# Electronic Supplementary Information (ESI) for:

## Frequency-dependent stimulated and post-stimulated voltage control of magnetism in transition metal nitrides: towards brain-inspired magneto-ionics

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### Compositional and structural characterization

High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and electron energy loss spectroscopy (EELS) were performed along the cross-sections of as-prepared 200 nm-thick CoN films and a 200 nm-thick CoN film electrolyte-gated at -25 V for 10 min. As shown in Fig. S1a and b, the as-prepared 200 nm-thick CoN films show a highly nanostructured morphology with homogeneous composition. As shown in Fig. S1b, upon gating at -25 V for 10 min, a well-defined interface between a N-free and highly porous Co top layer and the rest of unaltered CoN film can be observed both in the HAADF-STEM image and the EELS mappings, evidencing the existence of a planar N migration front in agreement with previous results[11].

As seen in Fig. S2, the $\theta/2\theta$ X-ray diffraction (XRD) pattern of an as-prepared 200 nm-thick CoN film reveals the presence of the (111) peak of CoN, indicating the growth of a textured phase. Upon subjecting a 200 nm-thick CoN film at -25 V for 15 min, the intensity of the (111) peak of CoN significantly decreases, in agreement with the denitriding caused but the transport of N ions towards the electrolyte.

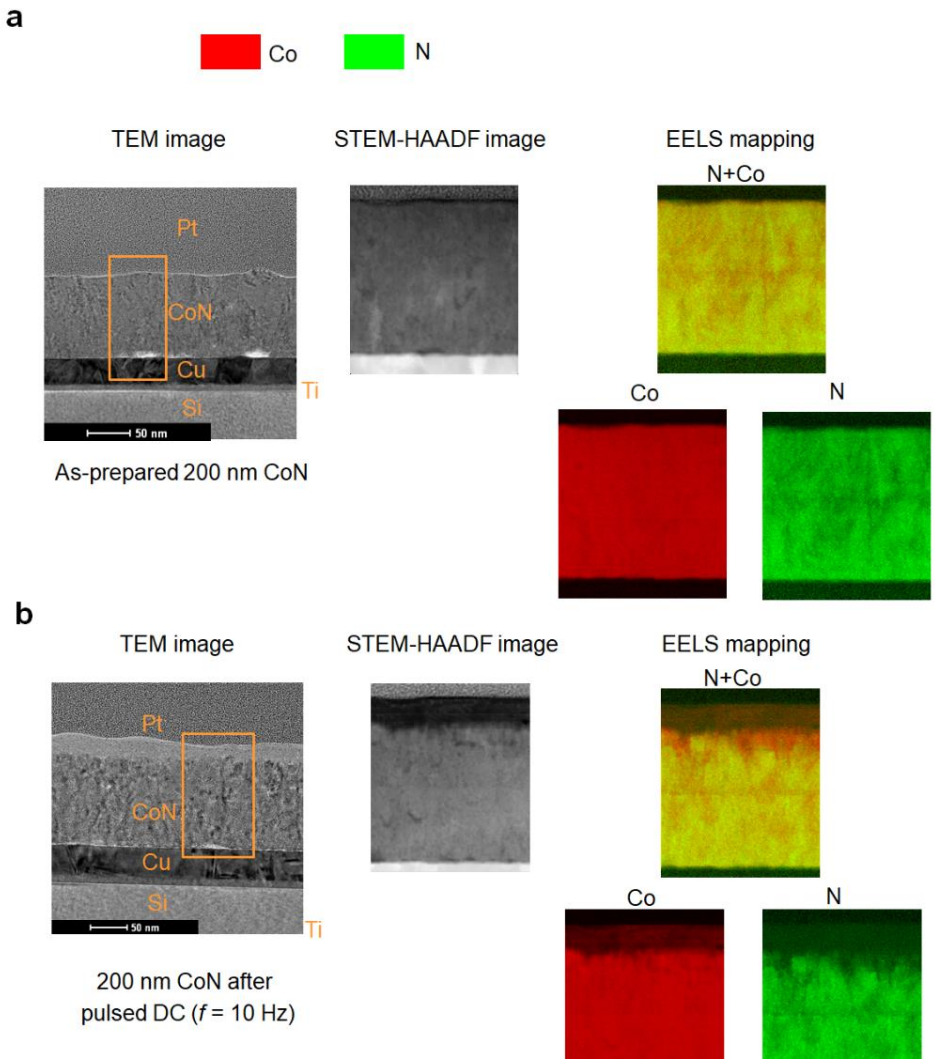


**Fig. S1.** Compositional characterization by high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and electron energy loss spectroscopy (EELS), and structural characterization by transmission electron microscopy (TEM). (a) TEM image of an as-prepared 200 nm-thick CoN film together with the HAADF-STEM image and corresponding Co and N EELS mappings of the area marked with a rectangle. (b**)** TEM image of a 200 nm-thick CoN film subjected to a -25 V for 600 s together with the HAADF-STEM image and corresponding Co and N EELS mappings of the area marked with a rectangle.

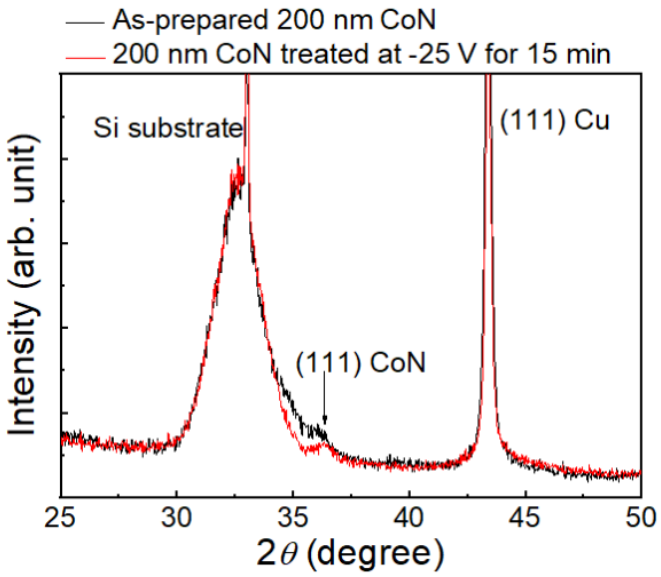


**Fig. S2.** Structural characterization by X-ray diffraction (XRD). $\theta/2\theta$ XRD diffraction patterns of an as-prepared 200 nm-thick CoN film and a 200 nm-thick CoN film after being treated at -25V for 15 min. For phase identification, the cards no. ICDD JCPDF 00-001-1241 and ICDD JCPDF 00-016-0116, were used for Cu and CoN, respectively.

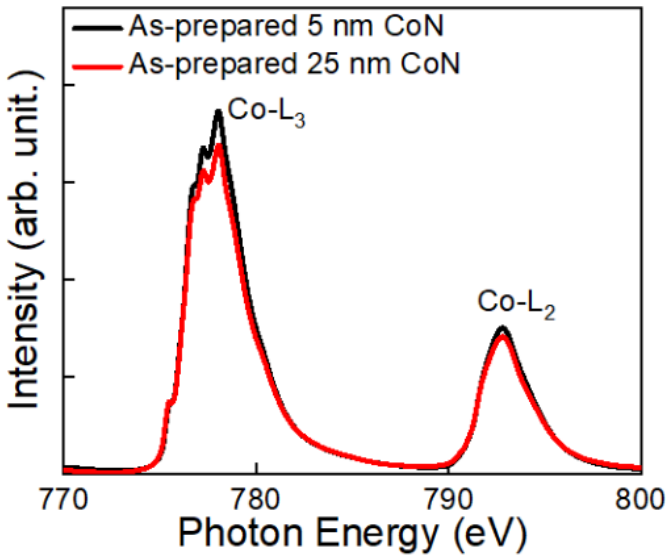


**Fig. S3.** Compositional characterization by X-ray absorption spectroscopy. Co $L_{2,3}$-edge X-ray absorption spectroscopy measurements of the 5 nm- and 25 nm-thick CoN films.

**Magneto-ionic characterization**

Fig. S4 shows the hysteresis loops of CoN films with thickness ranging from 5 to 200 nm, sequentially recorded while electrolyte-gated at -25 V until reaching a stable state. The as-prepared CoN-based heterostructures exhibit some traces of ferromagnetic signal (< 10 emu $cm^{-3}$), which could be ascribed to local deviations of CoN stoichiometry at the Cu-CoN interface.

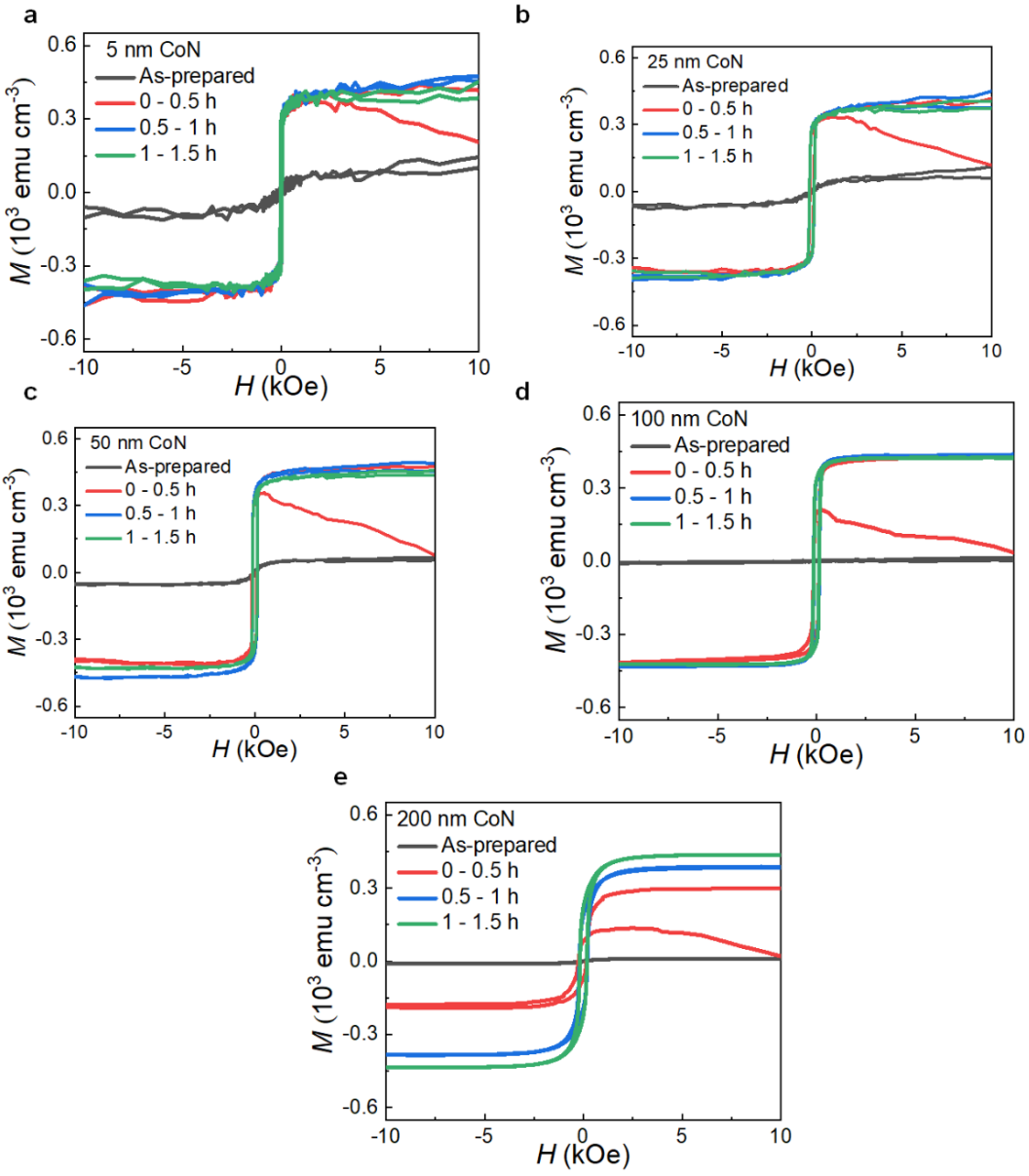


**Fig. S4.** Consecutive in-plane VSM hysteresis loops (each lasting 30 min) of the as-prepared film (black) and the films biased under -25 V: **a,** 5 nm, **b,** 25 nm, **c,** 50 nm, **d,** 100 nm, and **e,** 200 nm CoN films.

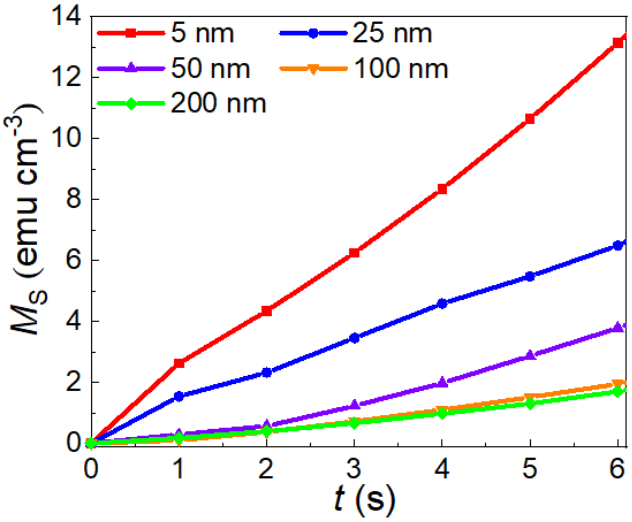


**Fig. S5.** Zoom of Fig. 1c in initial stages to highlight the generation of magnetization. Saturation magnetization ($M_S$) as a function of time $t$ from 0 to 6 s for a CoN films with thickness ranging from 5 to 200 nm under electrolyte-gating at -25 V while applying an external in-plane magnetic field of 10 kOe.

**Table S1.** Thickness-dependent magnetic and magnetoelectric parameters obtained from the consecutive hysteresis loops presented in Fig. S4 and $M_S$ *vs*. *t* plots of Fig. S5.

| Thickness (nm) | $\Delta M$ ($H$ = 10 kOe) (emu cm$^{-3}$) upon -25 V for 1 s | $\Delta M$ ($H$ = 10 kOe) ($10^3$ emu cm$^{-3}$) upon -25 V for 50 s | $\Delta M$ ($H$ = 10 kOe) ($10^3$ emu cm$^{-3}$) upon saturation [minimum time required to reach a stable $M_S$] | $H_C$ (Oe) |
|---|---|---|---|---|
| 5 | 2.6 | 0.235 | 0.297 [60 s] | 35 |
| 25 | 1.5 | 0.132 | 0.305 [130 s] | 132 |
| 50 | 0.3 | 0.068 | 0.379 [< 1 h] | 132 |
| 100 | 0.1 | 0.040 | 0.423 [< 1 h] | 143 |
| 200 | 0.1 | 0.037 | 0.507 [12 h] | 179 |

**Table S2.** $M_S$ attained once magneto-ionic process saturates (*i.e.*, maximum value), normalized by the time required to reach it ($\Delta t$) and by the absolute value of the applied voltage ($|\Delta V|$), from the current results and previous related works from the literature[3,11-14,17]

| Film | 85 nm-thick $Co_3O_4$ film | 85 nm-thick CoN film | 85 nm-thick FeN film | 5 nm-thick $Co_3O_4$ film | 30 nm-thick CoMnN film | 5 nm-thick CoN film (current work) |
|---|---|---|---|---|---|---|
| $M_S/(\Delta t\|\Delta V\|)$ (emu $cm^{-3}$ $h^{-1}$ $V^{-1}$) | 7.8 | 52.0 | 7.2 | 32.0 | 64.5 | 432.0 |

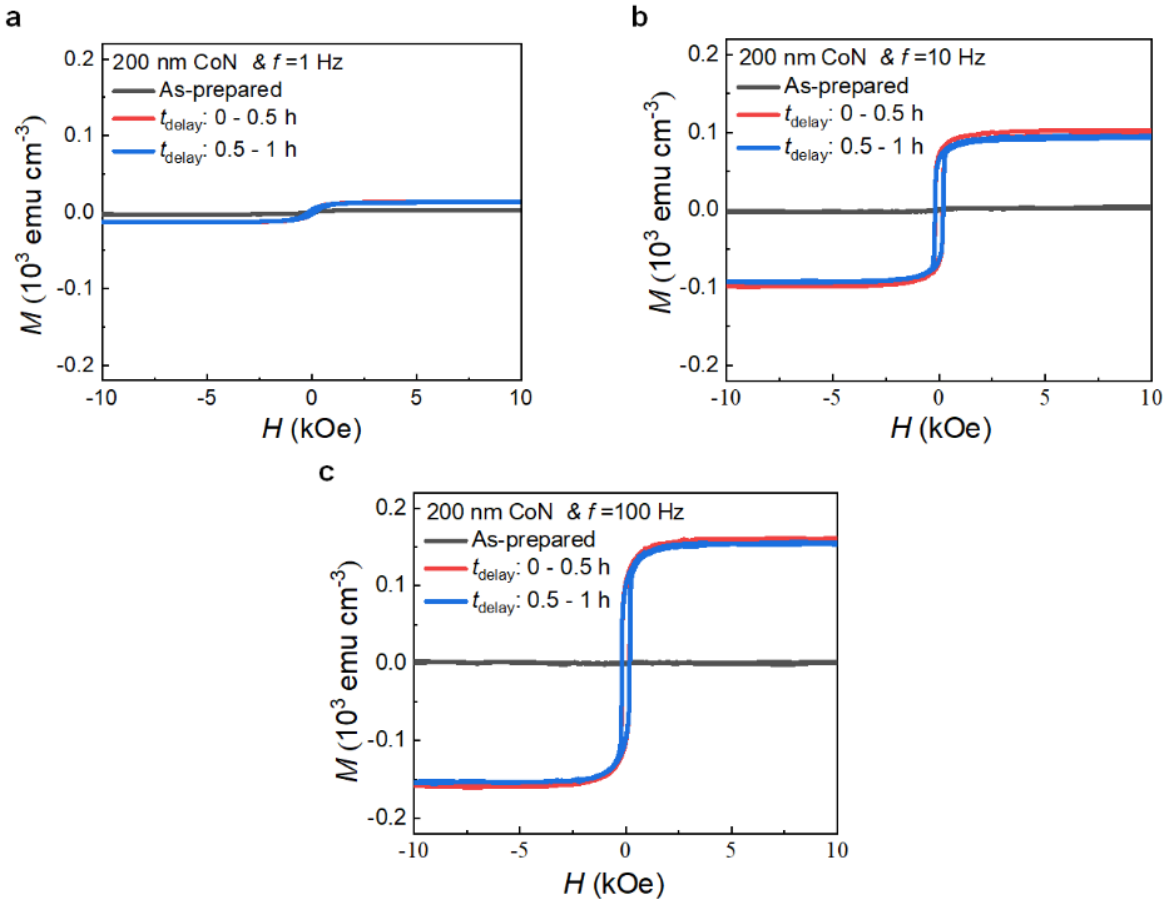


**Fig. S6.** In-plane vibrating sample magnetometry hysteresis loops (each lasting 30 min) of an as-prepared 200 nm CoN thin film and a 200 nm CoN film subjected to pulsed DC voltage

actuation for 0.5 and 1h at a frequency of (a) 1 Hz, (b) 10 Hz and (c) 100 Hz. Note that $t_{delay}$ refers to the time after the voltage has been switched off.

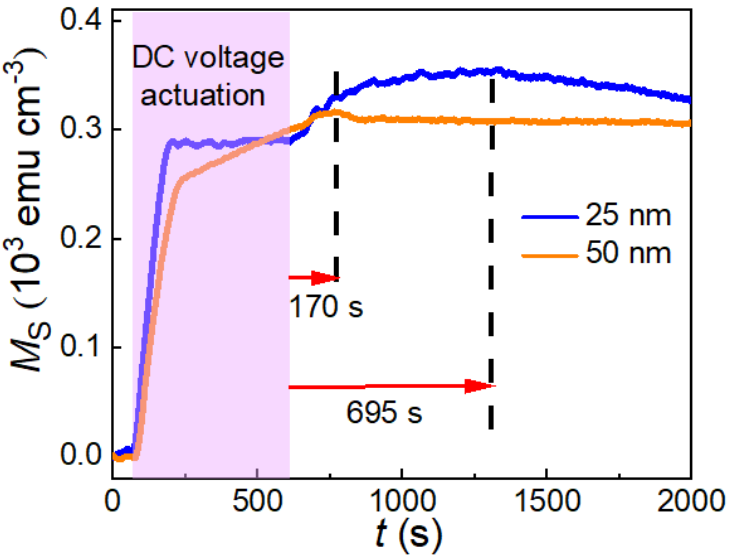


**Fig. S7.** Time evolution of the saturation magnetization ($M_S$ *vs*. $t$) during and after DC voltage actuation at -25V for 20 nm and 50 nm thick CoN films. Dashed lines mark the start of magnetization turning point.