Thin films of ferrimagnetic Mn$_4$N are candidate materials to host magnetic skyrmions that have demonstrated thermal stability up to 450°C. However, there are no experimental reports observing skyrmions in this system. Here, we discuss the results of sputter grown 15-17 nm Mn$_4$N thin films on MgO substrate capped with Pt$_{1-x}$Cu$_x$ layers. Vibrating sample magnetometry measurement of out-of-plane hysteresis loops confirmed that magnetic properties are insensitive to the cap layer composition. Imaging based on magnetic force microscopy measurements observed 300 to 50 nm sized skyrmions, as the Cu concentration was increased from $x = 0$ to 0.9. We performed density functional theory calculations and found that the interfacial Dzyaloshinskii-Moriya interactions (iDMI) follow a trend: Mn$_4$N/MgO(001) $<$ Cu/Mn$_4$N(001) $<$ Pt/Mn$_4$N(001). We infer from these calculations that $x$ in Pt$_{1-x}$Cu$_x$ capping layer can serve as a robust tuning knob to tailor the iDMI and control the skyrmion size. This work provides guidance to achieve smaller Néel-type skyrmions in Mn$_4$N thin films, which is an important step forward for building thermally stable skyrmionic devices.

As our society continues to increase its reliance on computing, efficient technologies are needed for processing and high-density data storage. Magnetic skyrmions are promising candidates to serve as information carriers in spintronics devices. Magnetic skyrmions are topological spin textures. They can be stabilized through the Dzyaloshinskii-Moriya interaction (DMI) or stray field. In particular, DMI-skyrmions arise from broken inversion symmetry, which occurs in bulk materials with non-centrosymmetric crystal structure or thin films due to the interfacial asymmetry. Recent studies have reported Bloch skyrmions in bulk B20 crystals with intrinsic DMI at low temperature. On the other hand, Néel skyrmions have been reported in heterostructures with heavy metal interfaces, such as Pt/CoFe/MgO, Ir/Fe/Co/Pt or stray field. For reliable high-density data storage, small (near 10 nm) and stable skyrmions at room temperature are necessary. In ferromagnetic multilayers, due to their large saturation magnetization, which leads to the large stray field, only large skyrmions (> 50 nm) have been observed at room temperature. On the other hand, close to 10 nm skyrmions have been reported in amorphous ferrimagnetic rare-earth transition metal (e.g., GdCo) thin films. However, their poor thermal stability presents a major challenge for device fabrication.

One promising candidate with superior thermal stability compared to the amorphous rare-earth transition metals is rare-earth-free Mn$_4$N. There are some key similarities and differences between the rare-earth transition metals and Mn$_4$N. Both are ferrimagnetic metals, and there have been successful experimental demonstrations of thin film growth in both systems. Both materials show perpendicular magnetic anisotropy (PMA) in the thin film geometry. However, unlike the amorphous structure in the rare-earth transition metals, Mn$_4$N is a crystalline compound that forms in the anti-perovskite crystal structure. Compared to the GdCo ferrimagnets, Mn$_4$N thin films offer key materials processing advantages for improving thermal stability due to the following two reasons: First, the Mn$_4$N films are deposited at 400-450°C and no structural transitions or loss of PMA has been reported after annealing or cooling to room temperature. This means that Mn$_4$N films are robust to temperature changes during device processing. Second, PMA has been reported in films thicker than 10 nm that will enhance the lifetime of skyrmions. Furthermore, recent experiment reports current-driven domain motion of up to 900 m/s for current density of $1.3 \times 10^{12} \text{A/m}^2$. Such high mobility is crucial for high speed skyrmions motion. Although Mn$_4$N thin films appear to possess many favorable characteristics for hosting skyrmions, to date there is no experimental report in the open literature of observing skyrmions in Mn$_4$N thin films. In this paper, we report the experimental observation of skyrmions in Mn$_4$N thin films. In a recent study, tuning of interfacial DMI (iDMI) was reported in Pt/Co/Gd/Pt$_{1-x}$W$_x$ through the dilution of Pt with W in the capping layer. Based on the simulated skyrmion phase diagram, such tuning of iDMI was predicted to allow the control of skyrmion size for desirable applications. In this work, we discuss the tuning of skyrmion size by diluting the Pt with Cu, which is a lighter element than W. Through imaging using magnetic force microscopy (MFM), skyrmions with sizes ranging from 300 to 50 nm have been observed in the MgO/Mn$_4$N/Pt$_{1-x}$Cu$_x$ heterostructures grown by magnetron sputtering. In addition, we also perform micromagnetic simulations and density functional theory (DFT) calculations to extract insights into the key characteristics that govern skyrmion size in the MgO/Mn$_4$N/Pt$_{1-x}$Cu$_x$ heterostructure. Similar to the Pt/Co/Gd/Pt$_{1-x}$W$_x$ work, our findings suggest that the Pt$_{1-x}$Cu$_x$ capping layer effectively reduces the iDMI as a function of $x$, which in turn leads to smaller skyrmions in the Cu-rich capping layer (eventually disappearing when $x = 0.95$). This work shows a path for tailoring smaller skyrmions in Mn$_4$N thin films, whose properties compete with the well-
studied amorphous GdCo and serves as a promising alternative for exploring skyrmion-based devices in thin film geometry with rare-earth-free elements.

We deposited 15±2 nm thick Mn₄N thin films on MgO(001) 5×5×0.5 mm substrate by reactive radio frequency (rf) sputtering at 450°C. We also deposited 3 nm thick cap layers of Ptₓ₋ₓCuₓ (where x = 1, 0.5, 0.1, 0.05) on the sputter grown Mn₄N layer at room temperature by co-sputtering of Pt and Cu targets to tune the iDMI, and 3 nm Pt film on top as the capping layer to prevent oxidation. Details of the deposition process were reported earlier[23]. X-ray diffraction (XRD) was measured with Rigaku SmartLab. MFM images were taken with Bruker atomic force microscopy. Before MFM imaging, the Mn₄N samples were demagnetized by alternating and reducing the magnitude of the applied magnetic field. MFM images were analyzed by Gwyddion[23] and magnetic moments were mapped out using the method described by Zhao et al.[23][24]. We quantified the skyrmion size as the average diameter around a boundary where magnetization is zero[25][26][27].

Spin-polarized electronic structure calculations were carried out in the DFT framework using the planewave pseudopotential code, QUANTUM ESPRESSO[28][29]. Core and valence electrons were treated using ultrasoft pseudopotential[30]. The exchange-correlation functionals were described using the Perdew-Burke-Ernzerhof parameterization of the generalized gradient approximation modified for solids[31]. The plane wave cutoff energy was set to 60 Ry. We explored four Γ-centered Monkhorst-Pack k-point meshes[32] to sample the Brillouin zone: 4×4×1, 10×10×1, 16×16×1 and 20×20×1. Our calculations revealed that we need at least a 16×16×1 k-mesh for convergence. Three separate interfaces were constructed to describe the MgO/Mn₄N/Ptₓ₋ₓCuₓ heterostructure: Mn₄N/MgO(001), Pt/Mn₄N(001), and Cu/Mn₄N(001). The in-plane lattice parameter was fixed to the MgO substrate with a lattice parameter of 4.14 Å for MgO. A vacuum of 22.5 Å was added above the slab for both systems to limit interactions from the periodic image. In the Mn₄N/MgO(001) interface calculation, we assumed that the first Mn₄N atomic layer during thin film growth will contain only the Mn-atoms. In both the Pt/Mn₄N(001) and Cu/Mn₄N(001) calculations, the interface was also assumed to contain only the Mn-atoms.

In both Mn₄N/MgO(001) and Pt/Mn₄N(001) calculations, the interfacial atoms, along with atoms in the layer directly above or beneath the interface, were allowed to relax in the z-direction to optimize the distance by minimizing the forces between the atoms. Since Mn₄N is a ferromagnet in the inverse perovskite structure, spins of the two crystallographically unique Mn-sites that are away from the interface are antiferromagnetically coupled in our calculations. In the Cu/Mn₄N(001) calculation, we substituted all Pt-atoms [shown in (a)] are substituted by Cu-atoms.

The DFT calculations for the N/Pt(001), Pt/Mn₄N(001) and Cu/Mn₄N(001) interfaces are shown in Fig. 1a and b, respectively. We quantified the skyrmion size as the average diameter around a boundary where magnetization is zero[25][26][27].

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Equation 3. We use the D
\[ \text{MgO/Mn} \]
tions is not to accurately model the physics behind the gate for the iDMI strength.

The superscript \( y \) is the number of magnetic layers considered in the simulation, while \( a \) is the lattice parameter. We used \( N_N = 4 \), \( N_L = 1 \), and \( a = 4.14 \) Å in Equation 3. We use the \( D^y_\mu \) (given in Equation 3) as a surrogate for the iDMI strength.

We note that the purpose of these DFT calculations is not to accurately model the physics behind the MgO/Mn\(_N\)/Pt\(_{1-x}\)Cu\(_x\) interface. Our objective is to extract insights that will shed light on the expected iDMI trend at the three interfaces.

Micromagnetic simulations were performed within the Object-Oriented Micromagnetic Framework (OOMMF). Atomistic simulations were performed by an in-house package. The details of the simulations were reported in a previous publication and supplementary materials. Although micromagnetic simulations do not take into account collinear spins in ferrimagnetic Mn\(_N\), it has been benchmarked with atomistic simulations for small skyrmions (< 40 nm) to verify its validity for a ferrimagnet. In a previous publication of sub-20 nm Mn\(_N\) films X-ray diffraction (XRD) showed only the MgO substrate peak and the Mn\(_N\) (002) peak in the 2θ-θ XRD profile. In the φ scan, four peaks at 90° interval confirms the epitaxial growth of Mn\(_N\) (001)[100] on MgO(001)[100].

Fig. 2(a) shows the out-of-plane hysteresis loops as measured by VSM. The data shows PMA in all four Mn\(_N\) films with four cap layer compositions. All samples have a similarly saturated magnetization (\( M_s \)) of 50 emu/cc. Such small \( M_s \) confirms the ferrimagnetic nature of Mn\(_N\). The remanent magnetization to \( M_s \) ratio remains consistently larger than 0.7, insensitive to cap layer concentrations. This is a key result because it shows that the cap layer has little to no effect on the \( M_s \), PMA, and remanent magnetization of the Mn\(_N\) films. Fig. 2(b) shows the XRD scans of MgO/Mn\(_N\)(15 nm)/Pt\(_{1-x}\)Cu\(_x\)(30 nm)/Pt(3 nm). Thicker Pt\(_{1-x}\)Cu\(_x\) cap layers are deposited to obtain an observable signal from the cap layers. Results show while the cap layers remain a face-centered cubic, lattice constant decrease as \( x \) increases from 0 to 0.9.

We next focus on the MFM images that were taken for all four samples after undergoing the demagnetization process as stated in the Methods section. Since the iDMI is related to the SOC, we expect the largest and smallest iDMI in the pure Pt and Pt\(_{0.05}\)Cu\(_{0.95}\) cap layers, respectively. From our previously published atomistic simulations work that explored the effect of iDMI on skyrmion size\(^{31} \), we found that the skyrmions size is proportional to the iDMI strength. Therefore, we anticipate the MgO/Mn\(_N\)/Pt and MgO/Mn\(_N\)/Pt\(_{0.05}\)Cu\(_{0.95}\) heterostructures to host the largest and smallest sized skyrmions, respectively.

Fig. 3(a)-(d) show the MFM images of skyrmions in Mn\(_N\) with different cap layers, and four different areas of a given sample are shown to demonstrate the distribution in skyrmion sizes. Fig. 3(e) shows the distribution of skyrmion size observed from different areas by MFM with Pt\(_{1-x}\)Cu\(_x\) (where \( x = 0.1, 0.9, 0.95 \)) cap layer composition. A blue to red color code maps the frequency shift of the tip due to the interaction with magnetic moments within the sample. Blue color corresponds to negative frequency shift and negative magnetization, while red corresponds to positive frequency shift and positive magnetization. These regions were benchmarked using a demagnetized MgO/Mn\(_N\)/Pt sample. The ferrimagnetic background of a magnetized Mn\(_N\) film would give either a blue or red region, depending on magnetized direction. In this experiment, the Mn\(_N\) films were first demagnetized then a down magnetic field is applied that gives the blue regions of ferrimagnetic backgrounds. Only spin textures that have a well-defined red core are considered skyrmions. For exam-
In Fig. 3(c), we show the MFM image for another heterostructure where we further increased the x to 0.9 (Cu-rich). We find that the average skyrmion size reduced to 100 nm in MgO/Mn₄N/Pt₀.₀₅Cu₀.₉₅. Finally, we increased the x to 0.95 resulting in a MgO/Mn₄N/Pt₀.₀₅Cu₀.₉₅ heterostructure with an almost pure Cu cap layer. In the MFM image shown in Fig. 3(d), only blue regions of ferrimagnetic background were observed, and no evidence for any skyrmions were found in MgO/Mn₄N/Pt₀.₀₅Cu₀.₉₅ heterostructure. There are at least two plausible explanations that can help describe the disappearance of skyrmions in Fig. 3(d): (1) The iDMI from Pt₀.₀₅Cu₀.₉₅ is too weak to stabilize a skyrmion. (2) The limited spatial resolution of MFM fails to image smaller skyrmions (< 50 nm). Further investigations with higher resolution imaging are needed to understand the complex physics at the Mn₄N/Pt₁₋ₓCuₓ interface. Nonetheless, these results show the unique potential of tuning skyrmions sizes in Mn₄N thin films by judiciously adjusting the cap layer composition. This outcome is expected to be critical for device applications, where smaller skyrmion sizes are desirable.

### Table I. DFT and DFT+U (≈3 eV) calculated \( D_{\text{tot}} \) (in meV) and \( D_{\text{y}} \) (in mJ/m²) for the three different interfaces explored in this work.

| Interface                | \( D_{\text{tot}} \) (meV) | \( D_{\text{y}} \) (mJ/m²) |
|--------------------------|-----------------------------|-----------------------------|
| Mn₄N/MgO (001)           | 0.290                       | 1.082                       |
| Mn₄N/MgO (001) (+U)      | 0.273                       | 1.017                       |
| Pt/Mn₄N (001)            | −3.20                       | −11.957                     |
| Pt/Mn₄N (001) (+U)       | −1.86                       | −6.969                      |
| Cu/Mn₄N (001)            | 2.602                       | 9.710                       |
| Cu/Mn₄N (001) (+U)       | 0.706                       | 2.633                       |

We now shift our attention to the DFT calculations. We obtained a significantly high \( D_{\text{tot}} \) and \( D_{\text{y}} \) in the Pt/Mn₄N(001) interface compared to the Mn₄N/MgO(001) interface. The data is given in Table I. Interestingly, the Pt/Mn₄N(001) and Cu/Mn₄N(001) interfaces carry opposite \( D_{\text{tot}} \) signs. Replacement of Pt with Cu in this interface resulted in a positive \( D_{\text{tot}} \) value, but with a slight reduction in the magnitude. When the Hubbard-\( U \) correction was added to electrons in Mn-3d orbitals, we found a reduction in the calculated iDMI in all three interfaces. The most significant drop was in the Cu/Mn₄N (001) interface, where the iDMI dropped to 2.6 mJ/m². Compared to Pt, Cu is a light metal where we do not expect a large iDMI. This highlights the importance of the Hubbard-\( U \) correction term in the iDMI calculations to address the self-interaction error. The change of DMI sign has been observed before in MnₓFe₉₋ₓGe thin films, where it was shown to alter the helix chirality. The consequence of the iDMI sign has been discussed previously in GdC. The spins in a skyrmion will rotate in an opposite direction, but it is not detectable with MFM imaging.

Our DFT+U calculated \( |D_{\text{y}}| \) value for Pt/Mn₄N(001) interface is also of similar magnitude to other Mn compounds reported in the literature. For example, Akanda et al reported a \( D_{\text{y}} \) value between ~11 and 17 mJ/m² in the antiferromagnetic MnP/W interfaces with a single magnetic layer configuration. In another work, Yuan et al reported a \( D_{\text{y}} \)
of ~0.91 and 6.7 mL/m² in the monolayers of MnSSe, MnSSTe and MnSeTe, respectively.\(^{53}\) This was an intriguing result from the context of this work because it shows the importance of the neighboring coordinating atoms (S/Se/Te) in affecting the \(D_\mu\) of the materials system. In Mn\(_2\)N, we have lighter, non-magnetic neighboring N-atoms that are in coordination with the magnetic Mn-atoms. More recently, Morshed et al reported a \(D_\mu\) between 0 and 4.5 mL/m² in the Pt(2)/GdCo(2)/Pt\(_{1-x}\)W\(_x\)(2) heterostructure, where (2) stands for the number of atomic layers in the simulation cell.\(^{53}\) This comparison is also important because it shows that our DFT+\(U\) parameter from DFT was calculated at 0 K, whereas the atomistic simulations and experimental measurements were performed at 300 K. Zhou et al and Schlottet al have shown that iDMI is sensitive to temperature by experimental measurements.\(^{61,62}\) (2) In our DFT calculations, we assumed an ideal interface of Mn-atoms. However, the actual atomic structure near the interface is not known. (3) Uncertainty in the four-state energy mapping analysis is not known and how it propagates to the micromagnetics simulations can impact the predicted skyrmion size. (4) Defects in the thin film sample due to experiments and materials processing can also lead to variations in iDMI, and thus the skyrmion size.\(^{15}\) Our study motivates the need for more experimental and theoretical studies to fully understand the intriguing behavior of rare-earth-free Mn\(_2\)N thin films for skyrmion-based applications.

In summary, thermally stable Mn\(_2\)N thin films have been investigated as a potential material for skyrmion-based spintronics. Magnetic skyrmions in Mn\(_2\)N films stabilized by varying iDMI are imaged by magnetic force microscopy. Skyrmion sizes can be tuned from 300 nm to 80 nm in MgO/Mn\(_2\)N/Pt\(_{1-x}\)Cu\(_x\) by increasing Cu concentration \((x)\) from 0 to 0.95. DFT calculations are employed to study the iDMI in three distinct interfaces. Micromagnetic and atomistic simulations informed by the experimental and DFT calculated parameters shed light on the plausible explanation behind tuning of the skyrmion size in Mn\(_2\)N thin films, and suggest a potential for further reduction in size to \(< 50\) nm with judicious interfacial engineering. These results provide a promising outlook for tailoring skyrmions in Mn\(_2\)N-based thin films, with implications in enabling future spintronics technologies.

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I. **SUPPLEMENTARY MATERIALS**

A. **Methods**

1. **Experiments**

For deposition of Mn4N, we used a base pressure of 7 × 10−8 Torr and deposited from a single Mn target. Flow rates of Ar:N2 ratio were maintained at 93:7. MgO substrates were wet-cleaned and heat-treated before loading into the vacuum chamber. Details of the pre-sputtering cleaning process and sputtering process were reported earlier. Fig. 5 shows an illustration of these heterostructures. The film thickness and epitaxial growth of Mn4N were verified by X-ray reflectometry and diffraction with Rigaku SmartLab. Magnetic properties were measured with VersaLab vibrating sample magnetometry (VSM).

2. **Micromagnetic Simulations**

Both micromagnetic and atomistic simulations were carried out at room temperature using Landau-Lifshitz-Gilbert (LLG) equation, also discussed in earlier work. Total simulated space is 300 nm × 300 nm × 15 nm in micromagnetic simulations, and each cell size is 5 nm × 5 nm × 5 nm. For these simulations, the exchange stiffness constant (A) is 1.5 × 10−11 J/m, estimated from Curie temperature of 745 K. Saturation magnetization Ms is 50 kA/m and anisotropy K is between 0.7 and 1.1 × 105 J/m², based on the previous measurement.

B. **Results**

1. **Additional Magnetic Properties of Mn4N**

Since anisotropy (K) plays a role in skyrmion formation, any changes in K will also affect the skyrmion size. To show that changes in skyrmion size observed are not due to K differences, in-plane hysteresis loop measurements of Mn4N with Pt1−xCu x cap layer are measured and shown in Fig. 6. Results show a similar anisotropy field (> 3 T) in Mn4N. This verifies that the anisotropy (K) of Mn4N is independent of cap layers. Thus, the change in skyrmion size is not due to the change in K.
FIG. 6. Room temperature in-plane hysteresis loop of Mn$_4$N with Pt$_{1-x}$Cu$_x$ cap layer.