

Supplementary Information

For

Large Perpendicular Magnetic Anisotropy in Ta/CoFeB/MgO on Full-Coverage Monolayer MoS₂ and First Principles Study of Its Electronic Structure

Ziqi Zhou^{1,2}, Paul Marcon³, Xavier Devaux², Philippe Pigeat², Alexandre Bouché², Sylvie Migot², Abdallah Jaafar², Rémi Arras³, Michel Vergnat², Lei Ren⁴, Hans Tornatzky⁴, Cedric Robert⁴, Xavier Marie⁴, Jean-Marie George⁵, Henri-Yves Jaffrès⁵, Mathieu Stoffel², Hervé Rinnert², Zhongming Wei^{1#}, Pierre Renucci^{4‡}, Lionel Calmels^{3†}, Yuan Lu^{2*}

¹*State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences & Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100083, China*

²*Institut Jean Lamour, Université de Lorraine, CNRS UMR7198, Campus ARTEM, 2 Allée André Guinier, BP 50840, 54011 Nancy, France*

³*CEMES, CNRS, Université de Toulouse, 29 rue Jeanne Marvig, BP 94347, F-31055 Toulouse, France*

⁴*Université de Toulouse, INSA-CNRS-UPS, LPCNO, 135 Av. Rangueil, 31077 Toulouse, France*

⁵*Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France*

Corresponding authors: *yuan.lu@univ-lorraine.fr; †lionel.calmels@cemes.fr; ‡renucci@insa-toulouse.fr; #zmwei@semi.ac.cn

Note1: Procedure to extract effective anisotropy energy from in-plane and out-of-plane M - H curves.

The effective anisotropy constant (K_{eff}) is an important parameter for characterizing the anisotropy of the sample, which can be calculated by $K_{\text{eff}}=M_{\text{s-eff}}\times\Delta/\mu_0$, where $M_{\text{s-eff}}$ is the effective saturation magnetization per volume, Δ is the difference in the area of the in-plane (IP) and out-of-plane (OOP) M - H curves in the first quadrant, as shown in **Figure S1** by the shaded area (blue lines).

From the fitting curve of thickness dependent magnetization (Figure 3b), $M_{\text{s-eff}}$ has been determined to be 1216 emu/cm^3 . Δ is extracted to be 0.09716 T in Figure S1. Furthermore, $\mu_0=4\pi\times 10^{-7} \text{ H/m}$ and $1 \text{ emu/cm}^3=4\pi\times 10^{-4} \text{ T}$. By injecting all parameters into the formula, the value of K_{eff} can be calculated to be: $K_{\text{eff}}=M_{\text{s-eff}}\times\Delta/\mu_0=(1216\times 4\pi\times 10^{-4})\times(0.09716)\div(4\pi\times 10^{-7})=118136.844 \text{ J/m}^3$.

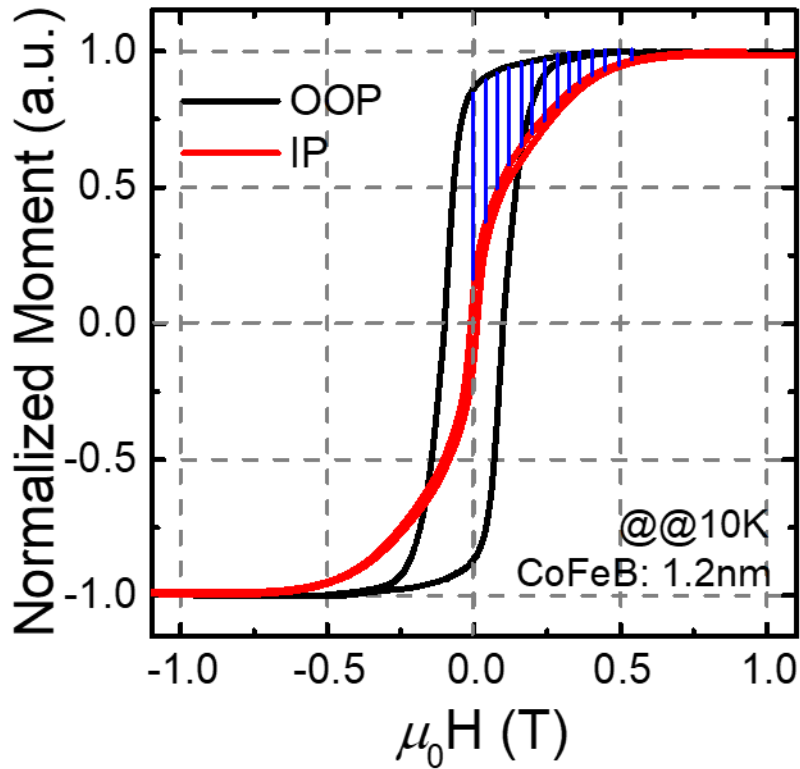


Figure S1: Out-of-plane and In-plane M - H curves for 1.2 nm CoFeB sample measured at 10K.

Note2: Details for extracting the effective anisotropy energy for the samples with different thickness of CoFeB.

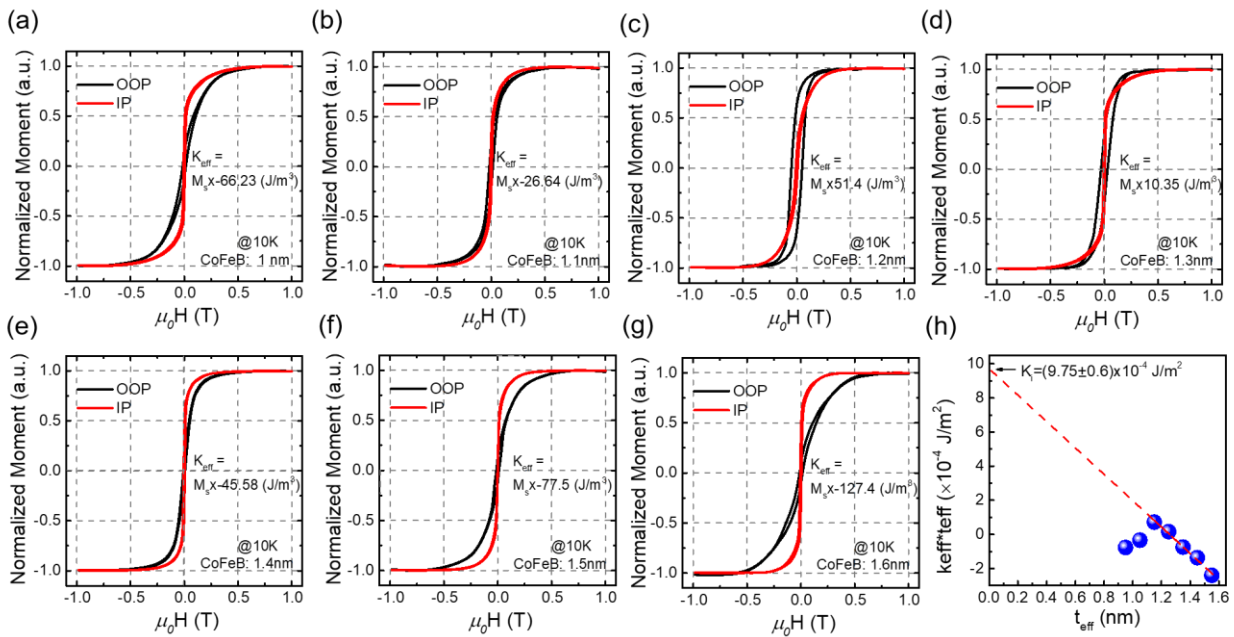


Figure S2: (a-g) Out-of-plane and in-plane M - H curves for samples with different CoFeB thicknesses with $T_{\text{an}}=250^\circ\text{C}$ measured at 10K. (a) 1nm, (b) 1.1nm, (c) 1.2nm, (d) 1.3nm, (e) 1.4nm, (f) 1.5nm, (g) 1.6nm. (h) t_{eff} dependence of the product of K_{eff} and t_{eff} , where the intercept to the vertical axis of the linear extrapolation corresponds to the interface anisotropy K_i .

Note3: Details for extracting the effective anisotropy energy for the samples with different annealing temperature.

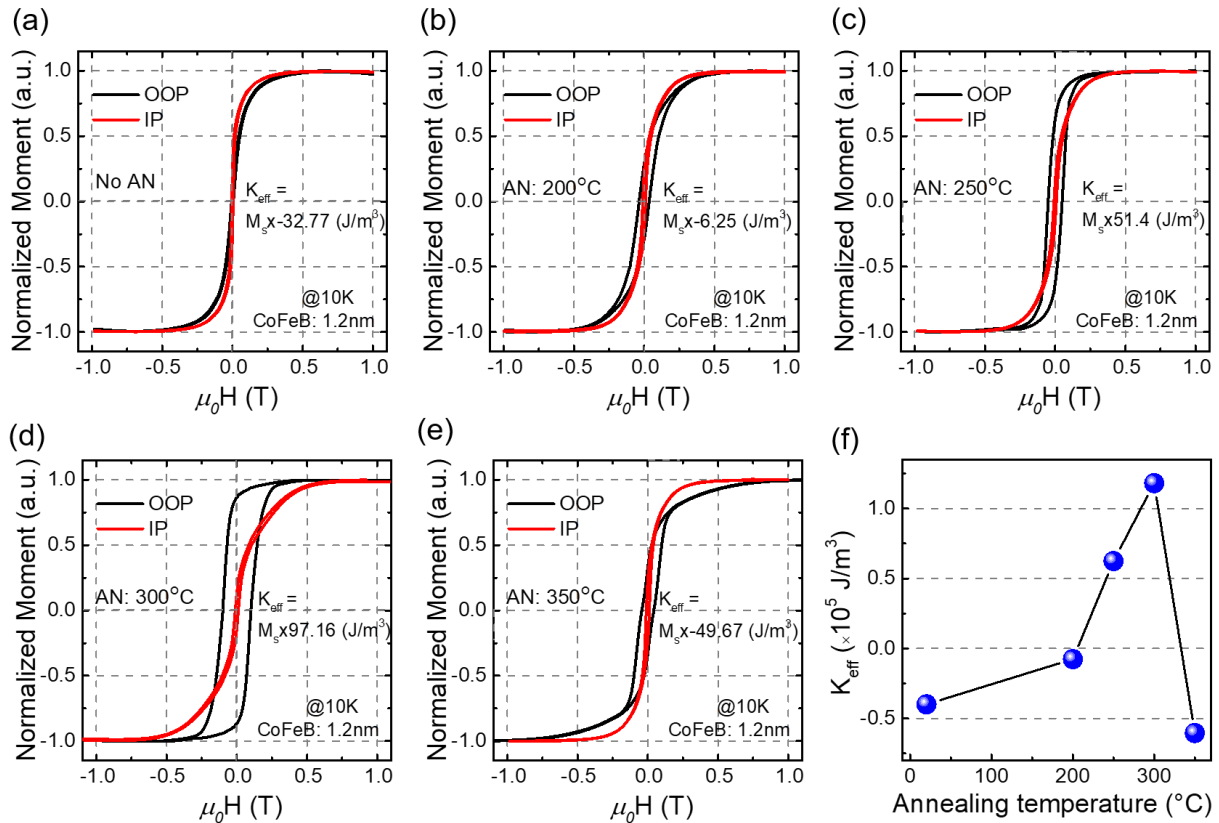


Figure S3: (a-e) Out-of-plane and in-plane M - H curves for the sample with 1.2nm CoFeB before and after different temperature annealing measured at 10K. (a) no annealing, (b) 200°C annealing, (c) 250°C annealing, (d) 300°C annealing and (e) 350°C annealing. (f) K_{eff} as a function of annealing temperature.

Note4: Element distribution profile and B diffusion in MgO for the sample capped with Pt.

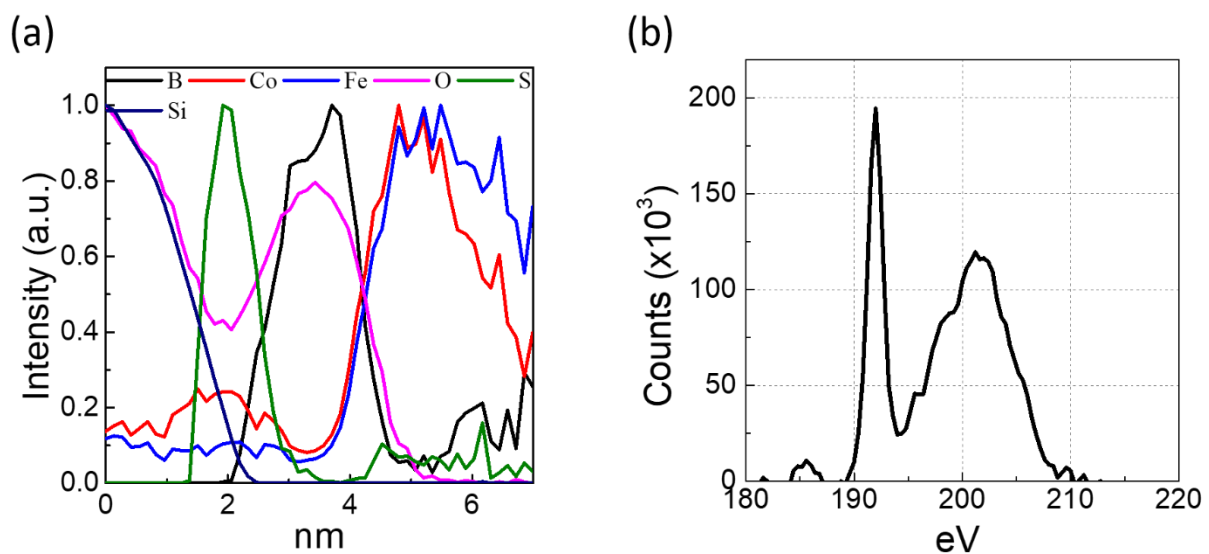


Figure S4: (a) The maximum-normalized element concentration profiles from EELS drawn perpendicularly to the MoS₂ layer shows that B diffuses completely inside the MgO barrier. (b) EELS spectrum of B_K edge extracted from spectrum image in the MgO layer. The energy loss near edge structure shows the fingerprint of B III, indicating the oxidation of boron simultaneously to its diffusion.

Note5: Band structure of the primitive (hexagonal) and conventional (rectangular) unit cells of a MoS₂ ML.

Figs S5a and S5b show the band structure of an isolated MoS₂ monolayer, when the crystal structure is respectively described with a hexagonal primitive cell and with a rectangular conventional cell similar to that used to study the Fe/MgO/MoS₂ multilayer. The same Γ -M-K- Γ -K'-M- Γ path has been used in the two figures, except that this path connects high symmetry directions shown with red letters in Figure 6a for the former case, and with black letters for the latter case. Comparison of the two figures shows how the band structure of MoS₂ is folded when the rectangular conventional cell is used. In particular, the electron states corresponding to some of the M points are folded at Γ in this representation.

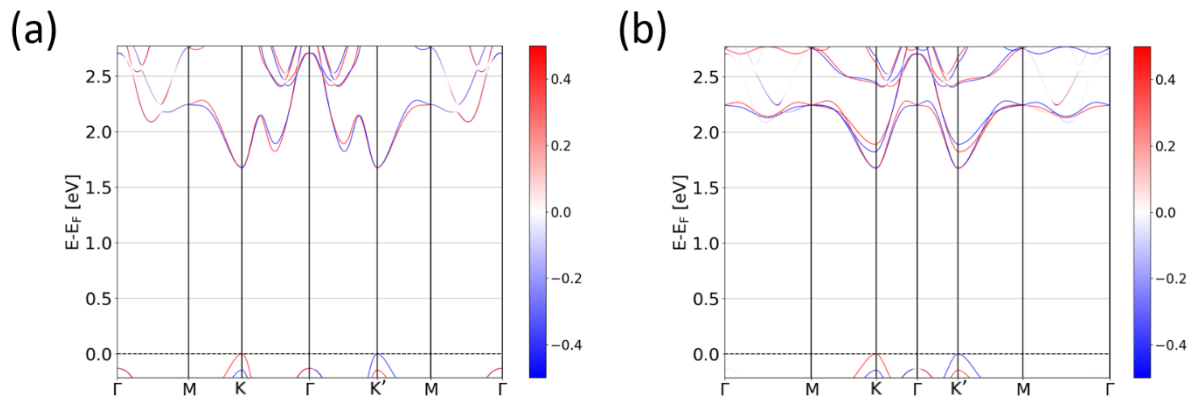


Figure S5: (a) Band structure of a MoS₂ monolayer calculated with a primitive hexagonal unit cell. (b) Band structure of a MoS₂ monolayer calculated with the conventional rectangular unit cell. The color bar shows the value of the spin component s_z for each of the Bloch states forming these bands.

Note6: First principle calculations for the MoS₂/3 MLs MgO/7 MLs Fe structure

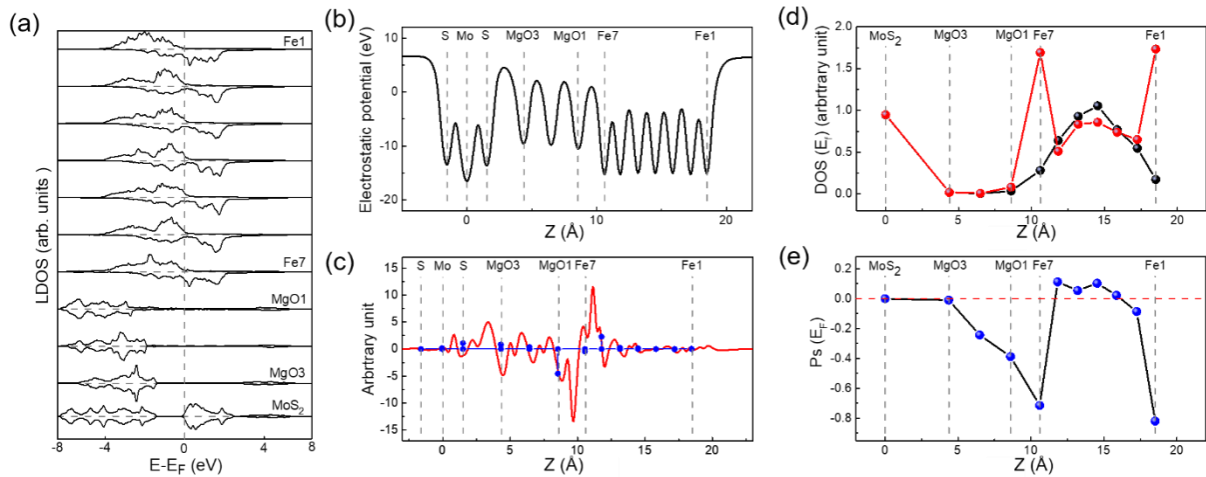


Figure S6: First principle calculations for the MoS₂/3 MLs MgO/7 MLs Fe structure. (a) Majority and minority spin DOS integrated over atomic spheres belonging to the successive atomic layers of the supercell. (b) z -variations of the xy -averaged electrostatic potential. (c) The red curve shows the variations of the electron charge density when the supercell is built from the MoS₂ ML, the 3 ML-thick MgO layer and the 7 ML-thick Fe layer. Each blue dot corresponds to the z -integration of the red curve between the two mid-perpendicular planes separating an atomic layer and its neighbors on the right and left sides. (d) z -dependence of the DOS at the Fermi level, calculated for majority (black line) and minority spin electrons (red line). (e) Profile of spin polarization at the Fermi level, calculated from the spin-resolved DOS.