On the precision of ELNES calculations for EMCD applications

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The recently discovered possibility of measuring atom-specific magnetic moments in the electron energy loss near edge structure (ELNES) [1] was followed by a fast development of the technique. The spatial resolution was improved down to 2nm [2] and there are indications that this is not yet the limit. The new technique, electron magnetic circular dichroism (EMCD), as an analogue of its x-ray counterpart – the x-ray magnetic circular dichroism (XMCD) – is however more intricate due to its use of the ELNES spectra and dynamical diffraction effects. First principles calculations seem to be an indispensable tool for optimizing the experimental conditions and understanding the sensitivity of the measurements. The theory was formulated in 2007 [3] and soon after it was followed by the derivation of sum-rules [4,5], which in principle allow to extract spin and orbital magnetic moments from experimental EMCD spectra. The first quantitative measurements using EMCD with detailed error analyses have been published only very recently [6].

On the theory side, to simulate the energy filtered diffraction patterns, one needs to perform rather demanding calculations involving elastic scattering before and after the inelastic event, summed over all possible scattering centers. The exit beam-electron wave-function is then projected on the mesh of outgoing wave-vectors, often consisting of thousands of pixels. Each such calculation can consist of a considerable number of evaluations of the inelastic scattering matrix element, the so called mixed dynamic form-factor (MDFF). Clearly, such calculations can be very demanding, depending on the accuracy goal.In the case of EMCD and simulations of the maps of the apparent orbital to spin moment ratios, one needs to perform extremely high-accuracy calculations in order to obtain meaningful results, because EMCD normally manifests itself as a small percentage variation of the heights of ELNES peaks. To demonstrate this fact, let's assume the case of 3d transition metals, where one typically measures the L_2 and L_3 ELNES. There are two steps of the calculation process, where the information has to come from small differences between two similar quantities: 1) the EMCD signal is a difference of two spectra; 2) in the expressions relating the EMCD signal with spin and orbital momentum [4,5], the positive and negative parts of the integral of the signal are added.

Previously, we have used an algorithm described in [3], which selected the most strongly excited reflections based on w, the dimensionless product of extinction distance and excitation error. In the domain of these, the most strongly contributing Bloch waves were identified. Subsequently, all possible octuple products and MDFFs for resulting momentum transfer vector diads were evaluated.

In this presentation we will describe a new optimized algorithm for calculating the electron energy loss spectra, including the dynamical diffraction effects. The algorithm aims to minimize the number of the summed terms and MDFF evaluations by reordering and sorting the Bloch coefficients and their products. The increased efficiency, accuracy and scaling of the resulting algorithm is significant.

Figure 1 demonstrates the convergence of diffraction patterns, difference maps and m_L/m_S maps as a function of the cutoff P_{min} for octuple products relative to the maximal term.

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Table 1 summarizes some more technical information, including the computational time. The algorithm can be further optimized by removing a certain constant offset in these calculations, since the initialization of the MDFF calculation and the diagonalization of the initial-beam secular matrix is repeated at each pixel, i.e., 51x51=2601 times. Evaluations based on the older method, such as those published in [6], reached accuracy comparable to Pmin=10^{-3} in ca 10 hours. Computing time however scaled very badly with cut-offs due to the eightfold products of Bloch coefficients in the expression for the scattering cross-section. To calculate converged mL/mS maps months of computing time would be needed.

We believe that our calculations will contribute to a better understanding of the convergence properties of Bloch-wave calculations and that our method will serve as an efficient starting point for more complete simulations including e.g. partial coherence in the convergent incoming beam set-ups.

1. P. Schattschneider, S. Rubino, C. Hebert, J. Rusz, J. Kunes, P. Novak, E. Carlino, M. Fabrizioli, G. Panaccione, G. Rossi, Nature **447**, 486 (2006).
2. P. Schattschneider, M. Stoger-Pollach, S. Rubino, M. Sperl, C. Hurm, J. Zweck, J. Rusz, Phys. Rev. B **78**, 104413 (2008).
3. J. Rusz, S. Rubino, P. Schattschneider, Phys. Rev. B **75**, 214425 (2007).
4. J. Rusz, O. Eriksson, P. Novak, P. Oppeneer, Phys. Rev. B **76**, 060408(R) (2007).
5. L. Calmels, F. Houdellier, B. Warot-Fonrose, C. Gatel, M. J. Hÿtch, V. Serin, E. Snoeck, P. Schattschneider, Phys. Rev. B **76**, 060409(R) (2007).
6. H. Lidbaum, J. Rusz, A. Liebig, B. Hjörvarsson, P. M. Oppeneer, E. Coronel, O. Eriksson, K. Leifer, Phys. Rev. Lett. **102**, 037201 (2009).

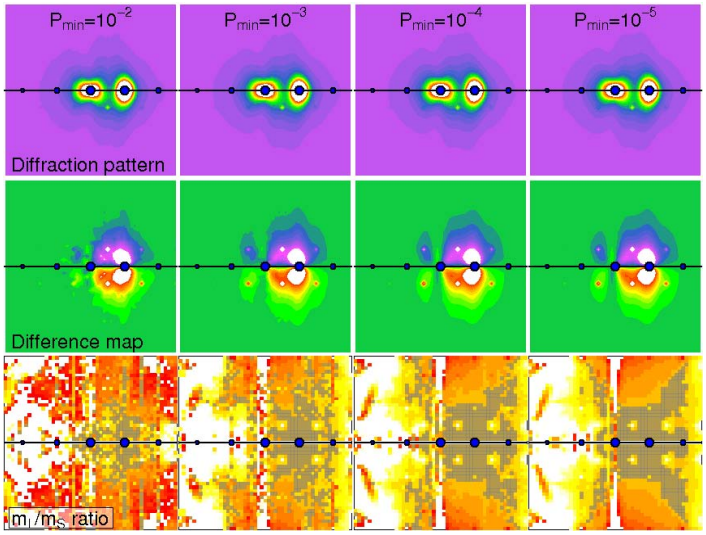

Figure 1. Diffraction patterns (top row), up-down difference maps (middle row) and apparent m_L/m_S ratio maps (bottom row) as a function of convergence parameter P_{min}. Calculations were performed for a 20nm layer of bcc iron in two-beam geometry with **G**=(200), at 300keV.

$P_{\min}$	$\langle N_2 \rangle$	$\langle N_4 \rangle$	$\langle N_8 \rangle$	$\langle N_{\mathbf{qq'}} \rangle$	t
10^{-2}	230	72	330	12	2h 16min
10^{-3}	1355	750	4710	110	3h 18min
10^{-4}	5545	5865	56830	705	11h 13min
10^{-5}	23060	39760	570250	3600	83h 35min

Table 1. Average lengths of double $\langle N_2 \rangle$, quadruple $\langle N_4 \rangle$ and octuple $\langle N_8 \rangle$ product lists, average number of momentum transfer diads per energy step $\langle N_{\mathbf{qq'}} \rangle$ and computing times for maps in Fig. 1 as a function of convergence parameter P_{min}. Time t refers to a calculation performed on a single Intel Pentium 4 Xeon processor at 2.5GHz.