Proposal for measuring magnetism with patterned apertures

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We propose a magnetic measurement method utilizing a patterned post-sample aperture in a transmission electron microscope. While utilizing electron magnetic circular dichroism, the method circumvents previous needs to shape the electron probe to an electron vortex beam or astigmatic beam. The method can be implemented in standard scanning transmission electron microscopes by replacing the spectrometer entrance aperture with a specially shaped aperture, hereafter called ventilator aperture. The proposed setup is expected to work across the whole range of beam sizes – from wide parallel beams down to atomic resolution magnetic spectrum imaging.

Nanotechnologies utilizing magnetic materials call for characterization techniques that allow to quantify magnetic properties with sufficient spatial resolution. Typically used methods, such as spin-polarized scanning tunneling microscopy [1, 2], magnetic exchange force microscopy [3], x-ray magnetic circular dichroism [4, 5] or electron holography [6], are either restricted to surface analyses or they lack sufficient spatial resolution.

An alternative measurement technique called electron magnetic circular dichroism (EMCD) is under development since its proposal in 2003 [7] and experimental confirmation in 2006 [8]. EMCD utilizes (scanning) transmission electron microscopes [(S)TEM], therefore it is a natural candidate to go beyond the previous limitations aiming for atomic spatial resolution magnetic measurements. Since 2006, significant improvements have been achieved in this direction, recently measuring antiferromagnets with astigmatic beams of atomic size [9], or utilizing aberration-free electron probes in experimental geometries with suitably oriented crystalline samples [10, 11]. Most recently, an alternative setup based on high-resolution TEM imaging has allowed for the detection of quantitative magnetic information from individual atomic planes [12].

EMCD measurements based on the initial experiment [8] use an off-axis aperture, most commonly a circular one built into the spectrometer, see e.g. [11, 13–19] among others. While many times successful, this approach has several shortcoming. Circular aperture collects only a small fraction of the inelastically scattered electrons and it is not an optimal shape for EMCD acquisition [20]. Its usual implementation then involves tilting the sample into two-beam or three-beam orientation, losing the view of individual atomic columns. In a STEM implementation it requires acquisition of several spectrum images from the same sample area [11, 13–15, 19]. This becomes progressively more challenging, when the desired spatial resolution increases together with convergence angles [21]. Alternative ways of detection were followed in Refs. [22, 23], though in these approaches it was not yet possible to extract EMCD spectra due to low signal to noise ratios.

In this Letter, we describe three crucial findings that have arisen from simulations of magnetically-sensitive inelastic scattering. First, we propose a new approach to optimize the EMCD signal collection in a wide range of crystallographic symmetries and scattering geometries. Using cubic systems as a test bed, we demonstrate that a custom-designed aperture can selectively allow for an optimal collection of either the positive or negative EMCD signal in a variety of high-symmetry zone axes. Second, we show that the signal collection with these apertures is robust across a large range of convergence angles, including those sufficiently large to focus an electron probe down to less than the width of an atomic column. Finally, we propose a strategy to allow for the collection of both positive and negative EMCD signal contributions simultaneously, negating the need for scanning the same sample area multiple times. These three findings represent a route to overcome the long-standing issues with EMCD acquisition.

Before discussing possible paths of practical realization of the scheme, we employ simulations to motivate our approach. The angle-resolved inelastic electron scattering cross-section is simulated using the combined multislice / Bloch waves method [24] as implemented in the code
with respect to all the symmetry axes (horizontal, vertical and both diagonal ones). In addition, with respect to the center of the diffraction pattern, the EMCD signal has a rotational symmetry whose order, however, depends on the crystal at hand.

These universalities lead us to propose a patterned detector aperture, which would have a regular rotationally symmetric shape dictated by rotational symmetry of the crystal and its orientation. In the case of [001] zone axis orientation of bcc Fe, it would be symmetric with respect to the rotations by 90 degrees, while in the three-beam orientation it is symmetric with respect to a rotation by 180 degrees. This leads to a characteristic ventilator-like shape, thus the name ventilator aperture.

Figure 1 shows a scheme of the principle. In Fig. 1(a) we present a set of simulations showing magnetic signal distribution in the [001] zone axis Fe-L₃ diffraction pattern for various convergence semi-angles \( \alpha \). The universal pattern of EMCD distribution can be recognized. In Fig. 1(b),(c) and Fig. 1(d),(e) we then show the ventilator apertures overlaid on the diffraction patterns in a zone axis and 3-beam orientation, respectively. Note how a ventilator shaped aperture permits selection of predominantly positive (red) or negative (blue) regions of the EMCD distribution by rotating 22.5 degrees and 90 degrees for zone axis and 3-beam orientation, respectively. (Suitable number of blades and the rotation angle obviously depend on the crystal symmetry and its orientation with respect to the electron beam, e.g., hexagonal crystals would require 3- or 6-blade apertures, etc.)

While the symmetry (number of blades) of the ventilator aperture is fixed by the symmetry of the crystal and its orientation with respect to the electron beam, the inner and outer collection angles, \( \Theta_{in} \) and \( \Theta_{out} \), are free parameters. A natural choice of these parameters would be such that maximizes the signal-to-noise ratio (SNR) of the EMCD signal, which can be expressed as [33]

\[
SNR = \frac{f_{red} M \sigma_{mag}}{\sigma_{nm}} \sqrt{\frac{2C_{L₃}}{1 + b}}
\]

where \( M/N \) is a material dependent property, \( C_{L₃} \) is count of electrons detected within the \( L₃ \)-edge energy range after the background subtraction and \( b \) is a ratio of the background electron counts to \( C_{L₃} \) within the same energy range. Finally, \( \sigma_{mag}/\sigma_{nm} \) is a ratio of normalized scattering cross-sections computed with mixed dynamical form-factor [34] set to \( S(q, q') = i(q_xq'_x + q_yq'_y) \) and \( S(q, q') = q_xq'_y \), representing EMCD due to magnetization parallel to \( z \)-axis and the non-magnetic component of the scattering cross-section, respectively [33].

Explicit optimization of the SNR for both zone-axis and 3-beam orientations is plotted in Fig. 2. Except for the thinnest samples considered in zone-axis orientation, the optimization over \( \Theta_{in} \) and \( \Theta_{out} \) leads to a very similar pattern, only weakly dependent on sample thickness \( t \) or convergence semi-angle \( \alpha \). This unforeseen finding

![Figure 1](image-url)
FIG. 2. Optimization of the signal to noise ratio (see Eq. 1) for a) zone axis and b) 3-beam orientations, respectively, as a function of inner and outer collection angles, $\Theta_{in}$ and $\Theta_{out}$, for a range of sample thicknesses $t$ and convergence semi-angles $\alpha$. Each panel shows the maximal SNR (in arbitrary units) as well as the inner/out collection semi-angles (in mrad) for which it was reached. All values are normalized to the same electron dose incident on the sample and the electron beam is always centered on an atomic column.

suggests that for each of the two symmetries (8-blade and 2-blade ventilator apertures, respectively) it should be possible to construct a universal ventilator aperture, which will be close-to-optimal for all convergence semi-angles and sample thicknesses. (Note though that its inner and outer collection angles depend on material and its Bragg scattering angles, nevertheless.) A global optimization is obviously an ambiguous procedure, due to the given (arbitrary) choice of convergence angles and thicknesses in our simulations, and also the weighting of the individual cases. Here we simply averaged all the SNRs over the panels presented in Fig. 2, from which we extracted the proposed optimal $\Theta_{in}$ = 19 mrad and $\Theta_{out}$ = 38 mrad for the zone axis case, and $\Theta_{in}$ = 6 mrad and $\Theta_{out}$ = 30 mrad for the 3-beam geometry.

We stress again that the exact values have little of meaning due to arbitrariness of the global optimization procedure. Nevertheless, some semi-quantitative observations can be made: 1) in the zone axis orientation the optimal collection of signal should happen at larger scattering angles, with $\Theta_{out}$ ≈ 2$\Theta_{in}$, 2) in 3-beam orientation the crystal scatters less strongly and consequently the $\Theta_{in}$ can be much smaller. On the other hand, $\Theta_{out}$ can be relatively large, therefore a much larger fraction of inelastically scattered electrons can be utilized for the EMCD measurements. This is reinforced by explicit comparison of the SNR values in Fig. 2 between zone-axis condition and 3-beam geometry, which suggests that for a fixed electron dose the 3-beam orientation often offers a significantly better SNR than the zone-axis orientation, except for thicker samples measured with larger convergence angles.

Figure 2 suggests that high SNRs can be obtained at rather large convergence semi-angles. Therefore, a natu-
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profilograph  


dis semblar to the Large Angle Convergent DIffraction (LACDIF) geometry previously used to acquire an EMCD signal [37], our proposed geometry allows to extract spectra with opposite signs of EMCD from the two half-planes of the CCD. While similar to the Large Angle Convergent DIffraction (LACDIF) geometry previously used to acquire an EMCD signal [37], our proposed geometry allows the probe to be fully converged on the sample. This experimental design thus permits simultaneous acquisition of both signs of EMCD with a single scan while acting on the zone axis of a magnetic material with a fully converged probe under electron optical conditions suitable for atomic column resolution. This strategy could be readily implemented by exchanging the spectrometer entrance aperture by a suitable patterned one, provided
that we can align the energy dispersion axis parallel to \( \theta_x \). Naturally, an (in this approach optional) physical rotation mechanism could simplify the alignment.

We have proposed a strategy for EMCD acquisition that should result in the ability to probe individual atomic columns. First, we use simulations to derive a series of optimized patterned aperture designs that can be implemented in any desired zone axis geometry. Second, we demonstrate that the EMCD signal is largely robust with respect to convergence angle, allowing for this strategy to be implemented for a wide variety of probe sizes ranging from above 10 nm down to sub-Ångström in diameter. Finally, we propose an aperture and spectrometer configuration that would allow for the simultaneous collection of the negative and positive EMCD contributions, negating the need for multiple scans.

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We have proposed a strategy for EMCD acquisition that should result in the ability to probe individual atomic columns. First, we use simulations to derive a series of optimized patterned aperture designs that can be implemented in any desired zone axis geometry. Second, we demonstrate that the EMCD signal is largely robust with respect to convergence angle, allowing for this strategy to be implemented for a wide variety of probe sizes ranging from above 10 nm down to sub-Ångström in diameter. Finally, we propose an aperture and spectrometer configuration that would allow for the simultaneous collection of the negative and positive EMCD contributions, negating the need for multiple scans.

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