

Electronic Supplementary Information:

Evidence of a strong perpendicular magnetic anisotropy in Au/Co/MgO/GaN heterostructures

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1. Optimization of MgO layer growth temperature on GaN template

The temperature for the growth of MgO on GaN has been optimized. Fig. S1(a)-(c) show the RHEED patterns for 3 nm MgO grown on GaN at different temperatures. The RHEED patterns show that MgO almost epitaxially grows on GaN at room temperature (Fig. S1(a)) and 200°C (Fig. S1(b)), while polycrystalline structures appear at 400°C with the ring features (Fig. S1(c)). To further check the MgO surface roughness, the MgO/GaN samples have been examined by atomic force microscopy (AFM). Fig. S1(d)-(f) display AFM images with tapping mode in a scanning area of $1 \times 1 \mu\text{m}^2$. The sample grown at 400°C shows the largest root mean square (RMS) roughness of 4.23Å. Both samples grown at RT and 200°C have smooth surface roughness. However, the MgO grown at RT exhibits small grain feature, while the atomic step feature can be clear observed in the sample grown at 200°C. Therefore, the MgO growth temperature at 200°C is an optimized temperature to obtain both high crystalline quality and very smooth surface.

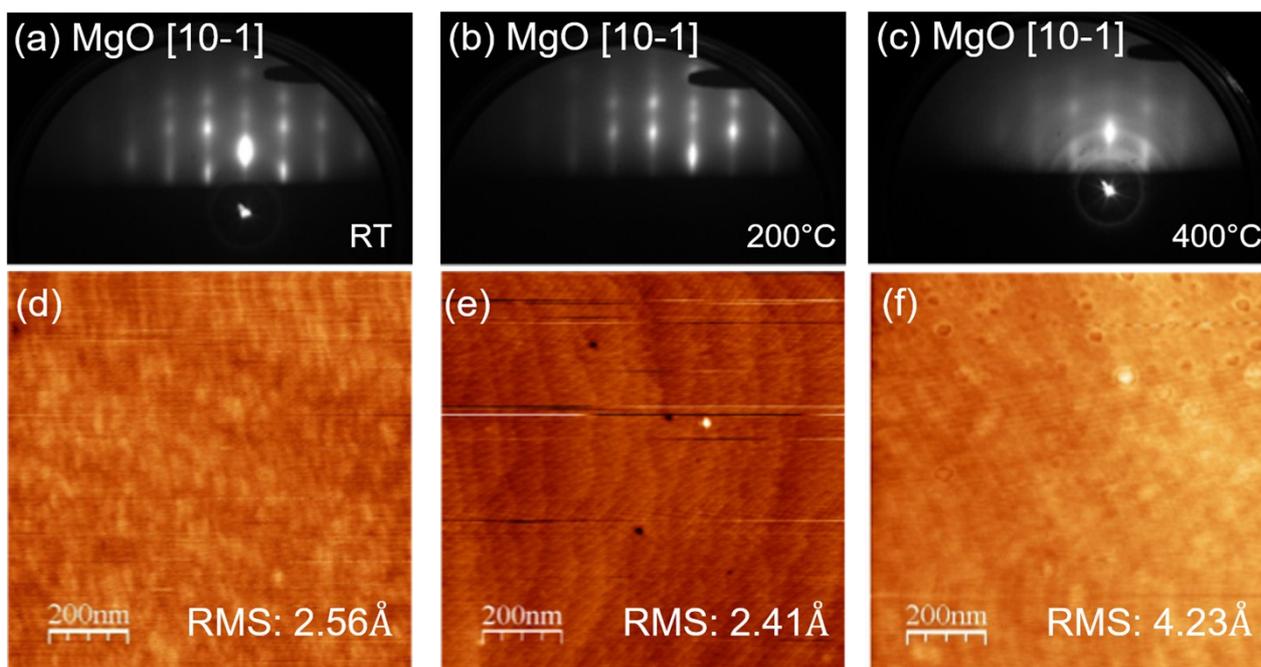


Fig. S1: RHEED patterns of MgO grown at (a) RT, (b) at 200°C and (c) at 400°C. RHEED patterns were taken at 30 kV beam energy along the MgO [10-1] azimuth. AFM images of MgO surface grown at (d) RT, (e) at 200°C and (f) at 400°C

2. Details for EELS mapping and profile analyses of chemical distribution in Co/MgO/GaN heterostructure

EELS spectrum images were recorded in STEM mode at 200kV with a probe current of 50 pA, a semi angle of convergence 24 mrad and a semi angle of collection angle of 83 mrad. Two spectrum images have

been recorded simultaneously with a dispersion of 0.5 eV, one containing the zero-loss peak, the second starting from a loss of 80 eV. Spatial drift control and correction were applied at the end of each line of spectra. The energy drift was corrected in the both spectrum images using the position of the zero-loss signal of each pixel. Then the spectrum images were denoised using a principal component analysis (PCA). After background subtraction and plural scattering correction, elemental maps were drawn from semi-quantitative analysis of the spectra.

Fig. S2(b) shows typical spectra after PCA processing and before the background subtraction. The spectra were extracted from the high-loss spectrum image in the zones indicated by the rectangular area spotted on the HAADF image (Fig. S2(a)) which was recorded simultaneously with the spectrum images. The positions of edges are indicated in the figure. One can note that even if the $\text{Au}_{\text{N}3,2}$ is a minor edge, the signal is enough strong to be used for gold identification and mapping.

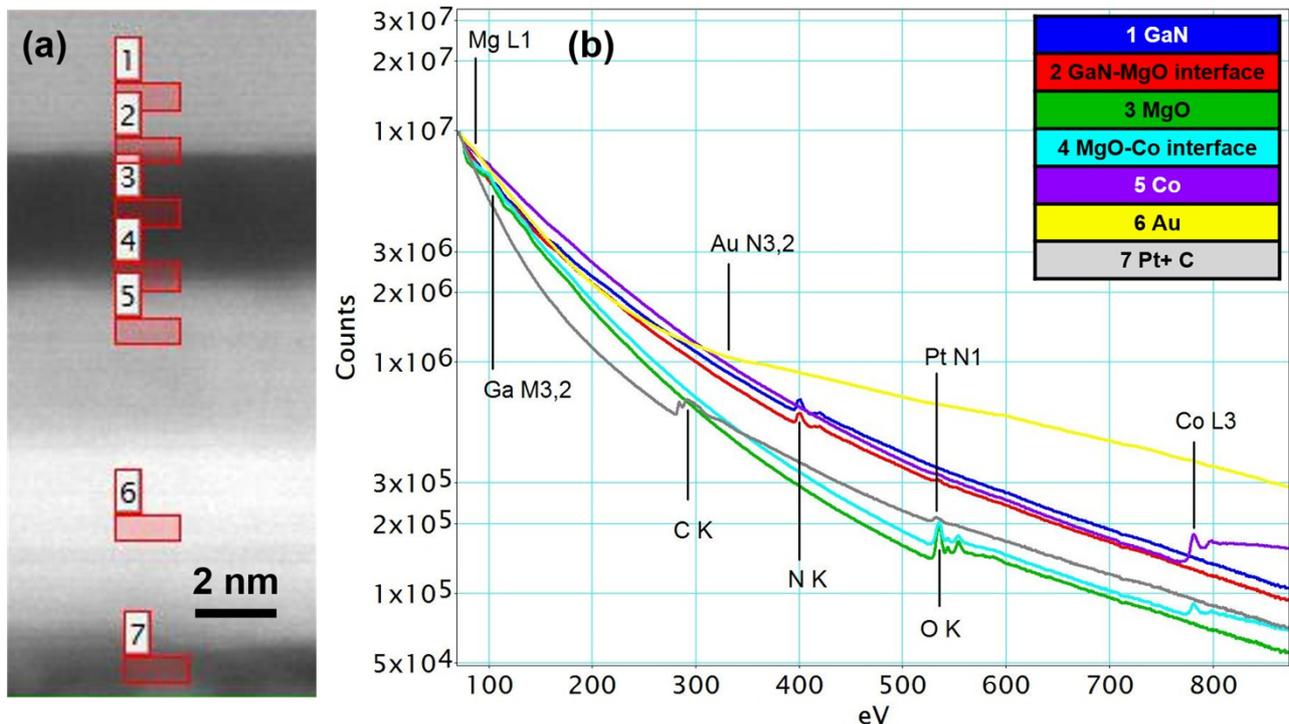


Fig. S2: (a) HAADF image recorded simultaneously with the EELS spectrum images. b) EELS spectra extracted from the spectrum images after PCA denoising in the area marked on the HAADF image. The position of edges are indicated in the figure.

3. Extraction of magnetic dead layer and effective magnetic anisotropy

To roughly estimate the magnetic dead layer in our structure, we have taken account of the sample with Co thickness of 4.6nm. The saturation magnetization per area is obtained to be $6.25 \times 10^{-4} \text{ emu/cm}^2$, which gives the saturation magnetization per volume (M_s) value of $1.358 \times 10^6 \text{ A/m}$. For the bulk hcp structure Co,

M_s is reported to be 1.422×10^6 A/m,^[1] which allows us to extract the real magnetic thicknesses of Co to be 4.4nm. Therefore, the magnetic dead layer (t_d) can be deduced to be approximately 0.2nm, which could be attributed to the inter-diffusion at Co/Au interface.

The effective anisotropy constant (K_{eff}) is calculated from $K_{\text{eff}} = M_{s\text{-eff}} \times \Delta / \mu_0$, where Δ is the difference in area between the magnetization loops measured when the magnetic field is applied perpendicular (OOP) and parallel (IP) to the layers, as shown in the zone with blue lines in Fig. S3. $M_{s\text{-eff}}$ is the effective saturation magnetization per volume after taking account of the magnetic dead layer t_d and μ_0 is the permeability in free space. By injecting all parameters into the formula, we obtain:

$$K_{\text{eff}} = M_{s\text{-eff}} \times \Delta / \mu_0 = 6.25 \times 10^{-4} \text{emu/cm}^2 / (4.6 \text{nm} - t_d) \times 0.154 \text{T} / \mu_0$$

With $1 \text{emu/cm}^3 = 4\pi \times 10^{-4} \text{T}$ and $t_d = 0.2 \text{nm}$, we obtain $K_{\text{eff}} = 1422 \times 4\pi \times 10^{-4} \times 0.154 / (4\pi \times 10^{-7}) = 2.2 \times 10^5 \text{J/m}^3$

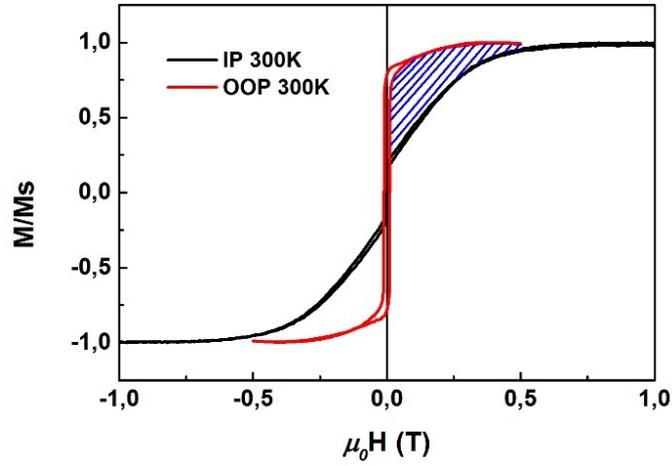


Fig. S3: M - H curves measured at RT by SQUID for Co (4.6 nm)/MgO/GaN structure with applied in-plane (IP) and out-of-plane (OOP) magnetic fields. The zone with blue lines presents the difference in area between the two magnetization loops, which is used for the extraction of K_{eff} .

4. Discussion on the contribution of PMA from Au/Co interface

We argue that the dominate PMA effect is from the Co/MgO interface but neither from Au/Co interface nor Co bulk with the following arguments.

- 1) We have summarized the available values of perpendicular interface anisotropy at Co/Au interface in the table listed below. The value of K_s varies with different growth techniques. For the best growth technique by electrodeposition, a value of 0.72mJ/m^2 was reported and a critical thickness of only 7.2 MLs (1.4nm) was obtained. This value is much smaller than what we obtained $K_s = 4.1 \text{mJ/m}^2$ in our Co(4.6nm)/MgO/GaN sample. This validates that the main contribution of PMA is from Co/MgO interface.

Structure and growth method	K_s (mJ/m ²)	Reference
Au/Co/Au(111) (MBE growth)	0.58	Ref. [2]
Cu/Co/Au(111) (Electrodeposition)	0.72	Ref. [3,4]
[Co 6 Å]/[Au 27 Å] ₂₇ multilayers (sputtering)	0.1	Ref. [5]

- 2) Our TEM results have revealed that an inter-diffusion exists at the Co/Au interface due to a large overlap of Co and Au element distribution. This inter-diffusion could possibly generate a magnetic dead layer, as we have mentioned above. Since a sharp interface is necessary to generate the PMA,^[5] the contribution of PMA from the diffused Co/Au interface should be limited.
- 3) To prove that Co/MgO interface has the main contribution to the PMA, we have designed one sample with the structure of GaN//MgO/Co(4.6nm)/MgO(5nm)/MgAlO_x(2nm), *i.e.* without the coverage of Au. Fig. S4 shows the $M-H$ loop measured at RT with out-of-plane magnetic field. The sample shows an evident perpendicular magnetic anisotropy with a M_R/M_s ratio of 0.64 and coercivity of 700 Oe, which is slightly different than our sample with Au coverage. This could be due to the increase of effective magnetic layer due to the suppression of interfacial diffusion at Au/Co interface. This gives a strong argument that the enhanced PMA character observed in our study is mainly due to the contribution from Co/MgO interface.

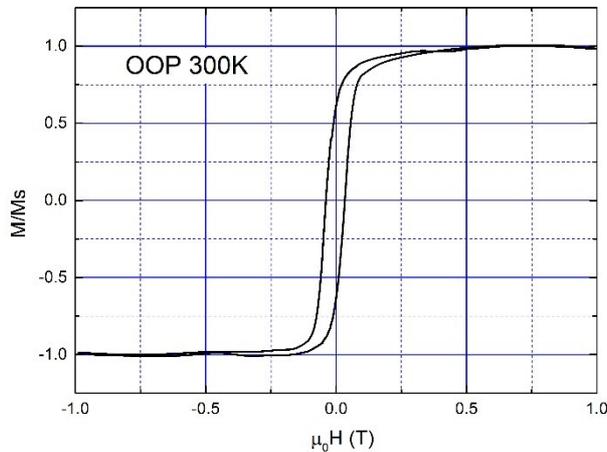


Fig. S4: $M-H$ curve measured at RT with out-of-plane magnetic field by SQUID for Co (4.6nm)/MgO/GaN structure covered with 2nm MgAlO_x/5nm MgO.

- 4) To exclude the possibility of contribution from Au/Co interface and Co bulk to the PMA, we have designed and fabricated one sample with the structure of GaN//Au(5nm)/Co(5nm)/Au(5nm). The Au and Co layers are grown at room temperature to avoid inducing a strong inter-diffusion at Au/Co interface. To make a careful comparison, the system designed keeps the same thickness of Co and Au top layer as the sample of MgO/Co/Au, while replacing the MgO bottom layer by a Au layer. In this sample, all

effects on magnetic anisotropy are from the Co/Au interface and Co bulk contribution. Fig.R1 shows the RHEED image of each layer after growth. The RHEED patterns show clear epitaxial relationship between Co[11-20]//Au[-110]//GaN[11-20] and Co[1-100]//Au[-1-12]//GaN[1-100]. From the streaky character of RHEED pattern of Co, the Co layer have almost identical crystalline quality as the sample of MgO/Co/Au, which is important for the comparison.

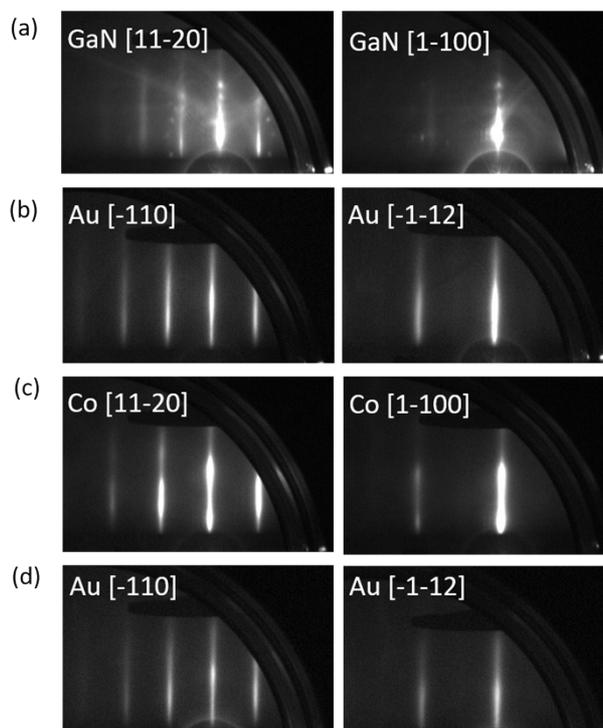


Fig. S5: In-situ RHEED patterns taken at 30 kV beam energy along the [11-20] and [10-10] azimuths of GaN for: (a) GaN surface after deoxidation at 600°C, (b) after 5 nm Au deposition at RT, (c) after 5 nm Co deposition at RT and (d) after 5 nm Au deposition at RT.

Fig.S6 show the SQUID measurement of M-H hysteresis loop with out-of-plane and in-plane magnetic field at RT. The sample shows a clear in-plan anisotropy with almost zero remanance of magnetization for the out-of-plane curve. For the in-plan curve, two components with different coercivities (4mT and 0.8T) appear. The large coercivity component could be due to the Au diffusion at the interface Au/Co, which is also consistent with our TEM results.

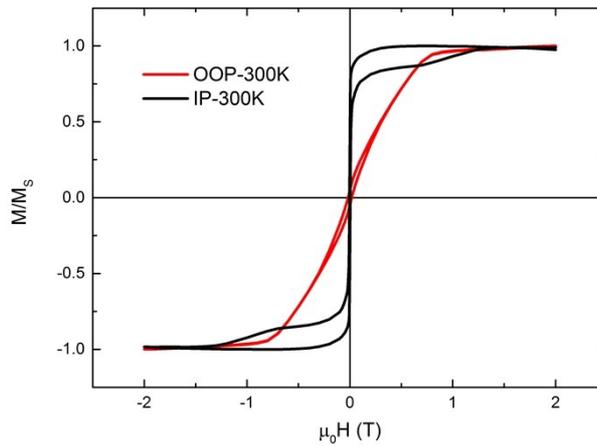


Fig. S6: M-H curves measured at RT by SQUID for GaN//Au 5nm/Co 5nm/Au 5nm structure with applied in-plane (IP) and out-of-plane (OOP) magnetic fields.

Above all these results, we can confidently argue that the PMA observed in our GaN//MgO/Co/Au system is originated from the Co/MgO interface. Neither Co/Au interface nor Co bulk can give a sizable contribution on the PMA.

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