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Theoretical Evaluation of Atomic-Resolution X-ray Analysis toward Quantification

M. Watanabe*
* Dept of Materials Science and Engineering, Lehigh University, Bethlehem. PA 18015.

The latest aberration-corrected scanning transmission electron microscopes (STEMs) in combination with the large solid-angle silicon-drift X-ray detectors (SDDs) improve limited efficiencies of signal generation and collections. It is no longer dream to acquire atomic resolution X-ray maps and to achieve single atom sensitivity in X-ray analysis by using the latest aberration-corrected instruments. Obvious next challenge is quantification of such atomic-resolution X-ray maps. There are several attempts to perform quantification of the atomic resolution X-ray maps [e.g. 1, 2]. However, quantified results are deviated from expected values from the structures. For example, Ga composition does not reach to 100% at Ga only columns in a [100]-projected GaAs compound [1]. Clearly, X-ray signals are generated from the specific atomic column where the incident electron is focused but also surrounding columns. For appropriate quantification, it is essential to determine how much X-ray signals are generated from the target and neighboring columns. In this study, X-ray signal generation in an oriented crystalline structure is simulated theoretically toward quantitative analysis.

In order to simulate X-ray generation, first, it is required to know how the incident electron probe propagates in an oriented crystalline material. The electron propagation, called the electron wave function, can be simulated by multislice calculation. In this study, the xHREM code [3] was used to simulate the electron wave function. Figure 1 shows the simulated electron propagation at a Ga column in the [100]-projected GaAs. An image at the bottom in Fig. 1 indicates the electron distribution at one of sliced planes. The electron distribution is not homogeneous but strongly related to atomic arrangements, which are scattering sources.

Once the electron distributions are simulated, X-ray signals can be simulated at each sliced plane. As shown in Fig. 2 (a schematic cross-section view of the GaAs structure), X-ray spectra were simulated at each sliced layer by porting the X-ray generation engine of legacy Desktop Spectrum Analyzer (DTSA) codes [4]. An X-ray spectrum at a certain depth from the top surface of the specimen was obtained by adding individual spectra. Since the DTSA codes were used, X-ray absorption is also taken care of. X-ray maps were constructed by repeating this process at various positions. In Fig. 3, a set of simulated results are summarized for different specimen thicknesses (10, 40, 80 and 160 unit-cell depths, corresponding to 5.6, 22.4, 44.8 and 89.6 nm, respectively): simulated high-angle annular dark-filed (HAADF) images in top row, simulated ratio maps of Ga K to total K intensities (Ga K and As K) in middle row, and normalized total K intensities in bottom row. Because X-ray generation and detection for the Ga K and As K line due to close atomic numbers of Ga (31) and As (33), the maps of Ga K/total intensity ratio approximately indicate the Ga concentration fraction. Although the individual atomic columns are still separated in the intensity ratio maps even at thicker region, more mixing can be seen in generated X-ray signals between atomic columns as the specimen thickness increases. Furthermore, the difference in X-ray intensities between on- and off-atomic columns is reduced as the specimen thickness increases. For Quantification of atomic-resolution X-ray maps, these X-ray generation behaviors must be modeled as a function of the specimen thickness.

References
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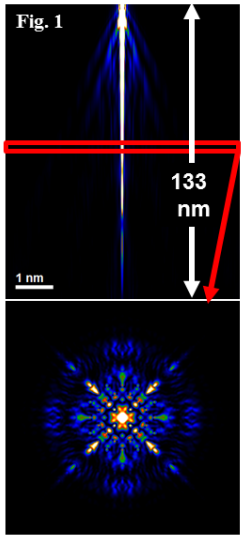
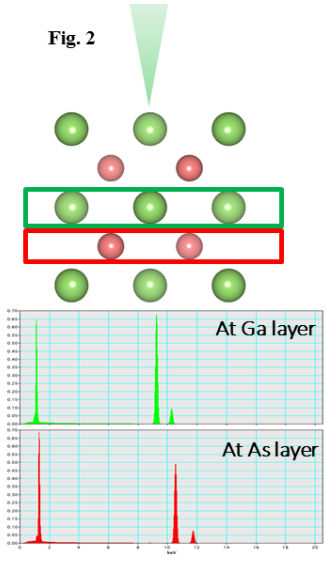
[5] The author wishes to acknowledge financial support from the NSF through grants DMR-0804528 and DMR-1040229.





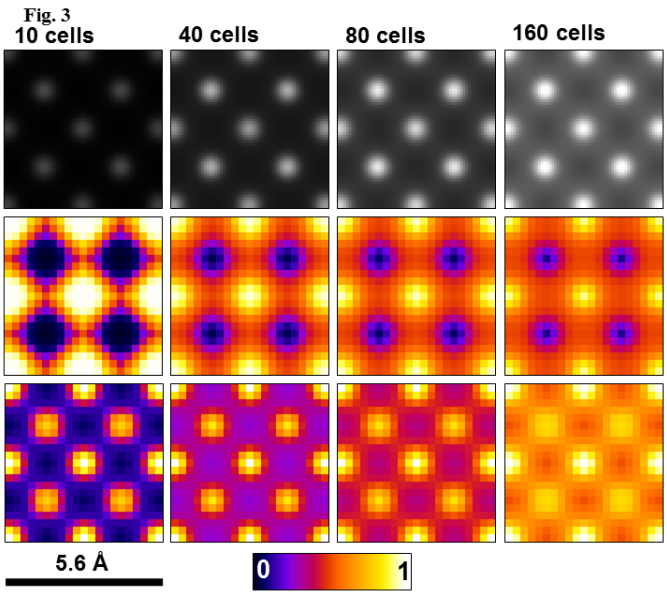


Figure 1: Simulated electron propagation at a Ga column position of [001]-projected GaAs (top) and an extracted intensity distribution (bottom).

Figure 2: A schematic cross-section view of the [001]-projected GaAs, showing individual slices for X-ray spectrum generation.

Figure 3: A set of simulated HAADF images (top), ratio maps of Ga K over total K intensities (middle), and normalized total K intensity maps (bottom) at different thicknesses.