

Ionically driven synthesis and exchange bias in $\text{Mn}_4\text{N}/\text{MnN}_x$ heterostructures

Cite as: Appl. Phys. Lett. **123**, 082403 (2023); doi: 10.1063/5.0165895

Submitted: 1 July 2023 · Accepted: 9 August 2023 ·

Published Online: 22 August 2023



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Export Citation



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Note: This paper is part of the APL Special Collection on Ferrimagnetic Spintronics.

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ABSTRACT

Ferrimagnets have received renewed attention as a promising platform for spintronic applications. Of particular interest is the Mn_4N from the ε -phase of the manganese nitride as an emergent rare-earth-free spintronic material due to its perpendicular magnetic anisotropy, small saturation magnetization, high thermal stability, and large domain wall velocity. We have achieved high-quality (001)-ordered Mn_4N thin film by sputtering Mn onto η -phase Mn_3N_2 seed layers on Si substrates. As the deposited Mn thickness varies, nitrogen ion migration across the $\text{Mn}_3\text{N}_2/\text{Mn}$ layers leads to a continuous evolution of the layers to $\text{Mn}_3\text{N}_2/\text{Mn}_2\text{N}/\text{Mn}_4\text{N}$, $\text{Mn}_2\text{N}/\text{Mn}_4\text{N}$, and eventually Mn_4N alone. The ferrimagnetic Mn_4N , indeed, exhibits perpendicular magnetic anisotropy and forms via a nucleation-and-growth mechanism. The nitrogen ion migration is also manifested in a significant exchange bias, up to 0.3 T at 5 K, due to the interactions between ferrimagnetic Mn_4N and antiferromagnetic Mn_3N_2 and Mn_2N . These results demonstrate a promising all-nitride magneto-ionic platform with remarkable tunability for device applications.

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22 August 2023 16:06:53

Ferrimagnets (FiMs) are characterized by antiparallel-coupled magnetic sublattices with different magnetic moments. They have gained tremendous interest recently as a promising platform for spintronic applications as they offer the combined benefits of ferromagnets (FMs) and antiferromagnets (AFs),^{1–4} such as easy manipulation with an external magnetic field and fast spin dynamics.⁵ Moreover, some FiMs such as rare-earth iron garnets,⁶ rare-earth-transition metal films,¹ and Mn_4N ⁷ also possess perpendicular magnetic anisotropy (PMA) under appropriate growth conditions, a desirable functionality for device applications. Among them, Mn_4N stands out due to its high Curie temperature ($T_C = 745$ K),⁸ PMA,^{7,9–11} small saturation magnetization (up to 145 emu/cm³),⁹ large domain wall velocity,¹² and possibility of hosting non-trivial spin textures.^{13,14} It can also be doped with other elements to change its magnetic properties and compensation point.^{15–17} As the only ferrimagnetic phase among the four stable manganese nitrides,¹⁸ namely, θ - MnN ,¹⁹ η - Mn_3N_2 ,^{20,21} ζ - Mn_2N ,²² and ε - Mn_4N ,⁸ Mn_4N has an anti-perovskite structure with two inequivalent and anti-aligned Mn sublattices at the corners and face centers, and a nitrogen atom at the body center.

Furthermore, there have been emerging interests in magnetoionics, where energy-efficient control of magnetic properties can be

achieved via ionic migration.^{23–40} Nitrogen-based magneto-ionics has been shown to exhibit good cyclabilities and fast ionic migration.^{34,41–43} Recently, nitride-based exchange bias has been reported in MnN/CoFe heterostructures, which can be manipulated via nitrogen ion migration in and out of the MnN layer using an electric field.⁴⁴ This motivates us to build an all-nitride magneto-ionic system taking advantage of the different magnetic phases in Mn nitrides.

Mn_4N thin films are typically grown onto SrTiO_3 or MgO substrates at elevated temperatures through molecular beam epitaxy, pulsed laser deposition, or reactive sputtering in a nitrogen environment.^{7,9–12,14} The film quality is susceptible to the nitrogen flow rate or partial pressure, and the optimum growth conditions vary from study to study.^{7,11} It is challenging to grow high-quality thin films of Mn_4N directly on Si substrates,^{45,46} which are CMOS compatible. In this work, we demonstrate that high-quality (001)-ordered Mn_4N thin films can be grown on Si substrates by directly sputtering pure Mn onto an Mn_3N_2 seed layer at elevated temperatures, resulted from the chemical reaction between Mn and the nitrogen in the Mn_3N_2 seed layer. In a nominally Mn_3N_2 (20 nm)/Mn (t_{Mn}) series of samples, by changing the deposited Mn thickness t_{Mn} from 0 to 50 nm, nitrogen ion migration gradually transforms the layers into $\text{Mn}_3\text{N}_2/\text{Mn}_2\text{N}/\text{Mn}_4\text{N}$,

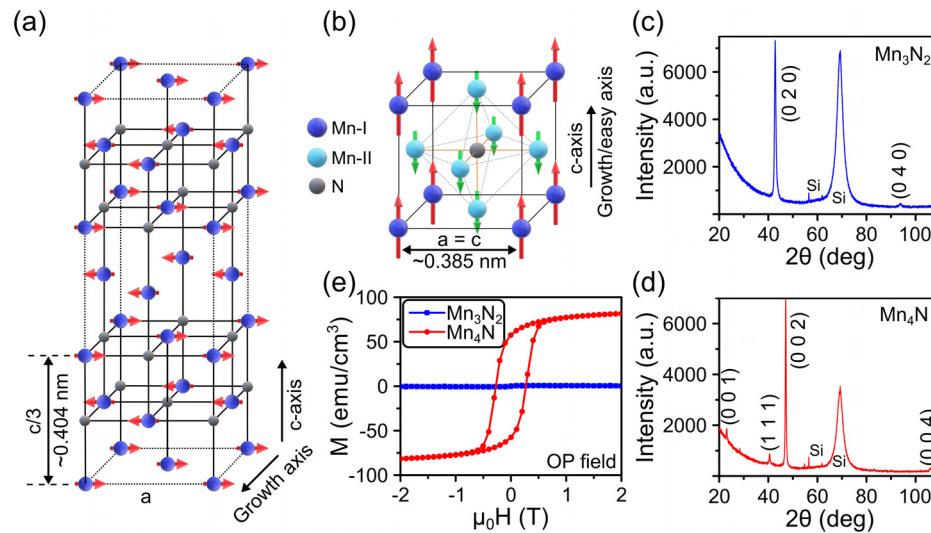
$\text{Mn}_2\text{N}/\text{Mn}_4\text{N}$, and eventually Mn_4N , confirmed by x-ray diffraction (XRD), transmission electron microscopy (TEM), and electron energy loss spectroscopy (EELS). The Mn_4N films are found to exhibit PMA. First-order reversal curve (FORC) measurements reveal that the Mn_4N forms with a nucleation-and-growth process. The nitrogen ion migration is also manifested in a significant exchange bias, up to 0.3 T at 5 K, due to the interaction between ferrimagnetic Mn_4N and antiferromagnetic Mn_3N_2 and Mn_2N .

Seed layers of 20 nm Mn_3N_2 were first reactive-sputtered onto Si substrate with 285 nm thermally oxidized SiO_2 layer from an Mn target using direct current (dc) in an ultrahigh vacuum chamber with a base pressure better than 5×10^{-8} Torr. The substrate temperature was kept at 450 °C, and the Ar:N₂ ratio was held at 1:1 with a 5 mTorr sputtering gas pressure. These Mn_3N_2 films were then left in vacuum for 30 min at the same substrate temperature to promote nitrogen reordering. Subsequently, 0–50 nm of Mn was deposited onto the Mn_3N_2 layer at the same 450 °C substrate temperature in an Ar-only environment. After deposition, substrate heating was immediately turned off, and the samples were cooled to room temperature before depositing a 5 nm Ti capping layer.

Structural characterizations were performed using XRD on a Panalytical X'Pert³ MRD with symmetric $2\theta-\omega$ and grazing incidence geometries. Sample microstructures and composition analysis were done using an FEI Titan Themes Cubed G2 300 (Cs Probe) TEM at KAUST.

Magnetic measurements were carried out using a Quantum Design superconducting quantum interference device (SQUID) magnetometer MPMS3. Exchange bias was measured at 5 K by first field-cooling the sample from 300 K in a 2 T magnetic field, all in the out-of-plane (OP) geometry. FORC measurements^{47–51} were done in a vibrating sample magnetometer at room temperature by saturating samples in a 1.5 T OP magnetic field and then measuring from a reversal field (H_R) back to saturation. This process was repeated at different H_R to fill the interior of the hysteresis loop, creating a family of FORCs. A FORC distribution was then calculated using the following equation:

$$\rho(H, H_R) \equiv -\frac{1}{2M_S} \frac{\partial^2 M(H, H_R)}{\partial H \partial H_R}, \quad (1)$$



where M_S is the saturation magnetization and $M(H, H_R)$ is the magnetization at the applied field H with reversal field H_R . The FORC distribution can also be represented in terms of local coercive field and bias field (H_C, H_B) defined by $H_C = \frac{1}{2}(H - H_R)$ and $H_B = \frac{1}{2}(H + H_R)$.

The η -phase Mn_3N_2 is chosen as the seed layer for Mn_4N growth because it provides the crystalline texture and nitrogen needed for the Mn_4N growth.⁵² As shown in Figs. 1(a) and 1(b), Mn_3N_2 is antiferromagnetic ($T_N \sim 925$ K), and its c -axis is about three times that of Mn_4N .^{21,53} XRD reveals the growth of the Mn_3N_2 phase, shown in Fig. 1(c), with a preferred orientation along the (010) direction and the c -axis in the film plane. Furthermore, grazing incidence XRD confirms that all peaks are from Mn_3N_2 (supplementary material, Fig. S1). Upon depositing 40 nm of Mn, XRD shows that the film is primarily the Mn_4N phase oriented along (001), with no appreciable Mn_3N_2 phase left, as shown in Fig. 1(d) and supplementary material, Fig. S1. Along with the structure changes, there is also a drastic change in the film magnetic properties. As shown in Fig. 1(e), the initial Mn_3N_2 layer does not exhibit any appreciable magnetic signal, consistent with its antiferromagnetic nature; interestingly, the sample deposited with Mn exhibits a square loop with a large coercivity (0.27 T) and a small M_S (85 emu/cm³) that are typical of Mn_4N films.^{7,9,10}

To understand how the film transforms from Mn_3N_2 to Mn_4N by only depositing Mn, we have investigated a series of samples starting with 20 nm Mn_3N_2 seed layer, but the deposited Mn nominal thickness varied from 0 to 50 nm with a 5 nm step size. From now on, each sample is referred by its deposited Mn thickness (t_{Mn}) unless otherwise stated. Figure 2(a) reveals how the Mn nitride phase evolves across the samples. Starting from $t_{\text{Mn}} = 0$ nm, which is the Mn_3N_2 layer, the only peak is the Mn_3N_2 (020). As t_{Mn} increases, a prominent peak emerges around 47.2°, corresponding to the Mn_4N (002), indicating Mn_4N formation as Mn is deposited. On the other hand, the Mn_3N_2 (020) peak diminishes and shifts to higher angles before eventually vanishing in the $t_{\text{Mn}} = 30$ nm sample. This trend indicates that the Mn_3N_2 phase is fading and is not as stable as Mn_4N at high temperatures, consistent with prior studies.^{19,54} Interestingly, as the Mn_3N_2 peak gets smaller, another peak emerges near 42.2° in the

FIG. 1. Schematics showing the lattice structure and spin orientation of (a) Mn_3N_2 and (b) Mn_4N . XRD $2\theta-\omega$ scans of (c) 20 nm Mn_3N_2 seed layer and (d) Mn_4N sample fabricated by depositing 40 nm Mn on top of 20 nm Mn_3N_2 . (e) Room temperature out-of-plane hysteresis loops for the same Mn_3N_2 and Mn_4N samples.

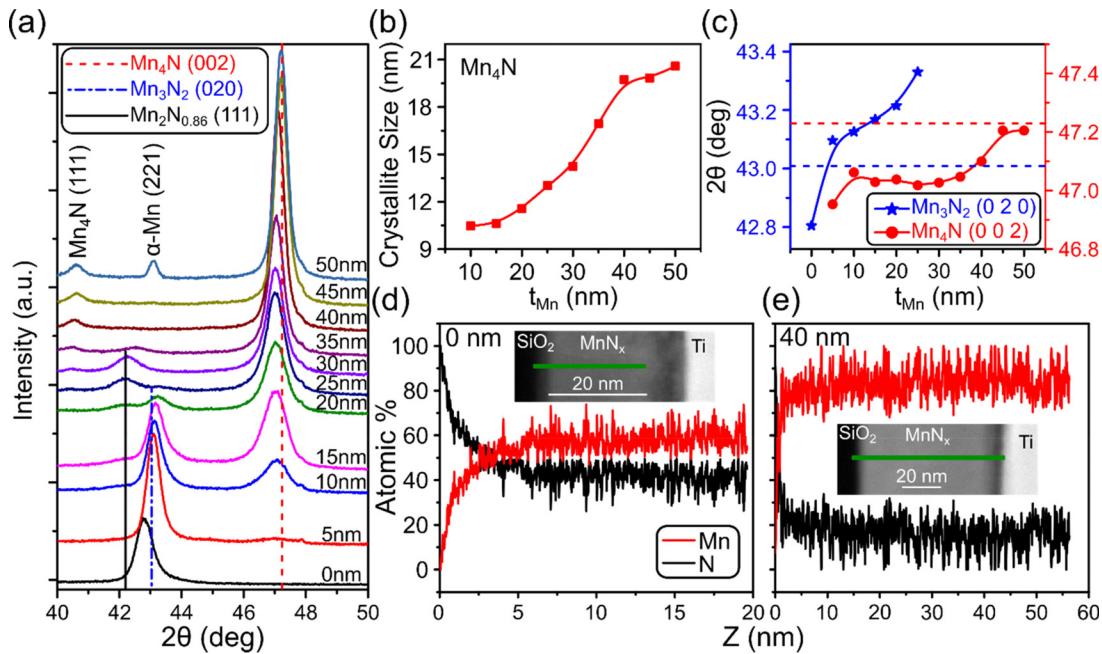


FIG. 2. (a) XRD 2θ - ω scans for nominally Mn_3N_2 (20 nm)/Mn (t_{Mn}) films, and t_{Mn} is the deposited Mn thickness listed next to each curve. Vertical lines show the expected peak locations of Mn_4N (002) (red), Mn_3N_2 (020) (blue), and $\text{Mn}_2\text{N}_{0.86}$ (111) (black). Trends showing (b) Mn_4N crystallite size and (c) Mn_4N (002) (red) and Mn_3N_2 (020) (blue) peak location variations. Solid lines are guides to the eye. Dotted horizontal lines are the expected peak locations for Mn_4N (002) (red) and Mn_3N_2 (020) (blue). EELS scans along the green line in the TEM insets showing Mn:N ratio across the sample for (d) $t_{\text{Mn}} = 0$ nm and (e) $t_{\text{Mn}} = 40$ nm, where $Z = 0$ is the starting point of the interface between the substrate and MnN_x layers.

$t_{\text{Mn}} = 20$ nm sample and grows larger before disappearing in the $t_{\text{Mn}} = 40$ nm sample. This peak is the (111) diffraction from the ζ -phase $\text{Mn}_2\text{N}_{0.86}$, which has a thermal stability and nitrogen content between η - Mn_3N_2 and ε - Mn_4N .^{18,19,54} In the $t_{\text{Mn}} = 40$ and 45 nm samples, both Mn_3N_2 and Mn_2N peaks have vanished, while the Mn_4N (002) peak gets even larger and closer to its expected location. Eventually, when t_{Mn} reaches 50 nm, the α -Mn (221) peak shows up near 43° , indicating that some deposited Mn remains unreacted as the entire nitride film is now Mn_4N . Grazing incidence scans are included in supplementary material, Fig. S1, consistent with Fig. 2(a), along with full range 2θ - ω scans (Fig. S2).

The Mn_4N crystallite size has been estimated from the full-width-at-half-maximum (FWHM) of the (002) peak, after instrument broadening correction, using the Scherrer equation.⁵⁵ The Mn_4N crystallite size nearly doubles as more Mn is deposited, reaching a plateau after $t_{\text{Mn}} = 40$ nm, as shown in Fig. 2(b). This is consistent with the trend in Fig. 2(a), where the Mn_4N peak becomes sharper and more prominent, and Mn_4N is the only phase after t_{Mn} reaches 40 nm. This crystallite size estimation is rather simplified, as it ignores peak width contribution from other factors such as inhomogeneities in d -spacing. As the film stoichiometry changes due to the nitrogen migration, any spread in N-content and the lattice parameters would lead to a broadening of the peak width. Interestingly, the overall narrowing trend of the Mn_4N peak width with increasing t_{Mn} suggests that the stoichiometry variation is suppressed at high t_{Mn} , which is consistent with the aforementioned observation of a single Mn_4N phase when t_{Mn} reaches 40 nm. Moreover, the peak locations shift to higher angles as t_{Mn}

increases, as shown in Fig. 2(c). Nitride phases' lattice constants are known to be very sensitive to nitrogen content,¹⁸ as interstitial nitrogen usually causes the lattices to expand. As Mn_3N_2 loses nitrogen to the deposited Mn, its lattice contracts, causing the Mn_3N_2 peak to shift to a higher angle until this phase is gone. The Mn_4N peak location, on the other hand, stays relatively constant before changing rapidly beyond $t_{\text{Mn}} = 40$ nm, likely caused by the nitrogen redistribution within the Mn_4N phase once the nitrogen from Mn_3N_2 and Mn_2N has been depleted. Thus, we postulate that as Mn is deposited onto the Mn_3N_2 layer at elevated temperatures, it reacts with the nitrogen coming from Mn_3N_2 and forms Mn_4N . While Mn_3N_2 loses nitrogen, it first turns into Mn_2N and eventually becomes Mn_4N . These reactions are summarized in Eq. (2a), and they can be combined into one chemical reaction [Eq. (2b)] since they are multistep reactions,



We also calculated the enthalpy of formation for the reaction shown in Eq. (2b) to be -110 kJ/mol using the standard enthalpy of formation for Mn_4N and Mn_3N_2 ,⁵⁴ indicating that this reaction is thermodynamically favorable.

Cross-sectional TEM and EELS line scans were performed to explore the microstructure and nitrogen content evolution as t_{Mn} varies. Figure 2(d) shows the EELS line scan along the film thickness (green line in the inset) for $t_{\text{Mn}} = 0$ nm. The Mn:N composition ratio

is determined to be 58:42, consistent with the nominal atomic ratio of Mn_3N_2 . As t_{Mn} increases to 40 nm, the EELS scan shown in Fig. 2(e) illustrates that the Mn:N ratio is now 84:16, also consistent with the nominal atomic ratio of Mn_4N . Interestingly, most of the nitride layers appear homogenous with constant Mn:N ratio from both the cross-sectional TEM and EELS, indicating that nitrogen in the Mn_3N_2 seed layer has redistributed to maintain a constant nitrogen concentration within the Mn nitrides after the Mn is deposited. These results further corroborate our postulation that nitrogen moves from the Mn_3N_2 seed layer into the Mn layer to form more stable Mn_4N .

We then investigate the magnetic properties of this series of samples. Figure 3(a) shows the room temperature hysteresis loops with in-plane (IP) and out-of-plane (OP) magnetic fields. The OP loops get more square and broader as t_{Mn} increases from 10 to 50 nm, while the IP loops stay relatively constant. These trends are further revealed by plotting the squareness, or ratio of remanence magnetization (M_r) over M_s , for each sample, Fig. 3(b). The OP and IP remanence are small and stay relatively constant for $t_{\text{Mn}} < 20$ nm, indicating the lack of a clear magnetic easy axis. For $20 \text{ nm} < t_{\text{Mn}} < 35$ nm, a sharp jump in OP remanence is observed, along with a drop in IP remanence, indicating a clear easy axis has been established in the OP direction. At $t_{\text{Mn}} > 35$ nm, the easy axis remains OP, while IP remanence increases slightly but remains low.

We have also calculated the uniaxial magnetic anisotropy constant (K_u , see the supplementary material). As shown in Fig. 3(c), K_u starts out to be negative for $t_{\text{Mn}} = 5$ nm and shows a clear switching from negative to positive, especially when $t_{\text{Mn}} > 20$ nm, further confirming the magnetic easy axis switching to OP as more Mn_4N is formed. Note that K_u exhibits the largest value (0.03 MJ/m³), or the film has the largest PMA when $35 \text{ nm} < t_{\text{Mn}} < 45$ nm. This is also consistent with the XRD result, which shows that Mn_4N is the only phase

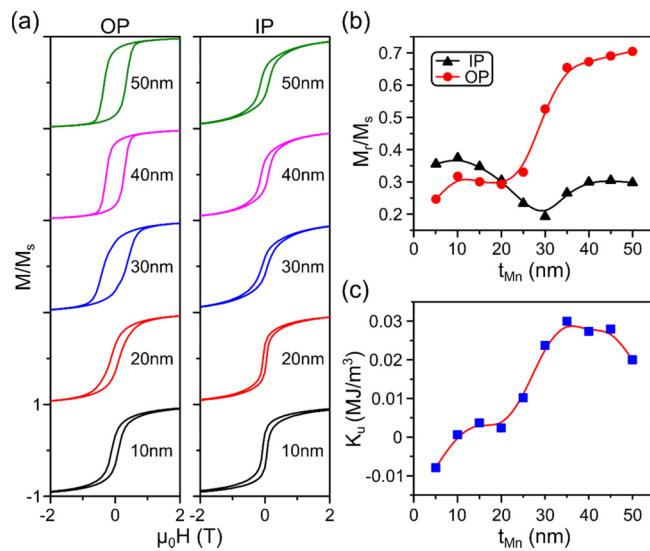


FIG. 3. (a) Room temperature hysteresis loop with out-of-plane (left) and in-plane (right) magnetic fields for nominally Mn_3N_2 (20 nm)/Mn (t_{Mn}) films, where t_{Mn} is the deposited Mn thickness listed next to each curve. Trends for the (b) in-plane and out-of-plane remanence and (c) K_u as t_{Mn} increases. Solid lines in (b) and (c) are guides to the eye.

for this t_{Mn} range. This K_u value is smaller than other reported values, which range from 0.05 to 0.2 MJ/m³.^{7,9–11,56,57} The uniaxial anisotropy has been attributed to the tetragonal distortion caused by in-plane tensile strains.^{7,9,10} However, in this study, the films that show the largest PMA have c/a close to 1 (Table 1 in the supplementary material). The lack of in-plane strain may be the reason for our somewhat smaller K_u values since the films are deposited onto an amorphous SiO_2 layer. Additionally, the sizeable PMA in our films may have come from other contributions such as the shape anisotropy of the Mn_4N grains.⁵⁸

To investigate how the Mn_4N phase evolves with t_{Mn} and the corresponding magnetization reversal, we have carried out FORC studies in the OP geometry at room temperature, as shown in Fig. 4. For the $t_{\text{Mn}} = 10$ and 20 nm samples, individual FORCs fill the major loops in a slanted fashion, Figs. 4(a) and 4(c), respectively. The corresponding FORC distributions exhibit a prominent vertical ridge centered around $H_C = 0$, which corresponds to reversible switching,^{51,59} and a smaller horizontal feature centered at $\mu_0 H_C = 120$ and 150 mT [Figs. 4(b) and 4(d), respectively]. This indicates that the Mn_4N film is mainly reversible and magnetically soft. Likely for this t_{Mn} range, the Mn_4N phase is just emerging in small clusters scattered in an antiferromagnetic matrix of Mn_3N_2 and Mn_2N . As more Mn_4N is formed, families of FORCs for $t_{\text{Mn}} = 30, 40$, and 50 nm are considerably different, as individual FORCs return to positive saturation in a more horizontal fashion, consistent with the establishment of a magnetic easy axis [Figs. 4(e), 4(g), and 4(i)]. Their FORC distributions are also strikingly different. The previous large vertical reversible ridge at $\mu_0 H_C = 0$ becomes smaller and eventually vanishes in the $t_{\text{Mn}} = 50$ nm sample. The horizontal feature along the H_C axis now becomes prominent and shifts to higher $\mu_0 H_C$ of 460, 310, and 390 mT [Figs. 4(f), 4(h), and 4(j), respectively]. The change in relative intensity of the horizontal and vertical FORC features likely indicates that Mn_4N forms via a nucleation-and-growth mechanism, similar to that reported previously in the ordering of L1_0 FeCuPt .⁵⁹

In this nominally Mn_3N_2 (20 nm)/Mn (t_{Mn}) series of samples, the evolution of the AF phase and the emergence of the FiM phase are also manifested in the exchange bias effect, which was studied at 5 K after cooling the samples from room temperature in a positive 2 T OP magnetic field. A significant horizontal shift to the negative field direction, up to 300 mT, and a coercivity enhancement can be seen, Fig. 5(a), typical of exchange bias systems.^{44,60–62} The t_{Mn} dependence of coercivity (H_C) and exchange field (H_E) both exhibit non-monotonic trends, with an intriguing peak around $20 \text{ nm} < t_{\text{Mn}} < 30$ nm, as shown in Fig. 5(b). These trends are likely a combined effect from the AF phase evolution as well as the FiM thickness and M_s variations. To further explore the exchange anisotropy independent of the FiM, we have evaluated the interfacial exchange energy (J_{int}) per unit area using the following equation:^{60,62}

$$J_{\text{int}} = M_{\text{FiM}} t_{\text{FiM}} H_E = m_{\text{FiM}} H_E / A, \quad (3)$$

where M_{FiM} , m_{FiM} , and t_{FiM} are the FiM saturation magnetization, saturation magnetic moment, and layer thickness, respectively, H_E is the exchange field, and A is the sample area. As shown in Fig. 5(c), the dependence of J_{int} on t_{Mn} exhibits a bell-shaped plot that peaks around 20–30 nm. J_{int} is small and increases continuously for t_{Mn} of 5–15 nm, where the dominating AF phase is Mn_3N_2 , as observed by XRD. Due to the high T_N of Mn_3N_2 , only a small fraction of the AF spins is aligned to pin Mn_4N by field-cooling from room temperature,

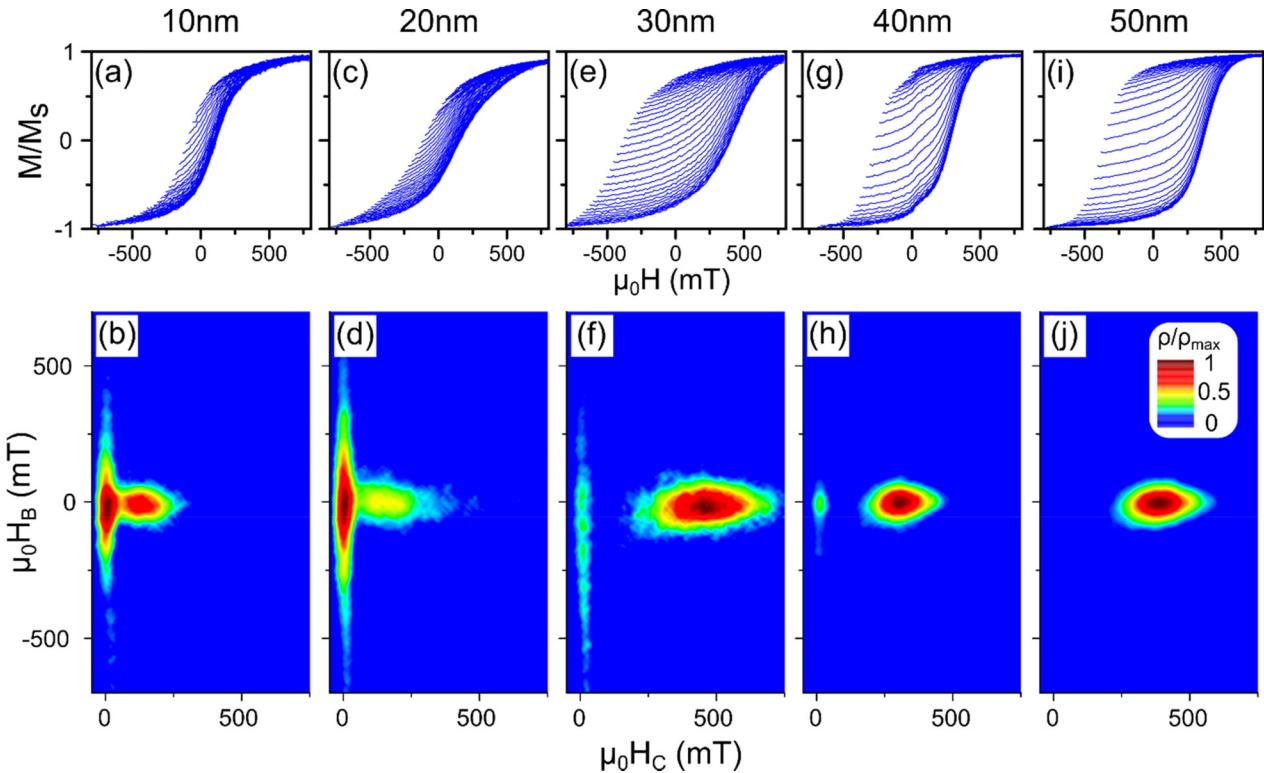


FIG. 4. Families of FORCs (top row) and FORC distributions (bottom row) for [(a) and (b)] $t_{Mn} = 10$ nm, [(c) and (d)] 20 nm, [(e) and (f)] 30 nm, [(g) and (h)] 40 nm, and [(i) and (j)] 50 nm. t_{Mn} is the deposited Mn thickness on top of the 20 nm Mn_3N_2 seed layer.

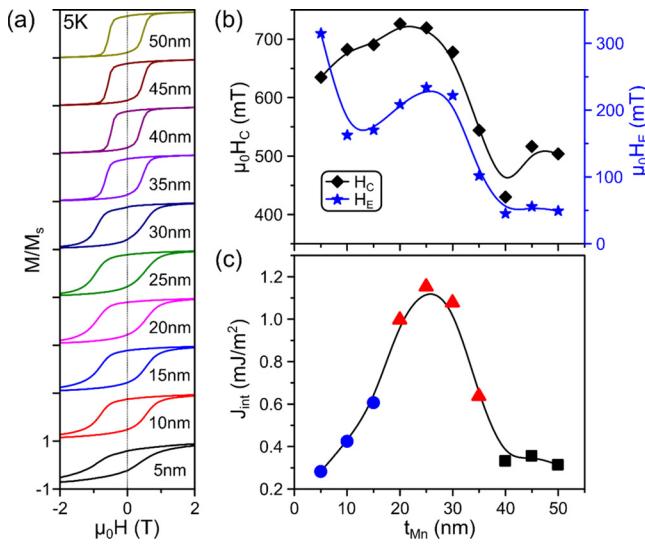


FIG. 5. (a) Hysteresis loops of the nominally Mn_3N_2 (20 nm)/Mn (t_{Mn}) series of samples measured at 5 K after 2 T field-cooling from 300 K, with the t_{Mn} listed for each sample. (b) Dependence of coercivity (black) and exchange field (blue) on t_{Mn} at 5 K. (c) Dependence of interfacial exchange energy on t_{Mn} at 5 K where the color represents the different dominating antiferromagnetic phases: Mn_3N_2 (blue), Mn_2N (red), and no antiferromagnets (black). Solid lines in (b) and (c) are guides to the eye.

resulting in a small J_{int} . However, for $20 \text{ nm} < t_{Mn} < 35 \text{ nm}$ samples, another AF phase, Mn_2N , starts to dominate. By field-cooling from 300 K, the Mn_2N is effectively coupled with Mn_4N , resulting in a significant exchange bias at 5 K. Exchange energy then quickly decreases as Mn_2N is turned into Mn_4N . By $t_{Mn} = 40$ nm, no AF phases can be identified from XRD, and J_{int} mostly vanishes.

In summary, we have achieved high-quality Mn_4N films growth by depositing pure Mn onto an Mn_3N_2 seed layer. By varying the Mn thickness t_{Mn} , the Mn phases in the starting Mn_3N_2 /Mn bilayers can be continuously tuned to be Mn_3N_2 / Mn_2N / Mn_4N , Mn_2N / Mn_4N , and eventually to Mn_4N alone, as observed by XRD and TEM/EELS. With increasing t_{Mn} , more Mn_4N is formed, with an increasing PMA reaching 0.03 MJ/m^3 . FORC measurements further reveal that Mn_4N forms via a nucleation-and-growth mechanism. A large exchange bias up to 0.3 T is found at 5 K in this all-nitride system. The variation of the exchange anisotropy is further attributed to the phase change of the antiferromagnets caused by nitrogen redistribution. These results demonstrate an effective all-nitride magneto-ionic platform for studying the properties of the emergent ferrimagnetic Mn_4N and its potential applications in spintronics.

See the supplementary material for grazing incidence and full range $2\theta-\omega$ x-ray diffraction patterns, Mn nitride phase evolution with layer thickness, cross-sectional TEM and EELS line scans, uniaxial magnetic anisotropy calculation, and additional sample information (PDF).

This work has been supported in part by the NSF (Nos. DMR-2005108 and ECCS-2151809), SMART (No. 2018-NE-2861), one of seven centers of nCORE, a Semiconductor Research Corporation program, sponsored by the National Institute of Standards and Technology (NIST), and by KAUST (No. OSR-2019-CRG8-4081). The acquisition of a Magnetic Property Measurements System (MPMS3), which was used in this investigation, was supported by the NSF-MRI program (No. DMR-1828420).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Zhijie Chen: Conceptualization (equal); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (equal); Validation (lead); Visualization (lead); Writing – original draft (lead); Writing – review & editing (equal). **Christopher J. Jensen:** Data curation (supporting); Formal analysis (supporting); Investigation (supporting); Methodology (equal); Validation (supporting); Writing – review & editing (equal). **Chen Liu:** Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Validation (equal); Visualization (equal); Writing – review & editing (equal). **Xiang Zhang:** Formal analysis (supporting); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Supervision (equal); Validation (equal); Writing – review & editing (equal). **Kai Liu:** Conceptualization (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its supplementary material.

REFERENCES

- ¹L. Caretta, M. Mann, F. Buttner, K. Ueda, B. Pfau, C. M. Gunther, P. Hessing, A. Churikova, C. Klose, M. Schneider, D. Engel, C. Marcus, D. Bono, K. Bagschik, S. Eisebitt, and G. S. D. Beach, *Nat. Nanotechnol.* **13**, 1154 (2018).
- ²R. Blasing, T. P. Ma, S. H. Yang, C. Garg, F. K. Dejene, A. T. N'Diaye, G. Chen, K. Liu, and S. S. P. Parkin, *Nat. Commun.* **9**, 4984 (2018).
- ³J. Finley and L. Liu, *Appl. Phys. Lett.* **116**, 110501 (2020).
- ⁴S. K. Kim, G. S. D. Beach, K.-J. Lee, T. Ono, T. Rasing, and H. Yang, *Nat. Mater.* **21**, 24 (2022).
- ⁵V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak, *Rev. Mod. Phys.* **90**, 015005 (2018).
- ⁶C. O. Avci, A. Quindeau, C. F. Pai, M. Mann, L. Caretta, A. S. Tang, M. C. Onbasli, C. A. Ross, and G. S. Beach, *Nat. Mater.* **16**, 309 (2017).
- ⁷K. Kabara and M. Tsunoda, *J. Appl. Phys.* **117**, 17b512 (2015).
- ⁸W. J. Takei, R. R. Heikes, and G. Shirane, *Phys. Rev.* **125**, 1893 (1962).
- ⁹Y. Yasutomi, K. Ito, T. Sanai, K. Toko, and T. Suemasu, *J. Appl. Phys.* **115**, 17a935 (2014).
- ¹⁰T. Hirose, T. Komori, T. Gushi, A. Anzai, K. Toko, and T. Suemasu, *AIP Adv.* **10**, 025117 (2020).
- ¹¹S. Isogami, K. Masuda, and Y. Miura, *Phys. Rev. Mater.* **4**, 014406 (2020).
- ¹²T. Gushi, M. Jovicevic Klug, J. Pena Garcia, S. Ghosh, J. P. Attane, H. Okuno, O. Fruchart, J. Vogel, T. Suemasu, S. Pizzini, and L. Vila, *Nano Lett.* **19**, 8716 (2019).
- ¹³T. Bayaraa, C. Xu, and L. Bellaiche, *Phys. Rev. Lett.* **127**, 217204 (2021).
- ¹⁴C. T. Ma, T. Q. Hartnett, W. Zhou, P. V. Balachandran, and S. J. Poon, *Appl. Phys. Lett.* **119**, 192406 (2021).
- ¹⁵K. Ito, Y. Yasutomi, S. Zhu, M. Nurmat, M. Tahara, K. Toko, R. Akiyama, Y. Takeda, Y. Saitoh, T. Oguchi, A. Kimura, and T. Suemasu, *Phys. Rev. B* **101**, 104401 (2020).
- ¹⁶S. Ghosh, T. Komori, A. Hallal, J. Pena Garcia, T. Gushi, T. Hirose, H. Mitarai, H. Okuno, J. Vogel, M. Chshiev, J. P. Attane, L. Vila, T. Suemasu, and S. Pizzini, *Nano Lett.* **21**, 2580 (2021).
- ¹⁷S. Isogami, M. Ohtake, and Y. K. Takahashi, *J. Appl. Phys.* **131**, 073904 (2022).
- ¹⁸N. A. Gokcen, *Bull. Alloy Phase Diagrams* **11**, 33 (1990).
- ¹⁹K. Suzuki, T. Kaneko, H. Yoshida, Y. Obi, H. Fujimori, and H. Morita, *J. Alloys Compd.* **306**, 66 (2000).
- ²⁰A. Leineweber, R. Niewa, H. Jacobs, and W. Kockelmann, *J. Mater. Chem.* **10**, 2827 (2000).
- ²¹H. Yang, H. Al-Brithen, A. R. Smith, J. A. Borchers, R. L. Cappelletti, and M. D. Vaudin, *Appl. Phys. Lett.* **78**, 3860 (2001).
- ²²Y. Liu, L. Xu, X. Li, P. Hu, and S. Li, *J. Appl. Phys.* **107**, 103914 (2010).
- ²³M. Weisheit, S. Fähler, A. Marty, Y. Souche, C. Poinsignon, and D. Givord, *Science* **315**, 349 (2007).
- ²⁴U. Bauer, L. Yao, A. J. Tan, P. Agrawal, S. Emori, H. L. Tuller, S. van Dijken, and G. S. D. Beach, *Nat. Mater.* **14**, 174 (2015).
- ²⁵C. 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, *Phys. Rev. Lett.* **113**, 267202 (2014).
- ²⁶D. A. Gilbert, J. Olamit, R. K. Dumas, B. J. Kirby, A. J. Grutter, B. B. Maranville, E. Arenholz, J. A. Borchers, and K. Liu, *Nat. Commun.* **7**, 11050 (2016).
- ²⁷D. A. Gilbert, A. J. Grutter, E. Arenholz, K. Liu, B. J. Kirby, J. A. Borchers, and B. B. Maranville, *Nat. Commun.* **7**, 12264 (2016).
- ²⁸K. Duschek, D. Pohl, S. Fähler, K. Nielsch, and K. Leistner, *APL Mater.* **4**, 032301 (2016).
- ²⁹J. Walter, H. Wang, B. Luo, C. D. Frisbie, and C. Leighton, *ACS Nano* **10**, 7799 (2016).
- ³⁰A. Quintana, E. Menendez, M. O. Liedke, M. Butterling, A. Wagner, V. Sireus, P. Torruella, S. Estrade, F. Peiro, J. Dendooven, C. Detavernier, P. D. Murray, D. A. Gilbert, K. Liu, E. Pellicer, J. Nogues, and J. Sort, *ACS Nano* **12**, 10291 (2018).
- ³¹A. J. Tan, M. Huang, C. O. Avci, F. Büttner, M. Mann, W. Hu, C. Mazzoli, S. Wilkins, H. L. Tuller, and G. S. D. Beach, *Nat. Mater.* **18**, 35 (2019).
- ³²L. Herrera Diez, Y. T. Liu, D. A. Gilbert, M. Belmeguenai, J. Vogel, S. Pizzini, E. Martinez, A. Lamperti, J. B. Mohammadi, A. Laborieux, Y. Roussigné, A. J. Grutter, E. Arenholz, P. Quarterman, B. Maranville, S. Ono, M. S. E. Hadri, R. Tolley, E. E. Fullerton, L. Sanchez-Tejerina, A. Stashkevich, S. M. Chérif, A. D. Kent, D. Querlioz, J. Langer, B. Ocker, and D. Ravelosona, *Phys. Rev. Appl.* **12**, 034005 (2019).
- ³³J. Zehner, R. Huhnstock, S. Oswald, U. Wolff, I. Soldatov, A. Ehresmann, K. Nielsch, D. Holzinger, and K. Leistner, *Adv. Electron. Mater.* **5**, 1900296 (2019).
- ³⁴J. de Rojas, A. Quintana, A. Lopeandía, J. Salguero, B. Muñiz, F. Ibrahim, M. Chshiev, A. Nicolenco, M. O. Liedke, M. Butterling, A. Wagner, V. Sireus, L. Abad, C. J. Jensen, K. Liu, J. Nogués, and J. L. Costa-Krämer, *Nat. Commun.* **11**, 5871 (2020).
- ³⁵G. Chen, A. Mascaraque, H. Jia, B. Zimmermann, M. Robertson, R. L. Conte, M. Hoffmann, M. A. González Barrio, H. Ding, R. Wiesendanger, E. G. Michel, S. Blügel, A. K. Schmid, and K. Liu, *Sci. Adv.* **6**, eaba4924 (2020).
- ³⁶J. Walter, B. Voigt, E. Day-Roberts, K. Heltemes, R. M. Fernandes, T. Birol, and C. Leighton, *Sci. Adv.* **6**, eabb7721 (2020).
- ³⁷G. Chen, M. Robertson, M. Hoffmann, C. Ophus, A. L. F. Cauduro, R. Lo Conte, H. F. Ding, R. Wiesendanger, S. Blügel, A. K. Schmid, and K. Liu, *Phys. Rev. X* **11**, 021015 (2021).
- ³⁸Y. Guan, X. Zhou, F. Li, T. Ma, S.-H. Yang, and S. S. P. Parkin, *Nat. Commun.* **12**, 5002 (2021).

³⁹Q. Wang, Y. Gu, C. Chen, F. Pan, and C. Song, *J. Phys. Chem. Lett.* **13**, 10065 (2022).

⁴⁰G. Chen, C. Ophus, A. Quintana, H. Kwon, C. Won, H. Ding, Y. Wu, A. K. Schmid, and K. Liu, *Nat. Commun.* **13**, 1350 (2022).

⁴¹J. de Rojas, J. Salguero, F. Ibrahim, M. Chshiev, A. Quintana, A. Lopeandia, M. O. Liedke, M. Butterling, E. Hirschmann, A. Wagner, L. Abad, J. L. Costa-Kramer, E. Menendez, and J. Sort, *ACS Appl. Mater. Interfaces* **13**, 30826 (2021).

⁴²J. de Rojas, A. Quintana, G. Rius, C. Stefani, N. Domingo, J. L. Costa-Krämer, E. Menéndez, and J. Sort, *Appl. Phys. Lett.* **120**, 070501 (2022).

⁴³Z. Tan, S. Martins, M. Escobar, J. de Rojas, F. Ibrahim, M. Chshiev, A. Quintana, A. Lopeandia, J. L. Costa-Krämer, E. Menéndez, and J. Sort, *ACS Appl. Mater. Interfaces* **14**, 44581 (2022).

⁴⁴C. J. Jensen, A. Quintana, P. Quarterman, A. J. Grutter, P. P. Balakrishnan, H. Zhang, A. V. Davydov, X. Zhang, and K. Liu, *ACS Nano* **17**, 6745 (2023).

⁴⁵K. M. Ching, W. D. Chang, T. S. Chin, J. G. Duh, and H. C. Ku, *J. Appl. Phys.* **76**, 6582 (1994).

⁴⁶S. Nakagawa and M. Naoe, *J. Appl. Phys.* **75**, 6568 (1994).

⁴⁷C. R. Pike, A. P. Roberts, and K. L. Verosub, *J. Appl. Phys.* **85**, 6660 (1999).

⁴⁸J. E. Davies, O. Hellwig, E. E. Fullerton, G. Denbeaux, J. B. Kortright, and K. Liu, *Phys. Rev. B* **70**, 224434 (2004).

⁴⁹D. A. Gilbert, G. T. Zimanyi, R. K. Dumas, M. Winklhofer, A. Gomez, N. Eibagi, J. L. Vicent, and K. Liu, *Sci. Rep.* **4**, 4204 (2014).

⁵⁰J. A. De Toro, M. Vasilakaki, S. S. Lee, M. S. Andersson, P. S. Normile, N. Yaacoub, P. Murray, E. H. Sánchez, P. Muñiz, D. Peddis, R. Mathieu, K. Liu, J. Geshev, K. N. Trohidou, and J. Nogués, *Chem. Mater.* **29**, 8258 (2017).

⁵¹E. C. Burks, D. A. Gilbert, P. D. Murray, C. Flores, T. E. Felter, S. Charnvanichborikarn, S. O. Kucheyev, J. D. Colvin, G. Yin, and K. Liu, *Nano Lett.* **21**, 716 (2021).

⁵²W. Li, X. Xu, T. Gao, T. Harumoto, Y. Nakamura, and J. Shi, *J. Phys. D* **55**, 275004 (2022).

⁵³M. Tabuchi, M. Takahashi, and F. Kanamaru, *J. Alloys Compd.* **210**, 143 (1994).

⁵⁴R. Yu, X. Chong, Y. Jiang, R. Zhou, W. Yuan, and J. Feng, *RSC Adv.* **5**, 1620 (2015).

⁵⁵A. L. Patterson, *Phys. Rev.* **56**, 978 (1939).

⁵⁶X. Shen, A. Chikamatsu, K. Shigematsu, Y. Hirose, T. Fukumura, and T. Hasegawa, *Appl. Phys. Lett.* **105**, 072410 (2014).

⁵⁷W. Zhou, C. T. Ma, T. Q. Hartnett, P. V. Balachandran, and S. J. Poon, *AIP Adv.* **11**, 015334 (2021).

⁵⁸A. Foley, J. Corbett, A. Khan, A. L. Richard, D. C. Ingram, A. R. Smith, L. Zhao, J. C. Gallagher, and F. Yang, *J. Magn. Magn. Mater.* **439**, 236 (2017).

⁵⁹D. A. Gilbert, J. W. Liao, L. W. Wang, J. W. Lau, T. J. Klemmer, J. U. Thiele, C. H. Lai, and K. Liu, *APL Mater.* **2**, 086106 (2014).

⁶⁰J. Nogués and I. K. Schuller, *J. Magn. Magn. Mater.* **192**, 203 (1999).

⁶¹S. M. Zhou, K. Liu, and C. L. Chien, *Phys. Rev. B* **58**, R14717 (1998).

⁶²R. L. Stamps, *J. Phys. D* **33**, R247 (2000).