Ultramicroscopy 193 (2018) 1–11

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Dr. Probe: A software for high-resolution STEM image simulation

J. Barthel[a,b,*]

[a] Central Facility for Electron Microscopy, RWTH Aachen University, 52074 Aachen, Germany
[b] Ernst Ruska-Centre (ER-C 2), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany

ARTICLE INFO

Keywords:
STEM
Image simulation
Multislice
Software

ABSTRACT

The Dr. Probe software for multislice simulations of STEM images is introduced, and reference is given of the applied methods. Major program features available with the graphical user interface version are demonstrated by means of a few examples for bright-field and dark-field STEM imaging as well as simulations of diffraction patterns. The numerical procedure applied for the simulation of thermal-diffuse scattering by the frozen-lattice approach is described in detail. Intensity variations occurring in simulations with atomic-column resolution due to frozen-lattice variations are discussed in the context of atom counting. It is found that a significant averaging over many lattice configurations with different random atomic displacements is required to prevent atom-counting bias from simulations. A strategy is developed for the assessment of the amount of required averaging based on the estimated signal variance and the expected signal gain per atom in a column.

1. Introduction

In the recent years high-resolution scanning transmission electron microscopy (STEM) has gained increasing interest with the commercial availability of aberration correctors. With this technique electron probes of below 100 pm size can be generated [1,2], to acquire structural information at a resolution sufficient to separate essentially all atomic distances materials. The very intuitive STEM modes of recording high-angle annular dark-field (HAADF) images providing *Z*-contrast [3] and the imaging of light atoms with annular bright-field (ABF) detectors [4] have attracted many labs world-wide to install and use aberration corrected STEM instruments.

While dark-field and bright-field STEM images can often be interpreted intuitively and directly on a qualitative level, the extraction of quantitative sample information at the atomic scale often requires researchers to reproduce the mechanisms determining the image contrast numerically in the computer. Careful determination of experimental parameters and in particular of the detector sensitivity has been shown to allow a comparison of experiment and simulation on the same absolute intensity scale with atomic column resolution [5]. Image simulations are applied to solve a large variety of problems occurring in local atomic structure analysis where other references are rare, difficult to obtain, or completely absent. Out of many, a few examples are referenced here demonstrating the use of image simulations for the determination of local sample tilt and thickness, the measurement of elemental concentrations in atomic columns, the clarification of atomic arrangements at interfaces, surfaces, and defects, and the analysis of structural disorder and relative column shifts [6-15].

There exist several computer programs implementing numerical image simulations, which differ partially in the applied methodological approaches, in the procedural concepts, and in the accessible computational power. The present paper gives an introduction and overview of the *Dr. Probe* simulation software, which is mainly focused on providing a quick and user-friendly access to quantitative STEM image simulations with commercial desktop computers. Simulations with Dr. Probe concern the imaging of high-energy electron diffraction signals with insignificant energy loss (quasi elastic) at the atomic scale. The applied simulation approach allows the user to quantitatively reproduce experimental data and is implemented in a flexible algorithm keeping the computational costs low without compromising much in terms of simulation quality.

The program is distributed free-of-charge for the academic community via download from website in form of executable object code [17]. The code runs on the central processing unit (CPU) of a computer node and allows spreading the calculation load in parallel over many processors. An intuitive graphical user interface (GUI) as shown in Fig. 1 is provided for Microsoft Windows operating systems to perform STEM image and diffraction simulations. In addition to the GUI, command-line tools are provided for Microsoft Windows, Linux, and Mac OS X allowing versatile, scripted and larger scale computations even across several computer nodes. Besides STEM image simulations, the command-line tools also offer capabilities to calculate high-resolution coherent transmission electron microscopy (TEM) images. However, the introduction given in this paper is meant to provide reference of the

* Corresponding author at: Central Facility for Electron Microscopy, RWTH Aachen University, 52074 Aachen, Germany.
E-mail address: ju.barthel@fz-juelich.de.

https://doi.org/10.1016/j.ultramic.2018.06.003
Received 18 April 2018; Received in revised form 29 May 2018; Accepted 3 June 2018
Available online 04 June 2018
0304-3991/

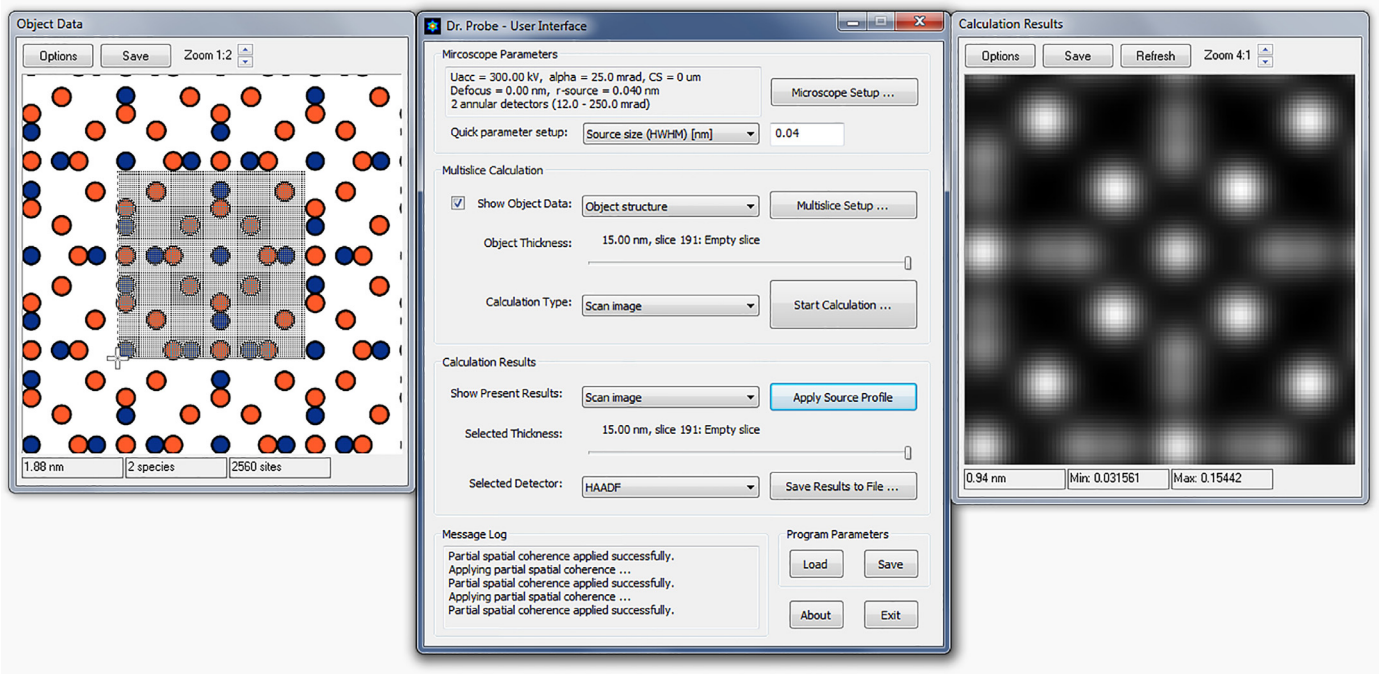

Fig. 1. Screenshot of the Dr. Probe graphical user interface showing, as an example, the structure model of Ru_3Sn_7 [16] in [001] projection and a corresponding HAADF image calculated for an aberration corrected STEM instrument with 80 pm resolution at 300 kV accelerating voltage and for a sample of 15 nm thickness.

particular methods, their implementation, and functionalities available with the graphical user interface for STEM simulations. Additional and more detailed information is presented on the Dr. Probe website [17] which contains an extensive documentation of all software features as well as introductory examples.

2. Methods

STEM image simulations with Dr. Probe apply the multislice method [18] to calculate the quasi-elastic forward scattering of the incident high-energy electron probes by the sample. While scanning an electron probe over positions distributed equidistantly in a rectangular frame, multislice electron-diffraction calculations are performed independently for each position, and the fractions of probe intensity falling into detector areas are registered. Atomic structure models are input to the simulations and provided in form of text lists similar to the CEL format of the EMS software [19] or by CIF structure files [20]. Information is required regarding the input cell dimension, angles, and symmetries together with fractional coordinates, occupancy factors, and thermal vibration parameters for each atomic site. STEM detectors are placed in a diffraction plane as disks for bright-field (BF), and as rings for annular bright-field, and annular dark-field imaging. Azimuthal segments of disks and rings are supported enabling the simulation of differential phase-contrast imaging [21,22]. A radial sensitivity profile may be specified for each detector, allowing a more accurate quantitative comparison between simulation and experiment [23]. With one simulation run, images are calculated for multiple detectors.

Image simulations with the Dr. Probe software have been tested for consistency with other simulation programs. The calculation of projected potentials agrees down to the numerical single-precision level (10^{-6}) with those of μSTEM [24] when using the same atomic form factors of Waasmaier & Kirfel [25]. HAADF STEM image intensities agree on the sub-percent level with μSTEM results also when form factors of Weickenmeier & Kohl [26] are used by Dr. Probe. HR-TEM image simulations with the Dr. Probe command-line tools have been checked to agree on an absolute scale to those of EMS [19] and MacTempas [27].

2.1. Electron-probe formation

Experimental parameters relevant for an image simulation essentially define the shape of the electron probe and how it propagates through an atomic structure. The most important instrumental parameters are

- the kinetic energy eU of the incident electrons, where U is the microscope's accelerating voltage and e is the elementary charge,
- the size of the probe-forming aperture in terms of the semi-convergence angle α,
- coefficients of coherent aberrations of the probe forming lenses (e.g. defocus $C_{1,0}$, two-fold astigmatism $C_{1,2}$, coma $C_{2,1}$, three-fold astigmatism $C_{2,3}$, spherical aberration $C_{3,0}$, etc.), and
- the effective diameter D_0 (FWHM) of the source in the object plane.

The kinetic energy of the incident electron determines its de Broglie wavelength according to the formula

$$\lambda = \frac{c\ h}{\sqrt{eU\,(eU + 2m_0c^2)}}, \tag{1}$$

where c is the speed of light in vacuum, h is Planck's constant, and m_0 is the rest mass of the electron. A convergent electron probe incident along the z axis on a point $\boldsymbol{R} = (x, y)$ of the object plane is calculated in a conjugate reciprocal-space plane with vectors $\boldsymbol{k} = (k_x, k_y)$ according to the expression

$$\psi_0(\boldsymbol{k}; \boldsymbol{R}) = A(\boldsymbol{k})\exp[-i\chi(\boldsymbol{k})]\exp[-2\pi i\ \boldsymbol{k}\cdot\boldsymbol{R}], \tag{2}$$

where $A(\boldsymbol{k})$ is an aperture function, and $\chi(\boldsymbol{k})$ describes the coherent aberrations of the probe. The vector $\boldsymbol{k}$ is the component of the wave vector $\boldsymbol{K}$ perpendicular to the z axis and $\lambda = 1/|\boldsymbol{K}|$. Within the usual approximation for small angles ($\theta \approx \lambda|\boldsymbol{k}| \ll 1$), the parallel component k_z is approximately constant with $k_z \approx |\boldsymbol{K}|$. Respective phase factors $\exp[2\pi i k_z z]$ are omitted in the probe wave function of Eq. (2). The aperture $A(\boldsymbol{k})$ blocks incident electrons with trajectories having angles $\theta > \alpha$ with respect to the z axis, and the aberration function is a polynomial of the form