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Electric field control on gated Pt/Co/SiO_2 heterostructure with insulating polymer

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Abstract

Due to its low-dissipative nature, electric field control on magnetic materials can greatly improve the energy efficiency of spintronic devices. Here, we demonstrate the use of a polymer, MaN2401 as an insulating layer for electric field control on a Hall cross structure comprising of a Pt/Co/SiO_2 heterostructure. The results show that electric field control is able to tune the coercivity of the magnetic Hall crosses up to percentage changes of −47 and +68%, corresponding to a change in thickness of the Co layer by ~0.4 nm. This provides evidence that validates the mechanism of the magneto-ionic effect from the thickness of the magnetic material instead of detecting the oxidation state at the Co/oxide interface. Dynamic studies conducted on the coercivity of the Hall crosses indicate that a thinner insulating polymer layer produces a higher rate of change in the coercivity of the device. The results presented here offer a fast and repeatable method to observe and study electric field control, paving the way towards the realization of a low-power spintronic device on a flexible substrate.

Keywords: spintronics, magneto-ionic, insulating polymer, electric field control, perpendicular magnetic anistropy, anomalous Hall resistance

S Supplementary material for this article is available online

(Some figures may appear in colour only in the online journal)

1. Introduction

Electric field control of spintronic devices in ferro-metallic materials has garnered a lot of attention from researchers as it opens up the possibility of developing novel magnetic recording devices with low power consumption [1–4]. A spintronics device with a high coercivity (H_c) is excellent for data retention, whereas a low H_c is preferred for data writing by means of magnetization switching. This dilemma between data retention and data writing can be elegantly solved with electric field control by decreasing or increasing the H_c for writing or storing data, respectively. The first experimental demonstration of altering the H_c of a ferro-metallic material resulted in a H_c change of −4.5 and +1% [5]. Since then, electric field control has been demonstrated in various types of spintronic devices. This is seen in magnetic tunnel junctions, where the H_c of a magnetic free layer can be reduced by electric field control before magnetization switching is achieved with lower energy [6–8]. In domain wall (DW) based devices, this can be further utilized in DW propagation [2, 9, 10]. Recently, it has been demonstrated that DWs propagated by spin–orbit torque (SOT) can achieve high DW propagation speeds [11–13]. The physics underpinning the mechanism of SOT [14, 15] is well studied. Hence, an application of electric field can potentially work in tandem with existing DW propagation methods to achieve highs speed DWs with low power consumption [2, 9, 16].

Due to the shielding effect of metals, an electric field can only influence a thin layer of ferro-metallic material by applying a gate voltage across an insulating layer directly on top of the ferro-metallic material. Without using piezo/ferroelectric materials, electric field control has been chiefly demonstrated with two different mechanisms. Firstly, voltage controlled magnetic anisotropy effect arises when the electric field affects the 3*d* orbitals of the ferro-metallic material at the interface, hence, altering the anisotropic properties of the ferro-metallic material [17]. Another approach to achieve electric field control is by the magneto-ionic effect. This is usually realized by altering the oxidation of the magnetic layer at the ferro-metallic/oxide interface which is done by applying a gate voltage [16, 18, 19]. The magneto-ionic approach is the focus of this work, where we investigate the correlation of the electric field and effective Co thickness from the H_c of the devices.

In the current state of art, the insulating layer used in electric field devices consist of high-κ dielectrics such as hafnium oxide [9, 20], zirconium oxide [21, 22] and gadolinium oxide [16, 18, 23] deposited using either atomic layer deposition (ALD), sputtering deposition [16], or pulse laser deposition techniques. Another insulating technique is by using a liquid electrolyte [3, 24]. However, these methods either require a long deposition duration or specialized techniques to encapsulate the liquid electrolyte to make it into a practical device. These issues can be solved by using an insulating polymer (IP) which can be spin coated on to a device. This is a much faster process than any conventional physical or chemical vapour deposition methods. A polymer layer also allows the possibility of future spintronic devices on flexible substrates.

In this work, we demonstrate that electric field control can be achieved with a polymer as the insulating layer. We investigate the H_c of the magnetic Hall crosses by varying the Co thickness (Co_t) and by using electric field control. These results support the mechanism of magneto-ionic effect which has been proposed due to the oxidation at the Co/oxide interface [16, 18, 19, 23, 25]. The results also indicate that a thinner polymer accelerates the magneto-ionic effect, leading to further possible optimization for high speed electric field control. This work provides experimental evidence on the mechanism of magneto-ionic effect and the use of polymer for faster fabrication process in future low-power spintronic devices on flexible substrate.

2. Material and methods

A trilayer stack consisting of Ta (3 nm)/Pt (3 nm)/Co (*t* nm) was grown on thermally oxidised silicon dioxide substrates by magnetron sputtering with a base pressure of $<3 \times 10^{-8}$ Torr. Nominal thicknesses of Co were deposited ranging from 0.8 to 2.3 nm. The Co layer was allowed to naturally oxidise at room temperature for ~10 min before it was capped with 5 nm of SiO_2 deposited by electron-beam evaporation. 500 nm width Hall crosses are fabricated using Ta (3 nm)/Pt (3 nm)/Co (0.9 nm) thin films, a combination of electron beam lithography, and ion-milling followed by lift-off techniques were used to for device patterning. For physical deposition, the deposition rate was calibrated using atomic force microscopy. The thickness of the IP is determined with direct step height measurement also using atomic force microscopy. The IP used in this work is MicroChem MaN2401, a polymer resist for electron beam lithography. The IP used in this work are baked at 100 °C for 1 min after spin coating. Figure 1(a) shows the device under scanning electron microscopy before the gate electrode was deposited. Figure 1(b) shows a schematic diagram of the measurement setup. An insulating layer of MgO was deposited to smoothen the step height from the Hall cross. The electrodes for the gate voltage and at the ends of the Hall cross consist of Ta (5 nm)/Cu (100 nm)/Au (5 nm) and were grown by using magnetron sputtering. All transport measurements done in this work were done at room temperature.

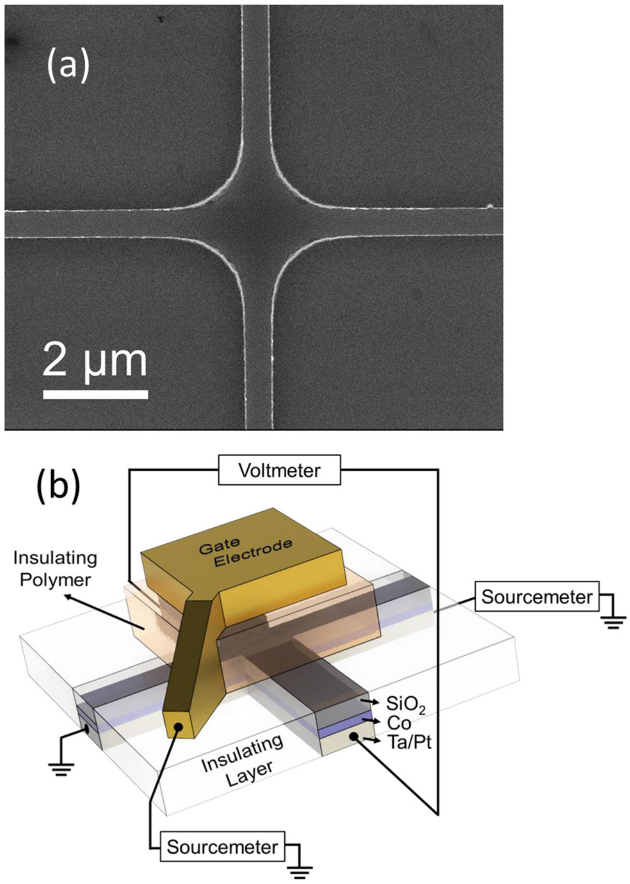

Figure 1. (a) Hall cross device under scanning electron microscpy before deposition of gate electrode. (b) Schematic diagram of Hall cross structure under electrical measurement for electric field control.

3. Results and discussion

Measurements done using vibrating sample magnetometer (VSM) with a sweeping magnetic field out-of-plane to the thin film indicate that the $Ta/Pt/Co/SiO_2$ thin films have perpendicular magnetic anisotropy (PMA). The VSM results are as shown in the insets of figure 2, the inset on the left (right) is the hysteresis loop for Co_t that is ⩽2 nm (⩾2 nm). From the VSM measurements, the H_c of the films is plotted in figure 2. It is evident that the H_c increases with thicker Co_t until it reaches 2 nm. When the Co_t is thicker than 2 nm, the H_c reduces significantly due to significant in-plane magnetic anisotropy from bulk anisotropy of the thick Co_t after the *fcc*/*hcp* growth [26]. The nominal thickness, Co_t presented in

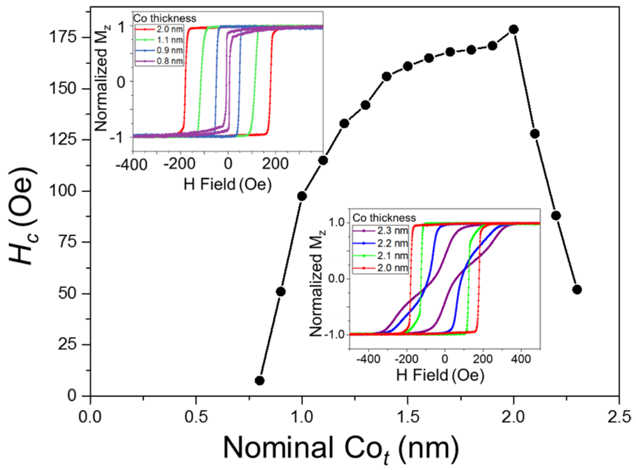

Figure 2. Coercivity of thin film stack Ta (3 nm)/Pt (3 nm)/Co (t)/ SiO_2 (5 nm). Insets shows the hysteresis loops measured by VSM.

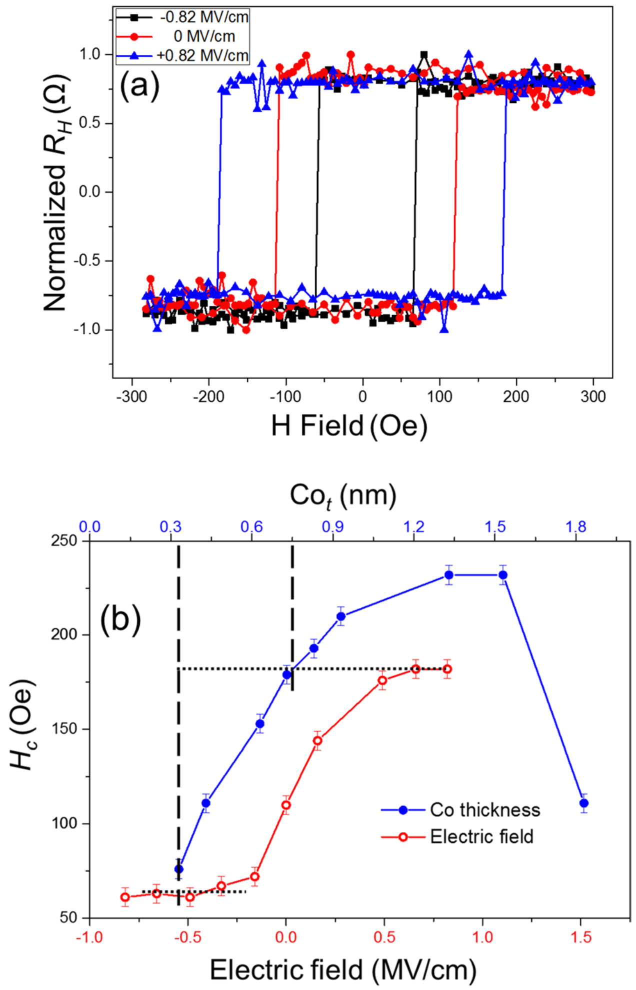

Figure 3. (a) Anomalous Hall effect measurement with a sweeping out-of-plane magnetic field with positive and negative gate voltage applied. (b) Direct comparison of H_c from a device (Co = 0.9 nm) under gate voltage and from devices with various Co thicknesses. The error bars arise from the instrument error of the Hall probe during measurement.

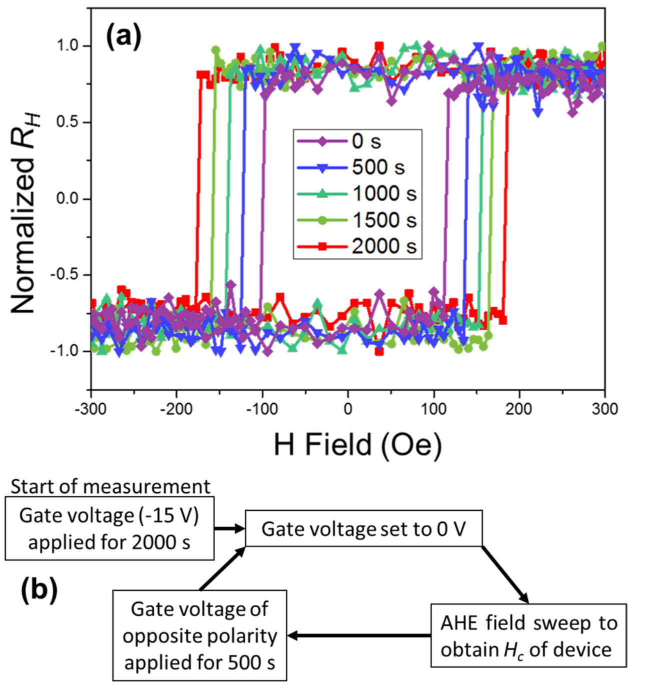

Figure 4. (a) Anomalous Hall effect measurement with applied gate voltage of +15 V (+0.85 MV cm^{-1}) across 2000 s on a 177 nm thick IP. (b) Flowchart of measurement process in figure (a).

figure 2 is an indicative thickness value based on the sputtering deposition rate, it does not derive from precise calibration of Co monolayer change.

An IP was chosen as an insulating layer as its viscosity ensures little to no major defects within the layer. Furthermore, after baking, the IP does not require encapsulation it to remain in place. The Hall cross device were fabricated as mentioned in section 2 and all the Hall resistance (R_H) measurements in this work was done with a sweeping out-of-plane magnetic field. The device has 152 nm of IP and measured in its pristine state without any gate voltage and measured again after applying ±12.5 V (±0.82 MV cm^{-1}) for 2000 s. A low current density (<1 × 10^{10} A m^{-2}) was used in all the R_H measurement in this work to avoid Joule heating which might affect the H_c of the device. The results of the R_H measurements are as shown in figure 3(a).

Figure 3(a) shows that the H_c of the device is larger (smaller) when +12.5 V (−12.5 V) gate voltage was applied. This results in a percentage change of +68 and −47% of the H_c for the negative and positive gate voltage, respectively. The R_H measurements were carried out at zero gate voltage after the application of ±12.5 V, demostrating the nonvolatility of the H_c of the device which lasted for at least ~48 h in room temperature.

Selected thin films of various Co_t were fabricated into 500 nm width Hall crosses and R_H measurements were done to obtain their H_c as shown in figure 3(b) with blue solid data points. Figure S1 (stacks.iop.org/JPhysD/51/365001/mmedia) in the supplementary material shows the full hysteresis measurement of the devices. The magnetic dead layer of the Pt/

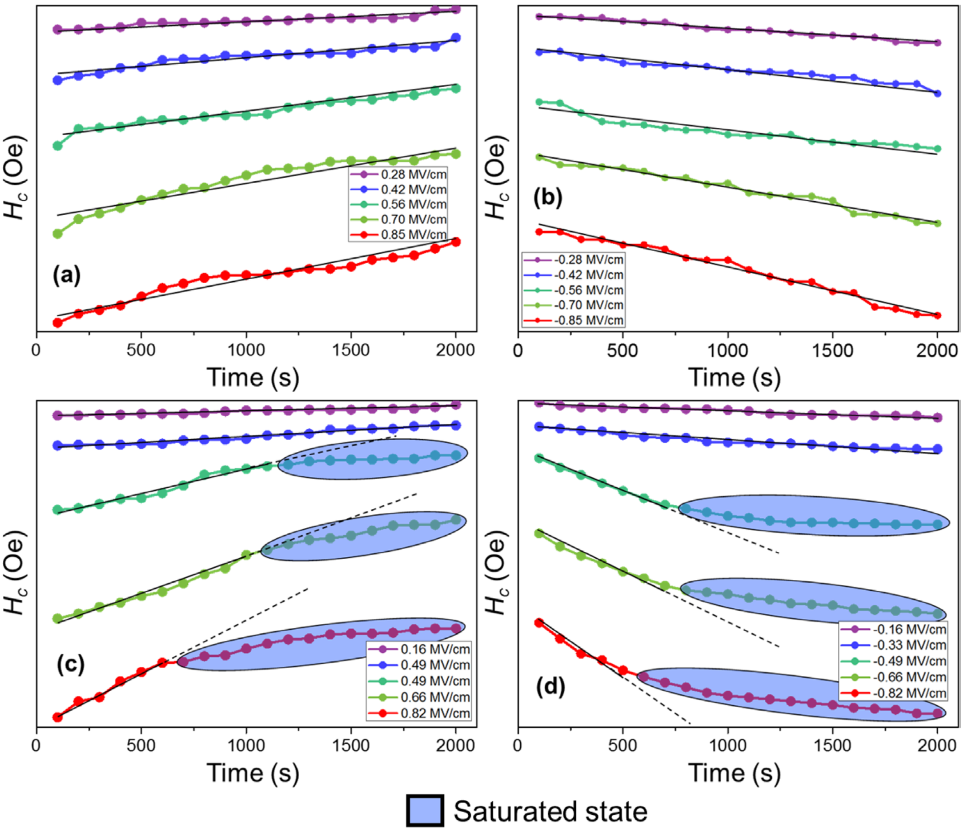

Figure 5. H_c of Hall cross with varying applied electric field, values are offset in the y axis with arbitrary units for visual representation. (a) Positive electric field with 177 nm of IP. (b) Negative electric field with 177 nm of IP. (c) Positive electric field with 152 nm of IP. (d) Negative electric field with 152 nm of IP.

Co/SiO_2 is found to be 0.47 nm from VSM measurements as shown in figure S2 in the supplementary material. This was subtracted from the nominal Co_t to provide the effective Co_t as shown in figure 3(b). The overall increase of the H_c compared to their thin films counterparts can be attributed to the lack of domain nucleation from surface irregularities or physical defects [27].

To compare the effect which the magneto-ionic effect and Co_t has on the device, the H_c change due to these effects are shown in figure 3(b). The range of H_c shared by the device and thin film is demarcated by dotted lines and the corresponding range of Co_t is demarcated by dashed lines. This suggests that the change in H_c of the device due to electric field control comes from a Co_t range of ~0.32 to ~0.75 nm. This supports and directly demonstrates the mechanism of the magneto-ionic effect. It is noted that due to experimental limits, the thinnest effective Co_t with PMA is ~0.32 nm, which resulted in a device H_c of 76 Oe. However, since the smallest H_c obtained by applying a negative electric field is 61 Oe. It is likely that the effective Co_t is thinner than 0.32 nm after the application of the negative electric field. In the supplementary material, figure S3 shows the resistance of the nanowire under various electric field. A thicker effective Co_t results in a lower resistance of the device, which also supports the magneto-ionic mechanism.

Due to the nonvolatility of its H_c, dynamic studies were carried out from a reference state. For the R_H measurement shown in figure 4(a), the reference state is reached when a gate voltage of −15 V (−0.85 MV cm^{-1}) was applied for 2000 s. During the R_H measurement, a gate voltage of opposite polarity, 15 V (0.85 MV cm^{-1}) is applied. This measurement process is illustrated as a flowchart in figure 4(b) as a reference for the measurements done to obtain the results in figure 4(a). Each R_H measurements was carried out after a gate voltage of +15 V (0.85 MV cm^{-1}) was applied for 500 s across a 177 nm thick IP layer. This procedure was repeated four times, achieving a total culmulative period of 2000 s during which the gate voltage was applied. The total culmulative period is not inclusive of the time taken during the R_H measurements with the sweeping magnetic field.

The thickness of the IP was varied by controlling the rotation speed during the spin coating process (2000–7000 RPM). In figures 5(a) and (b), the change of H_c at electric fields between ±0.85 MV cm^{-1} has a linear behaviour. This linear behaviour of the H_c change occurs for IP thickness between 157 to 233 nm for all the gate voltages used in this work. However, with a thinner IP layer of 152 nm and at an electric field of ±0.49, ±0.66 and ±0.82 MV cm^{-1}, the H_c appears to reach a saturation state where the $\frac{dH_c}{dt}$ is significantly different. In figures 5(c) and (d), this saturating behaviour is highlighted in blue, indicating that a shorter time is required to reach saturation with a thinner IP layer. This suggests that the polymer layer acts as a carrier for oxygen migration at the Co/SiO_2 interface, whereby a thinner IP layer proves to be more efficient due to the shorter distance involved. The $\frac{dH_c}{dt}$ at

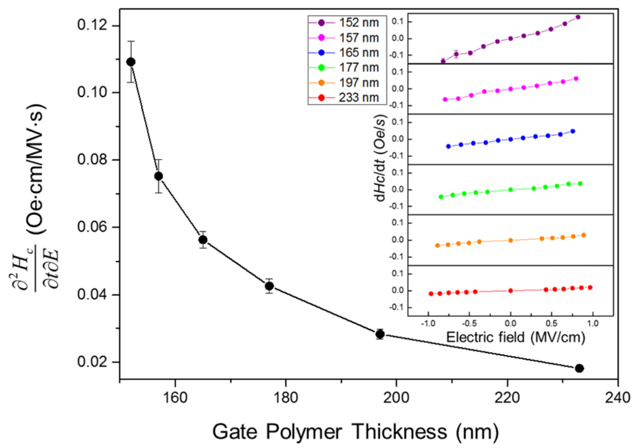

Figure 6. $\frac{\partial^2 H_c}{\partial t \partial E}$ is obtained from the gradient of the inset and plotted against the polymer thickness of the Hall cross device. The error bar reflects the standard error from the fitting of the inset. The value of $\frac{\partial^2 H_c}{\partial t \partial E}$ indicates how the electric field affects the H_c of the Hall cross device. Inset shows the obtained $\frac{dH_c}{dt}$ against electric field for the various polymer from the linear regime as shown in figure 5.

various electric field up to ± 0.96 MV cm^{-1} was obtained for the various IP thickness and the results are shown in figure 6.

In figure 6, the inset shows the obtained $\frac{dH_c}{dt}$ extracted from only the linear regime. The result in figure 6 shows that as the thickness of the IP gets reduced, the effect that the electric field has on the $\frac{dH_c}{dt}$ becomes greater. This trend of increasing $\frac{\partial^2 H_c}{\partial t \partial E}$ with decreasing IP thickness agrees with the observations from figure 5. The increased $\frac{dH_c}{dt}$ due to the thinner IP layer results in a faster saturation of the magneto-ionic effect. Thinner IP layers allow the possibility of optimizing spintronics devices using IP as an insulating layer by improving the spin coating process.

4. Conclusion

In conclusion, the viability of using an IP for electric field control has been demonstrated on nanoscale Hall crosses consisting of a Ta/Pt/Co/SiO_2. The magneto-ionic effect is reversible and has non-volatile retention of H_c. The applied electric field results in a percentage change in H_c of -47 and $+68\%$ which correlates to a Co_t change from 0.3 to 0.75 nm. This direct experimental evidence supports the mechanisms for the magneto-ionic effect. The dynamics of the H_c is characterized and it is indicative that a thinner layer of polymer results in faster magneto-ionic effect for tuning the H_c of the magnetic layer. Using an IP for electric field control of magnetic material provides an alternate approach which potentially has a higher throughput compared to conventional insulation deposition approaches.

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