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Evaluation of Frozen Phonons Models for Multislice Calculation of TDS

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The thermal 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 for 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, 6, 7] whereas the multislice approach based on the frozen phonons model has been considered to be 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: to what thicknesses multislice calculations are accurate? Is the Einstein model sufficient or is a more rigorous consideration of collective atoms movement necessary? Can HOLZ scattering be calculated correctly by multislice?

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 calculated by the multislice approach.

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 (46x46x300 $Å^3$) for parallel beam diffraction and 3×10^6 atoms (400 x 400 x 300 $Å^3$) for convergent beam electron diffraction (CBED) have been treated by MD and this scaling showed 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.

The correlation of atomic vibrations in the crystal results in TDS, being strongly peaked around Bragg reflections [13].

Fig. 1 demonstrates that this effect is very pronounced and cannot be neglected; consequently, the Einstein model is not satisfactory if diffraction intensity is to be treated quantitatively. We will show that the application of a classical multislice scheme, where the z-coordinate displacement of the atoms is neglected, leads to an over excitation of forbidden reflections and thus a slicing scheme has to be used, which is able to treat z-coordinates correctly. We also carry out a quantitative comparison of CBED patterns obtained for different phonon models to experimental data, showing the validity and limitations of the multislice calculations.

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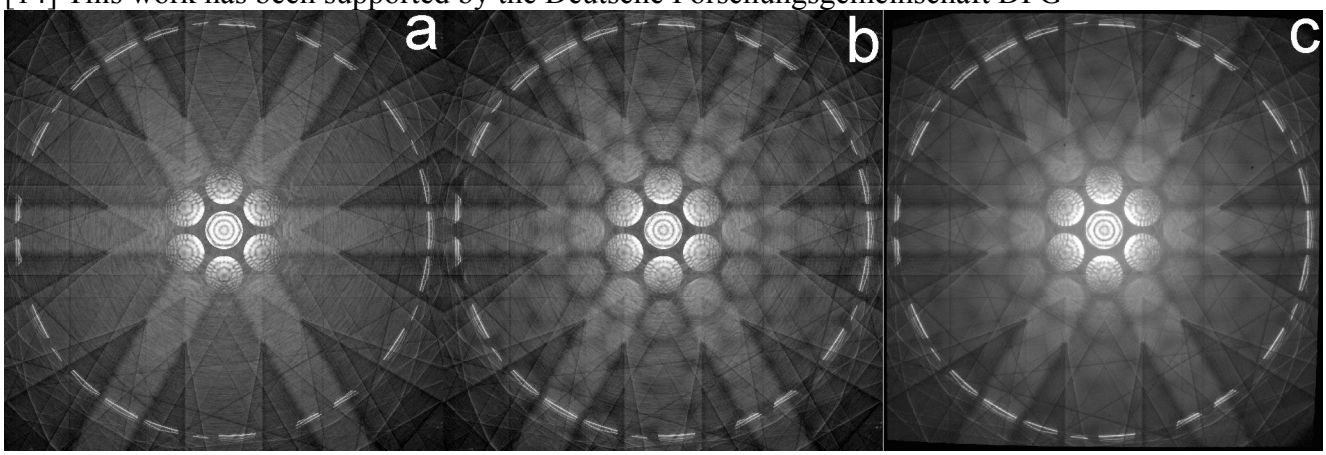

Fig.1. CBED pattern of Si[111], voltage 100.82kV, thickness 164nm
(a) Calculated pattern using uncorrelated frozen phonon configuration calculated by Einstein model
(b) Calculated pattern using correlated frozen phonon configuration calculated by Molecular Dynamics
(c) Experimental pattern (energy filtered, 20eV)