Supplementary Material

Nanoscale measurement of giant saturation magnetization in \( \alpha''\)-Fe\(_{16}\)N\(_2\) by electron energy-loss magnetic chiral dichroism

Xinfeng Chen\(^a\), Soma Higashikozono\(^b\), Keita Ito\(^b\), Lei Jin\(^c\), Ping-Luen Ho\(^a\), Chu-Ping Yu\(^a\)\(^d\), Nyan-Hwa Tai\(^d\), Joachim Mayer\(^c\)\(^e\), Rafal E. Dunin-Borkowsk\'\(\)\(^d\), Takashi Suemasu\(^b\), Xiaoyan Zhong \(^{a,*}\)

\(^a\) National Center for Electron Microscopy in Beijing, Key Laboratory of Advanced Materials (MOE), The State Key Laboratory of New Ceramics and Fine Processing, School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China.

\(^b\) Institute of Applied Physics, University of Tsukuba, Tsukuba, Ibaraki 305-8573, Japan.

\(^c\) Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany.

\(^d\) Department of Materials Science and Engineering, National Tsing-Hua University, Hsinchu 30013, Taiwan.

\(^e\) Central Facility for Electron Microscopy, RWTH Aachen University, 52074 Aachen, Germany.

Confirmation of absence of oxidation in both \( \alpha''\)-Fe\(_8\)N-dominant and mixed phase thin films

![Fe L\(_3\) and Fe L\(_2\) peaks in energy loss spectra](image)
**Supplementary Figure S1** | EEL spectra recorded from (a) the \(\alpha'\)-Fe\(_8\)N-dominant thin film and (b) the mixed phase thin film, respectively, showing the absence of detectable oxygen at \(\sim 532\) eV.

EEL spectra recorded from \(\alpha'\)-Fe\(_8\)N-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 \(\sim 532\) 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 \(\alpha'\)-Fe\(_8\)N-dominant thin film (shown in Fig. S2a) provides a value for \(t/\lambda\) of \(0.56 \pm 0.01\), while that recorded from the mixed phase thin film (shown in Fig. S2b) provides a value for \(t/\lambda\) of \(0.49 \pm 0.01\) based on a statistical average estimation, corresponding to values of \(49 \pm 1\) nm and \(43 \pm 1\) nm, respectively. The mean free path for inelastic scattering \(\lambda\) 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 \(\alpha'\)-Fe\(_8\)N-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 \(\alpha'\)-Fe\(_8\)N (Fig. S3a) and \(\alpha''\)-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 \(\alpha'\)-Fe\(_8\)N (Fig. S3c) and \(\alpha''\)-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 \(\alpha'-Fe_8\)N and \(\alpha''-Fe_{16}\)N\(_2\) was calculated to be 1.05.

3. Normalization by the isotropic spectra between \(\alpha'-Fe_8\)N and \(\alpha''-Fe_{16}\)N\(_2\) was achieved by using
the EELS for $\alpha''$-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) $\alpha'$-Fe$_8$N and (b) $\alpha''$-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) $\alpha'$-Fe$_8$N and (d) $\alpha''$-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) $\alpha'$-Fe$_8$N and (f) $\alpha''$-Fe$_{16}$N$_2$. 