Sub-nanoscale spatial mapping of magnetic moments by STEM-EMCD under symmetric 3-beam condition using convergent beam

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Since the concept of electron magnetic chiral dichroism (EMCD), the electron counterpart to X-ray magnetic circular dichroism (XMCD), was first proposed and subsequently experimentally measured, efforts have been dedicated mainly to solve the low signal-to-noise ratio (SNR), using sub-nanometer electron probe towards its atomic resolution measurements. Recent theoretical and experimental works have shown that a combination of a convergent electron probe and a detector aperture placed on specific positions of reciprocal space with the systematic row of reflection excited can provide atomically localized EMCD in principle with a sufficient SNR [1-3]. The ideas are schematically shown in Figures 1(a) and (b), showing the EMCD signal distributions in the diffraction plane under the symmetric 3-beam condition of BCC iron with **G** = 110. Figure 1 implies two alternative ways to effectively pick up the chiral plus/minus EMCD signals.

The experiment was done using a JEM ARM-200F STEM equipped with a GIF Quantum ER, operated at 200 kV. A rectangular collecting aperture was implemented in GIF for the spatially - resolved EELS (SR-EELS) option. The sample was a 30 nm thick polycrystalline BCC iron film. A typical ADF-STEM image and the corresponding electron diffraction pattern are shown in Figures 2(a) and (b), respectively. Since the rectangular aperture position is fixed in the detector, perpendicular to the EELS dispersion direction, we carefully selected grains having the appropriate orientation with **G** =110 parallel to the dispersion direction for Figure 1(a) (Classical EMCD), while perpendicular for (b) (APR-EMCD). The incident electron was scanned towards a grain boundary in the direction perpendicular to the boundary. For Classical EMCD the same area was scanned twice with the collecting aperture set at either side to pick up the signals of different chirality, recording q-selective 2D CCD image with the diffraction spectrum image option (Figure 2(c)). For APR-EMCD the grain was particularly selected parallel to the operating (110) planes for plane-by-plane analysis.

The obtained datasets (4D data by Classical EMCD, 3D datacube by APR-EMCD) were aligned by peak positions, denoised, followed by applying blind source separation techniques to extract the magnetic signals and their abundance map [4]. The extracted EMCD signal for Classical EMCD is shown in Fig. 3(a) as a function of distance from the grain boundary, where the step width was 0.5 nm. The sign of the magnetic signal is inverted across the grain boundary due to the nearly 90 degrees rotation of the operating diffraction vectors for the neighboring grains. On the other hand, the +/– EMCD signals appear on either side of the lattice planes in an alternating manner, as theoretically predicted (Figs. 3 (b) and (c)). We are now ready to tackle the problem to clarify the local magnetism in sub-nanometer scale, using the present experimental schemes.

[1] Rusz, J. et al, Nature Comm. 7, 12672 (2016).

[2] Thersleff, T. et al, Phys. Rev. 94, 134430 (2016).

[3] Rusz, J. et al, Phys. Rev. B 95, 174412 (2017).

[4] Spiegelberg, J. et al, submitted (2018).

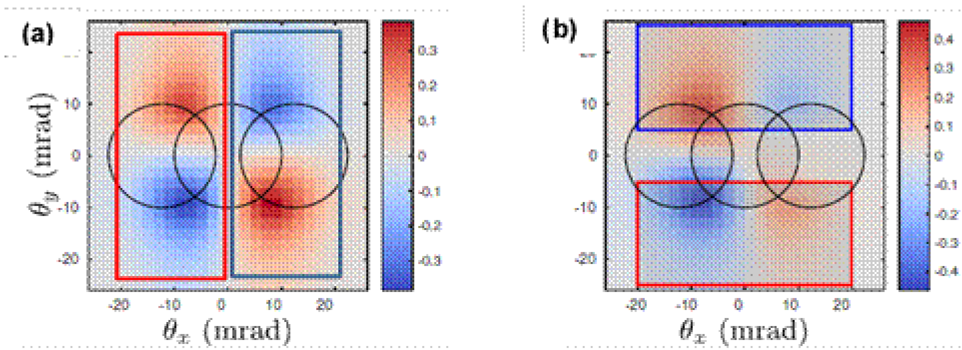

Figure 1 Theoretically calculated chiral plus (red)/minus (blue) EMCD signal in diffraction plane and two possible aperture positions (red and blue rectangulars) for collecting EMCD signal. (a) Classical EMCD case [1]. Chiral plus/minus spectra are recorded as ***q***-selective 2D CCD image using SR-EELS mode. (b) APR-EMCD case [2] Chiral plus/minus spectra are alternatingly collected, scanning the probe across the lattice planes.

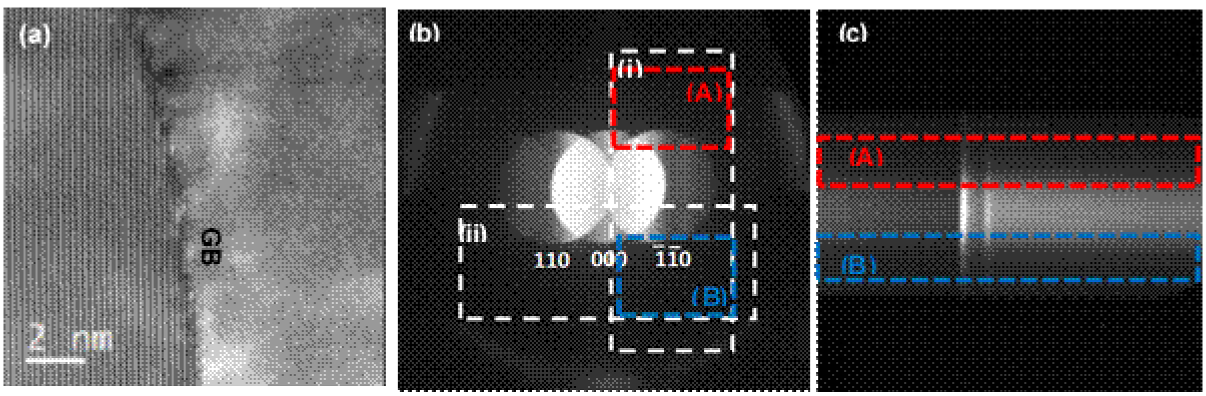

Figure 2 (a) Typical ADF-SEM image of BCC Fe including a grain boundary. 3-beam condition is set in the left grain. (b) Diffraction pattern with convergence semi-angle of 12 mrad and rectangular aperture positions (white broken frames: (i) Classical EMCD, (ii) APR-EMCD).(c) 2D CCD image of Fe-$L_{2,3}$ for Classical EMCD. Color-framed areas correspond to the areas with opposite chirality in (b) are used for EMCD signal extraction.

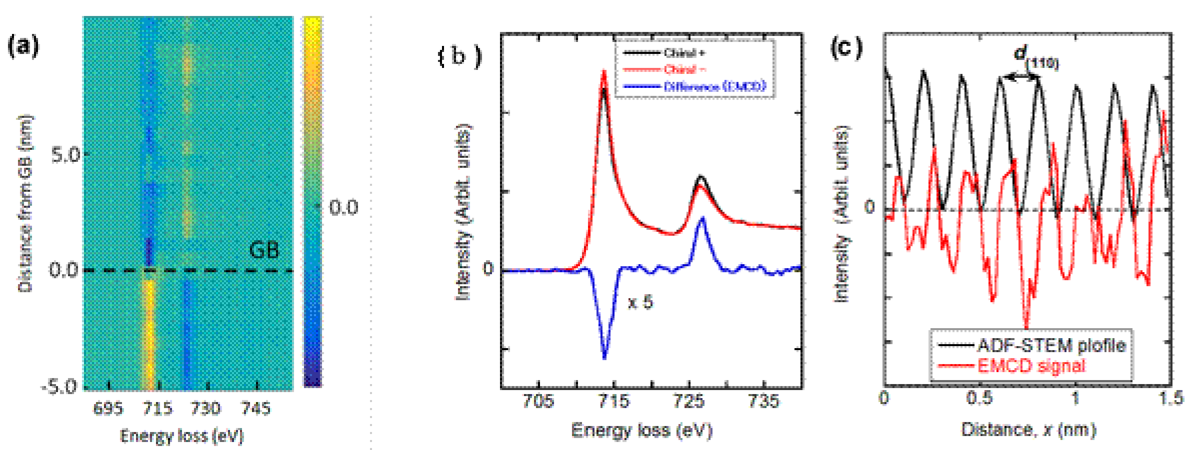

Figure 3(a) EMCD signal map as a function of distance from grain boundary by Classical EMCD. (b) Extracted chiral +/− spectra and EMCD profiles. (c) Projected EMCD signal distribution with respect to the 110 lattice planes.