Characterization of Nanostructured Interfaces

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Quantitative HRTEM investigation of an obtuse angle dislocation reaction in gold with a C_S corrected field emission microscope

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ABSTRACT

We investigate quantitatively the non periodic arrangement of atom columns surrounding an obtuse angle dislocation reaction in gold utilizing a CM200 FEG instrument equipped with a spherical aberration corrector. The in-plane component of the Burgers vector of the observed stair-rod dislocation is $\frac{1}{6}[110]$. Column positions are determined from single lattice images and compared with those from a reconstructed electron exit wave. We find that absolute position measurements of 1-3 pm require knowledge of the defocus to better than 1 nm which can be achieved by a reconstruction of the exit-plane wave. In contrast, a defocus value of 8.9 nm already leads to apparent displacements as large as 35 pm if single lattice images are considered. Such discrepancies are either caused by residual lens aberrations or by the superposition of delocalization effects at interfaces caused by defocusing of the objective lens. Commonly, however, only relative displacements are of interest. In this case the C_S corrector improves the interpretability of single defocused lattice images with a remarkable signal to noise ratio which is advantageous for in-situ experiments. As an example for analyzing in-situ experiments we determine displacements recorded in a time resolved experiment of radiation induced atom motion on surfaces.

INTRODUCTION

A quantitative investigation of dislocations and boundaries in crystalline solids often relies on a quantitative determination of atomic positions from lattice images. To date, high-resolution transmission electron microscopes that are equipped with a field emission source complicate the interpretation of single lattice images since they hold misphased information extending far beyond the Scherzer point resolution. Typically, this information can be accessed by a reconstruction of the electron exit-plane wave function from a focal series of lattice images [1,2]. Sub-Angstrom resolution can be obtained this way [3,4]. Another alternative consists of imaging with a C_S corrected microscope [5]. This approach allows for a direct comparison of single lattice images with the reconstructed electron exit wave since the contrast transfer function of such a microscope does not show any oscillations and ideally the point resolution extends up to the information limit [5]. Therefore, the interpretability of a single lattice image from a C_S corrected microscope is largely facilitated. In our study we utilized a gold [110] specimen to investigate the core structure of an obtuse angle dislocation and a surface reconstruction that varies with time. We compare results from single lattice images with those obtained by a reconstruction of the exit-plane wave in terms of the precision as to which absolute and relative column positions can be determined. Finally, it is demonstrated that in-situ experiments can largely benefit from a C_S correction.

EXPERIMENTAL DETAILS

For the TEM investigation a Philips CM200 (200kV) field emission gun (FEG) microscope with a spherical aberration (C_S) corrector was used that exhibits an information limit of ~ 0.13 nm [5]. The actual measured value $C_S = 2.7 \pm 2.4$ µm is orders of magnitude smaller than typical spherical aberration constants of about 1mm. Focal series of 20 lattice images were taken from the defocus interval 30 nm through –30 nm with typical steps of 3 nm between successive images. Other lens aberrations up to third order were on-line corrected prior each experiment exploiting Thon rings [6] to typical values of 59±76 nm for 2-fold astigmatism, 2.8±2.3 µm for 3-fold astigmatism and 51±39 nm for coma. The complex electron exit-plane wave was recovered from a focal-series of lattice images by the PAM/MAL-algorithm of the Philips/Brite-Euram software package [1,2]. Thus, a structure determination from single lattice images of the series can be directly compared with results obtained from the phase image of the exit-plane wave. A sample of gold in [110] zone axis orientation was investigated imaging an obtuse angle dislocation reaction. In-situ experiments were performed by recording a time series of lattice images at constant defocus instead of a focal series. Other details of the sample preparation procedure are published elsewhere [7]. To extract the position of the atomic columns from HRTEM images with pm precision we utilized the DARIP software package [8,9].

RESULTS

Figure 1 is a phase image of the electron exit-plane wave function of the gold sample in [110] zone axis orientation reconstructed from a focal-series of 20 HRTEM lattice images. The correction of the lens aberration leads to a superior signal-to-noise (S/N) ratio in single lattice images and resultantly in the reconstructed exit-plane wave phase image that is shown. Three stacking faults (marked with white lines, two on the left image side and one on the bottom) intersect to form an obtuse angle stair-rod dislocation in gold. The dislocation core is hollow. Its enlarged view in figure 2a was used to extract the column positions surrounding the dislocation core from intensity maxima (in pixels of the CCD camera, see figure 2b). Since maxima of the phase image coincide with the position of atomic columns, the procedure gives absolute column position to typically 2-3 pm of accuracy (σ value). Here the largest contribution comes from residual image distortions that reduce if identical areas in successive lattice images are considered. From the application of a Burgers circuit, the in-plane component of the Burgers vector is found as $\frac{1}{6}[110]$. However, since several partial dislocations are involved to produce this stair-rod dislocation and since a projection into the image plane is involved, an identification of the individual reaction components is not unique.

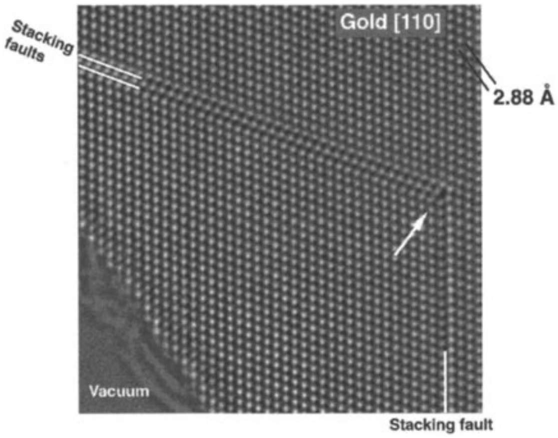

Figure 1. Phase image of the reconstructed exit-plane wave function from a focal-series of HRTEM lattice images of an gold wedge shaped specimen in [110] zone axis orientation, showing the stacking faults (white lines) and the core (arrow) of the obtuse angle stair-rod dislocation.

A direct interpretation of the reconstructed phase image is always possible since phase maxima mark the position of atomic columns. If over–focused lattice images are recorded, intensity maxima can also refer to column positions (white atom lattice images). In the C_S corrected microscope only a few nanometers of overfocus is required to produce such a lattice image and it is usually recorded in a focal series. The reconstructed phase image, on the other hand, can be intentionally defocused by its propagation through vacuum onto another exit plane [10]. This procedure is typically used to find the proper projection plane of zero defocus. Thus, it is feasible to study the effect of a finite defocus change on the determination of atomic positions from experimental images. This is done next.

Figure 3a shows the overlay of column positions extracted from a properly focused electron exit wave $\Delta f = 0 \pm 0.5$ nm with those recovered from an over-focused ($\Delta f = 8.9$nm) lattice image where atomic columns appear bright.

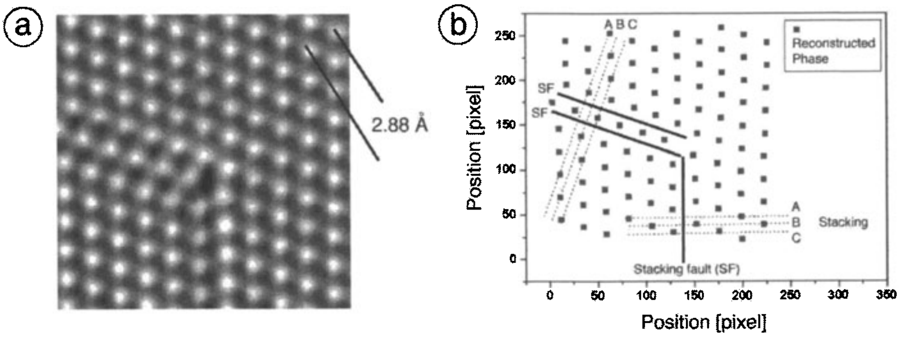

Figure 2. a) Enlarged view of figure 1 showing the phase image of the reconstructed exit-plane wave function of the specific dislocation core in gold, b) Plot showing the column positions around the dislocation core structure as recovered from intensity maxima in a). Typical errors of individual measurements are 2-3 pm if image distortions over a distance of 25 nm are included.

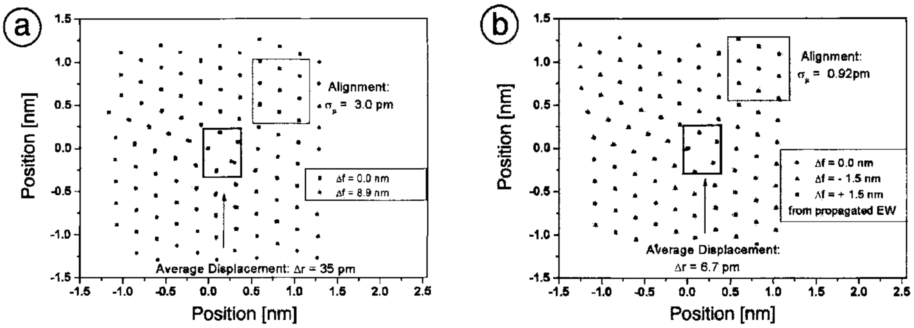

Figure 3. A comparison of the extracted column position from a) a defocused C_S corrected lattice image ($\Delta f = +8.9$ nm) with the properly reconstructed phase image of electron exit wave and b) defocused ($\Delta f = -1.5$nm and $+1.5$nm) phase images with the properly reconstructed phase image of electron exit wave. Images were aligned in the marked areas to the indicated precision of 3 and 0.9 pm respectively.

The patterns were aligned in the upper right corner of the image (indicated rectangle in figure 3) and in this region deviations were minimized to a smallest 3 pm residual value of mean deviations. In spite of the precision of this alignment, the overlay reveals an apparent displacement of 35 pm for column positions surrounding the dislocation core (squared in the center of figure 3a). Thus, due to contrast delocalization, defocus values below 10 nm already introduce substantial systematic errors if non periodic structures at interfaces are considered. In order to give an estimate about the precision of as to which an even smaller uncertainty in defocus determination affects a position measurement we propagated the electron exit wave from $\Delta f = 0$ to $\Delta f = -1.5$nm and $+1.5$nm and repeated the analyses as shown in figure 3b. Again the patterns were aligned in the upper right corner and an even better alignment precision of 1pm could be achieved. In the non periodic region of the dislocation core, however, apparent displacements up to 7 pm were measured.

Figure 4 summarizes the results obtained from the comparisons between defocused HRTEM

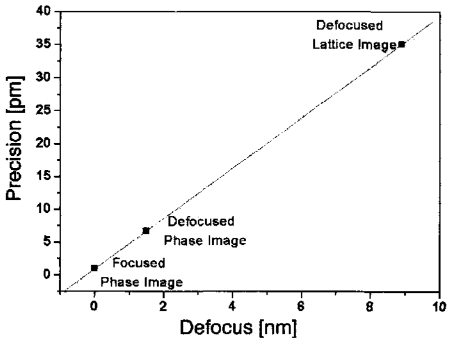

Figure 4. Defocus dependence of absolute position measurements from lattice images and reconstructed electron exit waves.

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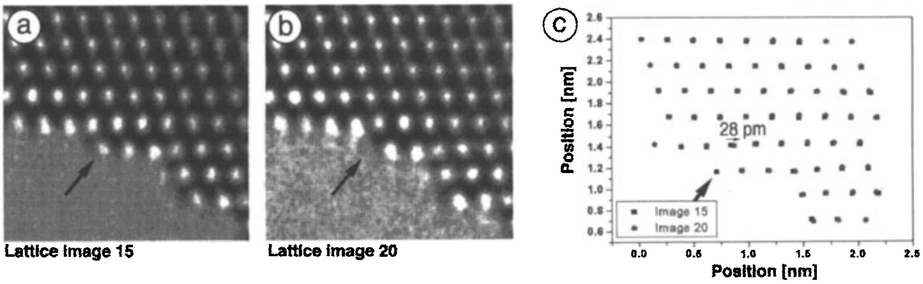

Figure 5. a) C_S corrected HRTEM lattice image #15 and b) C_S corrected HRTEM lattice image #20 recorded at the same defocus Δf= 9 nm 20 seconds later. c) Plot of atomic positions measured from a) and in b) showing the re arrangement of atoms before and after evaporation of a column.

lattice images and phase images of propagated exit-plane wave functions. It is seen that for small defocus variations the delocalization-induced systematic errors in the precision for the determination of column positions increase linearly with defocus. Consequently, an absolute position determination with a precision of 1-3 pm requires knowledge of the defocus to better than 1 nm. In particular we note that sample surfaces have to be flat to better than 1 nm if it is attempted to recover absolute column positions from non periodic objects to a precision of 1-3 pm. On the other hand it often suffices to determine relative displacements. Such situations typically occur if a time series of lattice images is recorded at constant defocus as it is the case in in-situ experiments.

In figure 5 we show two lattice images of a time series (figure 5a: image number 15, and figure 5b: image number 20) that were recorded with a time delay of 20 seconds. During this time interval the radiation of the electron beam leads to an evaporation of a gold column at the surface that is marked by an arrow in figure 5. As a result the surface columns rearrange in the surface region. In Figure 5c we compare the extracted column positions recorded before and after the evaporation of a single column. The comparison shows that one particular atom column changes its position by 28 pm in order to reduce surface energies. Apparent column positions are determined to a precision of about 3pm. The same image alignment procedure was used than in Figure 3 and the defocus is chosen such that bright spots mark column positions.

It is seen in this experiment that a relative determination of atom position suffices to observe the relevant displacements. A transformation of relative values into absolute positions, however, requires a comparison with image simulations to account for defocus settings and sample thickness since single lattice images are interferograms that only in simple structures may depict the crystal structure directly.

CONCLUSIONS

Using the TEM CM200 FEG equipped with a C_S corrector, single HRTEM lattice images are compared with phase images of the reconstructed electron exit wave function from a stair rod dislocation in gold with an in-plane Burgers vector of $\frac{1}{6}[110]$. Since column positions surrounding the dislocation core are arranged non periodically we utilized this structure to study the effect of defocus setting on a quantitative determination of absolute and relative column positions. We find that an absolute precision of ~ 3 pm can be achieved for the extraction of individual column positions which is made possible by the extraordinary S/N ratio in the lattice images from the aberration corrected microscope. Moreover, we find that a precision of 1-3 pm can only be achieved if the defocus can be set to better than 1nm. A determination of the proper zero defocus value may require the accumulation of focal series since individual lattice images may not be recorded at this specific defocus setting. Consequently, we find severe demands for sample preparation since surfaces need to be flat to better than 1 nm.

It has already been pointed out that C_S correction facilitates the orientation of samples along zone axes since the beam can be tilted more precise than a stage without introducing aberrations [5]. Here, we report that the signal to noise ratios can be high enough to access single column positions on a picometer level. If the determination of relative column positions is of relevance, only, the C_S correction facilitates the interpretation of single lattice images in addition. Thus, we find that a C_S correction offers advantages for in-situ experimentation and demonstrated this advantage by analyzing the rearrangement of gold columns on surfaces upon evaporation of a single atomic column.

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