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# Ferromagnet/Two-Dimensional Semiconducting Transition-Metal Dichalcogenide Interface with Perpendicular Magnetic Anisotropy

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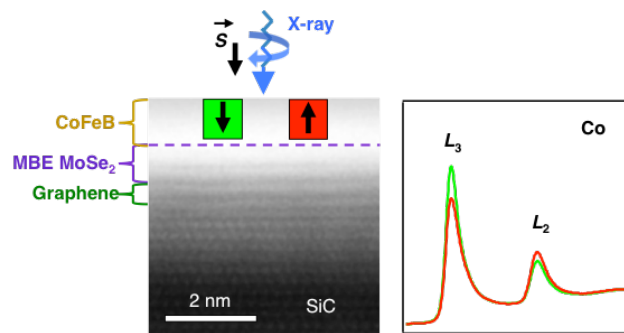
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TOC



ABSTRACT. Ferromagnet/two-dimensional transition-metal dichalcogenide (FM/2D TMD) interfaces provide attractive opportunities to push magnetic information storage to the atomically thin limit. Existing work has focused on FMs contacted with mechanically exfoliated or chemically-vapor-deposition-grown TMDs, where clean interfaces cannot be guaranteed. Here, we report a reliable way to achieve contamination-free interfaces between ferromagnetic CoFeB and molecular-beam epitaxial MoSe<sub>2</sub>. We show a spin re-orientation arising from the interface, leading to a perpendicular magnetic anisotropy (PMA), and reveal the CoFeB/2D MoSe<sub>2</sub> interface allowing for the PMA development in a broader CoFeB thickness-range than common systems such as CoFeB/MgO. Using X-ray magnetic circular dichroism (XMCD) analysis, we attribute generation of this PMA to interfacial *d-d* hybridization, and deduce a general rule to enhance its magnitude. We also demonstrate favorable magnetic softness and considerable magnetic moment preserved at the interface, and theoretically predict the interfacial band matching for spin filtering. Our work highlights the CoFeB/2D MoSe<sub>2</sub> interface as a promising platform for examination of TMD-based spintronic applications and might stimulate further development with other combination of FM/2D TMD interfaces.

KEYWORDS. two-dimensional materials, transition-metal dichalcogenides, spintronics, perpendicular magnetic anisotropy, x-ray magnetic circular dichroism, anisotropic orbital moment, interface

Magnetic interfaces are the building blocks of magnetic sensing and data storage technologies.<sup>1,2</sup> Many desirable effects, such as perpendicular magnetic anisotropy (PMA), spin-to-charge conversion and topologically protected magnetization textures, can be engineered by the interfaces.<sup>3,4</sup> In particular, recent theoretical predictions have triggered interest in hybrid interfaces between 3d ferromagnets (FMs) and two-dimensional transition-metal dichalcogenides (2D TMDs).<sup>5,6</sup> This is attributed to the potential of 2D materials in pushing magnetic information storage to the atomically thin limit, their high stability with strong in-plane bonds, as well as their ability to form hybrid interfaces without the need for lattice matching.<sup>7</sup> Other benefits of 2D TMDs include their diversity in elemental composition, attractive electronic band structures, valley (pseudospin), large spin-orbit coupling and broken inversion symmetry, thereby providing attractive opportunities for magnetization manipulation and spin-current generation at customized atomic interfaces.<sup>8,9</sup>

Existing experimental studies have focused exclusively on chemically-vapor-deposition-grown or mechanically exfoliated TMDs, which necessitate breaking vacuum or physical transfer (onto a substrate) prior to FM deposition.<sup>10,11</sup> Such approaches cannot provide an atomically clean interface, which is a key factor for achieving large spin signals.<sup>12,13</sup> For instance, in the so-called 2D magnetic tunnel junctions such as NiFe/MoS<sub>2</sub>/NiFe, Fe<sub>3</sub>O<sub>4</sub>/MoS<sub>2</sub>/Fe<sub>3</sub>O<sub>4</sub> and Co/MoS<sub>2</sub>/NiFe, only moderate tunneling magnetoresistance has been demonstrated so far.<sup>11,14,15</sup> The values are greatly below theoretical expectations, probably limited by interface degradation or oxidation caused by the air/wet processes.<sup>7</sup>

Here, we demonstrate a growth strategy in response to the above issue. Specifically, we utilize ultrahigh-vacuum molecular beam epitaxy (MBE) to obtain an ultraclean and large-scale TMD surface, with a selenium cap that can be conveniently removed by post-annealing at a moderate

temperature ( $>200^{\circ}\text{C}$ ). This enables (1) wafer-scale TMDs with full coverage on the substrate, guaranteeing no direct contact between the FM and the substrate, and (2) the subsequent FM deposition without interface contamination. Taking advantage of this growth strategy, we achieve a *clean* FM/2D TMD interface between magnetically soft CoFeB and semiconducting 2D MoSe<sub>2</sub>, either of which has been exclusively studied for their application potential in magnetic and spin storage.<sup>16-18</sup> Interestingly, we show a spin re-orientation arising from the CoFeB/2D MoSe<sub>2</sub> interface, leading to a perpendicular magnetic easy axis. Together with the well-retained magnetic moments and magnetic softness at the CoFeB/2D MoSe<sub>2</sub> interface, and theoretically predicted interfacial band matching for spin filtering, our work provides exciting opportunities for layered spintronic devices with atomic-level control of their interface properties and for further miniaturization of next-generation spintronic devices.

## RESULTS AND DISCUSSION

**Structural and electronic information.** Since the 2D MoSe<sub>2</sub> surface serves as a crucial growth front for subsequent CoFeB deposition, we used STM/STS to characterize the properties of the epitaxial TMD. The large-scale STM topography image in **Figure 1a** shows a full coverage of monolayer MoSe<sub>2</sub> on graphene, and triangular islands as the second layer. This coverage of MoSe<sub>2</sub> deposition is intended to prevent any direct contact between CoFeB and the graphene, which could otherwise complicate the present study. In the inset, the line profile indicates that the monolayer height is  $\sim 6.6$  Å, consistent with the expected thickness for one MoSe<sub>2</sub> layer. The semiconducting nature of the MoSe<sub>2</sub> is confirmed by STS measurement shown in **Figure 1b**, where the  $dI/dV$  spectrum taken on the first monolayer reveals a quasi-particle bandgap of  $\sim 2.18$  eV, in good agreement with previous reports.<sup>19</sup> The epitaxial growth of MoSe<sub>2</sub>

on graphene/SiC is confirmed by the cross-sectional high-resolution HAADF-STEM image of the MoSe<sub>2</sub>/graphene interface shown in **Figure 1c**, taken along the [2-1-10] zone axis of SiC. The close-up view of the interface is displayed in the inset. The STEM image of the Pt/CoFeB/MoSe<sub>2</sub> interface of stack 2, with *d*-spacing analysis, is presented in **Figure 1d**, and shows that the CoFeB layer grown on epitaxial MoSe<sub>2</sub> is polycrystalline with a body-centered cubic (bcc) structure.

**Tunable magnetic anisotropy at the CoFeB/2D MoSe<sub>2</sub> interface.** In order to determine the magnetically anisotropic behavior at the CoFeB/2D MoSe<sub>2</sub> interface, XMCD was performed at normal incidence (N.I.,  $\gamma = 0^\circ$ ) and grazing incidence (G.I.,  $\gamma = 60^\circ$ ) of the X-ray beam, as illustrated in **Figure 2a**. In each case, the applied magnetic field was parallel to the incident X-ray beam. **Figures 2b-c** display the angle-dependent XMCD hysteresis loops of the three stacks, obtained by recording the peak height of the Co *L*<sub>3</sub> signal at  $\sim 778$  eV divided by the Co *L*<sub>2</sub> signal at  $\sim 793$  eV, as a function of applied magnetic field. All loops feature a narrow hysteresis, as reflected by the tiny coercivity ( $H_C$ ) in **Figure 2d**, and easy-axis squareness higher than 0.8. Similar to those of other soft-magnetic CoFeB thin layers,<sup>20</sup> the  $H_C$  values are all smaller than 0.01 T, indicating that the three stacks are easily magnetized and demagnetized with low hysteresis loss. Such magnetic softness suggests 2D MoSe<sub>2</sub> as an alternative buffer material for CoFeB-based magnetic switching devices particularly driven by a low electrical switching current.<sup>21</sup> By comparing the N.I. and G.I. hysteresis loops, we assess the magnetic anisotropy of the stacks, which shows a tunability with different Co:Fe compositional ratio and different CoFeB thickness. For stack 1 and 3, the easy axis of magnetization is in the film plane, with the squareness of the G.I. loop much higher than that of the N.I. counterpart. In contrast, stack 2 has its easy axis tilted out of the film plane, with the squareness of the N.I. loop slightly higher than



that of the G.I. one. This observed PMA in stack 2, although small, is noteworthy, due to the following reasons: (i) It is not present in those CoFeB similarly grown on Pt buffer layer (see **Figure S1**). Moreover, the easy-axis redirection arises at the 3-nm-thick CoFeB, suggesting the critical FM thickness ( $t_{cr}$ ) for achieving PMA in CoFeB/2D MoSe<sub>2</sub> is  $\geq 3$  nm, more than double the values in common systems such as CoFeB/MgO and CoFeB/Ta ( $\leq 1.5$  nm).<sup>22-24</sup> Here,  $t_{cr}$  is defined as  $t_{cr} = -2K_i/K_b$  (where  $K_i$  is the interface contribution and  $K_b$  is the bulk contribution to the effective magnetic anisotropy energy),<sup>25</sup> above which the PMA will disappear. Hence, the large  $t_{cr}$  suggests large interface anisotropy energy and allows for the PMA development in a broad thickness-range of CoFeB grown on epitaxial MoSe<sub>2</sub>. (ii) The traditional approach to engineer PMA is to use noble metals or rare earth with high spin-orbit interaction, such as FM/heavy-metal interfaces and rare-earth/transition-metal alloys.<sup>26,27</sup> This usually enhances magnetic damping constant and strongly reduces spin-diffusion lengths, limiting magnetoresistance and precluding low threshold currents for magnetization reversal in spin-torque magnetic random access memories.<sup>28-30</sup> In this regard, our observed PMA is attractive in a view of maintaining weak spin-orbit coupling, which is reflected by the magnetic softness well preserved in the CoFeB layers grown on 2D MoSe<sub>2</sub>.

**Microscopic origin of the PMA at the CoFeB/2D MoSe<sub>2</sub> interface.** We provide atomic-level insight into the observed PMA at the CoFeB/2D MoSe<sub>2</sub> interface by XMCD analysis. **Figure 3** shows typical XA and XMCD spectra taken at the Co  $L_{2,3}$ -edges from stack 2, with no sign of oxide formation or interfacial intermixing which would otherwise lead to multiplet features in the spectra.<sup>31,32</sup> This is further verified by the Fe  $L_{2,3}$ -edge spectra of stack 2, as shown in **Figure S2**, confirming a clean CoFeB/MoSe<sub>2</sub> interface as aimed for in this study.

*Sum rules analysis.* — As shown in **Figure 3**, the XA spectra were taken with parallel ( $\mu^-$ ) and antiparallel ( $\mu^+$ ) alignment of the light helicity with respect to the applied magnetic field, and the XMCD was obtained by ( $\mu^- - \mu^+$ ). Atomic magnetic moments (per hole), including the orbital ( $m_L$ ) and spin ( $m_S$ ) magnetic moments, can be extracted within the framework of the XMCD sum rules as below,<sup>33</sup>

$$m_L = -\frac{4 \int_{L_3+L_2} (\mu^+ - \mu^-) dE}{3P \int_{L_3+L_2} (\mu^+ + \mu^-) dE}, \quad (1)$$

$$m_S + m_T = -\frac{1}{P} \frac{6 \int_{L_3} (\mu^+ - \mu^-) dE - 4 \int_{L_3+L_2} (\mu^+ - \mu^-) dE}{\int_{L_3+L_2} (\mu^+ + \mu^-) dE}, \quad (2)$$

where  $P$  is the X-ray circular polarization, and  $m_T$  corresponds to the magnetic spin dipole moment arising from the anisotropy of the atomic charge distribution. The latter is expected to vary with the incidence angles, which may cause an anisotropy in the spin distribution and thus needs to be taken care of when extracting the value of  $m_S$ . We first treat the ( $m_S + m_T$ ) as a whole and calculate its value from the XA/XMCD spectra using **eq 2**. The resulting ( $m_S + m_T$ ) value is found to be almost isotropic here, *i.e.*,  $(m_S + m_T)_{\text{NI}} \approx (m_S + m_T)_{\text{GI}}$ . Accordingly, the  $m_T$  term can be safely ignored in our case,<sup>34</sup> and the orbital-to-spin moment ratio,  $m_L/m_S$ , can thus be estimated as

$$m_L / m_S = \frac{1}{3 - 4.5 \left(\frac{P}{q}\right)}, \quad (3)$$

where  $p$  is the integral of the dichroic signal of the  $L_3$  peak alone, and  $q$  is the integrated dichroism over both the  $L_3$  and  $L_2$  edges, as marked by the arrows in **Figure 3**. Hereafter, we will focus on discussing the ratio of  $m_L/m_S$ , which can be taken as an estimate of  $m_L$  but provides more accurate information mainly because (*i*)  $m_L/m_S$  is independent of the  $d$ -band hole density in

CoFeB, degree of X-ray circular polarization and status of magnetization, and (ii)  $m_L/m_S$  is robust to the XA background subtraction during the sum rules analysis.<sup>35,36</sup>

*Anisotropic orbital moment versus magnetic anisotropy energy.* — In contrast to  $m_S$ ,  $m_L/m_S$  show a highly anisotropic behavior, as displayed in **Figure 4a**. In particular, the ratio at normal incidence,  $m_{L,NI}/m_{S,NI}$ , shows a much larger variation with Co:Fe ratio and CoFeB thickness, than that at grazing geometry,  $m_{L,GI}/m_{S,GI}$ . The  $m_L/m_S$  of Fe follows a similar trend, as shown in the inset of **Figure 4a**. This suggests that the perpendicular orbital moment plays an essential role in the anisotropic behavior. The resulting anisotropy of orbital moment is estimated by  $\Delta(m_L/m_S) = m_{L,NI}/m_{S,NI} - m_{L,GI}/m_{S,GI}$ .<sup>37</sup> As shown in **Figure 4b** and the inset, both stack 1 and 3 have a negative  $\Delta(m_L/m_S)$  at Co and Fe, while those for stack 2 have an opposite sign. The magnetic anisotropy energy (MAE) of Co, describing the magnetization tendency to align along specific directions,<sup>38</sup> is also plotted for comparison. The MAE is obtained from the difference between the XMCD hysteresis loops measured at different geometries, using  $MAE = \frac{\int_0^{m_L+m_S} (HdM_{NI} - HdM_{GI})}{\sin^2 60^\circ}$ , where  $H$  is the applied magnetic field, and  $M_{NI}$  and  $M_{GI}$  represent the normalized XMCD signal in the N.I. and G.I. loops, respectively.<sup>39,40</sup> Clearly, the MAE follows a consistent trend with the behavior of  $\Delta(m_L/m_S)$ , which is qualitatively in agreement with the perturbation-theory model proposed by Bruno, where MAE is given by a term proportional to  $\Delta m_L$ .<sup>41</sup> This suggests that the orbital moment, although small, has the ability to redirect the spin moment into a perpendicular direction due to the spin-orbit coupling. We have also prepared a wedged sample, as shown in **Figure 4c**, with the ~3-nm-thick CoFeB grown on MoSe<sub>2</sub> and graphene, respectively. With the circularly polarized X-ray beam spots hitting on the two different parts, the Co hysteresis loops are recorded in **Figure 4d-e**. Obviously, the CoFeB grown on graphene shows an in-plane easy

axis of magnetization, in contrast to that grown on MoSe<sub>2</sub>, confirming interfacial origin of the small PMA in CoFeB/MoSe<sub>2</sub>.

*Microscopic origin of the interfacial PMA.* — The above analysis suggests that the anisotropic orbital moment is responsible for the PMA, which would be caused by (i) the broken symmetry of the CoFeB layer as compared with the free layer, and (ii) the exchange interaction between CoFeB and MoSe<sub>2</sub> at the interface. The symmetry breaking and/or the interfacial hybridization lift the degeneracy of the 3*d* orbitals, leading to an energy splitting between those oriented in plane and those with out-of-plane character.<sup>42</sup> Splitting of the bands gives rise to the anisotropic orbital configuration and in turn the anisotropic orbital moments leading to the PMA.<sup>43,44</sup> Here, both Co(3*d*<sup>7</sup>) and Fe(3*d*<sup>6</sup>) have more than half-filled *d*-bands while the Mo<sup>4+</sup>(4*d*<sup>1</sup>) has a less than half-filled *d*-band, favoring *d-d* hybridization at the interface.<sup>45</sup> Similar hybridization effect might be expected for interfaces with similar FM/2D TMD combinations. For instance, W-based TMDs, such as WSe<sub>2</sub> and WS<sub>2</sub> with a less than half-filled *d*-band of W<sup>4+</sup> (5*d*<sup>0</sup>), could be selected and might lead to even stronger interfacial *d-d* hybridization than Mo-based TMDs do. In principle, 3*d* transition metals, including Co, Fe and even Ni could be suitable replacements for CoFeB. However, one has to note probably different growth mode and different magnetic softness from those of CoFeB. In contrast to Mo<sup>4+</sup>(4*d*<sup>1</sup>), the Pt (5*d*<sup>9</sup>) possesses a more than half-filled *d*-band like Co and Fe, which might not promote a strong interaction with the 3*d* metals. This could account for the absence of an interfacial PMA in the samples grown on Pt.

*Estimation of PMA enhancement.* — Unveiling the microscopic origin of the PMA allows us to estimate the critical parameters that could improve the magnitude of the PMA, based on spin re-orientation upon (i) increase of Co:Fe compositional ratio (from stack 1 to 2) and (ii) thickness reduction (from stack 3 to 2). Point (i) suggests that the higher Co:Fe ratio favors

PMA. This could be understood as follows: Compared to Co, which is a strong FM with its spin-up  $d$  band completely filled, Fe is less strongly ferromagnetic with both spin  $d$ -bands only partially filled, and thus would have an electronic structure and magnetic moment that is more sensitive to compositional changes of the alloy.<sup>46</sup> As shown in **Figure 4b**, the  $\Delta(m_L/m_S)$  of Fe shows a strong increase of 0.165, much higher than that of Co (0.097). Combined with the higher value of  $m_S$  in Fe (reflected in Table 1), it suggests that Fe carries a much higher MAE enhancement than Co does upon the compositional change. Point (ii) indicates that the reduced thickness favors PMA. This can be easily understood, as it is established that lower symmetry upon reduced layer thickness can lead to enhanced orbital anisotropy and in turn enhanced MAE.<sup>47</sup> The shift of the Fermi-level ( $E_F$ ) position may also play an important role in this enhancement,<sup>48</sup> as happens when the  $m_S$  changes from stack 3 to 2, which is reflected in Table 1. The  $m_L$  and the resultant MAE, depending strongly on the details of the occupied and unoccupied states near  $E_F$ , will change accordingly.<sup>49,50</sup> Based on the above two points, we therefore conclude that to use a high Co:Fe compositional ratio with a low CoFeB thickness might be an advantageous way to enhance the interfacial PMA in CoFeB/2D MoSe<sub>2</sub> hybrid structures.

*Total magnetic moments at the CoFeB/2D MoSe<sub>2</sub> interface.* — We show in Table 1 that the CoFeB films grown on MoSe<sub>2</sub> possess a comparable magnetic moment to those grown on metal or oxide buffer, topological insulator substrate, *etc.*<sup>51-53</sup> This further suggests that epitaxial MoSe<sub>2</sub> is a good candidate as buffer material for CoFeB growth, combined with the controllable interfacial PMA *via* proper Co:Fe compositional ratio and thickness.

**Theoretical prediction of interfacial band matching in stack 2.** As mentioned above, stack 2 is the most attractive among all three stacks due to the PMA formation, and accordingly we performed theoretical calculations for the electronic band structures of stack 2. **Figure 5a** shows

the complex band (left) and band structure (right) of monolayer MoSe<sub>2</sub>, where the tunneling probability of electrons is revealed to decay exponentially through the MoSe<sub>2</sub>. The evanescent states with  $\Delta_1$  symmetry, possessing the lowest decay rate, is marked in light green in the left panel. These states have the lowest barrier height, and thus will dominate in the electron tunneling of the FM/MoSe<sub>2</sub>/FM device. **Figure 5b** shows the spin-polarized band structure of bcc Co<sub>76</sub>Fe<sub>4</sub> (here we make an approximation of Co<sub>76</sub>Fe<sub>4</sub>  $\approx$  Co considering the very low Fe content), and the states matching the  $\Delta_1$  symmetry of MoSe<sub>2</sub> are labeled. We find that only one spin channel (spin-down state) with  $\Delta_1$  symmetry touches the  $E_F$ , and it is expected that electron tunneling exhibit a symmetry mismatch for the other spin channel. Specifically, the electron with  $\Delta_1$  symmetry in the FMs has the largest transmission when the two FMs are in parallel alignment while very small transmission when they are in antiparallel alignment due to the mismatching of the  $\Delta_1$  state in the spin-up channel of the electrodes.<sup>54</sup> Thus, the  $\Delta_1$  band of monolayer MoSe<sub>2</sub> may act as a  $\Delta_1$  symmetry filter for the electron spins of CoFeB, which is akin to the well-known spin-filtering effect of an MgO barrier with Fe or CoFeB,<sup>55</sup> providing an exciting prospect for achieving coherent tunneling effect and in turn considerable tunneling magnetoresistance in the CoFeB/2D MoSe<sub>2</sub> heterostructure.<sup>56</sup>

## CONCLUSIONS

We have exploited a growth strategy to obtain a *contamination-free* FM/2D TMD interface, with a ferromagnetic CoFeB thin layer sputtered onto MBE grown MoSe<sub>2</sub>. Angle-dependent XMCD analysis demonstrates a spin re-orientation at the 3-nm-thick-Co<sub>76</sub>Fe<sub>4</sub>B<sub>20</sub>/2D MoSe<sub>2</sub> interface, suggesting a larger critical thickness for achieving PMA than those in CoFeB/MgO, CoFeB/Ta, *etc.* and in turn allowing for the development of PMA in a broader FM thickness-

range. By a systematic XMCD sum rules analysis of different compositional ratios and thicknesses of CoFeB at the interface, we reveal that the observed PMA originates from interfacial  $d-d$  hybridization and more importantly, such a PMA is adjustable and is expected to possess a higher value by increasing the Co:Fe compositional ratio and reducing the CoFeB thickness. Together with the magnetic softness and considerable magnetic moment of the magnetic layer, the possible functionality of spin filtering predicted theoretically and of course the benefits of 2D materials (the ultimately reduced thickness and layer-by-layer control, natural sharp definition of the layers and interfaces, *etc.*), the CoFeB/2D MoSe<sub>2</sub> interface offers a promising platform for investigation and realization of technologically relevant magnetic and spintronic devices incorporating 2D TMDs. In particular, the CoFeB/2D MoSe<sub>2</sub> interface could be attractive in efficient generation of spin-orbit torques,<sup>57</sup> for the strong spin-orbit coupling and inversion symmetry breaking of monolayer TMD.<sup>58,59</sup> In addition, the monolayer TMDs, such as MoSe<sub>2</sub>, could also provide an advantageous platform for studying the engineering of spin-orbit torques at the 2D scale, in contrast to the relatively large thickness (~6 nm) required for topological insulators.<sup>60</sup>

## METHODS

**MBE growth of 2D MoSe<sub>2</sub>.** As first demonstrated by Koma *et al.*<sup>61</sup> and in a number of recent studies,<sup>62,63</sup> MBE has become an increasingly important route to fabricate atomically thin layered TMDs. Compared to other popular growth approaches involving low-vacuum and ambient-based chemical vapor deposition techniques,<sup>64,65</sup> MBE has distinct advantages of possible scaling up to wafer sizes and atomic-level controls over stoichiometry, crystallinity and thickness. In this work, a nitrogen-doped SiC(0001) substrate was first graphitized in ultrahigh vacuum by heating

to 1100°C, leading to a graphene-terminated surface.<sup>66</sup> 2D MoSe<sub>2</sub> was then deposited on the substrate in an in-house-built MBE chamber, with a base pressure of better than  $1 \times 10^{-9}$  mbar. High-purity Mo and Se were co-evaporated from an electron-beam evaporator and a standard Knudsen cell, respectively, with the substrate temperature kept at 500°C and a flux ratio of at least ~1:10. To protect samples from environmental contamination during *ex-situ* transport to other ultrahigh-vacuum measurement chambers, a 20-nm-thick Se cap was deposited onto the sample surface. Prior to subsequent sputter deposition of the CoFeB overlayer, this protective cap was desorbed by annealing at 200°C for 20 minutes.

**Sputter deposition of CoFeB.** Two CoFeB targets with different stoichiometry, *i.e.*, Co<sub>60</sub>Fe<sub>20</sub>B<sub>20</sub> and Co<sub>76</sub>Fe<sub>4</sub>B<sub>20</sub>, were used in this work. All depositions were performed at room temperature in a magnetron sputtering system with a base pressure of  $1.0 \times 10^{-5}$  Pa, and the sputter conditions were optimized at 0.4 Pa Ar pressure and 20 W DC power, in order to minimize damage to the epitaxial MoSe<sub>2</sub> layer. To prevent the CoFeB film from oxidation, a 6-nm-thick Pt film was utilized as a capping layer. After fabrication, all samples were heat treated at 380°C in vacuum for 1 hour. The general sample structure is, therefore, SiC/graphene/MoSe<sub>2</sub>/CoFeB/Pt, which we have further categorized, according to the CoFeB composition and thickness, as Co<sub>60</sub>Fe<sub>20</sub>B<sub>20</sub>(3 nm) for stack 1, Co<sub>76</sub>Fe<sub>4</sub>B<sub>20</sub>(3 nm) for stack 2, and Co<sub>76</sub>Fe<sub>4</sub>B<sub>20</sub>(6 nm) for stack 3.

**Scanning tunneling microscopy/spectroscopy (STM/STS) characterization.** STM measurements for 2D MoSe<sub>2</sub> were carried out in a multi-chamber ultrahigh-vacuum system housing an Omicron LT-STM interfaced to a Nanonis controller. The base pressure was better than  $10^{-10}$  mbar. A chemically etched tungsten tip was used, and the sample was kept at 77 K during the measurement. STM images were recorded in constant-current mode. For the  $dI/dV$



spectroscopic measurements, the tunneling current was obtained using the lock-in technique with a bias modulation of 40 mV at a frequency of 625 Hz.

**X-ray absorption/magnetic circular dichroism (XA/XMCD) measurements.** Conventional magnetic characterization tools, such as vibrating sample magnetometry (VSM) and superconducting quantum interference device (SQUID), cannot distinguish the intrinsic magnetic signal of CoFeB from that of the substrate. As shown in **Figure S3**, the MoSe<sub>2</sub>/graphene/SiC presents hysteresis as well, and in particular, the difference between the in-plane and out-of-plane loops shows a magnetic anisotropy, which would complicate the study. A similar phenomenon has been reported previously,<sup>67</sup> which has led to a controversy on the TMD magnetism, while subtracting the substrate contribution from the VSM/SQUID signal could be very tricky.<sup>68</sup> At this point, synchrotron radiation based XMCD is a perfect tool for investigating the *intrinsic* magnetism from the CoFeB, by selecting the X-rays within the energy range of the absorption edges of specific elements.<sup>69</sup> The Co and Fe  $L_{2,3}$  edge XA/XMCD spectra were collected in total-electron-yield mode,<sup>70</sup> where the sample drain current was recorded as a function of photon energy, with fixed helicity of 60% circularly polarized X-rays and opposite magnetic fields up to  $\pm 1.5$  T. The angle of incidence of the photon beam was set to  $0^\circ$  and  $60^\circ$ , respectively, with respect to the sample normal. After normalization to the incident beam intensity, the XMCD spectrum was obtained as the difference between the two XA spectra measured with opposite magnetization directions. XMCD hysteresis loops were measured by recording the peak heights of the Co  $L_3$  and  $L_2$  XA signals, respectively, as a function of applied magnetic field. Measurements were carried out at room temperature on beamline 6.3.1 at the Advanced Light Source, Berkeley, USA.

**Scanning transmission electron microscopy (STEM) analysis.** Cross-sectional high-resolution high-angle annular dark-field (HAADF)-STEM was performed using a JEOL JEM-ARM200F microscope equipped with an ASCOR aberration corrector and cold-field emission gun and operated at 200 kV. Electron transparent cross-sectional TEM samples were prepared by *in-situ* lift out using a FEI-VERSA-3d focused-ion beam machine. The HAADF images were acquired with a probe-forming aperture of 30 mrad and collection angle of 68-280 mrad. All HAADF images were filtered by radial Wiener filters to reduce the scanning noise.

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## FIGURES

**Figure 1.** STM/STS results for epitaxial MoSe<sub>2</sub> grown on graphene, and cross-sectional STEM analysis on the Pt/CoFeB/MoSe<sub>2</sub> interface. (a) Large-scale STM image of MoSe<sub>2</sub> terraces on graphene. Inset: Line profile indicating the height of monolayer MoSe<sub>2</sub> is  $\sim 6.6$  Å. (b) Spatially averaged STS spectrum indicating the semiconducting character of monolayer MoSe<sub>2</sub> with a bandgap of  $\sim 2.18$  eV. (c) Cross-sectional high-resolution HAADF-STEM image of the MoSe<sub>2</sub>/graphene interface taken along the [2-1-10] zone axis of SiC and a close up view of the interface (inset) showing the bilayer MoSe<sub>2</sub> and bilayer graphene at the interface. (d) Cross-sectional high-resolution HAADF-STEM image of the Pt/CoFeB/MoSe<sub>2</sub> interface showing *d*-spacing matching with (220) and (002) planes of bcc CoFeB.

**Figure 2.** Angle-dependent XMCD hysteresis loops taken at the Co  $L_{2,3}$ -edges of CoFeB thin layers grown on epitaxial MoSe<sub>2</sub>. (a) Schematic diagram of the experimental geometry for the XMCD measurements, with the applied magnetic field along the X-ray incidence direction. (b,c) Angle-dependent hysteresis loops taken at normal (N.I.,  $\gamma = 0^\circ$ , red) and grazing (G.I.,  $\gamma = 60^\circ$ , blue) incidences, respectively. The two insets display the loops of stack 2 in the same field scale of  $\pm 0.2$  T, indicating an easy axis pointing along the direction of the surface normal, in contrast to stack 1 and stack 3 where the preferred magnetization is in the film plane. (d) Values of the coercivity ( $H_C$ , *i.e.*, the reverse field required to demagnetize the samples after magnetization saturation) obtained from the hysteresis loops.

**Figure 3.** XA and XMCD spectra of stack 2 taken at the Co  $L_{2,3}$  edges for (a) N.I. and (b) G.I., respectively. The spectra are clean, with no sign of multiplet structure, suggesting no intermixing or diffusion at the Pt/CoFeB or CoFeB/MoSe<sub>2</sub> interface. The XA spectra were taken with parallel

( $\mu^-$ ) and antiparallel ( $\mu^+$ ) alignment of the light helicity with respect to the applied magnetic field, and the XMCD was obtained by ( $\mu^- - \mu^+$ ). The pink and aqua lines represent the integrals of the XMCD for N.I. and G.I., respectively. The arrows mark the parameters  $p$  and  $q$ , derived from the integral of the dichroic signal.

**Figure 4.** Anisotropic orbital moment *versus* magnetic anisotropy energy (MAE). (a) The  $m_L/m_S$  ratios extracted from Co and Fe (inset, with the same axis labels)  $L$ -edge XMCD spectra, respectively, for different geometries. (b) Anisotropy of orbital moment,  $\Delta(m_L/m_S) = m_{L,NI}/m_{S,NI} - m_{L,GI}/m_{S,GI}$ , at Co and Fe (inset, with the same axis labels), respectively, *versus* Co MAE calculated from the angle dependent Co XMCD hysteresis loops. (c) Illustration of step-by-step growth of the CoFeB/MoSe<sub>2</sub>/graphene/SiC hybrid structure, with the X-ray beam hitting on the areas of the CoFeB/MoSe<sub>2</sub> and CoFeB/graphene interfaces, respectively. (d,e) XMCD hysteresis loops, taken at the Co  $L_{2,3}$  edges, of the two abovementioned interfaces.

**Figure 5.** Theoretical calculations for bcc(110)-CoFeB/2D MoSe<sub>2</sub> interface. (a) Complex band (left) and band structure (right) of monolayer MoSe<sub>2</sub>, which is a semiconductor with a direct bandgap. The tunneling probability of electrons decays exponentially through MoSe<sub>2</sub>. The evanescent states with  $\Delta_1$  symmetry, possessing the lowest decay rate, is marked in light green in the left panel. (b) Spin-polarized band structure of bcc Co<sub>76</sub>Fe<sub>4</sub> (here we make an approximation of Co<sub>76</sub>Fe<sub>4</sub>  $\approx$  Co considering the very low Fe content), with the states matching the  $\Delta_1$  symmetry of MoSe<sub>2</sub> labeled. The Fermi level ( $E_F$ ) is set to zero.

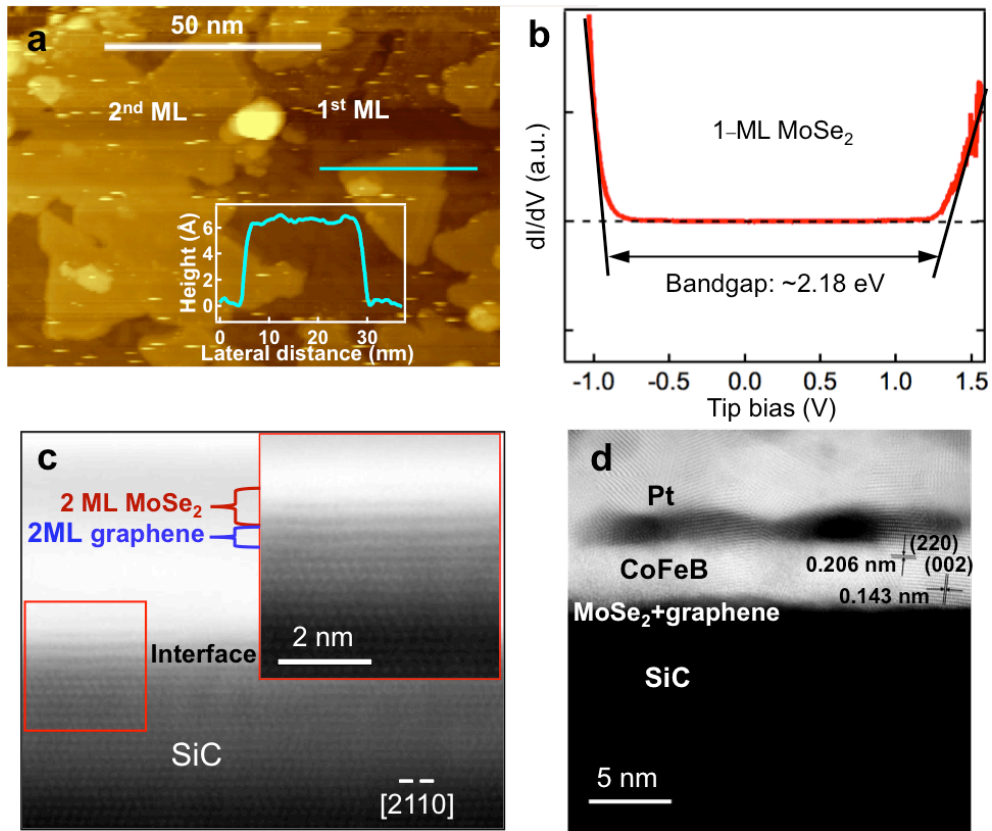


Figure 1

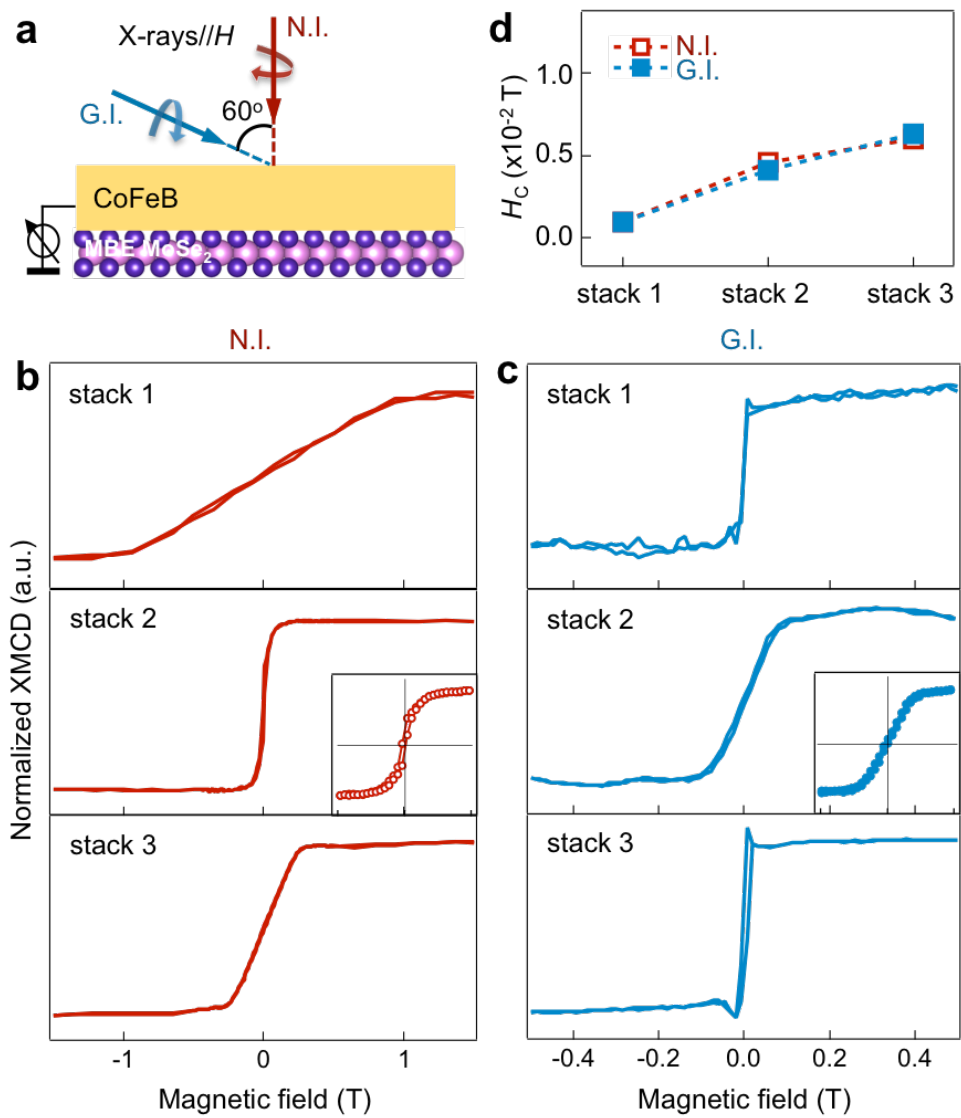


Figure 2

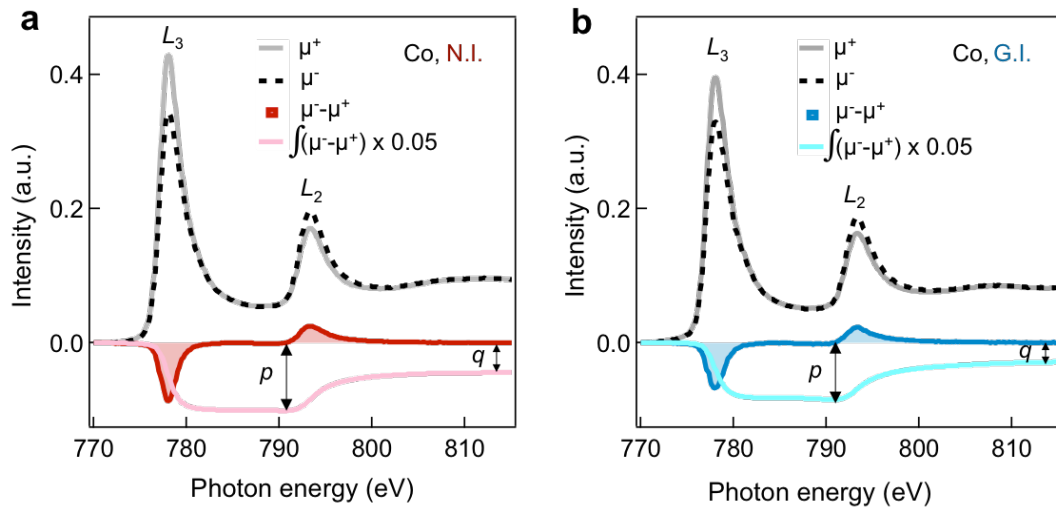


Figure 3

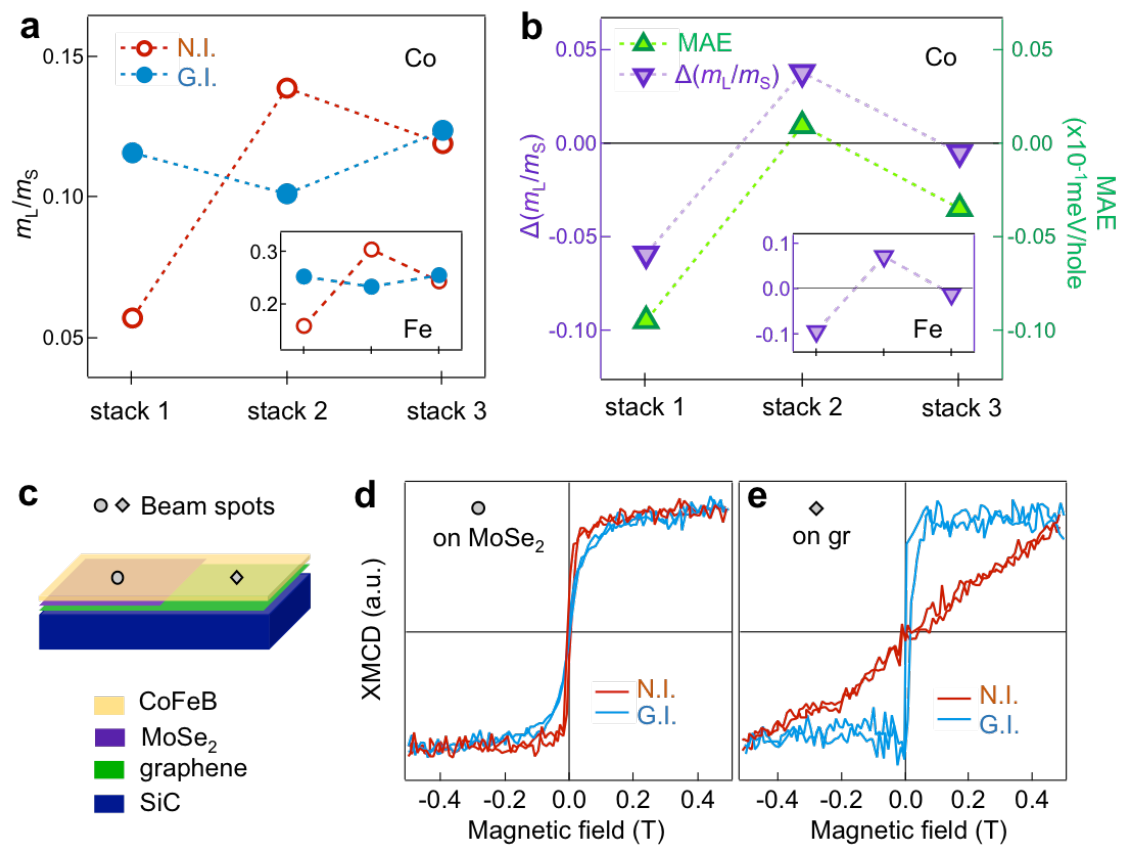


Figure 4



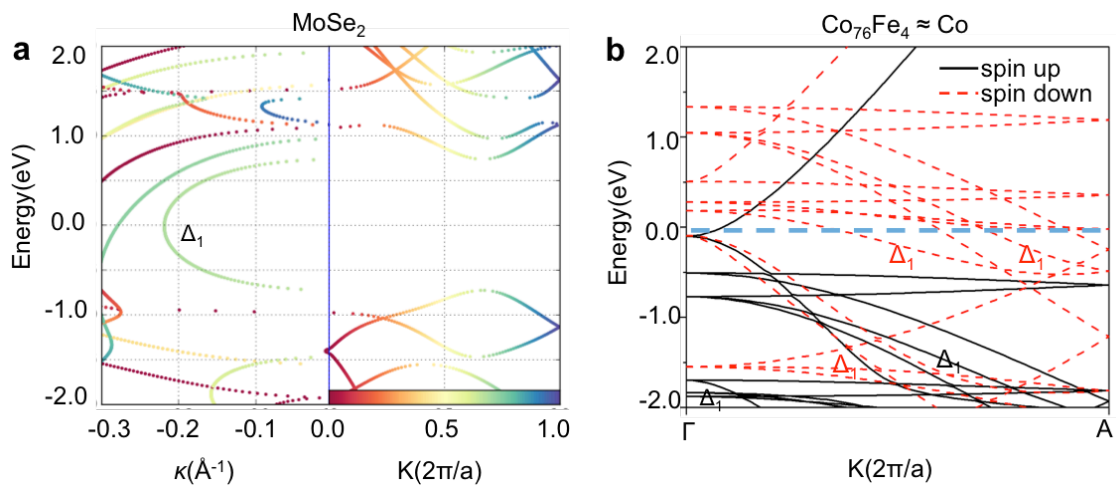


Figure 5

## TABLES

**Table 1.** Total magnetic moments,  $m_L + m_S$ , of Co and Fe extracted from Co and Fe  $L_{2,3}$ -edge XMCD spectra using the sum rules analysis.

CoFeB/2D-MoSe <sub>2</sub>	stack 1: Co <sub>60</sub> Fe <sub>20</sub> B <sub>20</sub> (3 nm)	stack 2: Co <sub>76</sub> Fe <sub>4</sub> B <sub>20</sub> (3 nm)	stack 3: Co <sub>76</sub> Fe <sub>4</sub> B <sub>20</sub> (6 nm)
$m_{L,Co} + m_{S,Co}$ ( $\mu_B$ /hole)	$0.40 \pm 0.04$	$0.39 \pm 0.04$	$0.61 \pm 0.06$
$m_{L,Fe} + m_{S,Fe}$ ( $\mu_B$ /hole)	$0.45 \pm 0.05$	$0.45 \pm 0.05$	$0.71 \pm 0.07$
References <sup>49</sup>	Co <sub>20</sub> Fe <sub>60</sub> B <sub>20</sub> (1.73 nm) on Ta buffer	Co <sub>40</sub> Fe <sub>40</sub> B <sub>20</sub> (2 nm) on MgO buffer	Co <sub>40</sub> Fe <sub>40</sub> B <sub>20</sub> (5 nm) on Bi <sub>2</sub> Te <sub>3</sub> substrate
$m_{L,Co} + m_{S,Co}$ ( $\mu_B$ /hole)	$\sim 0.55$	0.43	0.56
$m_{L,Fe} + m_{S,Fe}$ ( $\mu_B$ /hole)	$\sim 0.57$	0.60	0.51

## ASSOCIATED CONTENT

**Supporting Information Available:** XMCD hysteresis loops of CoFeB grown on Pt buffer layer; XA and XMCD spectra of stack 2 taken at the Fe  $L_{2,3}$  edges; Details of theoretical calculations; VSM hysteresis loops of MoSe<sub>2</sub>/graphene/SiC. (PDF) This material is available free of charge *via* the Internet at <http://pubs.acs.org>.

The authors declare no competing financial interests.

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