Measurement of structure factors by parallel and convergent beam electron nanodiffraction

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A quantitative understanding of transmission electron microscopic (TEM) images usually involves simulations as a central part. In the TEM community, the Bloch wave approach is frequently applied to simulate diffracted beam amplitudes in crystalline specimen. For example, the CELFA [1] technique compares the composition-dependent contrast in experimental (200) lattice fringe images with Bloch wave simulations to derive the local chemical composition x in $In_xGa_{1-x}As$ alloys. However, prior knowledge on the scattering properties of a crystal enters the Bloch wave method by means of structure factors (SF). Commonly, SF are calculated from isolated atom scattering data [2], so that the effect of a redistribution of electrons due to chemical bonding is neglected. Recent theoretical approaches overcome this problem by defining modified atomic scattering amplitudes (MASA [3]) which are calculated by density functional theory (DFT) methods.

In this work, we introduce a new technique to measure SF from TEM spot diffraction patterns acquired under parallel illumination. Starting with the SF for isolated atoms, Bloch wave refinement routines have been developed to minimize the R-value defined by

$$R = \sum_m \frac{(I_m^E - s \cdot I_m^S)^2}{\sum_m (I_m^E)^2} .$$

Here, the superscripts S and E denote simulated and experimental beam intensities, respectively, s is used to scale the simulation with respect to the experiment and can be included in the refinement. Nonlinear least-squares routines are used to find the minimum of R via variation of SF, specimen thickness and –orientation, as well as Debye parameters. As experimental diffraction patterns contain thermal diffuse (TD) background, a background subtraction is performed using the program GREED [4] for each Bragg spot.

Due to plane wave illumination, our method is highly efficient in computational respect, so that a conventional PC is usually sufficient. Moreover, the use of spot diffraction patterns allows for a study of large structures where adjacent reflections are located close to each other. For the refinement of atom positions, these circumstances have motivated the development of the program package ELSTRU by Jansen et al. [4].

First, to estimate inherent errors of the parallel beam electron diffraction (PBED) method, test diffraction patterns for GaAs in [053] zone axis have been simulated in frozen lattice approximation (thus they contain TD background) using the STEMSIM [5] program. To account for a slight misalignment of the microscope, a small beam convergence of 0.5mrad has been applied. As we work with FIB-fabricated condenser apertures of 5-10μm in

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diameter in order to restrict the probe diameter to a few nanometers in experiment, a Bessel-shaped probe with a full width at half maximum of 10nm was used in the simulations. All test patterns have then been propagated through the background subtraction and the refinement of the thickness, the orientation and the 200 SF. Comparison with the input for the frozen lattice simulations yields an accuracy of the SF refinement better than 0.02nm^{-2}. Furthermore, we derived rules to identify the correct initial guess for the specimen thickness entering the refinement using the simulated test patterns.

Second, we applied our method to 6 experimental GaAs [053] PBED patterns recorded unfiltered on imaging plates and 3 zero loss filtered patterns recorded with a Gatan imaging Filter. All patterns were first treated separately in the refinement of the 200 SF, the thickness and the orientation, resulting in an average of –0.252±0.019nm^{-2} (unrelativistic). No evidence for the need of energy filtering could be observed, which is an advantage from the experimental point of view. In a simultaneous refinement using all 9 data sets at the same time, Debye parameters have additionally been refined to B_{Ga}=0.65Å^2 and B_{As}=0.56Å^2.

Third, 7 convergent beam electron diffraction measurements have been performed for GaAs in the same zone axis due to the method of Tsuda et al. [6] to allow for a methodological comparison with a traditional method. The average refinement result for the 200 SF of –0.273±0.018nm^{-2} does not only confirm the PBED value, but also exhibits nearly the same precision. As in the PBED case, the refined 200 SF is about 30% larger than the isolated atom value but agrees with the DFT value from [3].

Finally, PBED measurements of the 200 SF for InAs have been performed with a mean result of 1.019±0.014nm^{-2}. Figure 1 shows the individual refinement results together with the R-value. Average and standard deviations are shown as solid and dashed lines, respectively. The encircled plus indicates the constraint SF refinement using all 8 data sets. It can be seen that our measurement is, as in the case of GaAs, in agreement with the MASA concept (solid triangle) and that the isolated atom value (light triangle) must be rejected.

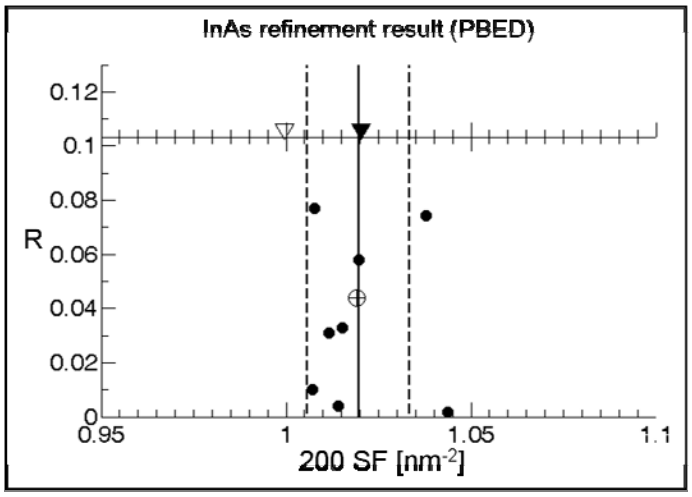

Figure 1. Refinement results for the 200 structure factor of InAs. Black circles indicate results of single refinements, the encircled plus sign is the result of a constraint refinement using all 8 data sets. It fall together with the average result indicated by the solid black line. The light and filled triangle corresponds to theoretical data from [2] and [3], respectively.

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