Efficient convergence of the Bloch waves method for calculation of orientation-sensitive ELNES

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The orientation dependence of the electron energy loss near-edge structures (ELNES) on crystals can be calculated by a three-step procedure: 1) elastic propagation of the probe electrons, 2) inelastic event matrix element (so called mixed dynamical form factor, MDFF [1]), 3) elastic propagation of the scattered probe electron out of the sample. We will deal with the Bloch waves theory of elastic propagation of the electrons. The formalism [1-3] leads to expressions

$$\frac{\partial^2\sigma}{\partial\Omega\partial E} = \sum_{\mathbf{ghg'h'}} \frac{1}{N_{\mathbf{u}}} \sum_{\mathbf{u}} \frac{S_{\mathbf{u}}(\mathbf{q},\mathbf{q}',E)}{q^2 q'^2} e^{i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{u}} \times \sum_{jlj'l'} Y^{jlj'l'}_{\mathbf{ghg'h'}} T_{jlj'l'}(t)$$

$$Y^{jlj'l'}_{\mathbf{ghg'h'}} = C^{(j)\star}_{\mathbf{0}} C^{(j)}_{\mathbf{g}} D^{(l)}_{\mathbf{0}} D^{(l)\star}_{\mathbf{h}} \times C^{(j')}_{\mathbf{0}} C^{(j')\star}_{\mathbf{g'}} D^{(l')\star}_{\mathbf{0}} D^{(l')}_{\mathbf{h'}}$$

where **q**,**q'** are momentum transfer vectors, E is energy loss, $S_{\mathbf{u}}$ is the MDFF at atom with basis vector **u**. The factor Y is an octuple product of so called Bloch coefficients C, D for incoming and outgoing beam, respectively. Vectors **g**,**h** denote beams, i,j are indices of Bloch waves. Finally, $T(t)$ is a function of sample thickness t.

The direct evaluation of the sum therefore scales with 8th power of the number of beams and therefore calculation very quickly becomes intractable, already for a few tens of beams. In [3] an algorithm was introduced for automatic selection of beams and Bloch waves on the basis of excitation error, extinction distance and a norm of the Bloch wave within a subspace of selected beams. Yet, as can be seen in the Fig. 1, already for 12/14 beams for incoming/outgoing probe electron and 5/9 most strongly excited Bloch waves, we are summing huge number of terms (in total ~57 millions) and vast majority of them have negligible magnitude. Pushing the convergence further quickly produces hundreds of millions of terms, while adding only few within the highest orders of magnitude.

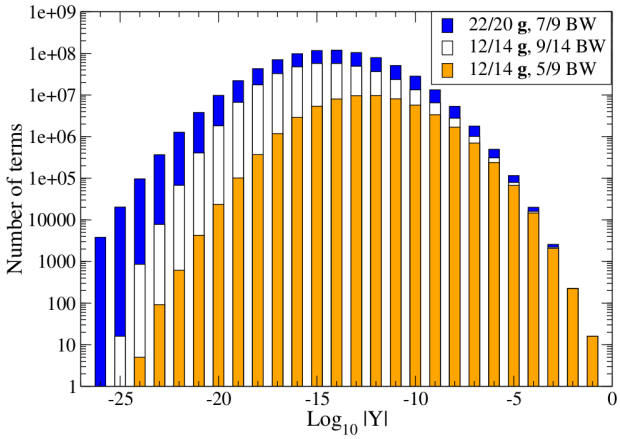

Fig. 1: Histogram of term magnitudes in ELNES calculation (see text).

Here we present a method named “modified automatic term selection” (MATS) that efficiently constructs only terms larger than given cut-off. The computational time grows inversely proportionally to the cut-off criterion and allows to solve the secular equation with hundreds of beams, yet being significantly faster than the direct summation.

In the Fig. 2, there is a comparison of four methods of calculating the energy-filtered diffraction pattern for a three-beam orientation tilted by ~10 degrees from (001) zone axis orientation, with **G**=(200), 300keV acceleration voltage, for bcc-iron crystal structure. Left column shows a systematic row approximation with 9 beams $\pm n\mathbf{G}$ with n up to 4. The secular equation leads to a matrix 9x9 and a complete summation of 9^8, i.e., 43 millions of terms is performed. Calculation of the complete diffraction pattern (201x201 pixels) took 561 CPU hours. For the 2nd column, we picked all beams for which a product of extinction distance and excitation error is below 300 and a norm of the eigenvector is above 0.01, see [3]. The resulting calculation shows additional features, such as Kikuchi bands in lower quadrants and some spot-like features around $(q_x,q_y)=(\pm 5,-1)$. This calculation took 2800 CPU hours. In the 3rd column there are results of the new summation method MATS picking only terms larger than 0.01. This calculation shows even more features, particularly the much better resolved Kikuchi bands passing from top corners of the diffraction pattern and crossing at the zone axis point (down, outside the range) at about 10 degrees tilt from the transmitted beam direction. The computing time required for this calculation was though only 43 hours. In the last column we show a result of the MATS algorithm considering all terms larger than 0.0001. Visually it is not easy to spot differences from a calculation with cut-off 0.01, which demonstrates that already such a low cut-off is close to a converged diffraction pattern. Note, that the more accurate calculation still took only moderate computing time 98 CPU hours. Most importantly, MATS calculations are substantially more accurate and yet they require an order of magnitude less time than even a simple systematic row approximation with 9 beams.

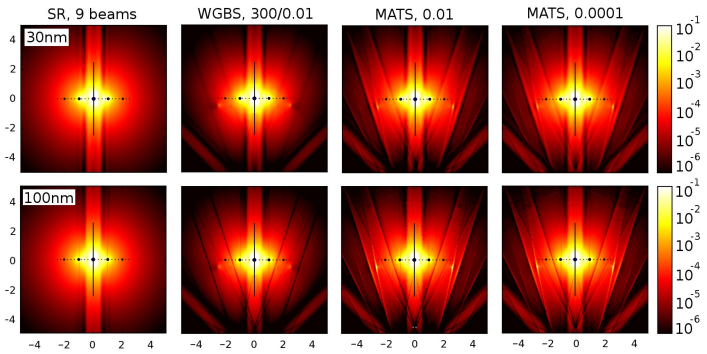

Fig. 2: Diffraction pattern of bcc-Fe in 3-beam orientation, G=(200), thicknesses 30nm (top) and 100nm (bottom), calculated using four different methods.

We will present a more detailed analysis of the scaling of MATS with the cut-off criterion and its performance in various situations, e.g., maps of magnetic signal [4,5], tilting from the zone axis towards a 3-beam orientation, electron channeling and comparison with ICSC code [6].

References

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