Supplementary Material

Nanoscale measurement of giant saturation magnetization in α''-$Fe_{16}N_2$ by electron energy-loss magnetic chiral dichroism

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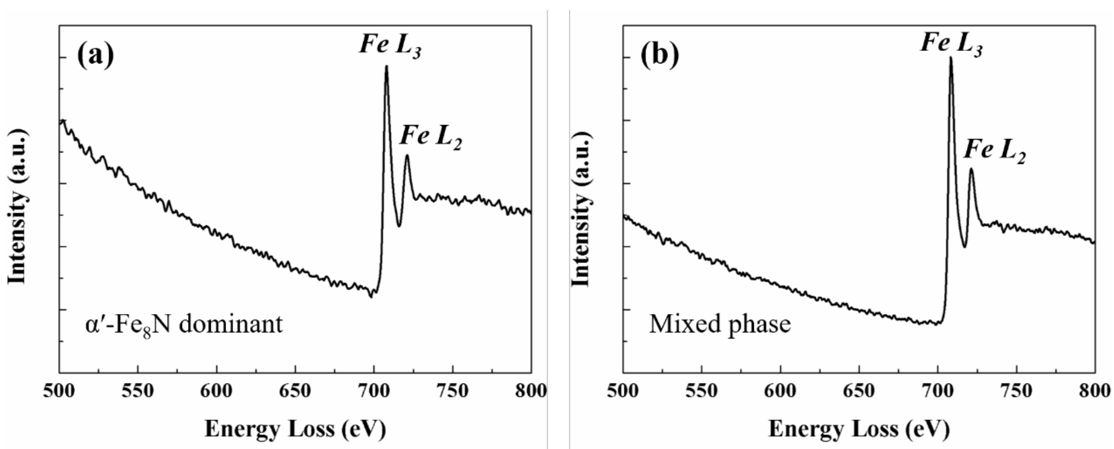
Confirmation of absence of oxidation in both α'-Fe_8N-dominant and mixed phase thin films

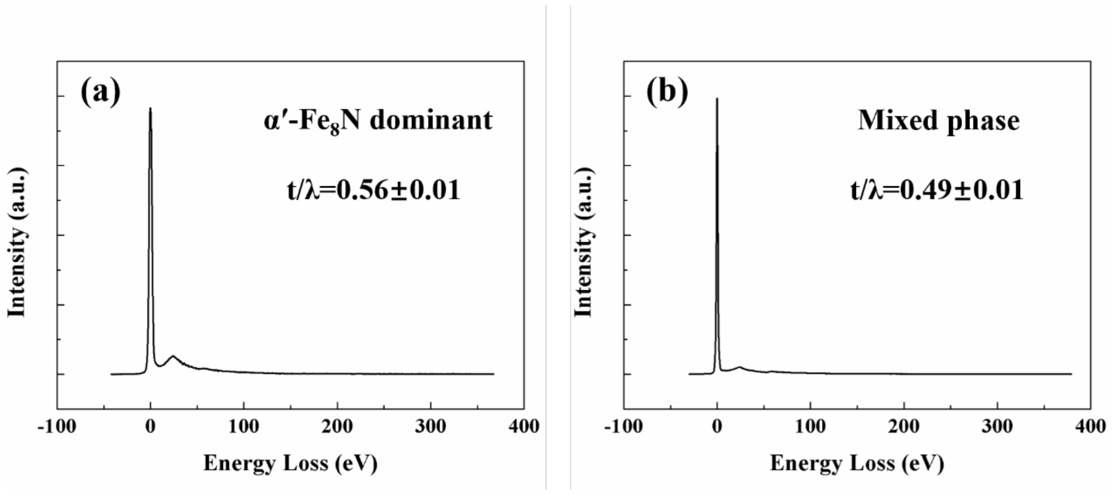


Supplementary Figure S1 | EEL spectra recorded from (a) the α'-Fe_8N-dominant thin film and (b) the mixed phase thin film, respectively, showing the absence of detectable oxygen at ~532 eV.

EEL spectra recorded from α'-Fe_8N-dominant thin film and mixed phase thin film confirm the absence of perceivable oxidation of both samples, as evidenced by the signals of O K edge started from ~532 eV.

Thickness measurements from the experimental areas

After core-loss spectra had been recorded at the " + " and " − " Thales positions during the EMCD experiments, corresponding low-loss spectra were recorded under the same experimental conditions, with the same detection aperture centered on the transmitted beam, in order to estimate the sample thickness. A typical low-loss EEL spectrum recorded from the α'-Fe_8N-dominant thin film (shown in Fig. S2a) provides a value for t/λ of 0.56±0.01, while that recorded from the mixed phase thin film (shown in Fig. S2b) provides a value for t/λ of 0.49±0.01 based on a statistical average estimation, corresponding to values of 49±1 nm and 43±1 nm, respectively. The mean free path for inelastic scattering λ can be determined using both theoretical calculations and convergent beam electron diffraction measurements.



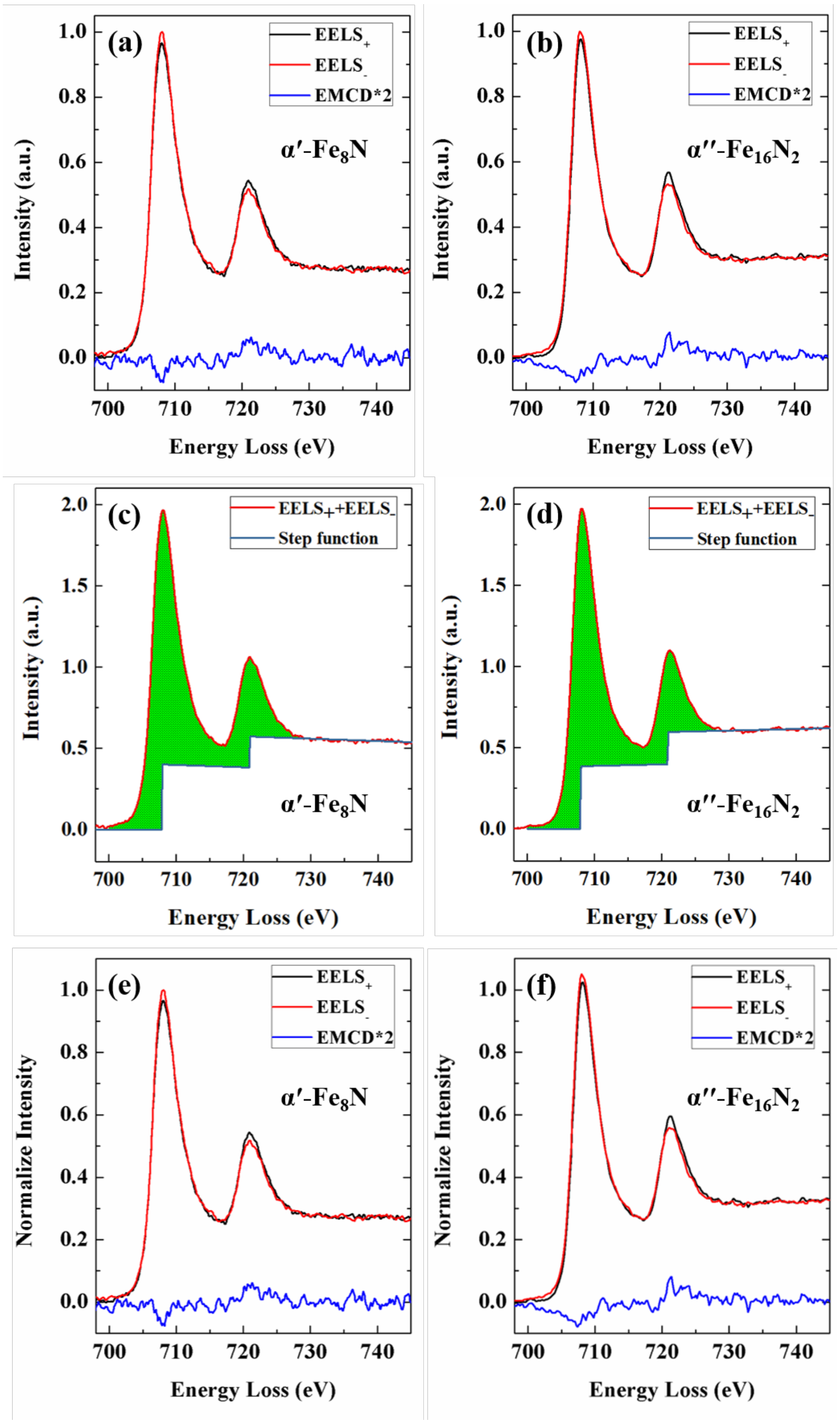
Supplementary Figure S2 | Representative low-loss EEL spectra recorded from (a) the α'-Fe_8N-dominant thin film and (b) the mixed phase thin film.

Details about the normalization procedure

1. Plot the EEL spectra obtained at the positive ($EELS_+$) and negative ($EELS_-$) positions and the corresponding EMCD spectra for α'-Fe_8N (Fig. S3a) and α''-$Fe_{16}N_2$ (Fig. S3b), with the peak values of both EEL_- spectra preliminarily normalized to "1".
2. Plot the sum ($EELS_+$+$EELS_-$) curves for α'-Fe_8N (Fig. S3c) and α''-$Fe_{16}N_2$ (Fig. S3d), based on the raw $EELS_+$ and $EELS_-$ spectra in Figs. S3a and S3b, respectively, and plot the corresponding step functions. In order to plot the curve of the step function, a straight line was fitted to the post edge range following the L_2 peak over a region of width 17 eV (from 728 eV to 745 eV) and extrapolated into the L_2 and L_3 region. This line was modified into a double step of the same slope with onsets at the maxima (721 eV and 708 eV). The ratio of the step

heights was chosen to be 2:1, in accordance with the multiplicity of the initial states (four $2p_{3/2}$ electrons and two $2p_{1/2}$ electrons). By subtracting step function from the integral over $L_{3,2}$ of the sum spectra, the area for normalization was obtained (marked in green). The ratio of the areas for α'-Fe_8N and α''-$Fe_{16}N_2$ to was calculated to be 1.05.

3. Normalization by the isotropic spectra between α'-Fe_8N and α''-$Fe_{16}N_2$ was achieved by using



the EELS for α''-$Fe_{16}N_2$ multiplied by the ratio 1.05. The results are shown in (e) and (f).

Supplementary Figure S3 | EEL spectra recorded at the positive ($EELS_+$ in black) and negative ($EELS_-$ in red) position and corresponding EMCD spectra (in blue) for (a) α'-Fe_8N and (b) α''-$Fe_{16}N_2$. (c) and (d) show the sums of $EELS_+$ and $EELS_-$ spectra (in red) with corresponding step functions (in blue) for (c) α'-Fe_8N and (d) α''-$Fe_{16}N_2$. The area for normalization is marked in green. (e) and (f) show normalized "+" (in black), "-" (in red) and EMCD spectra (in blue) for (e) α'-Fe_8N and (f) α''-$Fe_{16}N_2$.