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Blind identification of magnetic signals in electron magnetic chiral dichroism using independent component analysis

Jakob Spiegelberg[*,a], Dongsheng Song[b,c], Rafal E. Dunin-Borkowski[c], Jing Zhu[b], Ján Rusz[a]

[a] Department of Physics and Astronomy, Uppsala University, Box 516, S-751 20 Uppsala, Sweden

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

[c] Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons and Peter Grünberg Institute, Forschungszentrum Jülich, D-52425 Jülich, Germany

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ABSTRACT

Electron magnetic chiral dichroism (EMCD) is a promising technique to investigate local magnetic structures in the electron microscope. However, recognition of the EMCD signal, or also finding optimal parameter settings for given materials and sample orientations typically requires extensive simulations to aid the experiment. Here, we discuss how modern data processing techniques, in particular independent component analysis, can be used to identify magnetic signals in an unsupervised manner from energy filtered transmission electron microscopy (EFTEM) images. On the background of the recent advent of 4D scanning transmission electron microscopy, we discuss how this data processing may enable simultaneous tracking of all three spatial components of the magnetic momenta for arbitrary materials and several sample orientations without the previous need of complementary simulations.

Electron energy loss spectroscopy (EELS) offers a wealth of information of the sample down to atomic resolution on modern microscopes. For magnetic samples, the electron magnetic chiral dichroism (EMCD) [1,2] arises from the interaction of the electron with the magnetic moments of the sample in an EELS measurement. The magnetic signal is then obtained either by tuning the phase of the incoming electron, or by detecting electrons scattered into suitable angles. Thus, a successful EMCD measurement offers an opportunity to study nanoscale magnetic devices while at the same time retaining the advantages of EELS measurements, e.g., detailed information of the chemical environment, or site-specific fine-structure differences.

Despite experimental and theoretical advances in recent years pushing the achieved spatial resolution of EMCD measurements from several nanometers to few Ångström [3–13], experimental as well as conceptual challenges remain. Especially for measurements at high spatial resolutions, the low signal to noise ratio (SNR) of the inherently weak EMCD signal renders it difficult to detect (e.g., [4,7,10]). Dependent on multiple parameters, e.g., the sample thickness, convergence and collection angle, phase profile of the electron probe or exact shape and placement of a collection aperture in the objective lens's backfocal plane, the optimization of experimental parameters for a given sample (or material) to achieve the maximal SNR of the EMCD signal remains non-trivial to date.

Today, most studies focus on studying the out-of-plane magnetic moments assuming that the sample is magnetically saturated in the field of the objective lens. Going beyond this restriction to study also the in-plane components of the magnetic field adds an additional layer of complexity since the distribution of the three EMCD components in the backfocal plane overlap strongly. For many samples and crystal orientations, simple placement of a collection aperture at certain places in the backfocal plane may no longer be sufficient to fully distinguish all EMCD contributions.

In this article, we discuss how blind source separation (BSS) methods, in particular independent component analysis (ICA) (e.g., [14] or any other textbook on ICA), can be used to extract contributions to the EMCD signal in k-space, as contained in a series of energy filtered diffraction pattern covering the edge of interest, in an unsupervised manner. By applying ICA to both theoretically predicted and experimental data we evaluate the potential and practical applicability of our approach.

In Section 1, we discuss the usage of ICA for recognition of the EMCD signal in more detail. Furthermore, the method is applied to theoretically predicted data. Afterward, in Section 2, we demonstrate the proposed approach experimentally, followed by a brief discussion of the results in Section 3 and an outlook to extend the method to be applied to 4D-STEM data sets in Section 4.

* Corresponding author.
E-mail address: jakob.spiegelberg@physics.uu.se (J. Spiegelberg).

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1. Extraction of EMCD using ICA

In order to find a suitable extraction method for the EMCD signal it is useful to consider the mathematical characteristics of its k-space distribution. Experimentally, this distribution can be obtained by acquisition of a series of energy filtered transmission electron microscopy (EFTEM) images of the diffraction plane covering the EELS edge of the magnetic species of interest (see, e.g., [4,15]).

For this purpose, we have computed the k-space distribution of the EMCD signal of a Co crystal in (001) zone-axis (ZA), three beam case orientation (3BC, along the (018) axis) and two beam case orientation (2BC, obtained by successively tilting out of the 3BC so that G-spot and 0-spot have approximately equal intensities, which mimics the experimental setup of 2BCs) using the Mats.V2 code [16]. The crystal was chosen to be 21.72 nm (ZA) and 21.16 nm (3BC, 2BC) thick, the probe had an acceleration voltage of 200 kV. In order to compare EFTEM measurements to STEM experiments we compute both distributions using plane waves as incoming beam as well as convergent probes with a convergence angle of 5 mrad. The resulting distributions of the net non-magnetic and magnetic signals, i.e., the EMCD distributions assuming full polarization along x-, y- or z-axis, are shown in Fig. 1. The z axis was chosen parallel to the beam propagation direction, the sample tilt for the 3BC orientation followed the x axis.

At first sight, the k-space distributions of the EMCD signal are strongly orientation dependent. Additional factors, such as the crystal thickness or the convergence angle, are also known to affect these distributions. However, the z-component appears little correlated to the non-magnetic signal in all crystal orientations studied here. Furthermore, for 3BC and ZA computation, this uncorrelatedness extends to all three EMCD components. Comparing plane wave and convergent beam computations, one notices how the asymmetries of 2BC and 3BC become less pronounced at non-zero convergence angles and are negligible even at the moderate 5 mrad used here.

The origin of this asymmetry has been described before [17,18]. Since 2BC and 3BC imply tilted samples, the Ewald spheres are also correspondingly tilted so that an incoming plane wave will intersect it differently when scattered into the upper or lower half-plane (spanned by the systematic row reflections). When using convergent beams, the beam itself offers a selection of different incoming angles, each with slightly different intersections with the Ewald sphere. Hence, additional changes induced by the sample tilt become less pronounced.

These visual impressions are further corroborated by the correlation coefficients (i.e., Pearson correlation coefficients) shown in Fig. 2, which can easily be estimated from the vectorized component maps of Fig. 1. While the z-component is always uncorrelated to the non-magnetic signal, correlations remain in between the EMCD components for plane waves. At 5 mrad convergence angle, however, the observed correlations are attenuated. For the ZA calculation, all components are orthogonal to each other in either case.

Having recognized the uncorrelatedness of the components, ICA appears to be a suitable blind source separation technique for this unmixing problem. Particularly in cases where the source components, i.e., non-magnetic and EMCD signals, are known to be orthogonal and thus statistically independent, such as the ZA orientation or the 3BC for convergent electron probes, identifiability criteria of ICA are met, the method can be expected to extract the correct, physically meaningful source components. Note that uniqueness of ICA has to be understood as essential uniqueness, i.e., uniqueness up to permutation (ordering of the components) and scaling. Due to the scaling ambiguity, only relative amplitudes of the obtained components can be interpreted directly. To analyze also absolute amplitudes, further normalization with the associated scores is necessary. E.g., when applied to an EFTEM image, the amplitudes of the associated spectra need to be normalized before absolute amplitudes in the extracted k-space distributions can be compared.

A point of criticism of this approach is that while ICA can be expected to extract the EMCD components, at last, the user needs to identify them as such, which again presupposes knowledge of the EMCD distributions. We suggest to overcome this hurdle by using the following general criteria to identify unknown EMCD contributions

i. In contrast to non-magnetic signals, EMCD components are not centered around diffraction spots. In particular out-of-plane contributions fulfill this criterion well.
ii. EMCD contributions tend to be anti-symmetric with regard to some symmetry plane or rotationally symmetric with the rotation axis centered at the 0-spot. Both symmetries are dependent on the crystal orientation and distinguish EMCD contributions from other artifact components.
iii. Particularly in situations where EMCD contributions and non-magnetic signal are orthogonal, the EMCD contributions vanish when summed over large enough areas. Note that this criterion is likely violated when using electron probes with more general phase profiles, e.g., when using electron vortex beams.

We derive these criteria from our experience with simulations of k-space EMCD distributions across various samples and parameter settings, but point out that they may be violated for some experiment settings. In such cases, further simulations are needed. Yet, as we show below, using these criteria the EMCD contributions may now be identified without prior knowledge.

STEM-EMCD measurements, where the same sample region is scanned several times with different collection aperture placements obtaining spectrum images with EMCD signals of opposing signs, can be considered to yield a 4 dimensional data set, $\mathcal{X} \in \mathbb{R}^{x \times y \times E \times a}$ where x, y, E and a are the number of pixels along x- and y-axis, the number of energy channels and the number of aperture placements, respectively (the notation $\mathcal{X} \in \mathbb{R}^{a_1 \times a_2 \times \cdots \times a_n}$ denotes a n-dimensional tensor whose entries are real numbers and whose ith mode has the length a_i). One may thus wonder whether ICA, when applied to the matricized data $\mathbf{X} \in \mathbb{R}^{(x \cdot y \cdot E) \times a}$, can be used to extract the EMCD signal also here. Unfortunately, the typically low number of aperture placements a may be lower than the number of source spectra. The thus arising rank deficiency would prohibit ICA to fully separate magnetic and non-magnetic contributions. Furthermore, the averaging of large k-space regions, which is implicitly done by usage of a collection aperture, may destroy the statistical independence of the components (depending on the sample orientation and exact placement of the collection apertures). Hence, ICA applied to the k-space components may not offer a general route to EMCD extraction in STEM-EMCD measurements.

2. EMCD extraction on experimental data

In this section, we will demonstrate how ICA in combination with the above three criteria is able to identify the EMCD contributions in EFTEM images measured on a Co crystal tilted in two- (2BC) and three-beam case orientation and an Yttrium-iron garnet (YIG) tilted to ZA orientation. All three being typical crystal orientations used in EMCD measurements.

The samples used in the experiments are a Co nanoplate [13] and a YIG single crystal [19]. The energy filtered diffraction patterns for Co and YIG were acquired using a Gatan GIF spectrometer on a FEI Titan 80–300 operated at 300 kV with the energy step of 1 eV (slit width 2 eV). The acquisition time for each frame with a size of 1024 × 1024 is set to 10 s to ensure the signal-noise-ratio. The energy ranges in the experiments for Co and YIG are chosen around the L-edges of Fe and Co, respectively, i.e., 660–780 eV and 720–870 eV. The diameter of the (parallel) electron beam is about 100 nm. The thickness of the probed regions are approx. 20 nm for Co and approx. 45 nm for YIG. The three-dimensional data sets are consisting of the reciprocal k_x-k_y plane and electron energy loss. The artifacts and distortions are corrected for all the data sets. Firstly, the X-ray spikes are removed for all frames. Then,