

Supplementary Information for
Magnetic-Field-Assisted Assembly of CoFe Layered Double Hydroxide
Films with Enhanced Electrochemical Behavior and Magnetic
Anisotropy

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Preparation of CoFe LDH nanoplatelets

The CoFe LDH nanoplatelets with narrow size-distribution and high crystallinity were prepared by a modified coprecipitation method. Typically, the pH of the aqueous solution (150 mL) containing $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ (0.16 M) and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (0.055 M) was adjusted to 8.0 with NaOH solution (1.5 M), and it was aged in an autoclave at 130 °C for 24 h. The colloidal LDH suspension was obtained by thoroughly washing with water and then filtrating using a membrane filter (0.2 μm , Millipore). The deionized and decarbonated (purged with nitrogen) water was used in all the experimental processes. Elemental analysis: found (CoFe LDH) Co 36.1%, Fe 12.0%, O 46.0%, N 3.01%, H 2.81%.

Fabrication of the (CoFe LDH/TPPS)_n hybrid films

Prior to assembly, ITO substrates (3.0 cm×1.0 cm) were cleaned by sonication in a series of solvents: acetone, ethanol and deionized water for 10 min each time. Quartz glass slides were pretreated in a bath of methanol/HCl (1/1, v/v) and then concentrated H_2SO_4 for 30 min each to make the substrates surface hydrophilic and negatively charged.

The overall process of magnetic-field-assisted (MFA) layer-by-layer assembly of the

(CoFe LDH/TPPS)_n films consists of a cyclic repetition of the following steps: (a) dipping the pretreated substrates into the colloidal CoFe LDH suspension for 10 min and then rinsed with deionized water thoroughly (this procedure was carried out with a external magnetic field of 0.5 T and 0.3 T normal to the substrate respectively); (b) dipping into an aqueous solution of TPPS (1.0 g/L) for 10 min and washed extensively with deionized water. Subsequently, a series of these operations for LDH nanoplatelets and TPPS were repeated *n* times to obtain multilayer films of (CoFe LDH/TPPS)_n. The resulting films were finally rinsed with deionized water and dried at ambient temperature.

The conventional LBL deposition was carried out similar to the MFA LBL assembly without external magnetic field.

Sample characterization

X-ray diffraction (XRD) patterns were recorded by a Rigaku XRD-6000 diffractometer, using Cu K α radiation ($\lambda = 0.15418$ nm) at 40 kV, 30 mA. The UV-vis spectra were collected with a Shimadzu U-3000 spectrophotometer. The morphology of thin films was investigated using a scanning electron microscope (SEM Hitachi S-4700) with the accelerating voltage of 20 kV. Metallic elemental analysis was performed by atomic emission spectroscopy with a Shimadzu ICPS-7500 instrument. H, N content was determined using an Elementar vario elemental analysis instrument. The amount of water was analyzed by Thermogravimetry and differential thermal analysis (TG–DTA). The surface roughness was studied using the atomic force microscopy (AFM) software (Digital Instruments, Version 6.12). The electrochemical measurements were performed using a CHI 660C electrochemical workstation (Shanghai Chenhua Instrument Co., China). A conventional three-electrode system was used, including a

modified ITO glass as the working electrode, a platinum foil as the auxiliary electrode and a saturated Ag/AgCl electrode as the reference electrode. All electrochemical measurements were carried out in a NaOH solution (Milli-Q water: $>18 \text{ M}\Omega\cdot\text{cm}$) at room temperature (ca. 20°C). Magnetism of films was measured at room temperature on a LDJ 9600 vibration sample magnetometer.

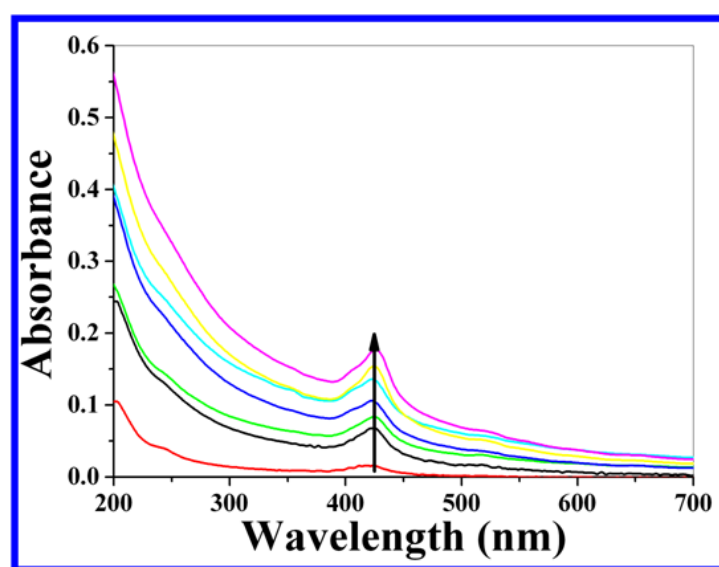


Fig. S1 UV-vis absorption spectra of the WMFA (CoFe LDH/TPPS) $_n$ films ($n=1-12$).

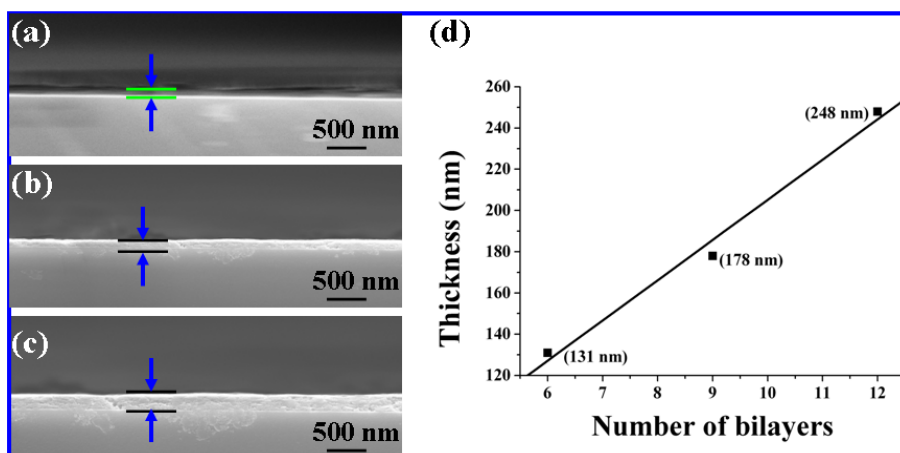


Fig. S2 Side view of SEM images for the MFA (CoFe LDH/TPPS) $_n$ films with a) 6, b) 9, c) 12 bilayers; (d) thickness of the MFA films as a function of n .

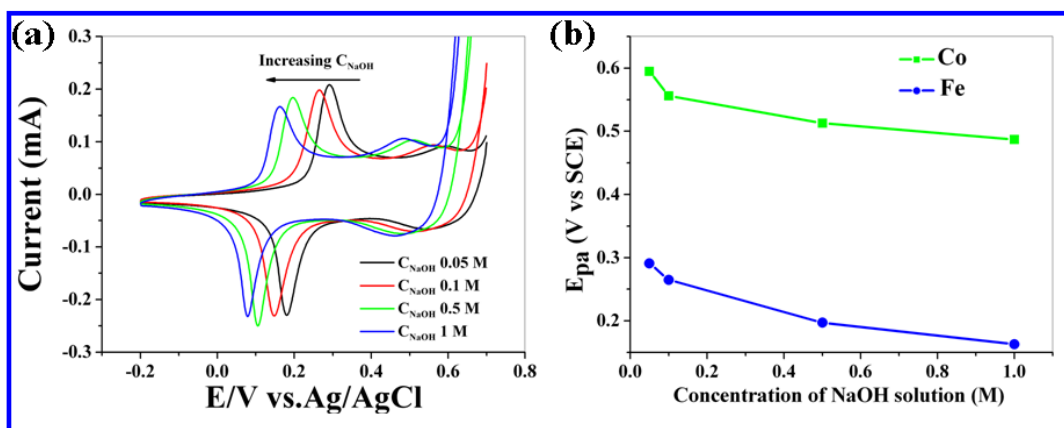


Fig. S3 (a) CVs of the MFA (CoFe LDH/TPPS)₆ modified ITO electrode in 0.05 M, 0.1 M, 0.5 M and 1.0 M NaOH solution at a scan rate of 0.1 V s⁻¹; (b) correlation of E_{pa} with NaOH concentration.

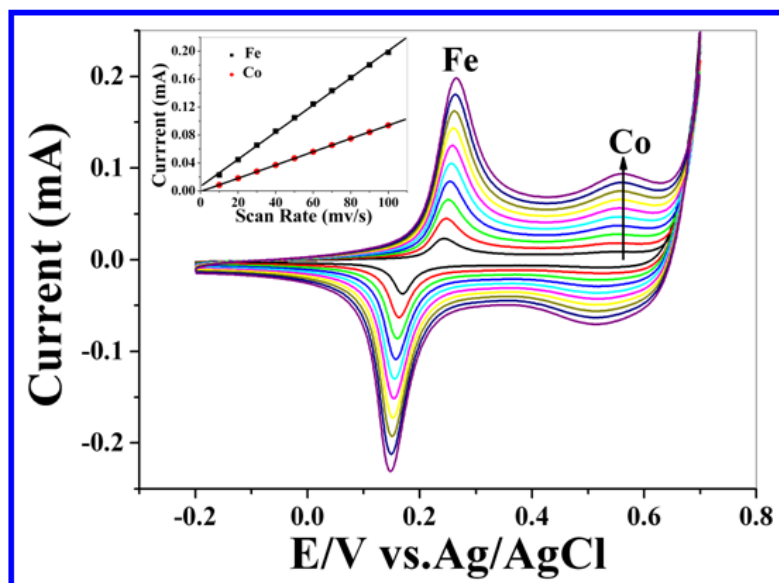


Fig. S4 CVs of the MFA (CoFe LDH/TPPS)₆ modified ITO electrode with scan rate ranging from 0.01 to 0.1 V s⁻¹; inset: plots of peak current vs. scan rate in 0.1 M NaOH.

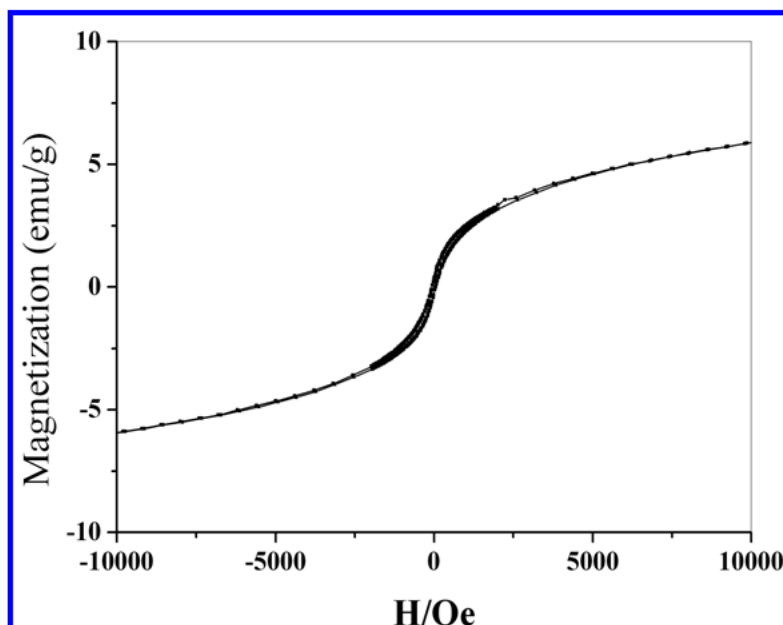


Fig. S5 Plot of magnetization M vs. applied magnetic field H at room temperature for the powder sample of CoFe LDH nanoplatelets.

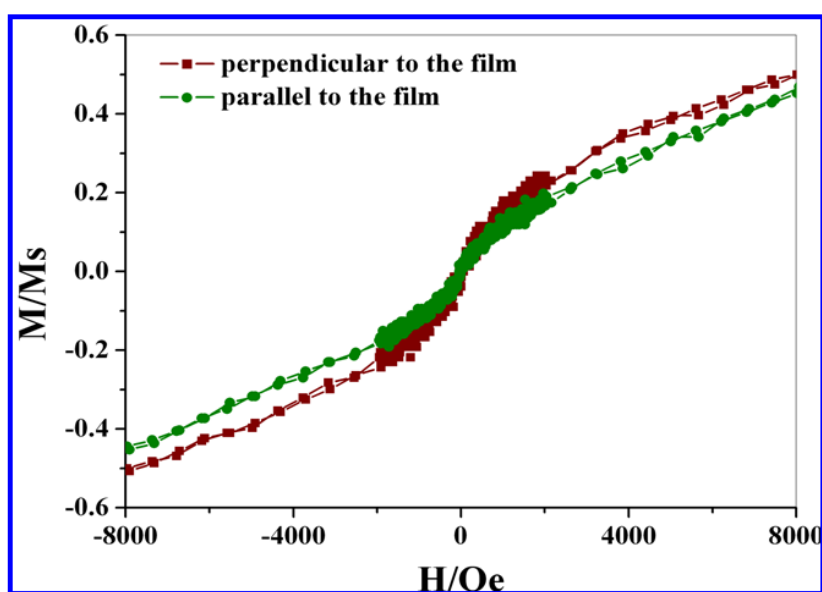


Fig. S6 Room temperature (300 K) hysteresis loops (0.3 T) at both orientations for the MFA (CoFe LDH/TPPS)₁₂ film. Magnetization values were normalized based on the corresponding saturation ones.

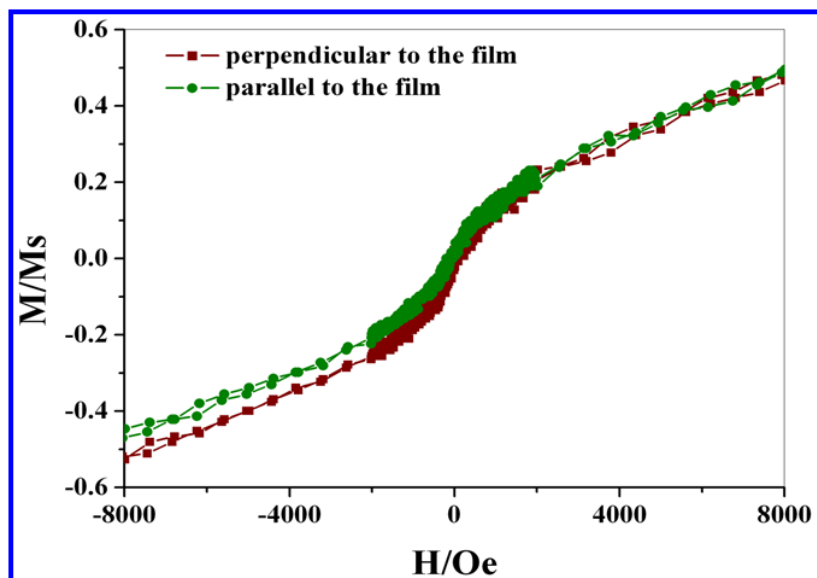


Fig. S7 Room temperature (300 K) hysteresis loops at both orientations for the WMFA (CoFe LDH/TPPS)₁₂ film. Magnetization values were normalized based on the corresponding saturation ones.