Quantitative Analysis of Nanoscale Magnets with Intrinsic Electron Magnetic Circular Dichroism

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Electron magnetic circular dichroism (EMCD) allows the quantitative, element-selective determination of spin and orbital magnetic moments [1], similar to its well-established x-ray counterpart, x-ray magnetic circular dichroism (XMCD). However, because of the low intensity of the EMCD signal, it has not yet been possible to obtain quantitative information from EMCD signals at the nanoscale. We develop a new approach to EMCD measurements that considerably enhances the outreach of the technique [2]. The statistical analysis introduced here yields robust quantitative EMCD signals. Moreover, we demonstrate that quantitative magnetic information can be routinely obtained using electron beams of only a few nanometres in diameter without imposing any restriction regarding the crystalline order of the specimen.

Rather than measuring a few energy-filtered diffraction patterns or a few spectra in a well-defined scattering geometry, we acquire a large number of spectra in a random alignment of incoming beam and detector orientations with respect to the lattice axes of an illuminated grain. The acquired large dataset forms the starting point for our statistical procedure leading to the extraction of the EMCD spectrum. In the measurement, we use an ultra-high-voltage (1 MV) electron beam to significantly reduce the effect of multiple scattering, which could otherwise distort the EMCD signal. A schematic diagram of our experimental setup is presented in Fig. 1a. The same acquisition procedure was repeated three times independently on different areas of the sample, yielding three independent datasets.

After pre-processing the measured spectra, we calculated a difference spectrum for every pair of the spectra present in the dataset. Approximately 20–25% of pairs out of the entire set of pairs always displayed signs of the EMCD signal. We averaged the accumulated EMCD signals to obtain the final EMCD spectrum, which is presented in Fig. 2a. The EMCD signal intensity fraction was estimated to be 2–2.3% of the Fe-$L_{2,3}$ signal intensity (Fig. 2a),

After extraction of an EMCD signal with a good SNR), the EMCD sum rules were applied to the signal to evaluate the orbital moment/spin moment ratio, given by:

$$\frac{m_{\mathrm{L}}}{m_{\mathrm{S}}} = \frac{2}{3}\frac{\int_{\mathrm{L}_3}\Delta\sigma(E)dE + \int_{\mathrm{L}_2}\Delta\sigma(E)dE}{\int_{\mathrm{L}_3}\Delta\sigma(E)dE - 2\int_{\mathrm{L}_2}\Delta\sigma(E)dE} = \frac{2q}{9p-6q}, \qquad (4)$$

where q is an energy integral of the EMCD spectrum $\Delta\sigma(E)$ over both edges and p is an energy integral over the L_3 edge only. Without loss of generality, we can rescale the EMCD spectrum or its integral such that $p = 1$ (Fig. 2b). Thus, the orbital to magnetic moment ratio m_L/m_S becomes a function of q only. We finally obtained $m_L/m_S = 0.0429 \pm 0.0075$, which is in good agreement with the value obtained by XMCD for bcc iron, i.e., 0.043 [3]. Our result represents the first quantitative EMCD detection performed on a polycrystalline film.

In summary, we conclude that the method introduced here leads to statistically significant EMCD spectra. Consequently, our approach allows quantitative EMCD studies of non-single crystalline samples on the nanoscale and hence paves the way for a new era of application of EMCD experiments in the field of nano-magnetism.

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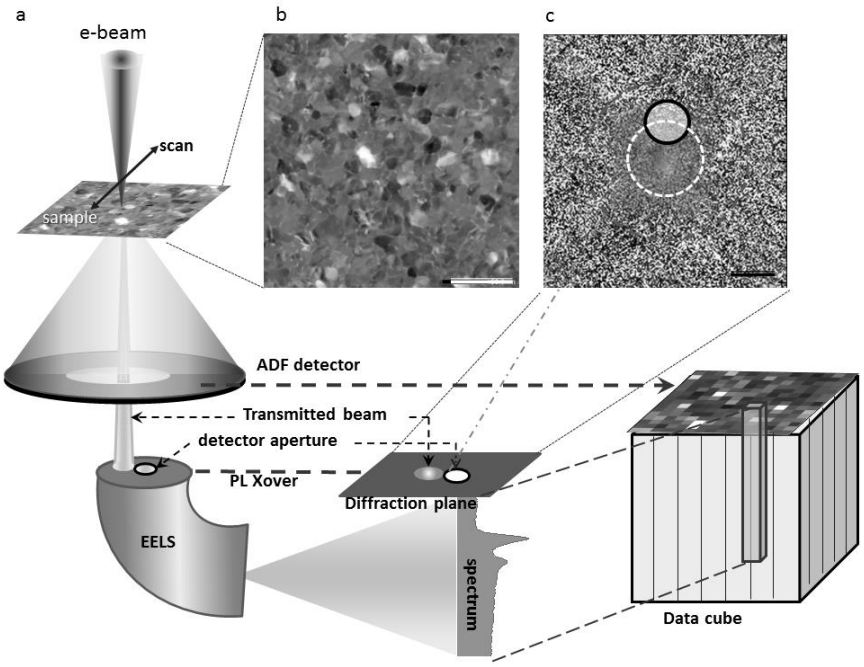

FIG. 1. **a**. Schematic drawing of the experimental setup and the data obtained. In the present STEM mode, the PL cross-over position is on the diffraction plane. **b.** A TEM image of the investigated polycrystalline iron film. Scale bar, 50 nm. **c.** Calculated EMCD signal intensity distribution of a polycrystalline iron film in the diffraction plane. The detector entrance aperture (solid circle) is located at the position of 0.4 $g_{(110)}$ away from the origin, and its diameter is 0.5 $g_{(110)}$. The white broken circle represents the possible aperture centre positions in the diffraction plane and blue broken circle corresponds to $g_{(110)}$ ring position for comparison. Scale bar, 2 nm^{-1}. The minimum (black) and maximum (white) EMCD values range from -3% to +3%.

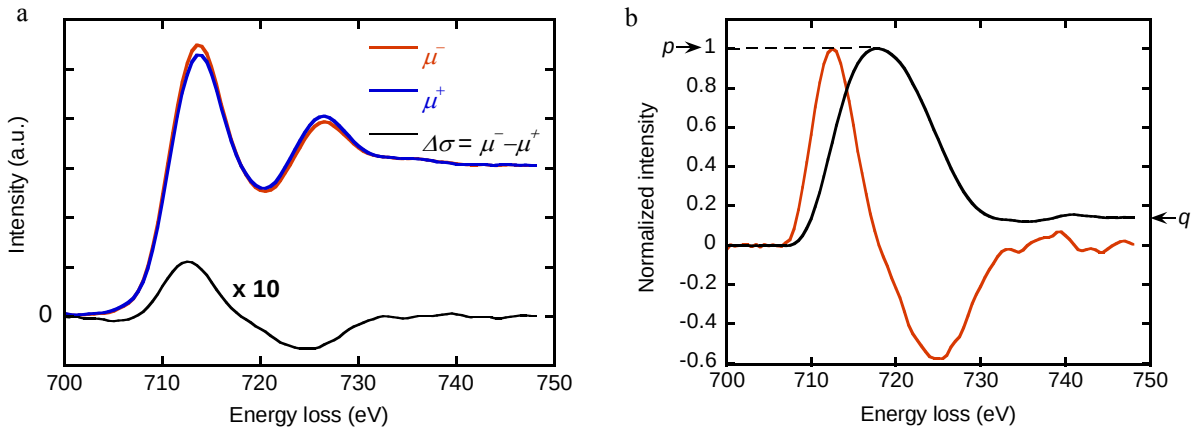

FIG. 2. **a**. A pair of Fe-$L_{2,3}$ EELS spectra (μ^+ and μ^-) extracted from the dataset and their difference spectrum (EMCD signal) $\Delta\sigma = \mu^- - \mu^+$. **b**. The averaged EMCD signal (red curve) and its cumulative sum (black curve), which are required to apply the sum rule. q is an energy integral of the EMCD signal over both edges, and p is an energy integral over the L_3 edge only. Without loss of generality, we can rescale the integral of the EMCD signal such that p = 1, as indicated, whereupon the orbital to magnetic moment ratio m_L/m_S is a function of q only, allowing an easy visual comparison of the energy integrals in the post-edge region.