Supplementary Material

Three-dimensional Measurement of Magnetic Moment Vectors using Electron Magnetic Chiral Dichroism at Atomic Scale

Dongsheng Song[1,2]* and Rafal E. Dunin-Borkowski[1]

1. Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons and Peter Grünberg Institute, Forschungszentrum Jülich, 52425 Jülich, Germany

2. Information Materials and Intelligent Sensing Laboratory of Anhui Province, Key Laboratory of Structure and Functional Regulation of Hybrid Materials of Ministry of Education, Institutes of Physical Science and Information Technology, Anhui University, Hefei 230601, China

*Corresponding author: songdongsheng1101@gmail.com

Description of the EMCD signals

The EELS signals in the diffraction plane, also evaluated as the double-differential scattering cross-section (DDSCS) of momentum-resolved inelastic electron scattering electrons, can be simply expressed as the linear superposition of nonmagnetic and magnetic EMCD signals within the dipole approximation for cubic crystal structure [34,35], that is $\partial^2\sigma/(\partial E\partial\Omega) \propto A * N(E) + B * M(E)$. $N(E)$ and $M(E)$ are the intrinsic nonmagnetic and magnetic EMCD signals, which are dependent on the electronic structure and set to one in the simulations. Therefore, the coefficients of A and B are used to represent the intensity of nonmagnetic and magnetic EMCD signals, respectively, which are composed of a product of Bloch coefficients and momentum transfers as described in Ref. [34,35]. The relative intensity of EMCD signal is defined as B/A in the Figures of manuscript. The magnetic signals (B) from three Cartesian directions can be separately calculated by modifying the terms of momentum transfers [34]. Finally, at a fixed electron beam position, the distributions of nonmagnetic and magnetic EMCD signals are calculated for each outgoing direction in the diffraction plane with the range of ± 50 mrad and the step of 2.5 mrad both in the x and y directions.

Details of EMCD simulation procedures and parameters

The orthogonal supercell of *bcc* Fe with [016] axis along the electron beam direction is taken in the simulations with the size of 2.80 Å *17.46 Å *17.46 Å, in order to avoid the large tilt angle of incident electron beam. First, the multislice calculations are conducted to calculate the propagation of incoming electron wave in the crystal [34,35]. The supercell is discretized on a grid of 32 × 180 × 72 grid points and repeated 24 × 4 times in x and y directions. All of the aberrations are set to zero except the defocus value. The accelerating voltage, beam position, convergence semi-angle, sample thickness and defocus are accordingly set in this step.

After that, the inelastic scattering calculations are conducted based on the MATS v2 software [34]. The convergence parameter was set to 10^{-6}. The energy loss of 708 eV for Fe L_3 edge is taken for the calculation of energy filtered diffraction patterns. The range of the diffraction pattern is ± 50 mrad by ± 50 mrad with the step of 2.5 mrad. At last, the MATS v2 software outputs the coefficients of A and B respectively for the nonmagnetic and magnetic EMCD components (including the signals from all of three Cartesian directions) at each outgoing direction in the diffraction plane as shown in the Figures in the manuscript.

Reference

[34] J. Rusz, Ultramicroscopy **177**, 20 (2017).

[35] J. Rusz, S. Bhowmick, M. Eriksson, and N. Karlsson, Phys. Rev. B **89**, 134428 (2014).

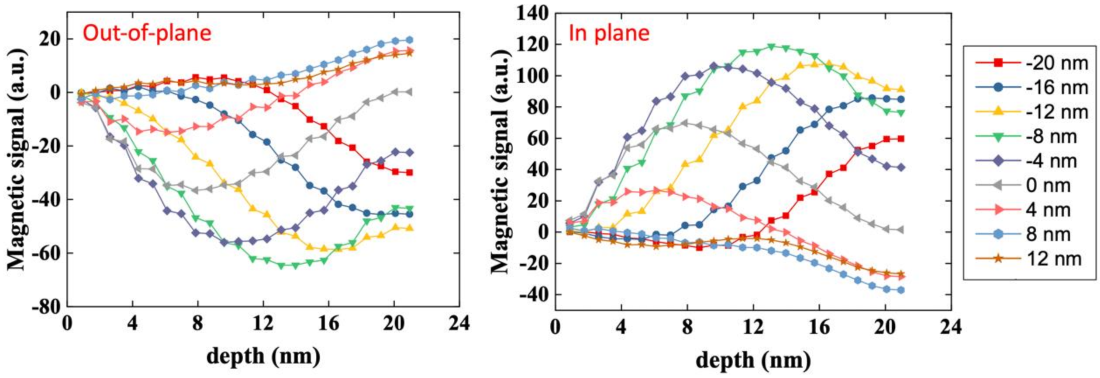

Fig. S1. Contribution of EMCD signals to total EMCD signals from different depths at different defocus values with V = 300 kV and CA = 15 mrad.

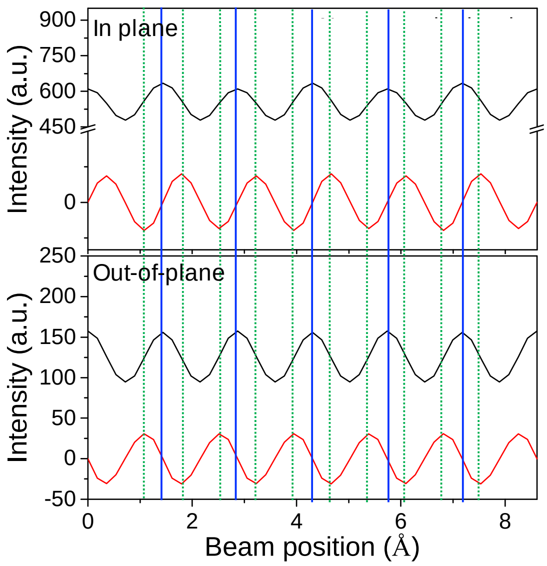

Fig. S2. Non-magnetic (black) and magnetic (red) signals plotted along the scanning direction (*x* axis) for Fig. 4 in the manuscript, respectively. The upper and lower panels show in-plane and out-of-plane signals, respectively. The vertical blue lines mark peaks in the non-magnetic signal and correspond to the positions of atomic planes. The green lines on either side of the blue lines mark the positions of maxima and minima in the magnetic signals. The maxima of the non-magnetic signals on atomic planes is approximately reduced by a factor of 4 between the in-plane (*on-axis* detector) and out-of-plane (*off-axis* detector) case owing to the different detector setups.