based materials have been reported for various applications due to their magnetostrictive properties^[33-37]. Our chosen alloy composition exhibits the optimum values of high saturation magnetization (which results in a higher flux) and a reasonably high magnetostriction (which leads to a large change in magnetization for small values of stress)^[38], which are key ingredients for harvesting energy from time-varying stress. We simulated a microwire with an induced magnetic anisotropy (K_u) in the transverse direction. In figure 1a, we show the domain patterns when no stress was applied. The magnetization lies along the transverse direction (set as easy axis of induced magnetic anisotropy) to the length of the wire. The domain walls lie parallel to the direction of easy axis. In order to simulate the stress applied along the hard axis, we increased the anisotropy along the hard axis (see supporting information). When such a stress was applied along the hard axis (figure 1b to 1f), the size of the domains varied, indicating a movement of the domain walls from their original positions. When stress applied along the hard axis increased (up to about 20% of K_u), the domains expanded (figure 1b and 1c). For large values of stress (100% of K_u), the domain walls disappeared, and the magnetization was found to be parallel to the hard axis.

For the intermediate values of stress anisotropy (between 20% of K_u to 40% of K_u), the domain width reduced with further increase in stress. Under this circumstance, the

magnetization and the domain walls were found to be tilted at an angle, resulting in a denser packing of domains in the wire (figure 1d). Nonetheless, with further increase in stress along the hard axis, multiple smaller domains rotate in to single direction forming large size domains (figure 1e). Further increase in the stress value resulted in a total diminishing of domain walls, bringing the microwire to a single domain state (figure 1e). Such domain wall motion with applied stress is promising for energy harvesting applications, if the stress varies as a function of time (e.g., ambient vibrations). The flux changes associated with the domain wall motion would induce a voltage in the pickup coils (Lenz Law). Therefore, this observation warranted further probing and we decided to carry out experimental investigations.

To demonstrate the stress induced domain wall motion experimentally, we sputter deposited $\text{Fe}_{65}\text{Co}_{35}$ thin film (henceforth, referred to as FeCo) in our first attempt (see experimental section). Figure 2b shows the hysteresis loop of the as-deposited film. Noticeably, there is no preferred magnetic anisotropy. In contrast, our simulation indicates that the presence of an anisotropy is a necessary condition to achieve stress-induced domain wall motion for harvesting energy. A close look at the hysteresis loops indicates that the coercivity of the films (80 Oe) is comparable to the anisotropy field (H_K) of the FeCo films (~ 100 Oe). Nevertheless, to achieve preferred anisotropy in the microwires, it was required that the coercivity must be considerably lower than the anisotropy field. Therefore, we decided to understand the origin of the higher coercivity and attempted to reduce it.

Figure 2c shows the scanning electron microscope (SEM) image of FeCo grains. We noticed that the grain size of FeCo was large (~ 30 nm) and speculated this as the reason for high coercivity (see supporting information). Therefore, we decided to decrease the coercivity of FeCo by reducing the grain size of FeCo ^[39] (see supporting information). We carried out a series of investigations to make use of underlayers (figure 2f) to tailor the grain size of FeCo

thin film, in order to tailor the anisotropy. We observed that thin underlayers such as Co or Ni ($\sim 3\text{nm}$) help in reducing the grain size and eventually chose CoNi underlayer to achieve an anisotropy. Figure 2e shows the hysteresis loop of a sample of the type “substrate/CoNi(3 nm)/FeCo(30 nm)/CoNi(3 nm)”. In this structure, top CoNi layer was deposited to protect FeCo from getting oxidized^[40]. The bottom CoNi layer plays a crucial role in reducing the coercivity through a reduction of grain size (see supporting information). As a result, we were able to notice the required magnetic anisotropy with the sandwiched structure (figure 2e), where an easy axis was set along the transverse direction of microwire by a fringing magnetic field between the two targets of the facing-targets sputtering system. Since, CoNi is a magnetic material, it may be argued that the magnetic anisotropy in the trilayer stack arises due to the magnetic anisotropy of CoNi. To eliminate this speculation, we measured the hysteresis loop of CoNi layer which showed that CoNi layer lacks magnetic anisotropy (supporting information). Hence, it corroborates the fact that the magnetic anisotropy in trilayer was achieved only through the reduction in the grain size of FeCo.

Figure 2. Hysteresis loops in the easy and hard axis and scanning electron microscope images of FeCo with different underlayers. a) and d) 0° represents the direction of the easy axis set by

the fringing magnetic field during deposition and 90° refers the direction transverse to the easy axis, both in film-plane. b) Hysteresis loops of the single-layered FeCo. The loop does not exhibit in-plane magnetic anisotropy. c) Scanning electron microscope image depicting grains of FeCo deposited without any underlayer. e) Hysteresis loops of the trilayer sample (CoNi/FeCo/CoNi) (shown in d), which exhibit a magnetic anisotropy. f) Scanning electron microscope image of FeCo deposited on Co and Ni underlayer. The grain size of FeCo with underlayers Co and Ni (2f) is smaller than the grain size of FeCo deposited without any underlayer (2c).

To observe the effect of the externally induced stress on domain wall motion, we fabricated microwires (see experimental section) and observed bitter patterns using a microscope to visualize the domain wall motion (figure 3b). This direction of easy magnetization was set by the direction of fringing magnetic field in the facing targets sputtering. The inset shows a close-up view of the domain patterns. In order to see the effect of applied stress on the deposited microwires, we used the apparatus shown in figure 3c (for more details see supporting information).

We observed an expansion of domain size (figure 3d and 3e) under externally applied compressive stress, which substantiates the results from simulations. When we applied more stress, the domain walls started diminishing (figure 3f), similar to our observations in simulations (figure 1e). Under much higher values of applied stress, the domain walls diminished as shown in figure 3g, quite similar to the simulated result in figure 1f. Surprisingly, we did not observe any tilting of domains experimentally, which was observed in simulations in figure 1d. In the experiments, the stress applied to the microwires during the process of taking images was static and varied in steps. The step size cannot be reduced. We speculate the tilting of domains might have occurred in-between two steps, which did not fall within the experimental observation window.

In figure 3h, we show the measured values of the average width of domains (for $25\text{ }\mu\text{m}$ wide micro-wire) as a function of externally applied compressive stress. We noticed that the

domain width increased with applied stress until about 2 GPa and from a stress of about 2.36 GPa, the domain walls started diminishing. This observation made the domain width unmeasurable for high stress values. The diminishing of domain walls with the increase of externally applied stress is speculated to be due to the change in direction and strength of the magnetic anisotropy under the applied stress. When the applied stress was removed, this stress-induced anisotropy vanishes, and the domains appear again with multiple domain walls. These observations indicate that the bending and unbending of the sample due to ambient sources of vibration will result in domain wall motion. The resultant change in magnetic flux due to this domain wall motion can be converted into electrical voltage using pick-up coils. Devices based on this phenomenon can be built to convert ambient forces to harvest energy.

Figure 3. Change of magnetic anisotropy of the micro-wires, as observed by a change in the domain wall positions using Bitter Patterns. The insets show zoomed-in views of the domains. a) Microscope image of as-deposited microwires. No mechanical roughness was observed on the edges of microwires. b) Domain walls in an as-deposited micro-wire which shows domains orientation transverse to length of the wires. Roughness seen on edges of microwire is magnetic roughness arising due to bitter pattern technique. c) Schematics of apparatus used to apply the stress on the as-deposited microwires. d), e), f) and g), Domain patterns under compressive stress. d) and e) Widening of domains under strain. f) Onset of diminishing of domain walls under higher value of stress. g) Diminished domain walls in a microwire. (Stress was calculated by using the formula mentioned in the supplementary information) h) Plot of domain width as function of applied stress.

From the observations that have been made so far, we speculated that the difference in the patterns of FeCo microwires under compressive stress is due to the changes in the easy axis of the anisotropy directions. To understand this further and to confirm our speculation, we measured hysteresis loops of CoNi/FeCo/CoNi thin film stack under compressive and tensile stress using a micro-Magneto-Optic Kerr Effect (MOKE) magnetometer. In both cases, we measured the loops along the easy axis (direction set by the sputtering process) and the hard axis (shown by pink and purple arrows respectively in figure 4a and 4d).

Figure 4. Hysteresis loops of films of CoNi/FeCo/CoNi measured using a Micro-MOKE. a) and d) Color code for direction of the measured MOKE hysteresis loops for no stress and under stress samples. Pink represents the easy direction set by the magnetic field and Purple, the transverse direction. b) and c) Hysteresis loops of trilayer stack along easy axis and hard axis respectively, without the application of any stress. e) and f) Hysteresis loops of trilayer stack along easy axis and hard axis respectively, under compressive stress. The bi-directional arrows in the plots represent the direction of applied external magnetic field. g) The bending method which was used to apply external stress during the Micro-MOKE measurements. The stress was applied along the hard axis by bending the samples over a curved surface of $R = 16$ mm along the hard axis. h) Hysteresis loops of sandwiched stack under different stresses, T – tensile stress, C – Compressive stress and for different radii of curvature (16 mm and 18 mm). Stress was applied in the magnetic hard axis direction of the samples (check supporting information for more details).

Figures 4b and 4c show the hysteresis loops of as-deposited FeCo sandwiched stack, where the FeCo layer was 40 nm thick, under no applied stress. During this measurement, we

applied magnetic field along the easy axis and hard axis respectively. These loops resemble those plotted in figure 2 (measured using vibrating sample magnetometer) and validate the effectiveness of micro-MOKE in measuring these properties. To study the effect of stress on the magnetization curves, we applied stress along the hard axis. Figures 4c and 4f show the hysteresis loops of the FeCo stack under externally applied stress. We can clearly observe a change of magnetization direction from the easy axis (set by fringing field) to the hard axis (due to the change in direction of magnetic anisotropy). The change in the direction of magnetization resulted into a rotation of magnetization in FeCo stack under externally applied stress, which was also predicted in the simulations. These results indicate that the applied stress changes the anisotropy direction, leading to a change in the direction of magnetization resulting in the motion of domain walls, which is exhibited in the figure 3 for microwires. Similar trend for domain walls under external stress were also observed in FeCo amorphous materials ^[29, 31].

To study the effect of different types of stress on the anisotropy of the stack, we measured the hysteresis loops of the trilayer stack under different types and values of applied stress using a micro-MOKE. Figure 4g shows the change in the hysteresis loops of FeCo stack under compressive and tensile stress, along with their magnitudes. The changes in the anisotropy directions with compressed stress are clear from this figure. It must be noticed that the change in the direction of anisotropy results into a change in the coercivity of material. The extent of the change in coercivity varies with the value of applied external stress ^[31]. In the case of a tensile stress, for unknown reasons, the easy axis remained unchanged. We did not see any significant changes in the coercivity for externally applied tensile stress. This result is in contrast with a drastic change reported in stress-annealed $\text{Fe}_{69}\text{B}_{12}\text{Si}_{14}\text{C}_5$ ^[28]. In the case of a compressive stress, the easy axis set by the sputtering process became the hard axis and hence, the loops exhibited a lower remanence. A field of more than 100 Oe was required to

bring the magnetization along the applied field direction. Similar results exhibiting the interchange between hard axis and easy axis are reported in some previous studies [27, 28].

Figure 5. Voltage harvesting from sandwiched FeCo microwires. a) A micrograph of a device used for energy harvesting and the schematic illustration of the method used to vibrate the device. b) Plot of waveforms obtained from the device shown in a) over a duration of 50 ms. c) Hysteresis loop of the trilayer stack with different thicknesses of FeCo layer. d) Schematic of energy harvesting from the trilayer stack. e) Plot of harvested voltage from the trilayer stack in a time span of 2.4 ms. f) M_t of trilayer stack of different thickness and the total energy harvested in the time span of 50 μs.

To show that the results demonstrated in this study can be used to harvest voltage for realization of this science in devices, we investigated the voltage harvesting performance of trilayer stack microwire deposited on flexible substrate. A preliminary device (figure 5a), consisting of microwire and pickup coil was fabricated (see supporting information). Acoustic waves from a speaker were used to vibrate the shown device. These acoustic waves, in a laboratory setting, mimic the ambient vibrations produced in real world applications. The waveform produced by the pick-up coil is shown in figure 5b. A sinusoidal waveform with a peak voltage of the order of mV in a 50 Ω load was observed, which clearly shows the promise of such a device for energy harvesting applications.

In energy harvesting devices based on this new mechanism, the flux produced will be proportional to the $M_r t$ of the device, where M_r denotes the remanent magnetization and t , the thickness of the film. Therefore, in order to increase the output signal, it is necessary to increase the thickness. To study this aspect in detail, we carried out a study on FeCo thickness dependent energy harvesting. We deposited trillayer stack thin films of $1\text{ cm} \times 1\text{ cm}$ with different thicknesses of FeCo (for more details see supporting information). Figure 5c shows the hysteresis of two film stacks with extreme thickness values of FeCo thin film in this study, indicating that the required anisotropy is achieved at the studied thickness values. For this study, we mimicked the ambient vibration from environment by using a piezoelectric stage to vibrate the samples at 3.7kHz. The samples were placed in the vicinity of a pick-up coil. Figure 5e shows the voltage pulses produced by the stack with 50 nm thick FeCo. In figure 5f, we plot the energy from stacks of different thicknesses, for an average time span of 50 μs . We noticed that the harvested energy increases linearly with the $M_r t$.

Although we show the feasibility of this study in energy harvesting, it must be noted that this article emphasizes a new observation in magnetism and focuses on energy harvesting using domain wall motion as a potential application. Increasing the density of the fabricated microwires and the thickness of the films to an optimum value is expected to increase the energy to potentially higher values. As a result, the reported results are useful in certain devices where solar power cannot be used.

3. Conclusion

We have demonstrated energy harvesting by the movement of domain walls in magnetic microwires entirely by stress. For this purpose, we have fabricated a trilayer magnetic stack that exhibits stress-dependent magnetic easy axis direction. We have made the easy axis to lie along the transverse direction of the microwires by sputtering. By applying stress, we have

demonstrated that the microwire magnetization axis can be changed from hard axis to easy axis. As a result, domain wall motion or collapse of domain walls occurs entirely by stress induced from the bending of substrate. Moreover, no damage to the samples was observed due to the induced stress. We have also demonstrated energy generation in the order of μJ in time span of $50\ \mu\text{s}$ using the deposited tri-layer magnetic stacks. It is to be noted that in the above experiment we only used a single microwire in the preliminary device to harvest energy. We speculate that by fabricating more microwires at an optimize distance, avoiding the magnetostatic interactions, a device with high density microwires can be fabricated. Hence, in future, by optimizing the layer stack, thickness, density of the microwires and the circuitry to generate voltage, further gain in power density can be made. This provides an opportunity for more scientific investigations and potential use as power source in IoT applications.

4. Experimental Section

We performed micromagnetic simulations in this study using the MuMax3 software^[41]. The simulated model was a nanotrack of dimension $7500\ \text{nm} \times 300\ \text{nm} \times 30\ \text{nm}$ with periodic boundary conditions in the x direction. The cell size was set as $5\ \text{nm} \times 5\ \text{nm} \times 5\ \text{nm}$. The exchange constant and saturation magnetization were fixed at $23\ \text{pJ/m}$ and $1950\ \text{kA/m}$, respectively. The easy-axis direction of the uniaxial magnetic anisotropy was set along the y direction, that is, (010). To study the effect of the external applied stress, we increased the anisotropy (K), which was varied from zero to $300\ \text{kJ/m}^3$ in x direction according to the relation $K = 3\sigma\lambda/2$ ^[42, 43]. More information regarding the simulation is provided in the

supporting information. The symbols σ and λ correspond to the applied stress and magnetostriction constant ($\sim 5.8 \times 10^{-5}$) of microwires, respectively^[38].

The experimentally studied samples were made as a stack of CoNi (3 nm)/FeCo (30 nm)/CoNi (3 nm) thin-films, which were grown by d.c. magnetron –facing targets sputtering at room temperature in an Ar pressure of about 0.24 Pa on two different substrates: glass substrates and 85 μm -thick Kapton (a flexible polyimide film) membranes^[44]. The use of facing targets sputtering facilitated the fringing magnetic field to achieve field-induced anisotropy in deposited magnetic film stack (see supporting information). The samples grown on Kapton membrane were used for studying the stress-dependent magnetic properties of thin-films stack and those on glass substrate were used for studying the properties under ambient conditions. We confirmed the induced in-plane axial magnetization anisotropy by measuring the magnetic properties of deposited samples with the vibrating sample magnetometer. In order to study the effect of trilayer on the structural properties, we also carried out the X-rays Diffraction (XRD) of FeCo, CoNi/FeCo/CoNi, and CoNi films (see supporting information).

We fabricated the CoNi (3 nm)/FeCo (30 nm)/CoNi (3 nm) microwires of varying width (4 μm to 40 μm), and of length 1mm on the Kapton (flexible polyimide) membrane using photolithography to study the effect of the applied external stress. For this purpose, a photoresist was spin coated on the membrane, exposed with a laser beam and developed to form trenches in the shape of microwires of defined dimensions. Subsequently, the trilayer magnetic stack was sputter deposited on the membrane using facing targets sputtering. Finally, isolated microwires were achieved by treating the sample with a resist remover to lift-off the unwanted deposited material. Bitter pattern technique^[45, 46] with a Kerr microscope was used to observe the effect of applied stress on the deposited thin-films stack. We applied

the stress in this case by holding one side of Kapton membrane strip firm and bending the other.

Figure 6. Schematics of steps involved in fabrication of the preliminary device. a) Coil shape trenches were formed using photolithography and Ta/Cu/Ti was sputter deposited consecutively. A lift off process was carried out to get the final pattern. b) An insulating layer of SiO_2 was deposited on the coil stack using photolithography. c) Trilayer magnetic stack was deposited on the SiO_2 layer. d) A second layer of SiO_2 was deposited atop of the trilayer stack to prevent from short circuiting. e) Final layer of the coil stack was deposited on the sample. Electrodes were connected to a resistive load and oscilloscope to measure the voltage.

To compare the changes in the magnetic properties of trilayer stack under ambient and applied stress, we used a custom made micro-MOKE ^[47]. The application of stress was done by pasting the samples grown on flexible membrane to the curved surfaces. We used concave or convex curved surfaces with the radii of curvature of 16 mm and 18 mm to measure the effect of stress under different compressive and tensile stresses, respectively. For this study,

the stress was applied on hard axis (by bending the sample along the length of microwire), i.e. in direction transverse to the direction of induced easy axis (transverse to the length of microwire). It must be noticed thickness of CoNi is only 3 nm as compared to the 30 nm thick FeCo. The CoNi was used to reduce the coercivity of FeCo to get magnetic anisotropy in the layered structure. Therefore, the magnetostrictive properties of CoNi was ignored during the studies.

To exhibit the potential of our reported science as an alternating approach to replace the power source for IoT devices, we fabricated a preliminary device. Figure 6 show the schematics of steps involved in the fabrication of the device. We spin coated the polyimide membrane with photoresist and used photolithography to make patterned trenches of coil shown in the figure 6a). Then we sputter deposited Ta(10 nm)/Cu(50 nm)/Ti(10 nm) on the sample. A lift off process was carried out to form patterned coil shape of one side (figure 6b). The photoresist was coated again and etched out using photolithography exposing the inner area, where 100 nm thick insulating layer of SiO₂ was deposited using RF sputtering (figure 6b). A lift off process was again carried out. Similarly, the trilayer magnetic stack (figure 6d) and successively another layer of SiO₂ was deposited (figure 6d). Figure 6e shows the cross section of device fabricated so far. To finish the fabrication of the device, sample was again coated with photoresist and etched using photolithography to deposit the other side of coil stack as shown in figure 6e. The fabricated device was then connected to a 50 Ω resistive load and an oscilloscope for measuring the harvested voltage.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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