Evaluation of Frozen Phonons Models for Multislice Calculation of TDS

Johannes Biskupek, Andrey Chuvilin, and Ute Kaiser

Electron Microscopy Group of Materials Science, Ulm University, Germany

contact: johannes.biskupek@uni-ulm.de

MOTIVATION

The thermo diffuse scattering (TDS) of electrons results in a diffuse background below the Bragg reflections. Accounting for this background is a key point in quantitative treatment of diffraction data [1], for example: the potential or charge density reconstruction [2]. The recent interest in TDS theories and calculations was caused by a wide spread of high resolution STEM imaging [3], because HAADF contrast is mostly determined by TDS [4]. There are a number of methods developed for the calculation of TDS [5-7] whereas the multislice approach based on the frozen phonons model has been most accurate and physically realistic so far [8]. There are a number of questions related to multislice TDS calculations, which were addressed [9,10], but not fully clarified so far: 1st To what thicknesses multislice calculations are accurate? 2nd Is the Einstein model sufficient or is a more rigorous consideration of collective atoms movement necessary? 3rd Can HOLZ scattering be calculated correctly by the multislice aproach?

In order to shed some more light on these and related issues, we are going to study the effect of phonon vibrations in Si derived from classical Molecular Dynamics (MD) and the influence of the treatment of the z-coordinates on parallel and convergent beam electron diffraction (CBED) patterns as well from high angle centered darkfield images calculated by the multislice approach.

METHODS

Phonon configurations have been calculated using a MD program based on the Tersoff potential [11]. The validity of these models has been proved by comparing phonon spectra to those found in the literature [12]. Models consisting of 3×10^4 atoms ($46\times46\times300$ Å^3) for parallel beam diffraction and high-angle centered darkfield (HACDF) [13] and 2×10^7 atoms ($400\times400\times1500$ Å^3) for CBED have been treated by MD and this scaling shown no influence on resulting phonon spectra. Phonon snapshots have been sequentially obtained from the same model at a time interval exceeding correlation time for atomic vibrations (10ps/snapshot). As scattering potential Doyle-Turner-potential have been used.

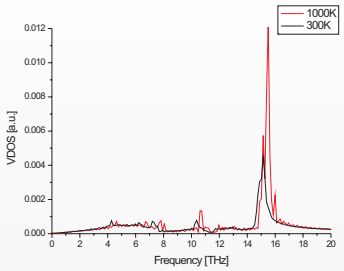

Fig.1. Vibrational density of states [VDOS] calculated by MD for Si at room temperature (300K) and 1000K. Higher temperatures lead to stronger vibrations and a shift towards higher frequencies of the major phonon modes.

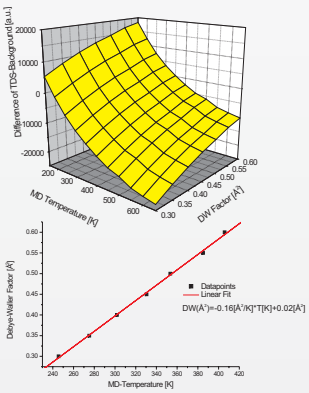

Fig.5 Correspondence between MD-Temperature and Debye-Waller-Factor obtained by comparisons of the intensity of thermal diffuse scattering.

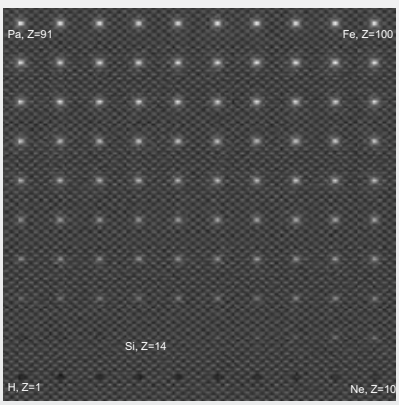

Fig. 7. Calculated Z-contrast HACDF-image of Si[110] with embedded foreign atom columns on Si-atom positions. Starting with Hydrogen H (Z=1) in the bottom left corner over Neon Ne (Z=10) in the bottom right corner to Proactinium Pa (Z=91) top left corner. Doyle-Turner scattering potential and Einstein model to calculate TDS have been used.

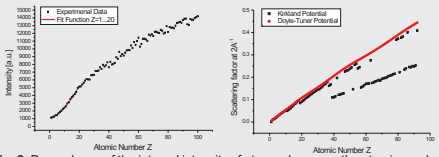

Fig. 8. Dependence of the integral intensity of atom columns on the atomic number (compare Fig. 7). For Z=1...20 a $Z^{1.7}$ -dependence can be shown using the fitting formula *Intensity* = $a+bZ^c$. For Z>20 Doyle-Tuner-scattering-potential is not valid for high angle scattering. TDS of heavy elemens is uncorrectly calculated.

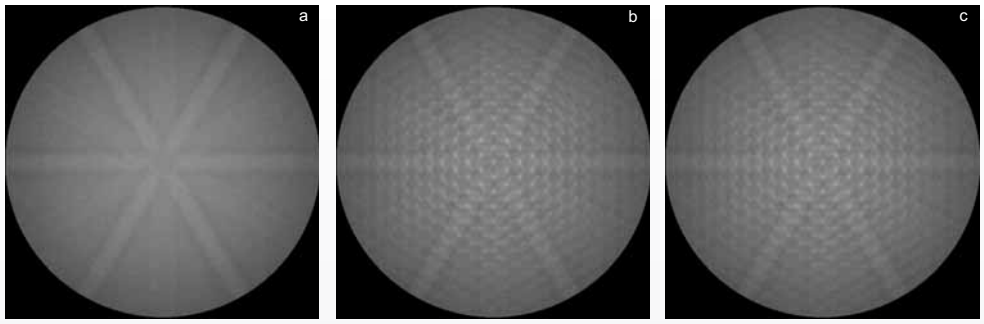

Fig.2. Parallel diffraction patterns of Si[111] with a thickness of 300Å obtained by differen frozen phonons models. (a) Einstein-model, (b) phonons obtained by MD but with not correct treatment of z-coordinates (atoms are aligned back to normal postions along z). (c) MD phonons with correct treatment of z-coordinates. A single slice was calculated for every z-cooridinate up to a precision of 10^{-4}Å. IDifferences to the Einstein-model (a) visible are clearly visible in (b) and (c). The correlated atom vibration are peaking around Bragg-reflections in (b) and (c)

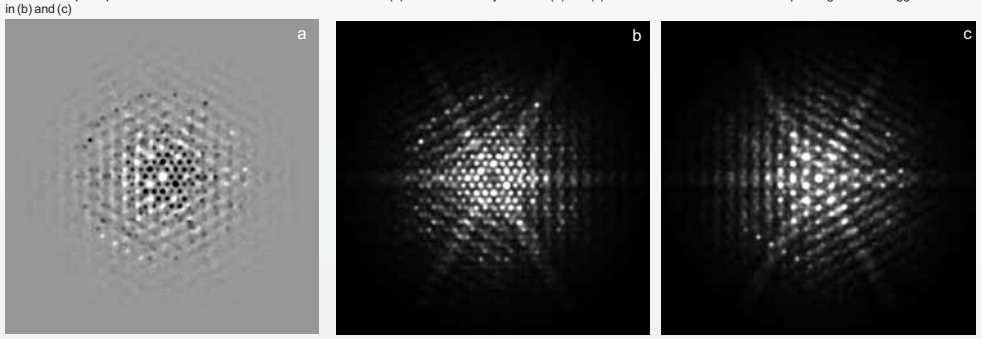

Fig. 4. Quantitative comparison of diffraction pattern Fig.1b and 1c. (a) is showing the simple difference image of Fig. 2b and Fig.2c. There is a difference in intensity of TDS. (b) is showing only the positive part of (a) while (c) is showing the negative part. It can be concluded that threefold symmetry is also present in the diffuse background but suppressed by aligning z-coordinates. Aligning of z-coordinates for the case of correlated atomic vibrations overexcites thermal diffuse scattering into forbidden reflections.

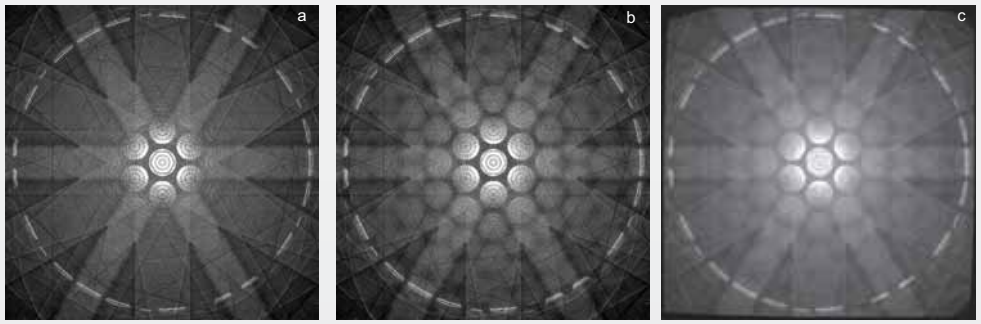

Fig. 6. Covergent electron diffraction pattern (CBED) for Si[111]. (a) Pattern calculated using TDS (Einstein model), (b) Pattern calculated using TDS (MD-phonons) (c) experimental zero-loss filtered (20eV) pattern . In (b) and (c) the correlated vibrations of phonons are visible.

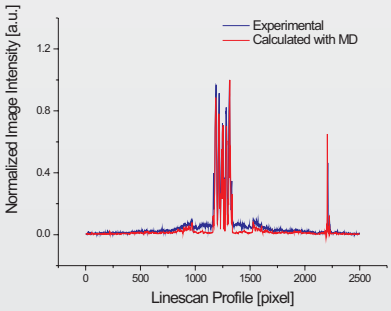

Fig. 9. The linescan profile along 110-reflections is showing the intensity of the experimental image (Fig.6c) and the calculated image (Fig.6b).

Summary

We have shown that image calculations of TDS based on the frozen phonons models can reproduce background intensity correctly if accompanied by MD. It was shown the importance of accounting of atoms shifts in z-direction for TDS calculations. The empirical $Z^{1.7}$ rule for Z-contrast imaging could be reproduced by numerical calculations.

Acknowledgement

We are grateful to Prof. K. Tsuda for providing energy filtered CBED pattern.

References

[1] Allen I. J., Josefsson T. W., Lehmpfuhl G., Uchida Y., Acta Cryst. A 53 (1997) 421-425 [2] Deininger C., Mayer J., Ruehle M., Optik 99 (4) (1995) 135. [3] Voyles P. M., Grazul J. L., Muller D.A., Ultramicroscopy 96 (2003) 251273 [4] Wang Z. L., Acta Cryst. A 51 (1995) 569-585

[5] Ishizuka K., Ultramicroscopy 90 (2002) 7183 [6] Watanabe K., Yamazaki T., Hashimoto I., Shiojiri M., Phys Rev B 64 (2001) 115432 [7] Rossouw C.J., Allen L.J., Findlay S.D., Oxley M.P., Ultramicroscopy 96 (2003) 299312 [8] Findlay S.D., Allen L.J., Oxley M.P., Rossouw C.J., Ultramicroscopy 96 (2003) 6581 [9] Chen J.H., Op de Beeck M., Van Dyck D., Microsc. Microanal. Microstruct. 7 (1996) 27-47 [10] Muller D. A., Edwards B., Kirkland E. J., Silcox J., Ultramicroscopy 86 (2001) 371380 [11] Tersoff J., Phys. Rev. B 39 (1989) 5566-5568 [12] Bernstein N., Mehl M. J., Papaconstantopoulos D.A., Papanicolaou N.I., Bazant M.Z., Kaxiras E., Phys. Rev B 62 (2000) 4477 [13] Kaiser U. & Chuvilin A., Microscopy & Microanalysis 9 (2002) 36