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Electric field control of domain wall propagation in Pt/Co/GdOx films

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The influence of a gate voltage on domain wall (DW) propagation is investigated in ultrathin Pt/Co/gadolinium oxide (GdOx) films with perpendicular magnetic anisotropy. The DW propagation field can be enhanced or retarded by an electric field at the Co/GdOx interface and scales linearly with gate voltage up to moderate bias levels. Higher gate voltage levels, corresponding to electric fields >0.2 V/\(\text{nm}\), produce a large irreversible change to the magnetic anisotropy that can enable nonvolatile switching of the coercivity.

Many proposed spintronic devices rely on domain wall (DW) dynamics to achieve logic and nonvolatile memory functionalities.\(^1\)\(^-\)\(^3\) So far, control of DWs in such devices has relied on magnetic fields and current-induced spin transfer torques which are often associated with prohibitively high power consumption. The introduction of voltage gated effects would allow low power operation while adding an additional degree of freedom to enable further device capabilities. Voltage control of the magnetization is most often achieved using complex materials such as multiferroic oxides,\(^4\) magnetic semiconductors,\(^5\)\(^,\)\(^6\) or strain coupled magnetostrictive/piezoelectric composites.\(^7\) However, direct electrical control of magnetic anisotropy\(^8\)\(^-\)\(^11\) has recently been realized in metal ferromagnets at room temperature. This effect has been exploited to realize magnetization switching in magnetic tunnel junctions\(^12\)\(^,\)\(^13\) and nonvolatile control of the magnetization vector in magnetoelectric charge trap memory cells.\(^14\) Control of magnetic anisotropy should also allow for electric field control of DW dynamics,\(^15\) which so far has received comparatively little attention.\(^16\)\(^,\)\(^17\)

In this letter, we report direct electric field control of DW propagation in Pt/Co/gadolinium oxide (GdOx) films with perpendicular magnetic anisotropy (PMA). We show that DW propagation can be enhanced or retarded by an electric field applied at the Co/GdOx interface via modulation of the activation energy barrier that governs DW creep dynamics. By contrast, no clear influence on DW nucleation is observed due to the stochastic nature of nucleation events throughout the film. We further identify two regimes of electric field effects in the Co/GdOx system. In the low-field regime (<0.2 V/\(\text{nm}\)), the DW propagation field varies linearly and reversibly with the applied electric field. However, higher electric fields (0.2–0.4 V/\(\text{nm}\)) produce a large irreversible change to the magnetic anisotropy that enables nonvolatile switching of the coercivity of the Co film.

Films were prepared by dc magnetron sputtering at room temperature under 3 mTorr Ar with a background pressure of 1 × 10\(^{-7}\) Torr. A schematic of the sample structure is shown in Fig. 1(a). First, a continuous Ta(4 nm)/Pt(3 nm)/Co(1 nm)/GdOx(40 nm) film was deposited on a Si(100) substrate with a 50 nm thick thermally grown oxide layer. The GdOx layer was deposited by reactive sputtering from a metal Gd target at an oxygen partial pressure of \(\sim 5 \times 10^{-5}\) Torr. After breaking vacuum, 100 \(\mu\text{m}\) diameter Ta(1 nm)/Au(5 nm) gate electrodes were deposited through a shadow mask. The metal gate was thick enough to provide good electrical contact but thin enough to allow optical access to the Co film.\(^18\) Magnetic properties were probed via the polar magneto optical Kerr effect (MOKE), using a 532 nm diode laser attenuated to 1 mW, focused to a \(\sim 5 \mu\text{m}\) diameter probe spot and positioned by a high resolution scanning stage. The switching field \(H_c\) and remnant to saturation magnetization ratio \(M_r/M_s\) were derived from hysteresis loops measured at a 17 Hz sweep rate. A mechanically compliant 15 \(\mu\text{m}\) diameter BeCu probe tip was used to apply a gate voltage \(V_g\) to the top electrode, while a 25 \(\mu\text{m}\) diameter blunt W microprobe was used to mechanically generate local DW nucleation sites as described below.

Pt/Co/M-oxide films (with \(M = \text{Al, Mg, etc.}\)) are known to exhibit strong PMA originating in part from Co-O coordination at the Co/M-oxide interface.\(^19\)\(^,\)\(^20\) Here, we have used GdOx as the gate oxide both because it has a high dielectric constant\(^21\) and because of the expected contribution to spin-orbit coupling at the interface through the presence of a high-Z rare-earth ion. The as-deposited Pt/Co/GdOx films exhibit strong PMA with a hard axis (in-plane) saturation field of >5 kOe and square out-of-plane hysteresis loops (Fig. 1(b)). The switching field \(H_c\) ranged from 240–280 Oe at a distance of \(\sim 50 \mu\text{m}\) from the location of the W tip, while the switching field distribution is greatly reduced. Moreover, \(H_c\) increases monotonically as a function of distance \(d\) from the W microprobe tip as shown in Fig. 1(d), indicating expansion of a circular domain centered at the contact point as the field is swept. This behavior can be explained by the creation of a local nucleation site with a reduced nucleation threshold.

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upon application of a local mechanical stress. In this case, $H_c$ is determined by the propagation field required to drive domain expansion via DW motion. The narrow spread in $H_c$ from cycle to cycle after creating the local nucleation site is consistent with DW creep through a fine scale disorder potential. In Fig. 1(d), inset, we show that $H_c$ and $d$ are related via a scaling relation $\ln(d) \sim H_c^{-1/4}$, which is identical to the scaling relation between DW velocity and applied field in the two-dimensional creep regime. The distance $d$ and the average DW velocity $\bar{v}$ during domain expansion are related by $d = \bar{v} \tau$, where $\tau$ is a time constant related to the field sweep rate. Thus, the observed scaling supports the identification of $H_c$ as the DW propagation field.

We find that if the W microprobe overdrive is not too great, the sample returns to its original higher $H_c$ state when the microprobe is lifted and the stress is relieved, which shows that no permanent damage arises from landing the W probe. This simple technique thereby provides a means to separately examine nucleation-limited and propagation-limited DW dynamics and the influence of a gate voltage on these processes.

In order to measure the voltage dependence of $H_c$ in the nucleation and propagation-limited cases, we have used a second microprobe to apply a gate voltage to the Au gate electrode (Fig. 1(a)). This second probe tip was a more mechanically compliant BeCu probe, which could be brought into electrical contact with the Au electrodes without causing any changes to the measured hysteresis loops. Figure 2 shows $H_c$ measured at the center of a Au gate electrode as a gate voltage $V_g$ was cycled from $+7.5 \text{ V}$ to $-7.5 \text{ V}$ before (Fig. 2(a)) and after (Fig. 2(b)) landing a W probe $\sim 80 \mu \text{m}$ away from the Au gate. Hysteresis loops were measured after every 0.5 V step by averaging 50 reversal cycles to account for stochasticity in the nucleation-limited case. In the DW nucleation limited regime, i.e., without landing the W microprobe, we observe a random fluctuation of $H_c$ of about 8 Oe throughout the measurements despite signal averaging, but with no clear correspondence to the applied gate voltage (see Fig. 2(a)). The lack of a clear correlation between $H_c$ and $V_g$ suggests that the distribution of random nucleation events throughout the Co film rather than within the area covered by the gate electrode dominates magnetization reversal.

When a local nucleation site is created by the W microprobe close to the gate electrode, a clear modulation of $H_c$ by $V_g$ becomes apparent (Fig. 2(b)). In this case, $H_c$ tracks $V_g$ linearly and reversibly over a range of $\sim 8 \text{ Oe}$, corresponding to a slope of $\sim 0.5 \text{ Oe/V}$. Negative $V_g$ reduces $H_c$ and therefore enhances DW propagation whereas positive $V_g$ increases $H_c$ and therefore retards DW propagation in the Co film. As DW propagation occurs by creep dynamics, the DW velocity $v$ can be expressed by an Arrhenius relation, $v \sim \exp(-E_a/kT)$, where $k$ is the Boltzmann constant, $T$ the temperature, and $E_a$ the activation energy for DW propagation. Moreover, $E_a \sim (H_{crit}/H)^{1/4}$ where $H$ is the applied magnetic field and $H_{crit}$ is a characteristic depinning field that

FIG. 2. Switching field $H_c$ in (a) DW nucleation and (b) DW propagation limited regime (i.e., W microprobe landed at $\sim 80 \mu \text{m}$) and (c) gate voltage $V_g$ plotted as a function of time. $V_g$ is ramped between $-7.5 \text{ V}$ and $+7.5 \text{ V}$ for 5 cycles and 50 MOKE hysteresis loops are averaged after every 0.5 V step in $V_g$. 

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scales with the DW elastic energy density \( \varepsilon_{el} \sim (AK_d)^{1/2} \).
Here, \( A \) is the exchange constant and \( K_d \) is the uniaxial anisotropy constant. Since \( E_s \) scales with \( K_d \), modulation of \( K_d \) by an electric field at the Co/GdOx interface correspondingly changes the DW creep velocity.

Voltage-induced changes to \( H_c \) are reversible as long as \( |V_g| < 8 \text{ V} \) (electric field < 0.2 \text{ V/nm}) which indicates that no significant charge trapping occurs in the GdOx layer or at the Co/GdOx interface.\(^{14,23} \) At higher \( V_g \), irreversibility appears in the \( H_c \) versus \( V_g \) response, with the change in \( H_c \) depending on \( V_g \) and the voltage dwell time. FIG. 3 shows \( H_c \) (FIG. 3(a)), \( M_r/M_s \) (FIG. 3(b)), and the leakage current \( I_L \) (FIG. 3(c)) measured as \( V_g \) was ramped to increasing positive and increasing negative voltage for two separate electrodes, respectively, beginning from the virgin state. For positive \( V_g \) up to 8 \text{ V}, we find that \( H_c \) increases linearly with \( V_g \), with \( M_r/M_s = 1 \) and \( I_L < 100 \text{ pA} \). Between +8 and +15 \text{ V}, \( H_c \) starts to deviate from its linear dependence on \( V_g \) and after an initially slow decrease, \( H_c \) drops dramatically by \( \sim 120 \text{ Oe} \) or \( \sim 60\% \) above +12 \text{ V}. At the same time, \( M_r/M_s \) decreases significantly from 1 to 0.2 and \( I_L \) increases up to 40 \text{ nA}. Above \( V_g = +15 \text{ V} \), we observe hard breakdown of the GdOx dielectric as can be seen from the jump in \( I_L \) to the externally set current limit of 0.5 \text{ \mu A}.\(^{24} \) After dielectric breakdown, \( H_c \) and \( M_r/M_s \) remain at their diminished values and no significant change is observed within hours after the breakdown occurred. Under negative \( V_g \) the devices show markedly different behavior, with no significant deviation from the linear dependence of \( H_c \) on \( V_g \) until dielectric breakdown occurs at \( V_g = -11 \text{ V} \). Also, no change in \( M_r/M_s \) is observed under negative \( V_g \).

From these observations, we separate the electric field effects into 3 regimes. In regime I, between \( V_g = -8 \text{ V} \) to +8 \text{ V}, DW propagation can be reversibly controlled by an electric field at the Co/GdOx interface, and \( H_c \) follows \( V_g \) linearly. In regime II, \( (V_g = +8 \text{ V} \text{ to } +15 \text{ V}) \), \( H_c \) and \( M_r/M_s \) decrease significantly and irreversibly. In Regime III, corresponding to \( V_g < -11 \text{ V} \) and \( V_g > +15 \text{ V} \), hard dielectric breakdown of the GdOx layer occurs, and no further changes in hysteresis characteristics are induced.

Since the strongest modifications of \( H_c \) and \( M_r/M_s \) in regime II occur at \( V_g \) close to regime III, it is likely that their origin is related to processes occurring during or directly preceding hard dielectric breakdown. Hard dielectric breakdown is typically preceded by a soft breakdown which is associated with trapped charge generation in the dielectric layer.\(^{24} \) Trapped charges have previously been shown to result in strong modifications of magnetic anisotropy in ultrathin Fe films and could therefore explain the irreversible character of the modifications of \( H_c \) and \( M_r/M_s \), observed here.\(^{14} \) However, changes in \( O^{2-} \) coordination at the Co/GdOx interface could also play an important role in the observed effects.\(^{24} \) Oxygen vacancy \( O^{2-} \) generation is often a precursor of dielectric breakdown in high-k gate oxides\(^{25} \) and \( O^{2-} \) would migrate predominantly to the Co/GdOx interface due to the high positive \( V_g \). Since hybridization between Co 3\( d \) and O 2\( p \) orbitals is expected to play a crucial role in the PMA of the Co film,\(^{19,20,26} \) the PMA should be very sensitive to changes in interface oxygen stoichiometry.\(^{19,20,27} \) Accumulation of oxygen vacancies at the interface could therefore significantly contribute to the observed loss of PMA in regime II. Moreover, hysteresis loops are only sensitive to changes at the interface between GdOx and the Co film but

![FIG. 3](image-url) (a) Modification of switching field \( H_c \) (i.e., \( \Delta H_c \)), (b) remnant to saturation magnetization ratio \( M_r/M_s \), and (c) leakage current \( I_L \) (clamped at 0.5 \text{ \mu A}) as a function of time while gate voltage \( V_g \) is ramped in steps of 0.5 \text{ V every } \sim 40 \text{ s} (d). Left and right half of figure correspond to two different devices, measured under increasing negative and positive \( V_g \), respectively. W microprobe is landed at \( 0.5 \text{ \mu m} \) and 250 MOKE hysteresis loops are averaged and \( I_L \) is measured after every voltage step. Effects in regime I are linear and reversible. Regime II (enlarged in inset of (a)) is marked by the onset of irreversibility and, at higher \( V_g \), a steep drop in \( H_c \) and \( M_r/M_s \). Regime III corresponds to hard dielectric breakdown. Dashed area highlights lack of regime II for negative \( V_g \). Insets in (b) show representative hysteresis loops at indicated points.

![FIG. 4](image-url) Evolution of MOKE hysteresis loops measured for Pt/Co/GdOx/Ta/Au structure with reduced GdOx thickness of 3 nm. (a) Before application of gate voltage \( V_g \), (b) after application of \( V_g = +1.8 \text{ V} \) for 60 s, and (c) after subsequent application of \( V_g = -1 \text{ V} \) for 15 s.
not to changes at the interface between GdOx and the gate electrode, which could explain why regime II is only observed at positive $V_g$.

Interestingly, the dramatic changes in $H_c$ and $M_r/M_s$ observed for positive $V_g$ in Region II can be partially or even fully reversed by application of a negative $V_g$ so long as hard breakdown has not been reached. In addition, the timescale and voltage levels necessary for these changes can be significantly reduced by reducing the GdOx thickness. Figure 4 shows a series of hysteresis loops measured for a Pt/Co/GdOx device with 3 nm thick GdOx. The (propagation-limited) $H_c$ is 235 Oe before application of a gate voltage (Fig. 4(a)). $V_g = +1.8$ V was then applied for 60 s and then set to zero, after which $H_c$ decreased to 160 Oe (Fig. 4(b)). This nonvolatile change in coercivity was retained until a negative gate voltage of $-1$ V was applied for 15 s, after which the coercivity returned to its initial state (Fig. 4(c)). It has been shown previously that the charge trapping and vacancy migration that occurs during soft breakdown of a dielectric can be reversed under appropriate conditions. This behavior might therefore be exploited as a nonvolatile mechanism for switching the magnetic state electrically.

In summary, we have demonstrated direct electric field control of DW propagation in ultrathin Co films with perpendicular magnetic anisotropy. We find that the DW propagation field linearly follows the applied gate voltage and can be enhanced and retarded by the presence of an electric field at the Co/GdOx interface. Moreover, we identify two different regimes of electric field effects in the Co film. The low field regime allows reversible control of DW propagation whereas the high field regime allows nonvolatile control of magnetic anisotropy. We believe that these findings provide an important step towards the realization of ultralow power, voltage gated spintronic logic and memory devices.

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