Ionitronic manipulation of current-induced domain wall motion in synthetic antiferromagnets

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Abstract

The current induced motion of domain walls forms the basis of several advanced spintronic technologies. The most efficient domain wall motion is found in synthetic antiferromagnetic (SAF) structures that are composed of an upper and a lower ferromagnetic layer coupled antiferromagnetically via a thin ruthenium layer. The antiferromagnetic coupling gives rise to a giant exchange torque with which current moves domain walls at maximum velocities when the magnetic moments of the two layers are matched. Here we show that the velocity of domain walls in SAF nanowires can be reversibly tuned by several hundred m/s in a non-volatile manner by ionic liquid gating. Ionic liquid gating results in reversible changes in oxidation of the upper magnetic layer in the SAF over a wide gate-voltage window. This changes the delicate balance in magnetic properties of the SAF and, thereby, results in large changes in the exchange coupling torque and the current-induced domain wall velocity. We also illustrate the potential of ionitronic spintronic logic devices with a 'knot' racetrack device.

Introduction

The current or voltage induced manipulation of magnetization is key to several spintronic technologies today including magnetic random access memories (MRAMs)¹⁻³ and magnetic racetrack memories^{4, 5}. In the racetrack memory, a series of magnetic domain walls, that encode digital data, are manipulated in magnetic nanowires via current pulses^{4, 5}. These nanowires have evolved from in-plane⁶ to out-of-plane⁷ magnetized materials, to heterostructures with strong interfacial spin-orbit coupling^{8, 9}, and, most recently, to synthetic antiferromagnetic (SAF) structures¹⁰. In the latter, the domain walls (DWs) can be moved very efficiently with velocities (v) that can exceed ~ 1 km/s by means of a giant exchange coupling torque (ECT) generated by the current¹⁰. Another means of manipulating magnetization are electric fields from gate voltages applied across insulating layers that have been shown, for example, to tune the perpendicular magnetic anisotropy of magnetic tunnel junctions and, thereby, allow for more energy efficient MRAM¹¹⁻¹³. To date, gate voltage induced DW velocity changes in racetracks formed from ferromagnetic metals are very small due to the limited modifications of, for example, the uniaxial anisotropy energy or interfacial Dzyaloshinskii Moriya interaction^{14-16, 25}. Here we show that, in a SAF structure, in which the lower and upper magnetic (LM and UM) layers are antiferromagnetically coupled via an ultrathin ruthenium layer^{17, 18}, gate voltages applied through ionic liquids can significantly affect the DW velocity with changes of several hundred m/s. We show that this is due to an electro-chemical process that is non-volatile but reversible and results from an enhancement or suppression of oxidation of the UM layer rather than from electrostatic changes in carrier density, as previously claimed^{14-16, 22, 23}.

Manipulation of current induced domain wall motion by ionic liquid gating

SAF structures have the basic structure of UL/LM/Ru/UM/CL where: UL is an underlayer; LM and UM are magnetic sub-layers that are coupled antiferromagnetically through a thin Ru layer, here 5 Å thick, unless otherwise stated; and CL is a capping layer. SAF structures were prepared by high vacuum magnetron DC

sputtering where UL= TaN(20)/Pt(15), LM=[Co(3)/Ni(7)/Co(1.5)], UM=[Co(5)/Ni(7)/Co(1.5)] and CL =TaN(15) (all thicknesses are in Å). Even though the individual layers are atomically thin, cross-section transmission electron microscopy (TEM) imaging shows the layers are well defined (see Fig. 1a). Both the LM and UM layers display perpendicular magnetic anisotropy (PMA), as shown via M - H loops that are measured with an out-of-plane magnetic field (Fig. 2b). Nanowire structures (3 μm wide and 50 μm long) with lateral gate electrodes, as schematically outlined in Fig. 1b, were fabricated using optical lithographic techniques. Current-induced DW motion (CIDWM) measurements under the application of a gate voltage (V_G) applied across an ionic liquid to the SAF racetrack were carried out: typical sequences of four magneto-optical Kerr microscope images of CIDWM corresponding to the successive application of individual 10 ns long pulses (current density $J=1.2 \times 10^8 \text{ A/cm}^2$) are shown in Fig. 1c ($V_G = 0 \text{ V}$, pristine state).

The dependence of DW velocity v on the injected current density under various V_G is shown in Fig. 1d. Firstly, when a positive V_G is applied, the CIDWM efficiency monotonically increases. When a negative V_G is subsequently applied, the CIDWM efficiency returns to its original value at -2 V , and further degrades at -3 V . The dependence of v on V_G at a fixed $J = 1.2 \times 10^8 \text{ A/cm}^2$ is shown in Fig. 1e. A very large change in v of up to 130 m/s is found between V_G of $+4 \text{ V}$ and -3 V in the SAF structure, which is much larger than previous reports of electric field induced changes in DW motion for single ferromagnetic layers^{14-16,25}. Interestingly, the threshold current density, below which the DWs do not move, is also slightly modified by V_G (See Fig. S1 in SI).

The nature of the coupling between the LM and UM layers can be inferred from the dependence of v on a magnetic field H_x that is applied in-plane along the racetrack (Fig. 1f). In these measurements, H_x is varied from -900 to 900 Oe and J is fixed at $1.2 \times 10^8 \text{ A/cm}^2$. In the pristine state, the DW velocity shows a non-monotonic response to the application of H_x for both up-down and down-up DW configurations (Fig. 1f and Fig. S1), which is a distinct character of DWs in a SAF that are driven by an ECT¹⁰. For positive V_G ($+2$ and $+4 \text{ V}$), v is highly sensitive to H_x with pronounced peaks at distinct values of H_x . Such changes are indicative of larger contributions from the ECT. On the other hand, for negative V_G (-2 V), the lower sensitivity of v to H_x provides evidence for a reduced ECT contribution. It is noteworthy that v displays an almost linear response with H_x for $V_G = -3 \text{ V}$. Such a behavior is characteristic of the response expected from a simple ferromagnetic racetrack.

The reversibility of ionic liquid gating effects on CIDWM is demonstrated by comparing data after gate voltages of -3 V and 4 V are applied, as illustrated in Fig. 1g and h. The CIDWM at a given current density ($1.2 \times 10^8 \text{ A/cm}^2$) robustly changes between a relatively low velocity of 110 m/s (-3 V) to a velocity of 240 m/s (at $+4 \text{ V}$) that is more than two times higher (Fig. 1h). This experiment was repeated several times with the same results, clearly showing large reversible and non-volatile changes in the CIDWM induced by ionic liquid gating.

Ionic liquid gating impact on magnetic properties

The non-volatile nature of the ILG effect was further investigated using *ex-situ* vibrating sample magnetometry (VSM) studies of the as-deposited films, as shown in Fig. 2a. The measured M - H loops of samples using an out-of-plane magnetic field for a sequence of different V_G s are plotted in Fig. 2b. Typical SAF-like loops are observed for all V_G except for -3 V. From these M - H loops, M_R/M_S and M_S are extracted and summarized in Fig. 2c, where M_R is the remnant magnetization at $H_Z = 0$, and M_S is the saturation magnetization at $H_Z = 15$ kOe. As V_G is increased from 0 to +4 V, M_R/M_S gradually decreases from 0.113 to 0.0008, while negative V_G results in an increase in M_R/M_S to a value of 1 at $V_G = -3$ V. Meanwhile, $M_S t$, where t is the total thickness of UM+LM, is enhanced from $\sim 1.20 \times 10^{-4}$ emu/cm² in the pristine state to $\sim 1.33 \times 10^{-4}$ emu/cm² for $V_G = +4$ V, but reduced to $\sim 0.666 \times 10^{-4}$ emu/cm² for $V_G = -3$ V.

Thus, the effect of ILG on the CIDWM of SAF structures can be understood as follows: since the ECT-driven CIDWM in the SAF structure depends sensitively on the value of M_R/M_S ¹⁰, the reduced M_R/M_S ratio by applying positive V_G results in an increase in the ECT, and thus, an enhancement of CIDWM as observed. By contrast, negative V_G suppresses v due to a reduced ECT that results from an increase in the M_R/M_S ratio. By further increasing the negative gate voltage, the SAF system is transformed into a FM system and a much reduced v is observed, that results from the vanishing of the ECT in a FM system. The variation of the DW velocity v versus M_R/M_S , at a fixed current density $J = 1.2 \times 10^8$ A/cm², can be well described by a 1D ECT analytical model, thereby substantiating these conclusions (see Supplementary Note 1 of SI for details of 1D model).

Electro-chemical mechanism of ionic liquid gating

The non-volatile characteristic of the ILG effects on the magnetization of the SAF films indicates an electro-chemical origin of the ILG process. In order to clarify this possibility we studied the chemical nature of several SAF structures using X-ray photoelectron spectroscopy (XPS) for various V_G . Since the XPS signal decays exponentially with depth from the surface of the film, the XPS spectra mainly arise from the UM and the TaN capping layer. Co- and Ni-2*p* core-level spectra are shown in Fig. 3a and b, respectively. The most important feature is the evolution of the satellite peaks at the Co- and Ni-2*p*_{3/2} edges. At the pristine state ($V_G = 0$ V), in addition to the dominant Co-2*p*_{3/2} (778.2 eV) and Ni-2*p*_{3/2} (852.6 eV) peaks, weak satellite peaks are observed at 780.7 eV for Co and 855.8 eV for Ni, which are attributed to a tiny degree of oxidation¹⁹⁻²¹. Remarkably, $V_G = +4$ and -3 V suppresses and enhances, respectively, such satellite peaks compared to the pristine sample. This indicates a weakening and strengthening, respectively, of the oxidation of Co and Ni. The reversibility is also manifested by XPS characterization of a sample as the gate voltage was varied from 0 V to +4 V, to -3 V, and back to +4 V. The first data and the last taken at +4 V are almost the same, illustrating the robustness of the reversibility of the gating process.

Considering the experimental findings presented above, we propose the following mechanism for the effect of the ILG. The application of V_G reversibly changes the oxidation state of the UM layer and, thus,

the magnetization of the UM. When the gate voltage reaches -3 V, the UM is heavily oxidized so that the SAF structure is effectively converted into a FM structure. The UM layer has a magnetization that is reduced by over 95%. Limited ionic liquid gate induced oxidation of a single layer FM structure has been reported using HfO_2 capping layers²⁵. However, the ability to tune the CIDWM in SAF structures by ILG is much greater than that previously found for FM layers^{15,16,25}. This is due to the fundamental differences in the primary mechanisms of CIDWM in FM and SAF structures, namely, chiral-SOT in the former, and ECT in the latter. Compared to chiral-SOT, the higher sensitivity of ECT to tiny changes in magnetic properties allows for a much greater impact of ILG in tuning the CIDWM and, thereby, gives rise to much larger changes in DW velocity.

Ionic liquid gate-induced modulation of passage of domain walls in a “knot” device

We give one example of how the large ILG effect on CIDWM that we have found for SAF racetracks can be used in a device. We consider a “knot” racetrack device that consists of a straight wire that is separated into two curved lanes, as depicted in Fig. 4a. In the knot device, an injected DW is split into the two curved lanes and exits the lanes as two individual DWs. We find a distinctly different behavior depending on V_G . At $V_G = -3$ V, as shown in Fig. 4c, when a down/up DW enters the knot device and is split into two individual DWs, the DWs exhibit highly asymmetric behavior: in the lower curved lane the DW shows a faster velocity than in the upper curved lane, and while the DW successfully passes through the lower curved lane into the straight wire region, the DW gets stuck when exiting the upper curved lane. On the other hand, at $V_G = +4$ V, the incoming down/up DW splits into two DWs showing a very symmetric behavior as the two DWs pass through both lanes at almost the same velocity and exit both curved lanes freely (Fig. 4d). For an up/down DW, as shown in Fig. S5 in SI, at $V_G = -3$ V, the DWs also shows an asymmetric characteristic but in an inverted manner; at $V_G = +4$ V, the DW behaves exactly as the down/up case with a symmetric behavior in both lanes.

These results are consistent with the discussion above. As illustrated in the straight wire case, ILG results in the reversible transformation between high ($+4$ V) and low (-3 V) CIDWM velocities, since the nature of the DW varies between a nearly compensated chiral DW to a simple chiral DW. It has previously been shown that DWs in a perfectly compensated SAF structure can move freely regardless of the track geometry, e.g. the curvature of the wires, by current, whereas in FM structures, the DW motion is strongly affected by local geometries and the DW configuration (up/down or down/up)^{26,27}. From this example of a “knot” device, we illustrate that by taking advantage of the large ILG induced effects on CIDWM in SAF structures, these effects have potential for novel logic devices using suitably designed racetrack device geometries.

In summary, we find that ionic liquid gating can be used to manipulate the current-induced domain wall motion in SAF structures with a significant change in DW velocity of up to 130 m/s. We show that the mechanism relies on changes in the exchange-coupling torque in SAF structures, which is fundamentally distinct from previous studies of FM structures^{15, 16, 25}. Such changes are realized by ionic liquid gating

via the manipulation of the magnetic moment of the upper magnetic sub-layer in the SAF structure, via ionic liquid gate induced oxygen ion migration and thereby chemical oxidation and reduction of the magnetic constituents of these layers. We have also demonstrated the possibility of using such an ionic liquid gate control of DW motion in SAF structures to allow for novel DW logic devices.

Methods

Sample preparation. Synthetic antiferromagnetic (SAF) structure of TaN(20)/Pt(15)/Co(3)/Ni(7)/Co(1.5)/Ru(5) /Co(5)/Ni(7)/Co(1.5)/TaN(15) deposited on SiO₂/Si substrates at room temperature by DC magnetron sputtering in a high vacuum deposition system. The deposition rates of the individual sputter sources were calibrated by an *in-situ* quartz crystal microbalance and by *ex-situ* x-ray reflection of individually deposited layers. For CIDWM and transport measurements, the thin films were respectively fabricated into wire structures (2 μm wide and 50 μm long) and Hall bars (effective channel: 100 μm wide and 400 μm long) with lateral Ru(50)/Au(500) gate electrodes, prepared by photolithography, Ar-ion milling, and deposition/lift-off processes. A drop of ionic liquid EMIM-TFSI [1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide] covered the racetrack and part of the gate electrode. V_G was applied between the gate and source electrodes.

Measurement set-up. The CIDWM measurements were carried out using *in-situ* V_G application. V_G was applied for 30 min before each measurement. CIDWM measurements were carried out using Kerr microscopy in a cryostat with a vacuum of $\sim 5 \times 10^{-6}$ mbar. Magnetization measurements were carried out *ex-situ* after removed of the ionic liquid after gating. Then V_G was set to zero and the samples were cleaned using first acetone and second ethanol before the corresponding measurement. This was possible because of the non-volatile ILG effect. Magnetization measurements were carried out in a superconducting quantum interference device-VSM (SQUID-VSM). XPS measurements were carried out in a K-AlphaTM system using an Al-K α X-ray source. The in-situ transport measurements were carried out in a physical property measurement system (PPMS) in a few Torr He. All the measurements were carried out at room temperature.

Declarations

Competing financial interests

The authors declare no competing financial interests.

Author contributions

S.S.P.P. conceived and directed the project. X.Z. and Y.G. prepared the samples and devices, respectively. Y.G. and F.L. carried out the transport, and magnetization, and the Kerr microscopy measurements. F.L. carried out the XPS measurements. T. M. built the Kerr microscope setup for CIDWM. S-H. Y. developed the 1D model. All authors participated in discussing the data and writing the manuscript.

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Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Figures

Figure 1

ILG effect on CIDWM in a SAF structure. a, Cross-section TEM image (left) and schematic structure (right) for a typical SAF structure. b, Sketch of racetrack device with gate electrode. c, Kerr microscopy images of a single DW moving along the nanowire at pristine state ($V_g = 0$ V): the images are taken after applying sequences of current pulses composed of 4'10 ns pulses (current density $J = 1.2 \times 10^8$ A/cm²). Blue and red arrows indicate the current injection direction, and dark and bright contrast indicates down and up

magnetization directions, respectively. d, DW velocity v as a function of current pulse density J at different V_G . e, v at fixed $J = 1.2 \times 10^8$ A/cm² versus V_G , the black arrows indicate the sequence of application of V_G . f, Dependence of v on H_x for various V_G , for down-up DWs at a fixed $J = 1.2 \times 10^8$ A/cm². g, The reversibility of ILG effects on CIDWM of SAF structures is shown by varying the gating voltage from -3 V to $+4$ V several times and the corresponding DW velocity at a given current density of 1.2×10^8 A/cm² for different V_G (h).

Figure 2

ILG induced magnetization changes in SAF structures. a, Schematic illustration of ionic liquid gating of the pristine SAF films; b, Magnetic hysteresis loops obtained from ex-situ SQUID measurements for several V_G applied in the following sequence: 0 V (pristine state), +2 V, +4 V, -2 V and -3 V. c, VG dependence of M_R/M_S (left axis) and MSt (right axis); d, DW velocity at a fixed current density (1.2×10^8 A/cm²) plotted as a function M_R/M_S . The dotted line corresponds to simulation results based on a 1D analytical model.

Figure 3

ILG induced ion migration in SAF structures. X-ray photoelectron spectra (XPS) of a, Co-2p, b, Ni-2p for SAF samples gated at various V_G applied in the following sequence: 0 V (pristine state), +4 V, +4 V $\rightarrow$ -3 V, and +4 V $\rightarrow$ -3 V $\rightarrow$ +4 V.

Figure 4

Knot devices controlled via ILG. a, Sketch of experimental set-up for a knot device controlled by ionic liquid gating. b, Optical micrograph of a typical knot device. Current-induced domain wall motion of a SAF knot device for $V_G =$ c, -3 V, and d, $+4$ V. Current pulses with a magnitude and length of $1.2 \cdot 10^8$ A/cm² and 50 ns, respectively, were used. The injected pulse numbers are labelled at the low right corner and the current DW position is marked with the red dashed line in each figure.

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