Applied Physics Letters

RESEARCH ARTICLE | FEBRUARY 23 2024

Magneto-ionic modulation of the interlayer exchange interaction in synthetic antiferromagnets

Maria-Andromachi Syskaki ; Takaaki Dohi ; Beatrice Bednarz ; Sergei Olegovich Filnov; Sergey Alexeyevich Kasatikov ; Mona Bhukta ; Alevtina Smekhova ; Rohit Pachat ; Johannes Wilhelmus van der Jagt ; Shimpei Ono ; Dafiné Ravelosona ; Jürgen Langer ; Mathias Kläui ; Liza Herrera Diez ; Gerhard Jakob

Appl. Phys. Lett. 124, 082408 (2024)
https://doi.org/10.1063/5.0198750

AIP Publishing

Applied Physics Letters ARTICLE pubs.aip.org/aip/apl

Magneto-ionic modulation of the interlayer exchange interaction in synthetic antiferromagnets

Cite as: Appl. Phys. Lett. **124**, 082408 (2024); doi: 10.1063/5.0198750
Submitted: 19 January 2024 · Accepted: 26 January 2024 ·
Published Online: 23 February 2024

Maria-Andromachi Syskaki,[1,2,a)] Takaaki Dohi,[2,3,b)] Beatrice Bednarz,[2] Sergei Olegovich Filnov,[2] Sergey Alexeyevich Kasatikov,[4] Mona Bhukta,[2] Alevtina Smekhova,[4] Rohit Pachat,[5] Johannes Wilhelmus van der Jagt,[6,7] Shimpei Ono,[8] Dafiné Ravelosona,[5,6] Jürgen Langer,[1] Mathias Kläui,[2] Liza Herrera Diez,[5] and Gerhard Jakob[2,c)]

AFFILIATIONS

[1]Singulus Technologies AG, 63796 Kahl am Main, Germany
[2]Institut für Physik, Johannes Gutenberg-Universität Mainz, Staudingerweg 7, 55128 Mainz, Germany
[3]Laboratory for Nanoelectronics and Spintronics, Research Institute of Electrical Communication, Tohoku University, Sendai, Japan
[4]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Hahn-Meitner Platz 1, 14109 Berlin, Germany
[5]Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, 10 Boulevard Thomas Gobert, 91120 Palaiseau, France
[6]Spin-Ion Technologies, C2N, 10 Boulevard Thomas Gobert, 91120 Palaiseau, France
[7]Université Paris-Saclay, 3 rue Joliot-Curie, 91190 Gif-sur-Yvette, France
[8]Central Research Institute of Electric Power Industry, Yokosuka, Kanagawa 240-0196, Japan

[a)]Author to whom correspondence should be addressed: msyskaki@uni-mainz.de
[b)]Electronic mail: takaaki.dohi.e5@tohoku.ac.jp
[c)]Electronic mail: jakob@uni-mainz.de

ABSTRACT

The electric-field control of magnetism is a highly promising and potentially effective approach for realizing energy-efficient applications. Recent interest has focused on the magneto-ionic effect in synthetic antiferromagnets, driven by its potential to enable high-density data storage devices with ultra-low power consumption. However, the underlying mechanism responsible for the magneto-ionic effect on the interlayer exchange coupling remains elusive. In our work, we find that the modulation of the interlayer exchange coupling is highly sensitive to the thickness of the ferromagnetic layer. We have identified that the changes in the interlayer exchange coupling induced by the gate voltage can be associated with the magneto-ionic effects on the top ferromagnetic layer of the synthetic antiferromagnet. The direct contact between the high ion mobility oxide and the top ferromagnetic layer plays a crucial role in facilitating these effects, largely modifying the anisotropy of the layers. Our findings highlight the important role of magneto-ionic control over the properties of the top ferromagnetic layer in governing the observed modifications in the interlayer exchange coupling. This study provides crucial insight into the intricate interplay between stack structure and magneto-ionic effect on magnetic properties in synthetic antiferromagnetic thin film systems.

Electric field control of magnetism in spintronic devices is one of the most promising, industrially realizable and energy-efficient routes to future applications, for instance in storage, due to its low power consumption.[1–3] Since the first demonstration using a ferromagnetic (FM) semiconductor,[4] it has been revealed that various magnetic properties can be modified by an electric field.[5,6] In metals, the high charge carrier density enables efficient screening of electric fields, but it also imposes limitations on their applications. However, recent advancements in spintronic devices have overcome these limitations by utilizing ultra-thin metal layers with thicknesses comparable to the screening length.[7] Consequently, in FM metals, electric field effects have been demonstrated for a variety of magnetic properties such as magnetic anisotropy,[8,9] Curie temperature,[10] damping constant,[11] proximity effects,[12,13] exchange interaction,[14,15] interfacial Dzyaloshinskii–Moriya interaction,[16,17] and g-factor.[18] Using voltages to control magnetic properties has the potential to pave the way for a technologically advanced class of computing devices with significantly lower energy consumption.[19] To be feasible for technological purposes, the changes induced by a voltage must have a significant effect on the magnetic properties of the system. The prospect of producing large effects with minimal voltages has led to a growing interest in magneto-ionic research.[20–22] Ionic liquid gating (ILG) offers several advantages over solid state gating methods, such as simple device fabrication and large gating areas.[23] The ionic liquids (ILs) can be engineered to have specific properties, such as low volatility and high thermal stability, enabling the use of this technique in a broad range of operations and environments. The ILG technique also has the potential to be used in conjunction with different classes of advanced materials and device structures, such as two-dimensional materials and nanostructures, to create innovative and improved electronic devices.[24,25]

Synthetic antiferromagnets (SyAFMs) have gained significant attention due to their unique properties and applications in the field of spintronics.[26–38] The SyAFMs are special systems in that their particular properties are governed by the interlayer exchange coupling (IEC),[39–43] which arises from exchange interactions between magnetic layers through a non-magnetic spacer layer. This interaction is mediated by the conduction electrons in the metal and is dependent on the density of states and distance between the magnetic layers, resulting in either FM or antiferromagnetic (AFM) coupling. Recent studies have shown the ability to dynamically control the IEC and the switching between FM and AFM coupling of out-of-plane magnetized Co/Pt-based stacks with a Ru interlayer through magneto-ionic gating with hydrogen ions,[44] ILG,[45,46] and solid-state Li-ion battery technology.[47] Furthermore, ILG on nanowires, composed of out-of-plane magnetized SyAFM Co/Ni layers with Ru spacer, can reversibly tune the velocity of domain walls and induce large changes in the exchange coupling torque and current-induced domain wall velocity due to the oxidation of the top magnetic layer.[48]

Our work showcases a remarkable capability to modulate the IEC in SyAFMs through magneto-ionic phenomena, specifically through the influence of gating on the top FM layer of the SyAFM. Additionally, we demonstrate that the modulation of IEC is dependent on the thickness of the top FM layer. These findings emphasize the crucial role of stack engineering in achieving efficient magneto-ionic control of IEC in synthetic antiferromagnetic structures.

The thin film material stacks were deposited on thermally oxidized Si/SiO$_2$ substrates at room temperature using a Singulus Rotaris magnetron sputtering tool. This tool offers exceptional reproducibility and sub-Angstrom thickness accuracy, maintaining a base pressure of 5×10^{-8} mbar. The metallic layers, including Ta, Pt, Ir, and Co$_{0.6}$Fe$_{0.2}$B$_{0.2}$ (CFB), were grown using DC-magnetron sputtering, while the HfO$_2$ layers were deposited using RF-sputtering from a composite target. The oxide layers serve as ionic reservoirs, facilitating magneto-ionic interactions within the metallic stack.[49,50] CoFeB was specifically chosen as a suitable material for its compatibility with magnetic random access memory technology in spintronics. It exhibits desirable properties such as high tunnel magnetoresistance, large perpendicular magnetic anisotropy (PMA), and low damping when incorporated into magnetic tunnel junctions.[51] The interlayer AFM coupling in our structure is mediated by a non-magnetic Ir spacer layer. The thickness of the Ir layer is chosen to be in the vicinity of the first peak of the IEC oscillation. As shown in Fig. 1(b), we denote the thickness of the top CFB layer as t_{CFB}, which is 0.8, 0.9, or 1.0 nm, respectively.

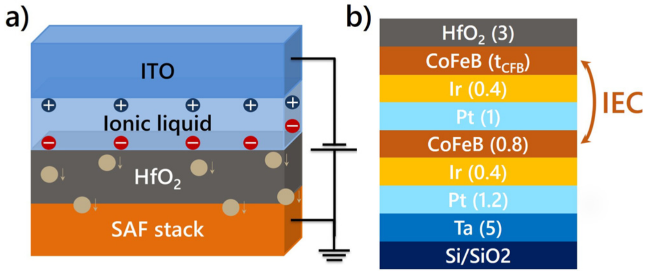

FIG. 1. (a) The schematic illustration shows the application of a negative gate voltage ($V_G < 0$), with mobile oxygen species (depicted in tan color), migrating toward the metallic stack. (b) The SyAFM stack, with t_{CFB} representing thicknesses of 0.8, 0.9, and 1.0 nm. The numbers in brackets denote the respective thicknesses in nm.

The as-grown samples exhibit magnetization with a perpendicular easy axis. The ILG technique was implemented using a sample area of 0.25 cm^2. The 1-ethyl-3-methylimidazolium-bis(trifluormethylsulfonyl)imide [EMIM]$^+$[TFSI]$^-$ was utilized as the IL on the sample surface,[52] and a glass substrate coated with indium tin oxide (ITO) was used as the top electrode. A schematic diagram illustrating the setup is shown in Fig. 1(a). The measurements were conducted at room temperature, and the polar magneto-optical Kerr effect (pMOKE) hysteresis loops were recorded at zero gate voltage (V_G) to exploit the nonvolatile nature of the magneto-ionic effects. To explore the ionic modulation of the interlayer exchange field, referred to as H_{int}, among various material stacks, we employed a successive, consistent, and fixed gating duration of 30 s with increasing V_G. This approach allows for a direct comparison of the effects observed.

In SyAFM stacks, schematically represented in Fig. 1(b), the H_{int} represents the magnetic field at which the AFM–FM transition with a spin flip-like behavior occurs, indicating antiferromagnetic coupling between the two FM layers through the non-magnetic interlayer[26] at low magnetic fields. The H_{int} has commonly been used as an indicator of the modulation of the interlayer exchange interaction (J_{int}).[44,46,47] Previous studies have demonstrated the AFM–FM transition, which corresponds to a sign change in J_{int}, using ILG.[44,45,48] However, it should be noted that changes in the total magnetization of the stack also contribute to H_{int}, which can potentially affect the accurate assessment of the J_{int} modulation under gating.

To elucidate the influence of the magneto-ionic effect on the modulation of H_{int}, we conduct a systematic investigation by exploring the relationship between the thickness of the top FM layer and the varying negative V_G. Subsequently, we record the relative changes in the pMOKE measurements following each 30 s gating step.

Our systematic study of the modulation of H_{int} as a function of V_G has revealed a non-monotonic behavior, graphically depicted in Fig. 2(a) for the 0.9 nm CFB SyAFM stacks as a function of the applied magnetic field. The Kerr loops for t_{CFB} 0.8 and 1.0 nm are presented in Sec. 2 of the supplementary material (S2). Observing the change in thickness from 0.8 nm to 0.9 and 1.0 nm for the top FM layer, a consistent pattern is noted across all three thicknesses in the variation of

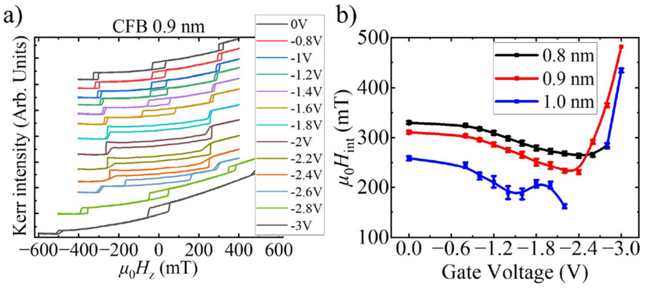

FIG. 2. (a) The non-monotonic behavior of the H_{int} as a function of the magnetic field was investigated for t_{CFB} of top FM equal to 0.9 nm, using pMOKE magnetometry. Hysteresis loops were recorded successively after each 30-s application of V_G. Curves are vertically shifted. (b) H_{int} as a function of V_G for different thicknesses of CFB. B-Spline interpolation lines are included as a guide to the eye.

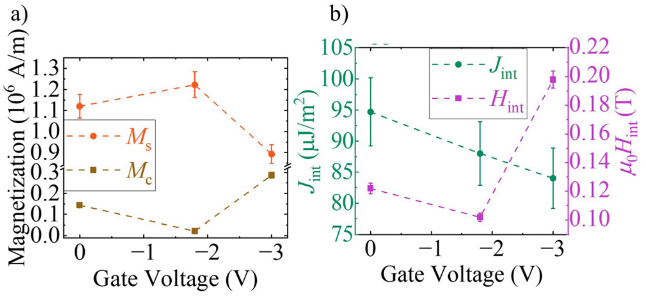

FIG. 3. *Ex situ* SQUID measurements were conducted on the SyAFM stack with a top FM layer thickness of $t_{CFB} = 0.9$ nm as a function of V_G. (a) Saturation magnetization M_s and uncompensated magnetization M_c, as well as (b) H_{int} and IEC strength J_{int} as a function of V_G.

H_{int}, depicted in Fig. 2(b). Initially, there is a decrease followed by an increase. Specifically, both the 0.8 and 0.9 nm stacks reveal a single minimum, while the 1.0 nm stack displays a local minimum at lower V_G, followed by a non-monotonic behavior in H_{int}. Analyzing the switching curves, the 0.8 nm system maintains the typical three-step switching behavior expected for a SyAFM for all applied V_G. Conversely, the 0.9 nm CFB shows a transition from the typical three-step pattern to a two-step one at intermediate V_G, returning to the three-step pattern at maximum V_G, as seen in Fig. 2(a). For the 1.0 nm CFB SyAFM stack, this evolution of the curves occurs twice, as in Fig. S2(c). This systematic progression highlights the significant influence of the top FM layer's thickness in modulating the qualitative aspects of the magneto-ionic effect on H_{int}.

The dependence of gate effects on the thickness of the top FM layer suggests that the modulation induced by magneto-ionic gating in the H_{int} is not singularly attributed to that of the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. Instead, it indicates that there might exist a potentially intricate interplay between the RKKY interaction and the modulation of quantum interference,[53] further allowing us to rationalize the observed thickness-dependent effects. Theoretical computations, providing insights into the unique nature of the IEC, have substantiated that the qualitative oscillation of J_{int} is influenced by both the spacer's thickness and the FM layer's thickness.[54,55] These factors significantly impact the system's quantum interference.[42] In this context, the magneto-ionic effects observed in our study deviate from the anticipated conventional trend for H_{int} and the RKKY interaction.[43] It appears that these effects are influenced by both the spatial separation between magnetic moments and the thickness of the FM layer.

We performed *ex situ* SQUID measurements for a subset of the gating voltages to clarify the impact of gating on different properties, such as saturation magnetization (M_s), uncompensated magnetization (M_c), H_{int} and J_{int}, as shown in Figs. 3(a) and 3(b). While observing the modulation of J_{int} with varying V_G, it is evident that it displays a different trend compared to the changes in M_s induced by alterations in V_G. The *m*–*H* curves and the J_{int} calculation, as well as further supporting material are presented in Sec. 3 of the supplementary material (S3).

To gain a deeper understanding of the non-monotonic behavior of H_{int}, we investigated the influence of magneto-ionic effects on the modulation of PMA within the single-layer FM stack. PMA in thin films of FM materials can be induced through two distinct types of interfaces: one with a heavy metal (HM) and another with an oxide. In the case of the HM interface, PMA is induced by spin–orbit coupling,[56] and its magnitude is significantly affected by the sharpness or thickness transition at the interface.[57,58] On the other hand, the origin of PMA at the oxide interface arises from the hybridization between the O-p_z and the FM's $3d_{z^2}$ orbitals hybridized with $d_{xz(yz)}$ orbitals.[59,60] Regarding the magneto-ionic effects on the single FM layer, previous studies of CoFeB/HfO_2[61] systems have demonstrated that under negative V_G, these stacks undergo a transition from in-plane anisotropy to PMA and back to in-plane. This transition can be attributed to the modulation of the oxidation level of the FM layer.[62]

In our work, the as-grown state of the FM stacks demonstrates PMA at 0 V, as observed in the square hysteresis loops shown in Fig. 4(a). The Kerr loops for t_{CFB} 0.8 and 1.0 nm are presented in Sec. 4 of the supplementary material (S4). For the 0.9 nm CFB stack, the H_C shows no significant change in the low range of V_G until −1.6 V. At this V_G, the square hysteresis loop undergoes a transformation, turning into a distorted sigmoidal curve that approaches the spin re-orientation transition (SRT). This behavior is similar to what was observed in the 0.8 nm stack. At −2.4 V, the stack loses PMA, which is then recovered at −2.8 V. Figure 4(b) illustrates the variation of H_C as a function of V_G for all three FM stacks, which exhibits a non-monotonic dependence, revealing the modulation of the anisotropy in all three cases.

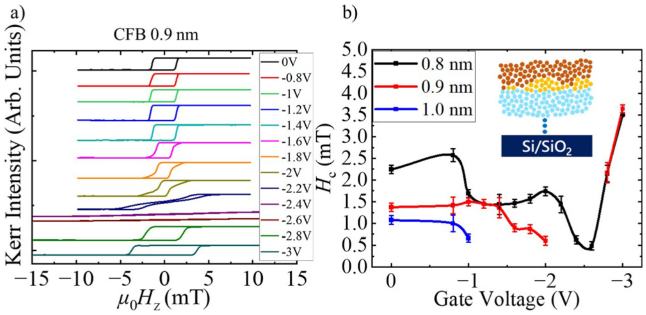

FIG. 4. (a) The non-monotonic behavior with increasing V_G of single FM layers as a function of the magnetic field was investigated for t_{CFB} equal to 0.9 nm, using pMOKE magnetometry. Hysteresis loops were recorded after each successive 30-s application of V_G. (b) The coercive field H_C as a function of V_G for the single FM stacks. Modified Bezier interpolation lines are included as a guide to the eye. The inset provides a schematic representation of the island growth of Ir on Pt.

The magneto-ionic response demonstrated by the individual FM stacks maintains consistent behavior across all three thicknesses, portraying a transition from the initially robust PMA state to an intermediate state situated between the SRT point and the in-plane anisotropy state. This intermediate state eventually reverts to PMA solely under the influence of a negatively polarized V_G. For the 0.8 nm thickness, this response encompasses a sequence starting from robust PMA, transitioning to a reduced PMA state near the SRT point, and finally returning to PMA. In the case of the 0.9 nm thickness, where this intriguing behavior is more pronounced, the transition from the initial PMA state leads to an easy-plane anisotropy state before eventually reverting to strong PMA. This behavior is analogous for the 1.0 nm thickness, with the exception that it does not return to the PMA state. This transition from PMA to easy-plane anisotropy and back to PMA can be linked to the non-monotonic behavior observed in a similar range of negative V_G values for the SyAFM stacks.

The inclusion of a 0.4 nm Ir layer beneath the FM layer can lead to the development of a non-continuous layer at the lower FM interface. This non-continuous Ir layer, resembling island-like growth on the Pt layer, as schematically shown in the inset of Fig. 4(b), could exert a noteworthy influence on the anisotropy profile, consequently affecting the switching behavior of the system upon application of a V_G. Given that the FM layer comprises regions with anisotropy contributions from both Pt and Ir/Pt interfaces, these regions collectively contribute to the overall switching behavior modulation under gating. The intricate relationship between the switching behavior and magneto-ionic gating can be attributed to the interplay between the anisotropies of the two interface types and their dependencies on the gating process, and possibly to the domain formation during the gating process.

To understand the changes in magnetic properties, we investigated the modifications in chemical and electronic structure occurring due to the ILG process using near-edge x-ray absorption fine structure (NEXAFS) spectroscopy and x-ray photoemission spectroscopy (XPS) measurements. These measurements were performed at the RGL-PES station of the Russian–German beamline at the synchrotron-radiation facility BESSY II, operated by Helmholtz-Zentrum Berlin [see supplementary material 1 (S1) for more details].

The NEXAFS spectra recorded at the Co and Fe $L_{2,3}$-edges of both the as-grown state and the gated state, obtained after applying $V_G = -3$ V for 30 s, are presented in Figs. 5(a) and 5(b). The spectra recorded at the O K-edge, as well as Pt, Ir, and Hf 4f levels and Hf 5p level, and O 1s core level are presented in S1. The overall shape of the O K-edge spectrum is consistent with that of HfO_2, as reported in previous studies.[63] The presence of a distinctive pre-edge feature at approximately 531 eV in the gated state spectrum indicates the existence of Fe–O or Co–O bonds, signifying oxidation of Fe or Co atoms.[64]

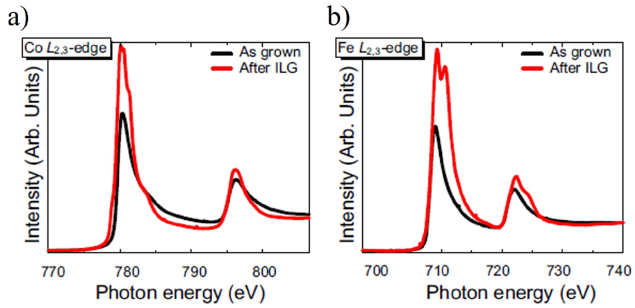

FIG. 5. Normalized NEXAFS spectra for (a) Co $L_{2,3}$-edges and (b) Fe $L_{2,3}$-edges.

The oxidation process is further corroborated by the prominent features observed in the Fe and Co $L_{2,3}$-edges spectra, which are characteristics of α-Fe_2O_3 and CoO, respectively, indicating the presence of Fe^{3+} and Co^{2+} ions.[65–67] The presence of a rich multiplet structure at the Co L_3-edge line in the gated state spectrum reveals the admixture of higher valence states of Co to its metallic state,[68,69] as reported for Co/HfO_2[70] systems. The appearance of a secondary peak at higher energies relative to the main metallic component in the Fe $L_{2,3}$-edge spectrum of the gated state signifies the co-existence of α-Fe_2O_3 and nearly metallic Fe.[69,71] The enhanced intensity of the $L_{2,3}$-edge spectra for Fe and Co in the gated state suggests a decrease in the average occupancy of $3d$ states,[72] supporting the existence of Co and Fe atoms with higher valence states.

Concluding, our findings reveal an intriguing non-monotonic behavior of the H_{int} in the SyAFM stacks, providing valuable insight into the interplay between IEC, M_s, and V_G. Through a systematic study, we have identified the key role of the thickness of the FM layer in shaping the influence of the magneto-ionic effect on the H_{int}. The results obtained from the SyAFM systems demonstrate a non-monotonic modulation of H_{int}, consistent with the changes induced by the thickness variation of the top FM layer. The observed non-monotonic modulation of the H_{int} cannot be described only by considering the changes in the effective magnetization, which indicates that a modulation of interlayer exchange interaction also takes place. Furthermore, we propose that the observed interplay between the in-plane and out-of-plane anisotropy, modulated by the application of V_G, could be justified by the potential formation of islands of Ir on Pt, which potentially imparts different anisotropy contributions. These contributions could potentially influence the observed magneto-ionic response in the SyAFM systems. The observed effects are primarily governed by changes in the oxidation states of the top FM layer and the chemical environment of the interfaces, as confirmed by the conducted spectroscopy measurements on the SyAFM stack. These findings are crucial for understanding the role of ILs in controlling the IEC and hold potential for advancing the development of highly efficient spintronic devices. The ability to achieve magneto-ionic control in SyAFM structures may generate possibilities for innovative applications in spintronics.

See the supplementary material for comprehensive details about the experimental measurements and data analysis. Additionally, it includes pMOKE and SQUID loops for both the SyAFM stacks and the single FM stacks, along with spectroscopy measurements for the SyAFM stack.

This project received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie Grant Agreement No. 860060 "Magnetism and the effect of Electric Field" (MagnEFi) and the Horizon Europe Project No. 101070290 (NIMFEIA). This work was partly supported by JSPS KAKENHI (No. 23K13655), as well as funded by the Deutsche Forschungsgemeinschaft (DFG, German Research

Foundation)—TRR 173/2-268565370 Spin+X (Projects B02 and A01). Helmholtz-Zentrum Berlin für Materialien und Energie is acknowledged for provision of access to synchrotron radiation facilities and allocation of synchrotron radiation. S. Kasatikov acknowledges the financial support from the Helmholtz Association (Helmholtz Initiative for refugees) and Helmholtz-Zentrum Berlin.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Maria-Andromachi Syskaki: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Validation (lead); Visualization (lead); Writing – original draft (lead). **Takaaki Dohi:** Data curation (supporting); Formal analysis (lead); Investigation (supporting); Methodology (supporting); Supervision (lead); Validation (lead); Visualization (equal); Writing – review & editing (lead). **Beatrice Bednarz:** Data curation (supporting); Formal analysis (supporting); Methodology (supporting); Validation (supporting); Writing – review & editing (supporting). **Sergei Olegovich Filnov:** Data curation (supporting); Formal analysis (supporting); Investigation (supporting); Methodology (supporting); Validation (supporting). **Sergey Kasatikov:** Data curation (supporting); Investigation (supporting); Validation (supporting); Visualization (supporting). **Mona Bhukta:** Investigation (supporting); Methodology (supporting); Validation (supporting); Visualization (supporting); Writing – review & editing (supporting). **Alevtina Smekhova:** Investigation (supporting); Methodology (supporting); Validation (supporting). **Rohit Pachat:** Data curation (equal); Investigation (supporting); Methodology (lead); Validation (equal); Writing – review & editing (supporting). **Johannes Wilhelmus van der Jagt:** Data curation (supporting); Investigation (supporting); Validation (supporting); Writing – review & editing (supporting). **Shimpei Ono:** Methodology (lead). **Dafiné Ravelosona:** Funding acquisition (supporting); Methodology (supporting); Supervision (supporting); Writing – review & editing (equal). **Juergen Langer:** Funding acquisition (lead); Resources (lead); Supervision (supporting); Writing – review & editing (supporting). **Mathias Kläui:** Funding acquisition (lead); Resources (lead); Supervision (equal); Validation (equal); Writing – review & editing (equal). **Liza Herrera Diez:** Conceptualization (equal); Funding acquisition (lead); Methodology (lead); Supervision (lead); Validation (supporting); Visualization (lead); Writing – review & editing (lead). **Gerhard Jakob:** Formal analysis (equal); Funding acquisition (lead); Project administration (equal); Resources (lead); Supervision (lead); Validation (lead); Writing – review & editing (lead).

DATA AVAILABILITY

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

REFERENCES

[1] F. Matsukura, Y. Tokura, and H. Ohno, "Control of magnetism by electric fields," Nat. Nanotechnol. **10**, 209 (2015).

[2] T. Nozaki, "Recent progress in the voltage-controlled magnetic anisotropy effect and the challenges faced in developing voltage-torque MRAM," Micromachines **10**, 327 (2019).

[3] A. Molinari, H. Hahn, and R. Kruk, "Voltage-control of magnetism in all-solid-state and solid/liquid magnetoelectric composites," Adv. Mater. **31**, 1806662 (2019).

[4] H. Ohno, D. Chiba, F. Matsukura, T. Omiya, E. Abe, T. Dietl, Y. Ohno, and K. Ohtani, "Electric-field control of ferromagnetism," Nature **408**, 944 (2000).

[5] D. Chiba, M. Yamanouchi, F. Matsukura, and H. Ohno, "Electrical manipulation of magnetization reversal in a ferromagnetic semiconductor," Science **301**, 943 (2003).

[6] D. Chiba, M. Sawicki, Y. Nishitani, Y. Nakatani, F. Matsukura, and H. Ohno, "Magnetization vector manipulation by electric fields," Nature **455**, 515 (2008).

[7] M. Weisheit, S. Fähler, A. Marty, Y. Souche, C. Poinsignon, and D. Givord, "Electric field-induced modification of magnetism in thin-film ferromagnets," Science **315**, 349 (2007).

[8] T. Maruyama, Y. Shiota, T. Nozaki, K. Ohta, N. Toda, M. Mizuguchi, A. A. Tulapurkar, T. Shinjo, M. Shiraishi, S. Mizukami, Y. Ando, and Y. Suzuki, "Large voltage-induced magnetic anisotropy change in a few atomic layers of iron," Nat. Nanotechnol. **4**, 158 (2009).

[9] M. Endo, S. S. Kanai, S. Ikeda, F. Matsukura, and H. Ohno, "Electric-field effects on thickness dependent magnetic anisotropy of sputtered MgO/$Co_{40}Fe_{40}B_{20}$/Ta structures," Appl. Phys. Lett. **96**, 212503 (2010).

[10] D. Chiba, S. Fukami, K. Shimamura, N. Ishiwata, K. Kobayashi, and T. Ono, "Electrical control of the ferromagnetic phase transition in cobalt at room temperature," Nat. Mater. **10**, 853 (2011).

[11] A. Okada, S. Kanai, M. Yamanouchi, S. Ikeda, F. Matsukura, and H. Ohno, "Electric-field effects on magnetic anisotropy and damping constant in Ta/CoFeB/MgO investigated by ferromagnetic resonance," Appl. Phys. Lett. **105**, 052415 (2014).

[12] Y. Hibino, T. Koyama, A. Obinata, K. Miwa, S. Ono, and D. Chiba, "Electric field modulation of magnetic anisotropy in perpendicularly magnetized Pt/Co structure with a Pd top layer," Appl. Phys. Express **8**, 113002 (2015).

[13] A. Obinata, Y. Hibino, D. Hayakawa, T. Koyama, K. Miwa, S. Ono, and D. Chiba, "Electrical control of the ferromagnetic phase transition in cobalt at room temperature," Sci. Rep. **5**, 14303 (2015).

[14] T. Dohi, S. Kanai, A. Okada, F. Matsukura, and H. Ohno, "Effect of electric-field modulation of magnetic parameters on domain structure in MgO/CoFeB," AIP Adv. **6**, 075017 (2016).

[15] F. Ando, H. Kakizakai, T. Koyama, K. Yamada, M. Kawaguchi, S. Kim, K.-J. Kim, T. Moriyama, D. Chiba, and T. Ono, "Modulation of the magnetic domain size induced by an electric field," Appl. Phys. Lett. **109**, 022401 (2016).

[16] K. Nawaoka, S. Miwa, Y. Shiota, N. Mizuochi, and Y. Suzuki, "Voltage induction of interfacial Dzyaloshinskii-Moriya interaction in Au/Fe/MgO artificial multilayer," Appl. Phys. Express **8**, 063004 (2019).

[17] T. Srivastava, M. Schott, R. Juge, V. Křižáková, M. Belmeguenai, Y. Roussigné, A. Bernand-Mantel, L. Ranno, S. Pizzini, S.-M. Chérif, A. Stashkevich, S. Auffret, O. Boulle, G. Gaudin, M. Chshiev, C. Baraduc, and H. Béa, "Large-voltage tuning of Dzyaloshinskii–Moriya interactions: A route toward dynamic control of skyrmion chirality," Nano Lett. **18**, 4871 (2018).

[18] H. Mizuno, T. Moriyama, K. Tanaka, M. Kawaguchi, T. Koyama, D. Chiba, and T. Ono, "Electric field effect on spectroscopic *g*-factor and magnetic anisotropy in a Pt/Co/MgO ultrathin film," Jpn. J. Appl. Phys., Part 1 **61**, 103001 (2022).

[19] C. Song, B. Cui, F. Li, X. Zhou, and F. Pan, "Recent progress in voltage control of magnetism: Materials, mechanisms, and performance," Prog. Mater. Sci. **87**, 33–82 (2017).

[20] M. Nichterwitz, S. Honnali, M. Kutuzau, S. Guo, J. A. Zehner, K. Nielsch, and K. Leistner, "Advances in magneto-ionic materials and perspectives for their application," APL Mater. **9**, 030903 (2021).

[21] S. Manipatruni, D. E. Nikonov, C.-C. Lin, T. A. Gosavi, H. Liu, B. Prasad, Y.-L. Huang, E. Bonturim, R. Ramesh, and I. A. Young, "Scalable energy-efficient magnetoelectric spin-orbit logic," Nature **565**, 35 (2019).

[22] J. de Rojas, A. Quintana, G. Rius, C. Stefani, N. Domingoa, J. L. Costa-Krämer, E. Menéndez, and J. Sort, "Voltage control of magnetism with magneto-ionic approaches: Beyond voltage-driven oxygen ion migration," Appl. Phys. Lett. **120**, 070501 (2022).

[23] C. Leighton, "Electrolyte-based ionic control of functional oxides," Nat. Mater. **18**, 13 (2018).

[24] U. Bauer, L. Yao, A. J. Tan, P. Agrawal, S. Emori, H. L. Tuller, S. van Dijken, and G. S. D. Beach, "Magneto-ionic control of interfacial magnetism," Nat. Mater. **14**, 174 (2015).

[25] N. Lu, P. Zhang, Q. Zhang, R. Qiao, Q. He, H.-B. Li, Y. Wang, J. Guo, D. Zhang, Z. Duan, Z. Li, M. Wang, S. Yang, M. Yan, E. Arenholz, S. Zhou, W. Yang, L. Gu, C.-W. Nan, J. Wu, Y. Tokura, and P. Yu, "Electric-field control of tri-state phase transformation with a selective dual-ion switch," Nature **546**, 124 (2017).

[26] R. A. Duine, K.-J. Lee, S. Parkin, and M. D. Stiles, "Synthetic antiferromagnetic spintronics," Nat. Phys. **14**, 217 (2018).

[27] S. S. P. Parkin, N. More, and K. P. Roche, "Oscillations in exchange coupling and magnetoresistance in metallic superlattice structures: Co/Ru, Co/Cr, and Fe/Cr," Phys. Rev. Lett. **64**, 2304–2307 (1990).

[28] R. Lavrijsen, J.-H. Lee, A. Fernández-Pacheco, D. C. M. C. Petit, R. Mansell, and R. P. Cowburn, "Magnetic ratchet for three-dimensional spintronic memory and logic," Nature **493**, 647 (2013).

[29] S.-H. Yang, K.-S. Ryu, and S. Parkin, "Domain-wall velocities of up to 750 m s^{-1} driven by exchange-coupling torque in synthetic antiferromagnets," Nat. Nanotechnol. **10**, 221 (2015).

[30] T. Dohi, S. DuttaGupta, S. Fukami, and H. Ohno, "Formation and current-induced motion of synthetic antiferromagnetic skyrmion bubbles," Nat. Commun. **10**, 5153 (2019).

[31] T. Dohi, M. Weißenhofer, N. Kerber, F. Kammerbauer, Y. Ge, K. Raab, J. Zàzvorka, M.-A. Syskaki, A. Shahee, M. Ruhwedel, T. Böttcher, P. Pirro, G. Jakob, U. Nowak, and M. Kläui, "Enhanced thermally-activated skyrmion diffusion with tunable effective gyrotropic force," Nat. Commun. **14**, 5424 (2023).

[32] M. Bhukta, T. Dohi, V. K. Bharadwaj, R. Zarzuela, M.-A. Syskaki, M. Foerster, M. A. Niño, J. Sinova, R. Frömter, and M. Kläui, "Homochiral antiferromagnetic merons, antimerons and bimerons realized in synthetic antiferromagnets," arXiv:2303.14853 (2023).

[33] R. Chen, Q. Cui, L. Liao, Y. Zhu, R. Zhang, H. Bai, Y. Zhou, G. Xing, F. Pan, H. Yang, and C. Song, "Reducing Dzyaloshinskii-Moriya interaction and field-free spin-orbit torque switching in synthetic antiferromagnets," Nat. Commun. **12**, 3113 (2021).

[34] K. Kobayashi, K. Hayakawa, J. Igarashi, W. A. Borders, S. Kanai, H. Ohno, and S. Fukami, "External-field-robust stochastic magnetic tunnel junctions using a free layer with synthetic antiferromagnetic coupling," Phys. Rev. Appl. **18**, 054085 (2022).

[35] J. Z. Sun, C. Safranski, P. Trouilloud, C. D'Emic, P. Hashemi, and G. Hu, "Easy-plane dominant stochastic magnetic tunnel junction with synthetic antiferromagnetic layers," Phys. Rev. B **108**, 064418 (2023).

[36] M. Ishibashi, Y. Shiota, T. Li, S. Funada, T. Moriyama, and T. Ono, "Switchable giant nonreciprocal frequency shift of propagating spin waves in synthetic antiferromagnets," Sci. Adv. **6**, eaaz6931 (2020).

[37] A. Fernández-Pacheco, E. Vedmedenko, F. Ummelen, R. Mansell, D. Petit, and R. P. Cowburn, "Symmetry-breaking interlayer Dzyaloshinskii–Moriya interactions in synthetic antiferromagnets," Nat. Mater. **18**, 679 (2019).

[38] D.-S. Han, K. Lee, J.-P. Hanke, Y. Mokrousov, K.-W. Kim, W. Yoo, Y. L. W. van Hees, T.-W. Kim, R. Lavrijsen, C.-Y. You, H. J. M. Swagten, M.-H. Jung, and M. Kläui, "Long-range chiral exchange interaction in synthetic antiferromagnets," Nat. Mater. **18**, 703 (2019).

[39] M. A. Ruderman and C. Kittel, "Indirect exchange coupling of nuclear magnetic moments by conduction electrons," Phys. Rev. **96**, 99–102 (1954).

[40] T. Kasuya, "A theory of metallic ferro- and antiferromagnetism on Zener's model," Prog. Theor. Phys. **16**, 45–57 (1956).

[41] K. Yosida, "Magnetic properties of Cu-Mn alloys," Phys. Rev. **106**, 893–898 (1957).

[42] P. Bruno, "Recent progress in the theory of interlayer exchange coupling," J. Appl. Phys. **76**, 6972 (1994).

[43] P. Bruno, "Interlayer exchange coupling: A unified physical picture," J. Magn. Magn. **121**, 248–252 (1993).

[44] A. E. Kossak, M. Huang, P. Reddy, D. Wolf, and G. S. D. Beach, "Voltage control of magnetic order in RKKY coupled multilayers," Sci. Adv. **9**, eadd0548 (2023).

[45] Q. Yang, L. Wang, Z. Zhou, L. Wang, Y. Zhang, S. Zhao, G. Dong, Y. Cheng, T. Min, Z. Hu, W. Chen, K. Xia, and M. Liu, "Ionic liquid gating control of RKKY interaction in FeCoB/Ru/FeCoB and (Pt/Co)$_2$/Ru/(Co/Pt)$_2$ multilayers," Nat. Commun. **9**, 991 (2018).

[46] Q. Yang, Z. Zhou, L. Wang, H. Zhang, Y. Cheng, Z. Hu, B. Peng, and M. Liu, "Ionic gel modulation of RKKY interactions in synthetic anti-ferromagnetic nanostructures for low power wearable spintronic devices," Adv. Mater. **30**, 1800449 (2018).

[47] M. Ameziane, R. Rosenkamp, L. Flajman, S. van Dijken, and R. Mansell, "Electric field control of RKKY coupling through solid-state ionics," Appl. Phys. Lett. **122**, 232401 (2023).

[48] Y. Guan, X. Zhou, F. Li, T. Ma, S.-H. Yang, and S. S. P. Parkin, "Ionitronic manipulation of current-induced domain wall motion in synthetic antiferromagnets," Nat. Commun. **12**, 5002 (2021).

[49] C. Bi, Y. Liu, T. Newhouse-Illige, M. Xu, M. Rosales, J. W. Freeland, O. Mryasov, S. Zhang, S. G. E. te Velthuis, and W. G. Wang, "Reversible control of Co magnetism by voltage-induced oxidation," Phys. Rev. Lett. **113**, 267202 (2014).

[50] A. Fassatoui, J. P. Garcia, L. Ranno, J. Vogel, A. Bernand-Mantel, H. Béa, S. Pizzini, and S. Pizzini, "Reversible and irreversible voltage manipulation of interfacial magnetic anisotropy in Pt/Co/Oxide multilayers," Phys. Rev. Appl. **14**, 064041 (2020).

[51] S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H. D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, and H. Ohno, "A perpendicular-anisotropy CoFeB-MgO magnetic tunnel junction," Nat. Mater. **9**, 721 (2010).

[52] S. Ono, S. Seki, R. Hirahara, Y. Tominari, and J. Takeya, "High-mobility, low-power, and fast-switching organic field-effect transistors with ionic liquids," Appl. Phys. Lett. **92**, 103313 (2008).

[53] P. Bruno, "Theory of interlayer magnetic coupling," Phys. Rev. B **52**, 411–439 (1995).

[54] A. O. Leon, J. d'Albuquerque e Castro, J. C. Retamal, A. B. Cahaya, and D. Altbir, "Manipulation of the RKKY exchange by voltages," Phys. Rev. B **100**, 014403 (2019).

[55] J. d'Albuquerque e Castro, D. Altbir, A. O. Leon, and J. C. Retamal, "Phase-shift control of the exchange coupling between magnetic impurities," Nanotechnology **31**, 355002 (2020).

[56] P. Bruno, "Tight-binding approach to the orbital magnetic moment and magnetocrystalline anisotropy of transition-metal monolayers," Phys. Rev. B **39**, 865–868 (1989).

[57] T. Devolder, "Light ion irradiation of Co/Pt systems: Structural origin of the decrease in magnetic anisotropy," Phys. Rev. B **62**, 5794–5802 (2000).

[58] S. Bandiera, R. C. Sousa, B. Rodmacq, and B. Dieny, "Enhancement of perpendicular magnetic anisotropy through reduction of Co-Pt interdiffusion in (Co/Pt) multilayers," Appl. Phys. Lett. **100**, 142410 (2012).

[59] B. Rodmacq, S. Auffret, B. Dieny, S. Monso, and P. Boyer, "Crossovers from in-plane to perpendicular anisotropy in magnetic tunnel junctions as a function of the barrier degree of oxidation," J. Appl. Phys. **93**, 7513–7515 (2003).

[60] H. X. Yang, M. Chshiev, B. Dieny, J. H. Lee, A. Manchon, and K. H. Shin, "First-principles investigation of the very large perpendicular magnetic anisotropy at Fe|MgO and Co|MgO interfaces," Phys. Rev. B **84**, 054401 (2011).

[61] R. Pachat, D. Ourdani, J. van der Jagt, M.-A. Syskaki, A. Di Pietro, Y. Roussigné, S. Ono, M. Gabor, M. Chérif, G. Durin, J. Langer, M. Belmeguenai, D. Ravelosona, and L. H. Diez, "Multiple magnetoionic regimes in Ta/Co$_{20}$Fe$_{60}$B$_{20}$/HfO$_2$," Phys. Rev. Appl. **15**, 064055 (2021).

[62] A. Di Pietro, R. Pachat, L. Qiao, L. Herrera-Diez, J. W. van der Jagt, S. Picozzi, D. Ravelosona, W. Ren, and G. Durin, "*Ab initio* study of magneto-ionic mechanisms in ferromagnet/oxide multilayers," Phys. Rev. B **107**, 174413 (2023).

[63] F. Frati, M. O. J. Y. Hunault, and F. M. F. de Groot, "Oxygen K-edge x-ray absorption spectra," Chem. Rev. **120**, 4056–4110 (2020).

[64] J. P. Singh, S. Gautam, W. C. Lim, K. Asokan, B. B. Singh, M. Raju, S. Chaudhary, D. Kabiraj, D. Kanjilal, J.-M. Lee, J.-M. Chen, and K. H. Chae, "Electronic structure of magnetic Fe/MgO/Fe/Co multilayer structure by NEXAFS spectroscopy," Vacuum **138**, 48–54 (2017).

[65] D. H. Kim, H. J. Lee, G. Kim, Y. S. Koo, J. H. Jung, H. J. Shin, J.-Y. Kim, and J.-S. Kang, "Interface electronic structures of BaTiO$_3$@X nanoparticles ($X = \gamma$-Fe$_2$O$_3$, Fe$_3$O$_4$, α-Fe$_2$O$_3$, and Fe) investigated by XAS and XMCD," Phys. Rev. B **79**, 033402 (2009).

[66] X.-C. Liu, E.-W. Shi, Z.-Z. Chen, B.-Y. Chen, W. Huang, L.-X. Song, K.-J. Zhou, M.-Q. Cui, Z. Xie, B. He, and S.-Q. Wei, "The local structure of Co-doped ZnO films studied by x-ray absorption spectroscopy," J. Alloys Compd. **463**, 435–439 (2008).

[67] Z. Lin, W. Lai, Z. Wu, J. Liu, and Y. An, "Investigations of the valence states, cobalt ion distribution, and defect structures in Co-doped ITO films," J. Mater. Res. **33**, 2336 (2018).

[68] B. Liu, M. M. van Schooneveld, Y.-T. Cui, J. Miyawaki, Y. Harada, T. O. Eschemann, K. P. de Jong, M. U. Delgado-Jaime, and F. M. F. de Groot, "In-situ 2p3d resonant inelastic x-ray scattering tracking cobalt nanoparticle reduction," J. Phys. Chem. C **121**, 17450–17456 (2017).

[69] T. J. Regan, H. Ohldag, C. Stamm, F. Nolting, J. Lüning, J. Stöhr, and R. L. White, "Chemical effects at metal/oxide interfaces studied by x-ray-absorption spectroscopy," Phys. Rev. B **64**, 214422 (2001).

[70] L. H. Diez, Y. Liu, D. Gilbert, M. Belmeguenai, J. Vogel, S. Pizzini, E. Martinez, A. Lamperti, J. Mohammedi, A. Laborieux, Y. Roussigné, A. Grutter, E. Arenholtz, P. Quarterman, B. Maranville, S. Ono, M. S. E. Hadri, R. Tolley, E. Fullerton, L. Sanchez-Tejerina, A. Stashkevich, S. Chérif, A. Kent, D. Querlioz, J. Langer, B. Ocker, and D. Ravelosona, "Nonvolatile ionic modification of the Dzyaloshinskii-Moriya interaction," Phys. Rev. Appl. **12**, 034005 (2019).

[71] A. S. Vinogradov, A. B. Preobrajenski, S. A. Krasnikov, T. Chasse, R. Szargan, A. Knop-Gericke, R. Schlogl, and P. Bressler, "X-ray absorption evidence for the back-donation in iron cyanide complexes," Surf. Rev. Lett. **9**, 186 (1990).

[72] C. T. Chen, Y. U. Idzerda, H.-J. Lin, N. V. Smith, G. Meigs, E. Chaban, G. H. Ho, E. Pellegrin, and F. Sette, "Experimental confirmation of the x-ray magnetic circular dichroism sum rules for iron and cobalt," Phys. Rev. Lett. **75**, 152–155 (1995).