Slice-by-Slice Dynamical Simulation Scheme for High-Angular Resolution Electron Channeled X-ray Spectroscopy

Masahiro Ohtsuka[1], and Shunsuke Muto[2]

[1]Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan
[2]EcoTopia Science Institute, Nagoya University, Nagoya 464-8603, Japan

X-ray spectral mapping as a function of the incident electron beam angle is called as high-angular resolution electron channeled X-ray spectroscopy (HARECXS), and is known as an element/site-selective analysis technique. Different X-ray spectra are collected with respect to the incident angle variation, which accordingly changes the symmetries and amplitudes of the Bloch waves excited, propagating preferentially along particular atomic sites. The technique is currently being extended to ionization channeling pattern (ICP) recording by two-dimensional beam rocking (scanning reciprocal space).

In order to interpret the ICP quantitatively, detailed comparison between experimental results and the corresponding dynamical simulation is indispensable. A dynamical HARECXS/ICP simulation has been established based on the inelastic scattering cross-section and the Bloch-wave method [1, 2]. Such a scheme, however, includes time-consuming processes, particularly in calculating 2D ICPs. In this study, we have developed an efficient algorithm for calculating HARECXS/ICP patterns. In the new algorithm, two kinds of absorption potentials are introduced as was done in an annular dark-field STEM image simulation [3]: the absorption potential U_{TDS}, the effect of thermal diffuse scattering (TDS), and $U_{TDS+EDX} = U_{TDS} + U_{EDX}$, which incorporates the additional term U_{EDX} expressing the core excitation effect associated with the following X-ray emission. The corresponding wavefunctions Ψ_{TDS} and $\Psi_{TDS+EDX}$ are calculated with taking U_{TDS} and $U_{TDS+EDX}$ into account, respectively. The difference in the total intensities between these two wavefunctions is related to the X-ray intensity, I_{EDX}.

$$I_{EDX} \propto \int |\Psi_{TDS}(\mathbf{R},t)|^2 \, d\mathbf{R} - \int |\Psi_{TDS+EDX}(\mathbf{R},t)|^2 \, d\mathbf{R} \quad , \tag{1}$$

where $\mathbf{R}$ is a two-dimensional position vector in a plane parallel to the entrance surface, and t is the depth of the exit surface from the entrance surface. This scheme is found to drastically accelerate the calculation speed compared to the conventional method using the inelastic cross section, though it yields serious error at exit surface. Figure 1 shows calculated the thickness dependence of the relative Sr-L line intensities of $SrTiO_3$ at the exact [001] incident, respectively using Eq. (1) (solid black line) and the conventional method (open circles). The black line is significantly deviated from the results by the conventional method. This error should arise from the additional potential U_{EDX}, which overestimated the attenuation of the electron densities propagating through the particular atomic columns.

In order to overcome the difficulty, we introduced a slice-by-slice method which divides the sample into many thin slices. Eq. (1) is thus rewritten, for an n slices system, as

$$I_{EDX} \propto \sum_{j=1}^{n} \left[\int |\Psi_{TDS}^j(\mathbf{R},t_j)|^2 \, d\mathbf{R} - \int |\Psi_{TDS+EDX}^j(\mathbf{R},t_j)|^2 \, d\mathbf{R} \right] \quad , \tag{2}$$

where $\Psi^{j}_{TDS}(\mathbf{R}, t_j)$ and $\Psi^{j}_{TDS+EDX}(\mathbf{R}, t_j)$ are the corresponding wavefunctions at the depth t_j corresponding to the exit surface of j-th slice. In order to reduce above-mentioned error, $\Psi_{TDS+EDX}$ in j-th slice is connected with Ψ_{TDS} in j–1-th slice by applying the boundary condition, $\Psi^{j-1}_{TDS}(\mathbf{R}, t_{j-1}) = \Psi^{j}_{TDS+EDX}(\mathbf{R}, t_{j-1})$. For instance, dividing the sample of 150 nm thick into 769 slices, whose thickness corresponds to the half of unit-cell length, followed by applying Eq. (2) provided the result consistent with the conventional method, as also shown in Fig. 1 (red line). Figure 2 shows calculated two-dimensional ICPs, for the 150 nm tilted around the [001] axis. It is seen that the calculated ICP with the sufficient number of slices is nearly identical to that of the conventional method. The present scheme is easily extended to the multislice method with the inelastic scattering incorporated, which is particularly suitable for the cases where the system of interest contains lattice defects. The calculation details are presented on the poster.

References

[1] M. P. Oxley, and L. J. Allen, *Acta Cryst.*, **A56** (2000) 470.
[2] M. P. Oxley, and L. J. Allen, *J. Appl. Cryst.*, **36** (2003) 940.
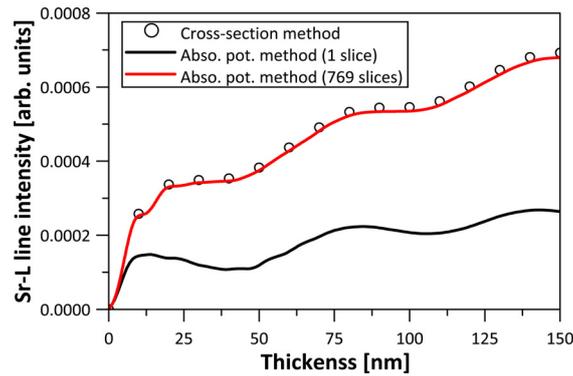[3] T. Yamzaki *et al.*, *Acta Cryst.*, **A62** (2006) 233.

FIG. 1: Through-thickness simulated Sr-L line intensities of $SrTiO_3$ with the incident-beam angle parallel to [001] axis, calculated by using present method at two different conditions (solid lines), and conventional inelastic cross-section method (open circles).
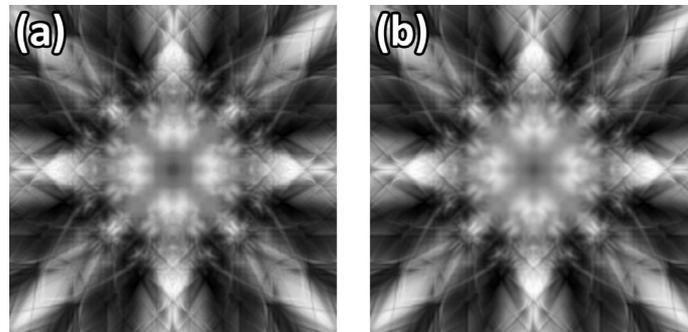

FIG. 2: Simulated two-dimensional Sr-L ICPs around [001] orientation of 150 nm thick $SrTiO_3$, calculated by using the conventional inelastic cross-section method (a), and present method with 769 slices (b), respectively.