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Temperature dependence of the Dzyaloshinskii-Moriya interaction in Pt/Co/Cu thin film heterostructures

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Magnetic materials that exhibit chiral domain walls are of great interest for spintronic devices. In this work, we examine the temperature-dependent behavior of the Dzyaloshinskii-Moriya interaction (DMI) in Pt/Co/Cu thin film heterostructures. We extract the DMI strength, D, from static domain spacing analysis between 300 K and 500 K and compare its temperature dependence to that of the magnetic anisotropy, K_u, and saturation magnetization, M_s. Consistent with expected scaling in thin films, M_s exhibits Bloch-law temperature scaling and K_u scales as $M_s^{2.1\pm0.1}$. However, D varies more strongly with temperature than expected, scaling as $D \propto M_s^{4.9\pm0.7}$, indicating that interfacial DMI is more sensitive to thermal fluctuations than bulk magnetic properties. We suggest that this may be related to the temperature dependence of locally induced magnetic moments in the Pt underlayer and the 3*d*-5*d* orbital interactions at the interface. While we observe stable domain widths in the studied temperature range, a strongly temperature dependent DMI may have significant consequences for potential devices based on the chiral domain wall or skyrmion motion. *Published by AIP Publishing*. https://doi.org/10.1063/1.5038353

Chiral magnetic thin film heterostructures have been of great interest in recent years for their potential for fast domain wall motion[1,2] stabilized by the Dzyaloshinskii-Moriya interaction (DMI) as well as the possibility of hosting room temperature skyrmions in racetrack-based spintronic devices.[3–5] The Dzyaloshinskii-Moriya interaction (DMI) is an anti-symmetric exchange interaction that promotes chiral spin textures in ferromagnetic materials.[6,7] In bulk materials, it can arise from spin-orbit coupling (SOC) acting as a high-order perturbation of the direct exchange interaction,[7,8] or it can result from exchange between two ferromagnetic atoms and a third nonmagnetic atom with large spin-orbit coupling, such as at heavy metal/ferromagnet interfaces.[9] Clarifying the origins of DMI and its relation to other magnetic and environmental parameters is critically important for understanding and designing spin-orbitronic materials for potential applications.

Fundamentally, interfacial magnetic anisotropy (IMA) and DMI in ferromagnet/heavy metal systems both arise from broken inversion symmetry and interfacial spin-orbit coupling (SOC). However, few studies have examined the correlation between DMI and IMA in thin film heterostructures and their relative dependence on temperature, which could provide insights into their common origins. Recent work has suggested that the orbital moment anisotropy responsible for IMA[10] plays a critical role in establishing DMI, based on a correlation between the temperature dependence of the former and latter.[11] While the temperature dependence of IMA has been well-studied theoretically and experimentally,[12–14] much less is known in the case of DMI. Rózsa *et al*. derived the temperature dependence of DMI using a Green's function formalism and found that it scales similarly to Heisenberg exchange.[15] This result is consistent with experiments examining the temperature dependence of the wavelength of spin spirals in Mn on W(110).[16] Barker and Tretiakov concluded a strongly temperature-dependent DMI based on a study of antiferromagnetic skyrmions,[17] but a detailed analysis of the related micromagnetic parameters was lacking.

In this letter, we use static domain spacing analysis and conventional magnetometry to characterize the temperature dependence of DMI, anisotropy, and magnetization and identify the corresponding scaling laws. We find that in the studied heavy-metal ferromagnetic multilayers, the DMI varies significantly with temperature and scales much more strongly than a mean-field model would suggest. This is in contrast to the magnetization and anisotropy, which agree well with existing theory. Despite the strong temperature dependence of all micromagnetic parameters, we find that the domain spacing varies relatively weakly with temperature up to at least 500 K, which would be beneficial in devices in which the bit size and spacing should be robust to temperature variations. However, given the important role of DMI in stabilizing chiral domain walls and skyrmions and its influence on their dynamics,[18] this temperature dependence may have significant consequences for devices dependent on domain wall motion.

We studied Ta(3 nm)/[Pt(2 nm)/Co(1.1 nm)/Cu(1 nm)]$_{15}$/Pt(2 nm) films [Fig. 1(a)] grown on thermally oxidized Si via DC magnetron sputtering. The base chamber pressure was 1.6×10^{-8} Torr, and samples were grown under an Ar pressure of 4 mTorr. Material growth rates were calibrated using *ex situ* X-ray reflectivity which was also used to confirm the final sample thickness and structure. The Pt layers were used

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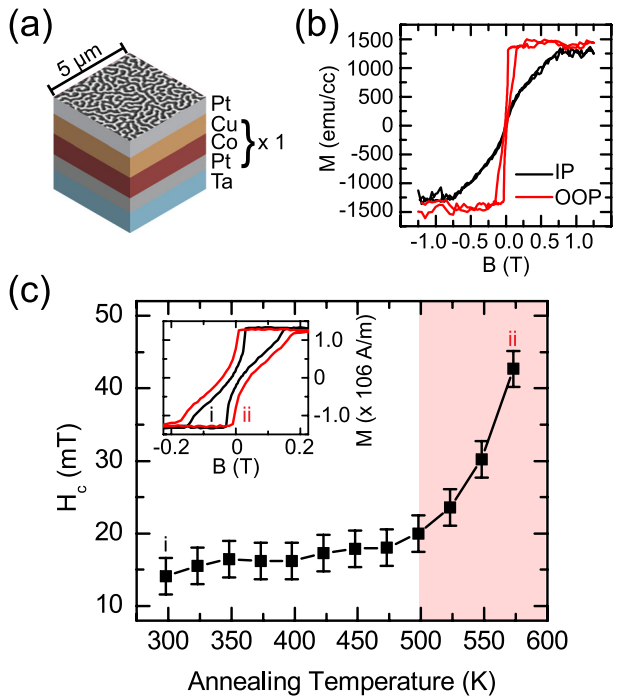

FIG. 1. (a) Schematic representation of the Ta (3)/[Pt (2)/Co (1.1)/Cu (1)]$_{15}$/Pt (2) sample (thicknesses in nm), with a room temperature magnetic force microscopy (MFM) image overlaid. (b) Out of plane (OOP) and in plane (IP) hysteresis loops taken by vibrating sample magnetometry (VSM). (c) Coercivity, H_c, as measured from OOP hysteresis loops after annealing at each temperature point for approximately 1 h. H_c begins to increase at approximately 500 K, which is the onset of annealing in this sample.

as a source of DMI and IMA, while the Cu layers were used to break inversion symmetry without contributing significantly to SOC-induced interfacial effects. We particularly chose to use Cu as a spacer layer because of its immiscibility with Co, which has previously been shown to prevent Co/Pt intermixing at the upper interface.[19] Additionally, Cu has been shown to strengthen perpendicular magnetic anisotropy (PMA) in Pt/Co film systems.[19–21] In-plane (IP) and out-of-plane (OOP) hysteresis loops measured using vibrating sample magnetometry (VSM) [Fig. 1(b)] confirm strong PMA in the film. The sheared OOP loop character with low remanence is characteristic of a multidomain demagnetized state,[22] which is confirmed by magnetic force microscopy (MFM) in the inset of Fig. 1(a).

To determine the DMI constant D, we measured the domain width d in the labyrinth domain state and used a static domain spacing model based on the magnetostatic energies within the system. The analysis accounts for the competition between magnetostatic energy and domain wall energy, with the latter treated using an effective medium model which includes the transverse anisotropy of the domain wall, as proposed by Lemesh *et al.*[23] Similar experimental analysis has been presented by Meier *et al.*[24] From d, one can extract D with knowledge of the saturation magnetization M_s, uniaxial anisotropy constant K_u, and exchange stiffness, A.

Prior to performing temperature-dependent measurements, we determined the temperature threshold above which irreversible changes to the sample properties occur, due to processes such as crystallization or interdiffusion at the interfaces,[25,26] as interfacial DMI is particularly sensitive to such structural changes.[27–29] Figure 1(c) shows the coercivity H_c as a function of annealing temperature (T) for an annealing time of 1 h. The data were obtained on a single sample subjected to sequentially increasing annealing temperatures and cooled back to 300 K between each step to measure an OOP hysteresis loop by VSM. H_c remains nearly constant up to approximately 500 K, beyond which a significant increase is observed. We hence restrict our measurements to temperatures below this threshold.

To measure $d(T)$, we performed MFM at variable temperatures using an Asylum MFP-3D with a sample heating stage (stability ± 0.5 K). Fourier transforms of the labyrinth domain images were used to extract d; an example Fourier transform of an image taken at room temperature is shown in Fig. 2(a), and the corresponding Gaussian fit to the Fourier transform is shown in Fig. 2(b). Prior to acquisition of each image, an *in situ* AC demagnetization cycle was applied at temperature to ensure that the magnetic domains were in their true equilibrium width. The MFM tip was then allowed to reach thermal equilibrium with the sample before imaging for approximately 10 min. For each measurement, the heating, thermal equilibrating, and imaging cycle took approximately 1 h.

MFM was also used to verify that no irreversible structural changes were occurring as the sample was heated. Figure 2(c) shows a labyrinth domain image taken at 300 K after 8 h of imaging at temperatures up to 450 K. The initial value of d at 300 K is 161 ± 2 nm, while after annealing, it is 167 ± 2 nm. The lack of significant variation of d after annealing indicates that the likelihood of large structural changes occurring is low.

Figure 2(d) shows a subset of images taken at several temperatures, and Fig. 2(e) shows the measured $d(T)$. The spike in $d(T)$ at 425 K appears to be an anomaly. As the value of d is carried through to D, this point does not follow the general trend of the entire dataset. For this reason, we believe that there may have been an issue with the AC demagnetization cycle for this data point, resulting in domain widths that were not at equilibria.

M_s and K_u were measured versus T using VSM. Rise times were at a maximum 5 min, and the sample equilibrated at temperature for another 5 min before the measurement cycle. M_s was determined from saturation in the OOP loops, measured to a maximum field of 1.25 T. The area between OOP and IP hysteresis loops was used to determine the effective magnetic anisotropy, $K_{eff} = K_u - \mu_0 M_s^2/2$, which then provides K_u. As shown in Fig. 3(a), M_s is well-fitted in the measured temperature range by Bloch's law, $M_s = M_s(0)(1 - (T/T_C)^x)$, with $x = 2.06$, $M_s(0) = 1.44 \times 10^6$ A/m, and $T_C = 1150$ K. The deviation of x from the expected value of 3/2 can be attributed to the granular nature of sputtered films.[30] Both M_s and K_u vary markedly with T between 300 K and 500 K [Fig. 3(b)], whereas the domain spacing varies rather weakly [Fig. 2(e)], implying that the exchange energies must vary in such a way as to keep the balance between magnetostatic and domain wall energies nearly constant.

The exchange stiffness, A, and its temperature dependence were estimated from the literature. Micromagnetic simulations often take A to be approximately $1.3–1.5 \times 10^{-11}$ J/m (Ref. 31) or $2.2–2.5 \times 10^{-11}$ J/m,[32,33] while experimentally Brillouin light scattering experiments have reported values of

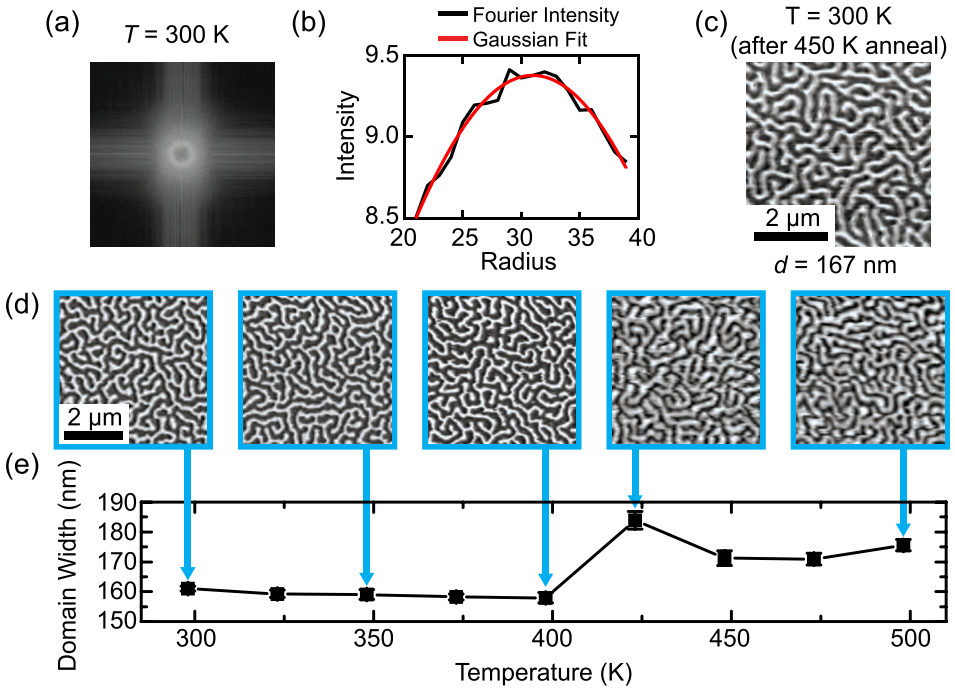

FIG. 2. (a) A sample Fourier transform of an MFM domain image taken at 300 K and (b) the corresponding Gaussian fit to the radial average peak intensity of the Fourier transform. (c) An MFM image taken at 300 K after imaging at temperature steps up to 450 K (roughly 8 h of imaging), with $d = 167 \pm 2$ nm, comparable to the starting value of $d = 161 \pm 2$ nm. (d) A selection of images taken at 300, 350, 400, 425, and 500 K while (e) shows calculated domain widths using Fourier transforms of the MFM images.

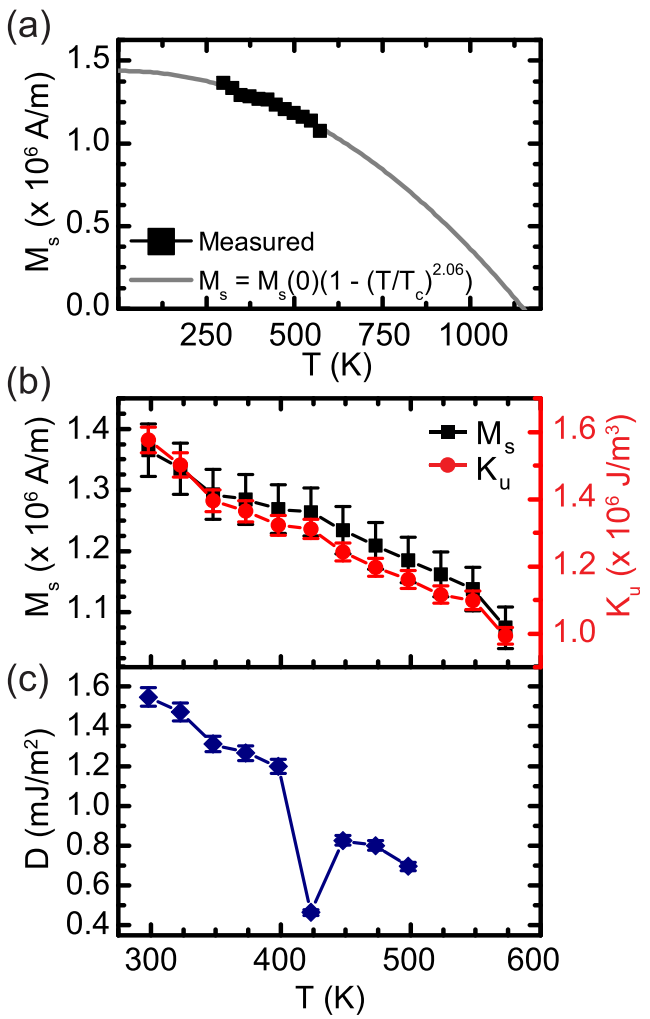

FIG. 3. (a) Measured saturation magnetization, M_s, taken between 300 and 575 K via vibrating sample magnetometry up to 1.25 T. $M_s(T)$ fits well to the Bloch law, such that $M_s(T) = M_s(0)(1 - (T/T_C)^{2.06})$. (b) Magnetocrystalline anisotropy, K_u, overlaid on the previous M_s data and (c) the Dzyaloshinskii-Moriya interaction strength, D.

2.5×10^{-11} J/m or 3.6×10^{-11} J/m, respectively.[34,35] For simplicity, we assumed that $A(300\,\mathrm{K}) = 2.0 \times 10^{-11}$ J/m. Additionally, we model $A(T)$ in accordance with work by Moreno *et al.*,[36] which indicated that in bulk Co, $A(T) \propto M_s^{1.8}$, rather than $\propto M_s^2$ as reported elsewhere.[37,38] We find that the analysis carried out below of $D(T)$ does not depend strongly on the choice of the assumed scaling exponent for $A(T)$, since the latter enters the domain wall energy through its square root, $\sigma = 4\sqrt{AK}$.[23]

From the measured K_u and M_s and the above assumption about $A(T)$, we extracted the DMI strength D from the measured domain spacing d. Here, we used the total energy expression $\sigma(d, A, K_u, M_s, D)$ for the multidomain state that was derived in Eq. (31) of Ref. 23 using an effective medium treatment of the multilayer. D was determined by numerical minimization of σ with respect to d, and the extracted $D(T)$ data are shown in Fig. 3(c). We evaluate this behavior and that of K_u by considering their scaling with M_s. Per Callen and Callen, $K \propto M^{l(l+1)/2}$, which over most temperature ranges results in $K \propto M^3$.[12] We find a best fit, however, with $K_u(T) \propto M_s(T)^{2.2\pm0.1}$, shown in Fig. 4(a). As these films are thin enough to support PMA, interfacial strain effects are likely contributing to the temperature dependence of anisotropy and changing its temperature scaling with M_s.[39,40] Additionally, given evidence that DMI induces some anisotropy in heavy metal/ferromagnet interfaces,[15] their interrelation may influence the net $K_u(T)$.

Neglecting Callen-like fluctuation corrections, one expects $D \propto M_s^2$ in a mean-field approximation similar to the mean-field scaling $A \propto M_s^2$.[15,41] Rather, we find that $D(T) \propto M_s(T)^{4.9\pm0.7}$, as shown in Fig. 4(b), indicating that DMI is substantially more sensitive to temperature than bulk magnetic properties. This may be due to the 2-dimensional nature of interfacial DMI, as it has been shown that DMI in these types of thin film systems is a truly interfacial effect.[42–44] We expect that lattice strains at material interfaces are playing a role in the behavior of $D(T)$. Thermal

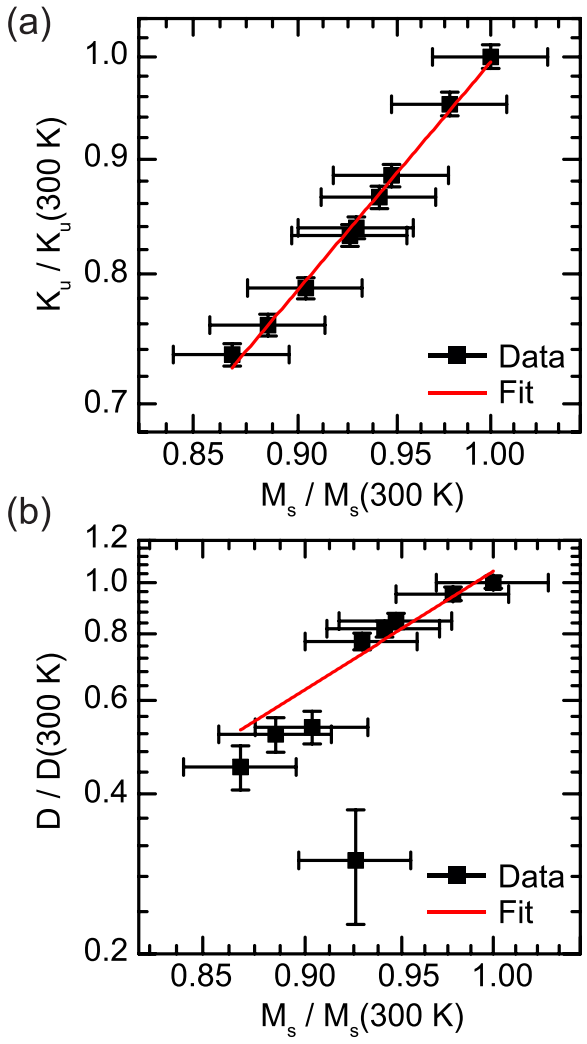

FIG. 4. (a) Log-log plot of the normalized $K_u/K_u(300\,\mathrm{K})$ versus $M_s/M_s(300\,\mathrm{K})$ fit with $K_u \propto M_s^{2.2\pm0.1}$ and (b) a log-log plot of the normalized $D/D(300\,\mathrm{K})$ versus $M_s/M_s(300\,\mathrm{K})$ fit with $D \propto M_s^{4.9\pm0.7}$. Fits were weighted in both x and y, and the fit in (b) excluded the anomalous point at 425 K.

lattice expansion is a primary source of temperature dependence of Heisenberg exchange,[45] and it has been shown that even 5% strain can significantly affect *d–d* hybridization.[46] Disentangling structural changes such as atomic diffusion and lattice strain from the underlying physics of DMI will be essential in future work.

Assuming that the crystal structure was stable during our measurements, we suggest several possible sources for the physical origin of the observed behavior. According to Kim *et al.*,[11] electron hopping occurs between orbitals in noncentrosymmetric systems. The resulting electron-phonon interactions and magnetization fluctuations cause a broadening of the Fermi energy.[11] Our results thus indirectly suggest a correlation between the electron hopping due to inversion symmetry breaking and DMI and additionally with the Rashba field.[47]

We further posit that the strong temperature dependence of the DMI may be indicative of the role of proximity-induced magnetic moments in Pt and the corresponding interaction of the 3*d* and 5*d* orbitals at the Pt/Co interface. Computational work by Belabbes *et al.* has shown that the strength and sign of DMI in 3*d*–5*d* films are highly dependent on the hybridization of the 3*d* and 5*d* orbitals near the Fermi level.[48] Additionally, the locally induced magnetic moments of 5*d* metals in 3*d*/5*d* systems are strongly affected by temperature, an effect which has been shown to influence the temperature dependent behavior of *K* in 3*d*–5*d* alloys[49] and which we can similarly expect to influence the temperature dependence of DMI.

In summary, we measured the temperature dependence of the Dzyaloshinskii-Moriya interaction in Pt/Co/Cu thin film heterostructures using vibrating sample magnetometry and magnetic force microscopy and determined DMI via magnetostatic energy minimization. We find that domain spacing shows only a weak temperature dependence at elevated *T*, which could be a useful attribute for domain wall or skyrmion devices. Further, we found that the behavior of the magnetocrystalline anisotropy deviated from the expected relation, $K_u \propto M_s^3$, but was consistent with similar measurements in the literature. However, DMI scales more strongly with M_s than either *A* or K_u. This work raises questions about the specific sources of the temperature dependence of interfacial DMI, the impact of structural changes on the behavior of DMI, and the relation of the behavior of DMI to that of Heisenberg exchange.

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