

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

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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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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/cm^3),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 magneto-ionics, 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 $\text{Mn}_3\text{N}_2/\text{Mn}_2\text{N}/\text{Mn}_4\text{N}$,

Mn₃N/Mn₄N, and eventually Mn₄N, confirmed by x-ray diffraction (XRD), transmission electron microscopy (TEM), and electron energy loss spectroscopy (EELS). The Mn₄N films are found to exhibit PMA. First-order reversal curve (FORC) measurements reveal that the Mn₄N 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 Mn₄N and antiferromagnetic Mn₃N₂ and Mn₂N.

Seed layers of 20 nm Mn₃N₂ were first reactive-sputtered onto Si substrate with 285 nm thermally oxidized SiO₂ 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 Mn₃N₂ 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 Mn₃N₂ 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 2θ - ω 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 Mn₃N₂ is chosen as the seed layer for Mn₄N growth because it provides the crystalline texture and nitrogen needed for the Mn₄N growth.⁵² As shown in Figs. 1(a) and 1(b), Mn₃N₂ is antiferromagnetic ($T_N \sim 925$ K), and its c -axis is about three times that of Mn₄N.^{21,53} XRD reveals the growth of the Mn₃N₂ 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 Mn₃N₂ (supplementary material, Fig. S1). Upon depositing 40 nm of Mn, XRD shows that the film is primarily the Mn₄N phase oriented along (001), with no appreciable Mn₃N₂ 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 Mn₃N₂ 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 Mn₄N films.^{7,9,10}

To understand how the film transforms from Mn₃N₂ to Mn₄N by only depositing Mn, we have investigated a series of samples starting with 20 nm Mn₃N₂ 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 $t_{Mn} = 0$ nm, which is the Mn₃N₂ layer, the only peak is the Mn₃N₂ (020). As t_{Mn} increases, a prominent peak emerges around 47.2°, corresponding to the Mn₄N (002), indicating Mn₄N formation as Mn is deposited. On the other hand, the Mn₃N₂ (020) peak diminishes and shifts to higher angles before eventually vanishing in the $t_{Mn} = 30$ nm sample. This trend indicates that the Mn₃N₂ phase is fading and is not as stable as Mn₄N at high temperatures, consistent with prior studies.^{19,54} Interestingly, as the Mn₃N₂ peak gets smaller, another peak emerges near 42.2° in the

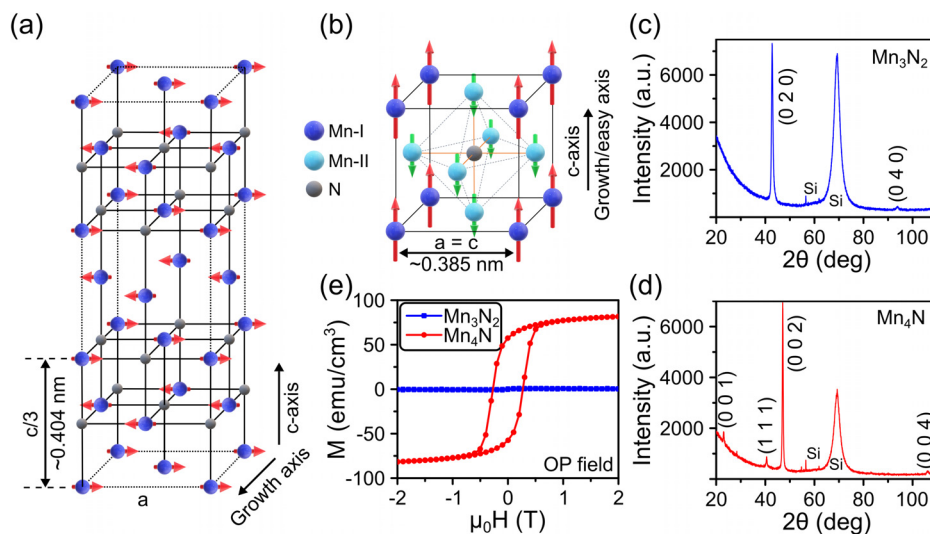


FIG. 1. Schematics showing the lattice structure and spin orientation of (a) Mn₃N₂ and (b) Mn₄N. XRD 2θ - ω scans of (c) 20 nm Mn₃N₂ seed layer and (d) Mn₄N sample fabricated by depositing 40 nm Mn on top of 20 nm Mn₃N₂. (e) Room temperature out-of-plane hysteresis loops for the same Mn₃N₂ and Mn₄N samples.

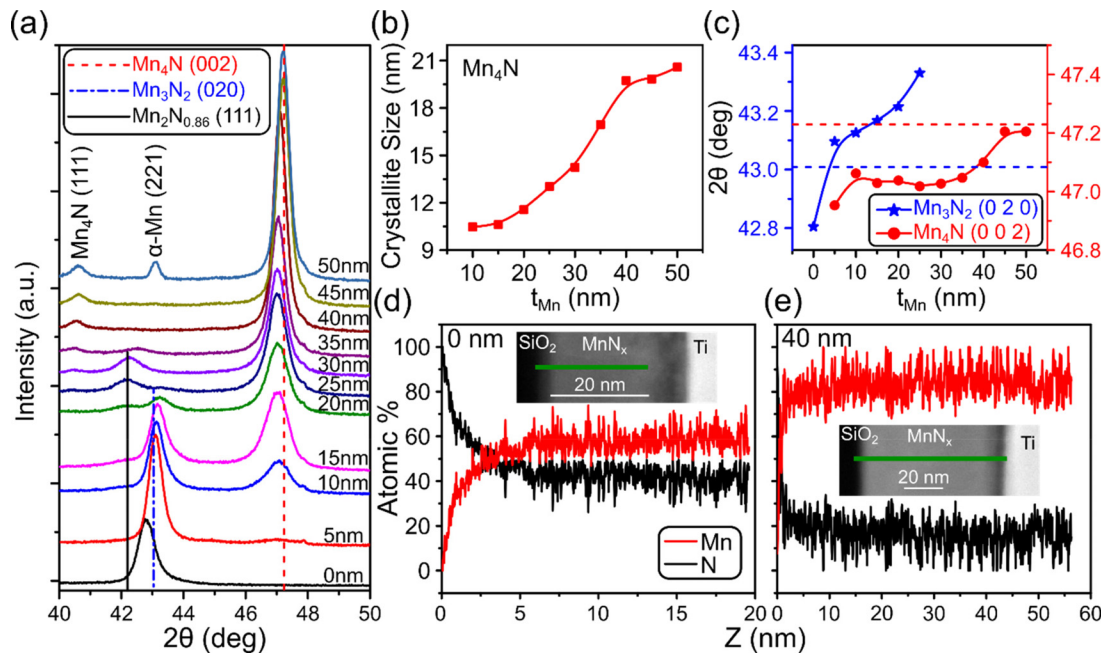
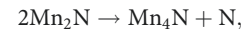


FIG. 2. (a) XRD 2θ - ω scans for nominally Mn_3N_2 (20 nm)/Mn (t_{Mn}) films, and t_{Mn} is the deposited Mn thickness listed next to each curve. Vertical lines show the expected peak locations of Mn_4N (002) (red), Mn_3N_2 (020) (blue), and $\text{Mn}_2\text{N}_{0.86}$ (111) (black). Trends showing (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/m^3), 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

for this t_{Mn} range. This K_u value is smaller than other reported values, which range from 0.05 to 0.2 MJ/m^3 .^{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 has 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,

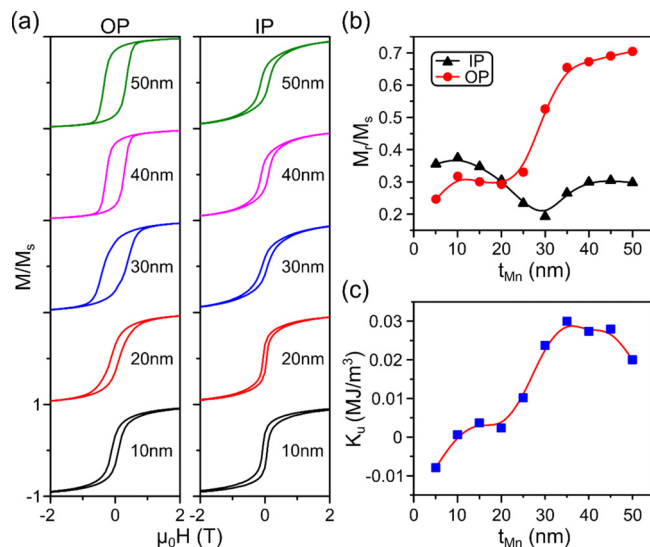


FIG. 3. (a) Room temperature hysteresis loop with out-of-plane (left) and in-plane (right) magnetic fields for nominally Mn_3N_2 (20 nm)/Mn (t_{Mn}) films, where t_{Mn} is the deposited Mn thickness listed next to each curve. Trends for the (b) in-plane and out-of-plane remanence and (c) K_u as t_{Mn}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

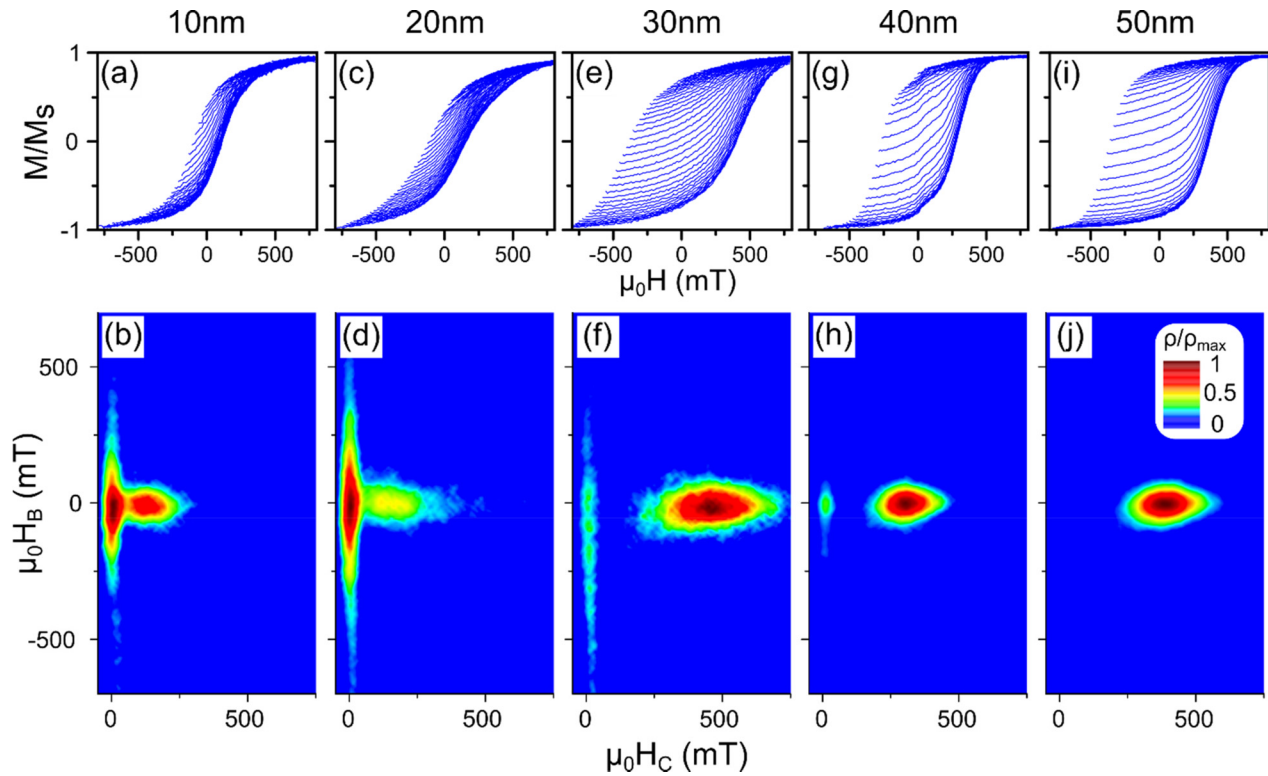


FIG. 4. Families of FORCs (top row) and FORC distributions (bottom row) for [(a) and (b)] $t_{Mn} = 10$ nm, [(c) and (d)] 20 nm, [(e) and (f)] 30 nm, [(g) and (h)] 40 nm, and [(i) and (j)] 50 nm. t_{Mn} is the deposited Mn thickness on top of the 20 nm Mn_3N_2 seed layer.

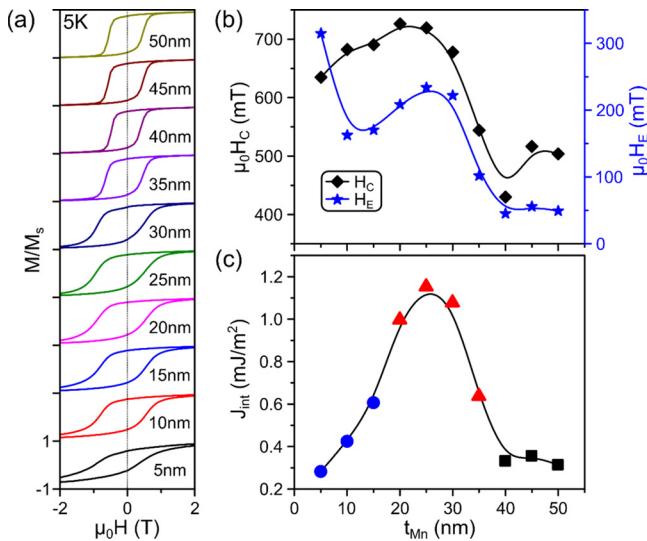


FIG. 5. (a) Hysteresis loops of the nominally Mn_3N_2 (20 nm)/ Mn (t_{Mn}) series of samples measured at 5 K after 2 T field-cooling from 300 K, with the t_{Mn} listed for each sample. (b) Dependence of coercivity (black) and exchange field (blue) on t_{Mn} at 5 K. (c) Dependence of interfacial exchange 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 /Mn₂N/ Mn_4N , Mn_2N /Mn₄N, 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).

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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). **Xixiang 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.

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