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Influence of spatial and temporal coherences on atomic resolution high angle annular dark field imaging

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ABSTRACT

Aberration-corrected (scanning) transmission electron microscopy ((S)TEM) has become a widely used technique when information on the chemical composition is sought on an atomic scale. To extract the desired information, complementary simulations of the scattering process are inevitable. Often the partial spatial and temporal coherences are neglected in the simulations, although they can have a huge influence on the high resolution images.

With the example of binary gallium phosphide (GaP) we elucidate the influence of the source size and shape as well as the chromatic aberration on the high angle annular dark field (HAADF) intensity. We achieve a very good quantitative agreement between the frozen phonon simulation and experiment for different sample thicknesses when a Lorentzian source distribution is assumed and the effect of the chromatic aberration is considered. Additionally the influence of amorphous layers introduced by the preparation of the TEM samples is discussed.

Taking into account these parameters, the intensity in the whole unit cell of GaP, i.e. at the positions of the different atomic columns and in the region between them, is described correctly. With the knowledge of the decisive parameters, the determination of the chemical composition of more complex, multinary materials becomes feasible.

1. Introduction

Modern semiconductor devices are usually built from complex compound materials. In the field of III/V semiconductors the binary materials GaN, GaP and GaAs are alloyed with one or more additional elements from the group III or V of the periodic table to meet the demands of the respective application. For example (GaIn)N, Ga(NAsP) and (GaIn)(NAs) can be used for LED, Laser and solar cell applications, respectively [1–5].

In all these materials the exact control of the chemical composition on each sublattice is crucial to achieve the desired functionality. Hence, adequate methods to determine the composition are needed, as e.g. conventionally used X-ray diffraction alone is not applicable anymore for materials consisting of more than three constituents. Moreover, its lateral resolution is limited and local fluctuations in composition cannot be detected.

In this task, (scanning) transmission electron microscopy ((S)TEM) has proven as a valuable tool to deliver the desired information. Due to the intuitive interpretation in terms of Z contrast, the high angle annular dark field (HAADF) technique is frequently applied [6]. In the last decade sub-angstrom resolution became possible due to the introduction of aberration correctors [7,8]. Nevertheless, the absolute composition of a sample is not always derivable directly in both low and high resolution imaging. This makes an adequate simulation of the scattering process inevitable. Here the multi slice algorithm in the frozen phonon (FP) approximation has shown to reproduce the experimental data very accurately [9].

By normalizing the experimental data to intensity of the impinging beam, e.g. via a detector scan [10], it is possible to compare simulations and experiment on the same intensity scale. Taking into account the sensitivity of the used detector quantitative results could be derived for various material systems [11–13].

Nevertheless, when high resolution information is sought, some additional challenges need to be faced. Depending on the studied specimen and its thickness, cross scattering from neighboring columns occurs and the measured HAADF intensity does not necessarily reflect the chemical composition at a certain position [14,15]. Moreover, amorphous layers due to sample preparation become increasingly important in the thin samples investigated in high-resolution STEM (HRSTEM) [16].

Most importantly, the resolution and especially the contrast are higher in the simulated than in the experimental data, even if the present geometric aberrations are included. This discrepancy is

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caused by the partial spatial and temporal coherence of the electrons. The partial temporal coherence is mainly caused by the chromatic aberration and energy spread of the electrons in the TEM [17]. The influence of the chromatic aberration on the STEM has been investigated for example in [18,19].

The spatial coherence is related to the source size [20], i.e. the fact that the real electron source is not a point like emitter but has a finite dimension. The effect of the partial spatial coherence can be treated by convoluting the simulated images with an effective source distribution; see e.g. [21–23]. The shape of the source distribution is discussed controversially in the literature, as Gaussian, Lorentzian or combinations of both are used [21,24]. Nevertheless, the exact shape can have a huge impact on the appearance of the HAADF images and the application of a wrong distribution can lead to a wrong quantification of the chemical composition at an atomic scale.

If all the influencing parameters are accounted for in the simulation, the question remains how to compare simulated and experimental results with each other? Often only qualitative comparisons are carried out or intensity profiles are compared to simulations. In other approaches, some of the spatially resolved information available in a HRSTEM image is neglected by averaging the experimental intensity over a certain region, e.g. the Voronoi cell of an atomic column [25–27].

In this contribution we present a method to derive quantitative two-dimensional data, i.e. a representative unit cell of the investigated crystal, from experimental images, which can be compared to complementary simulations. We show the influence of the chromatic aberration and the choice of the source distribution on the simulated HAADF intensity. By comparing simulated and experimental data quantitatively for different sample thicknesses we deduce which source distribution has to be applied to the simulations to reproduce the experimental data correctly. Furthermore, we investigate how the presence of amorphous layers due to sample preparation affects the drawn conclusions. Although the influences of temporal coherence, spatial coherence and amorphous layers on STEM investigations have been investigated individually before, the interplay of these effects has not been studied yet. With the correct consideration of all of them we achieve excellent agreement throughout the whole two-dimensional unit cell.

We chose binary GaP as model material because of its inherently fixed stoichiometric composition. Moreover, because of the relative big difference in atomic number of the two constituents, the contrast within a unit cell is higher than for materials consisting of elements with similar atomic number like for example GaAs. Therefore, assumptions in the simulation can be checked and amended.

The conclusions drawn from the simple GaP system can then be used for future evaluation of more complex ternary or multinary materials.

2. Material and methods

Commercially available GaP wafers (Crystec) were used for this study. Electron transparent samples were prepared by conventional mechanical grinding and consecutive argon ion milling using the Gatan PIPS. The incident angle of the ions was chosen to 4° for bottom and top side, resulting in a wedge-shaped sample. The final ion energy was reduced to 1.7 kV in order to minimize the thickness of the amorphous layers at the sample surfaces introduced by the ion bombardment. Please note that the final ion energy of 1.7 kV is not suitable to create a sample fully free from amorphous layers, this energy was rather chosen to create a layer with a defined thickness to investigate its influence on the HAADF intensity. A more detailed description of the applied sample preparation and the resulting (sub-) surface damage is given in [28]. The GaP [010] zone axis was chosen as viewing direction because in this projection the group III and group V sub-lattice exhibit the largest spatial separation of around 0.19 nm.

The STEM investigation was carried out in a double C_S-corrected JEOL JEM 2200FS operating at 200 kV. A condenser aperture of 24 mrad and a detector range of 73–173 mrad were chosen for the HAADF measurements. Atomic-resolution image series were acquired at different positions of the sample exhibiting different TEM sample thicknesses. The individual images of the series were aligned non-rigidly utilizing the 'Smart align'-software [29] in order to reduce the influence of experimental noise and scan distortions on the images. The aligned images were normalized with respect to the impinging beam following the approach described in [30] to be able to compare experiment and simulation on an absolute intensity scale. The radial sensitivity of the used HAADF detector was determined via a detector scan [10] and taken into account.

Complementary image simulations were carried out utilizing the frozen phonon approximation of the STEMSIM code [31]. A residual spherical aberration of 2 μm was measured during the experiments with the help of the corrector software and taken into account in the simulations. A value of 5 mm was assumed for the fifth order spherical aberration coefficient C_5. Additionally the influence of the chromatic aberration was taken into account by summing over a simulated defocus series with 7 different defocus values. The weight of each defocus was determined by a Gaussian function with a full width half maximum (FWHM) of Δz_{CC}, which is connected to the chromatic aberration coefficient C_C by [18]

$$\Delta z_{CC}=2C_c\frac{dE}{E_0}\sqrt{(2log2)}. \quad (1)$$

For the JEOL JEM 2200FS microscope with a C_C of 1.5 mm and a dE of 0.42 eV follows a Δz_{CC} of 7.5 nm at 200 kV acceleration voltage. For each defocus 10 phonon configurations were used. The total number of 70 phonon configurations showed to be a good trade-off between the convergence of the HAADF intensity and the simulation time.

The supercell for the intensity simulations consisted of 5×5 GaP unit cells in the *x*-*y*-plane and 92 unit cells in the z direction corresponding to a final thickness of approximately 50 nm.

Additionally, the intensity of amorphous GaP was simulated. Here a supercell of the same dimensions as for crystalline case was derived by the software to simulate the atomic packing in Ideal Amorphous Solids (IAS) [32–34]. This software generates the spherical amorphous structure that satisfies the basic requirements for amorphous solids. It contains none of the elements of symmetry such as mirror, rotation or glide and retains the stoichiometry. The density of amorphous material was adjusted by uniform deformation of IAS sphere and was chosen to 4.94 10^{22} atoms/cm^3, i.e. the same value as for the crystalline GaP. Finally, the supercell of desired rectangular geometry was cut from generated IAS sphere. More realistic models for amorphous layers require very complicated and time-consumable calculations in frames of Molecular Dynamic (MD) simulations [35]. Furthermore, the resulting structure is extremely sensitive to the initial conditions such as the initial temperature and the quenching rate. We believe that the theory of IAS is a sufficient approach for adequate theoretical interpretation of STEM images obtained in our experimental studies. Experimental and simulated data were compared utilizing MATLAB scripts as will be discussed in the result sections.