High-resolution imaging with an aberration-corrected transmission electron microscope

M. Lentzen*, B. Jahnne, C.L. Jia, A. Thust, K. Tillmann, K. Urban

Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany

Received 11 July 2001; received in revised form 28 January 2002

Abstract

Recently an electromagnetic hexapole system for the correction of the spherical aberration of the objective lens of a 200 kV transmission electron microscope has been constructed by Haider and coworkers. By appropriately exciting the hexapole elements it is possible to adjust specific values of the spherical aberration coefficient ranging from the value of the original uncorrected instrument over zero even to negative values. In the first part of the paper the consequences of the tunable spherical aberration are investigated. New imaging modes are available: By adjustment of an optimum value for the spherical-aberration coefficient, the point resolution of phase-contrast imaging can be extended to the information limit. Phase-contrast imaging can be improved by a reduced level of contrast delocalisation. For zero aberration contrast delocalisation does not occur. In this case high-resolution investigations are carried out under amplitude-contrast conditions, where the local image intensity of crystalline objects is controlled by electron diffraction channelling. The defocus and spherical aberration values related to the new imaging modes are given. In the second part novel applications of the instrument to semiconductor heterostructures and ceramic grain boundaries are examined. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Transmission electron microscopy; High-resolution imaging; Aberration correction; Spherical aberration; Delocalisation

1. Introduction

In transmission electron microscopy, high-resolution imaging of smallest object structures is hampered by the influence of aberrations of the lens system, in particular the strong spherical aberration of the objective lens. As a result of spherical aberration, the image intensity originating from an object point is spread over an area whose radius is dependent on the spherical aberration and the defocus, and whose minimum radius is proportional to the spherical aberration coefficient. This so-called contrast delocalisation is particularly strong for microscopes equipped with a field-emission electron gun providing high spatial coherence of the illumination system [1,2].

Recently following a suggestion by Rose [3] a double-hexapole system was constructed by Haider et al. [4] which permits to correct the spherical aberration of the objective lens. This corrector was adapted to a Philips CM 200 FEG ST electron microscope equipped with a field emission gun. First results on the optical alignment and of the performance of this new instrument were reported in Refs. [5–7].

*Corresponding author.
By appropriately exciting the hexapole elements, it is possible to adjust specific values of the spherical aberration coefficient of the combined system consisting of objective lens and corrector element, which range between the aberration value of the original uncorrected instrument over zero towards negative values. In the first part of the present paper, we briefly explore the consequences of an adjustable spherical aberration for contrast transfer as well as for the conditions under which experimental high-resolution investigations are carried out. In the second part, some new experimental results obtained in high-resolution materials science applications are presented.

2. Aspects of contrast theory

In transmission electron microscopy with electrons of a given wavelength \( \lambda \) and a fixed spherical aberration coefficient of the objective lens \( C_s \), different modes of contrast transfer can be achieved by proper adjustment of the lens defocus value \( Z \). In particular, three settings of \( Z \) are important:

(A) Scherzer’s defocus: In this case the defocus value is given by [8]

\[
Z_{\text{Sch}} = -\sqrt{\frac{1}{2}} C_s \lambda \tag{1}
\]

and the phase contrast of a weak-phase object is maximised, up to a point resolution characterised by the maximum spatial frequency

\[
g_{\text{Sch}} = \frac{1}{\sqrt{\frac{\pi}{16} C_s \lambda^3}} \tag{2}
\]

(B) Lichte’s defocus of least confusion: The contrast delocalisation is quantified by the radius of the point-spread function [9]

\[
R = \max \left| \frac{\partial \hat{g}}{\partial g} \right|, \quad g \in [0, g_{\text{max}}] \tag{3}
\]

where \( g_{\text{max}} \) is the information limit and \( \hat{g} \) is the aberration function given by

\[
\hat{g}(\bar{g}) = \frac{1}{2} Z \lambda \bar{g}^2 + [1 - \cos(2\pi \bar{g})] \tag{4}
\]

As shown by Lichte [1] setting the defocus to

\[
Z_L = -\frac{1}{2} C_s \lambda g_{\text{max}} \tag{5}
\]

minimises contrast delocalisation to a value given by

\[
R_L = \frac{1}{4} C_s \lambda^3 g_{\text{max}}^3 \tag{6}
\]

(C) Minimum phase-contrast defocus: For a defocus value given by

\[
Z_{\text{min}} = -\sqrt{\frac{1}{4} C_s \lambda} \tag{7}
\]

phase contrast reaches a minimum [9] and at the same time amplitude contrast is maximised up to a spatial frequency

\[
g_{\text{min}} = \frac{1}{\sqrt{\frac{\pi}{16} C_s \lambda^3}} \tag{8}
\]

In the conventional transmission electron microscope, due to the strong spherical aberration of the objective lens, none of these special defocus settings is ideal. Scherzer’s defocus allows to image a wide band of space frequencies at nearly the zero-phase shift. However, the point resolution obtained at Scherzer’s defocus is lower than the information limit \( g_{\text{max}} \) which is limited by spatial and temporal coherence of the electron source. Up to the point resolution given by Eq. (2) the delocalisation amounts to \( R_{\text{Sch}} = \frac{1}{4} g_{\text{Sch}} \). At Lichte’s defocus (5) the reduced contrast delocalisation is obtained at the cost of the great disadvantage that the phase contrast transfer function, given by \( -\sin(2\pi \bar{g}) \), exhibits a large number of rapid contrast oscillations at medium spatial frequencies, and the corresponding contrast reversals make the resulting image very difficult to interpret. At the minimum-contrast setting (7) the defocus aberration and the spherical aberration do not cancel properly at higher spatial frequencies, already well below the point resolution. With the new degree of freedom offered in the new instrument by the adjustable spherical aberration coefficient it is worth while to reconsider the above conditions.

For exact compensation of the aberration of the objective lens, \( C_s \), and \( Z = 0 \), and according to Eq. (6) contrast delocalisation vanishes. The phase contrast transfer function (4) adopts the value 0 while the amplitude-contrast transfer function, given by \( \cos(2\pi \bar{g}) \), adopts the maximum value 1. Under these conditions electron microscopy is carried out under pure amplitude-contrast
conditions. High-resolution imaging under amplitude-contrast conditions has hardly been discussed in the literature since this imaging mode is not available in the standard uncorrected electron microscope. For the focus of minimum phase and maximum amplitude contrast (7), much of the resolution of the microscope is wasted if an objective aperture is used which excludes spatial frequencies higher than the maximum value (8). Under amplitude-contrast conditions the image intensity \( I(\vec{r}) \) is given by

\[
I(\vec{r}) = |\psi(\vec{r}) \ast p(\vec{r})|^2
\]

with \( \psi(\vec{r}) \), the exit plane wave function and \( p(\vec{r}) \), the point-spread function of the objective aperture or the virtual aperture originating from the partial coherence of the illuminating electron beam. Fig. 1 shows a map of simulated through-focus and through-thickness series of images of Ge [1 1 0] calculated for \( C_\text{s} = 0 \), a wavelength of 2.5 pm, and an information limit of 7.3 nm\(^{-1}\). The images at the Gaussian focus, \( Z = 0 \), exhibit a local intensity distribution originating from electron diffraction channelling [10–12]. The image contrast is pronounced at a thickness of 7.5 nm, which is half the extinction distance \( \xi \) of the two strongest Bloch waves of Ge [1 1 0]. Under these conditions and for specimen thicknesses which are odd multiples of \( \xi/2 \), the electrons are strongly localised at the atom positions and the location of the intensity maxima directly represents the projected atom column structure.

For \( C_\text{s} = 0 \) phase contrast can still occur, due to the defocus aberration term in Eq. (4). In particular, the defocus value

\[
Z_0 = -\frac{1}{2\lambda g_{\text{max}}^2}
\]

provides a pass band up to the information limit \( g_{\text{max}} \), at the expense of a delocalisation \( R_0 = 1/g_{\text{max}} \). Taking advantage of the tunable spherical-aberration coefficient we can, according to Eq. (2), extend the point resolution at Scherzer’s defocus by reducing \( C_\text{s} \). However, it is not reasonable to increase \( g_{\text{Sch}} \) beyond the value of \( g_{\text{max}} \). This defines the following value of the spherical aberration constant at which \( g_{\text{Sch}} = g_{\text{max}} \):

\[
\frac{g_{\text{Sch}}}{g_{\text{max}}} = \frac{16}{3} \frac{1}{\lambda^2 g_{\text{max}}^2}
\]

and the appertaining defocus

\[
Z_{\text{Sch, ext}} = \frac{8}{3} \frac{1}{\lambda^2 g_{\text{max}}^2}
\]

This setting allows phase contrast taking advantage of the higher point resolution possible with

Fig. 1. Through-focus and through-thickness series of images of Ge [1 1 0] for \( C_\text{s} = 0 \). The defocus runs from 0 to –25 nm, from left to right; the thickness runs from 1.5 to 15.0 nm, from top to bottom.
reduced $C_S$, however, at the cost of a contrast
delocalisation of $R_{\text{Sch, ext}} = \frac{8}{7} R_{\text{max}}$.

Producing phase contrast by the defocus aberration,
the spherical aberration, or both, is generally
linked to introducing a delocalisation. Therefore,
no strict optimum exists for having both maximum
phase contrast and low delocalisation. Contour
plots of the amount of phase contrast (Fig. 2) and
of the amount of delocalisation (Fig. 3) versus
spherical aberration and defocus, simulated for a
wavelength of 2.5 pm at an acceleration voltage of
200 kV and aninformation limit of 7.3 nm$^{-1}$,
show the combined effect of both aberrations and
illustrate the connection of relations (1)-(10). As a
measure for phase contrast we take the integrated
area under the phase-contrast transfer function.
The contour plot of the amount of phase contrast
exhibits a ridge starting at $C_S = 0$ from defocus
(10), following roughly relation (1) of Scherzer’s
defocus for $C_S > 40 \mu m$, and culminating at a
point given by relations (11) and (12). The contour
plot of the delocalisation exhibits a valley with
slopes rising towards increasing spherical aberra-
tion and increasing defocus, with the bottom of the
valley following relation (5) of Licht’s defocus of
least confusion.

A compromise between a high amount of phase
contrast and a reduced delocalisation can be found

\[
C_{S_{\text{opt}}} = \frac{64}{27} \frac{1}{\lambda^2 R_{\text{max}}} \\
Z_{\text{opt}} = \frac{16}{9} \frac{1}{\lambda^2 R_{\text{max}}}^2
\]

accompanied by a delocalisation of $R_{\text{opt}} = \frac{16}{27} R_{\text{max}}$,
which is $\frac{2}{7}$ times smaller than the delocalisation for
the extended Scherzer pass-band, relations (11)
and (12). For a wavelength of 2.5 pm and an
information limit of 7.3 nm$^{-1}$ the amount of phase
contrast is reduced by only 10%, which can be
deduced from the contour plot shown in Fig. 2.

For the Philips CM 200 FEG ST microscope
equipped with the aberration correction system
the different settings for defocus and spherical
aberration, Eqs. (11)-(14), are, using a wavelength
of 2.5 pm and an information limit of 7.3 nm$^{-1}$:

$C_{S_{\text{Sch}}} = 0.120 \, \mu m$, $Z_{\text{Sch, ext}} = -120.0 \, \mu m$, $R_{\text{Sch, ext}}$

$= 0.365 \, \mu m$, $Z_{\text{opt}} = -7.5 \, \mu m$, $R_{\text{opt}} = 0.137 \, \mu m$, $C_{S_{\text{opt}}}$

$= 0.053 \, \mu m$, $Z_{\text{opt}} = -13.3 \, \mu m$, $R_{\text{opt}} = 0.081 \, \mu m$.

Significantly higher values for the spherical
aberration with respect to optimum phase-contrast
imaging are found by den Dekker et al. [13], due to
a stronger weighting of lower spatial frequencies.
The consequences for high-resolution imaging of smallest object structures using the extended Scherzer pass-band, Eqs. (11) and (12), or the extended pass-band with reduced delocalisation, Eqs. (13) and (14), can be estimated very well without excessive image simulation studies. Both settings produce a phase contrast transfer without contrast reversals, leading to the same type of structure image known from the Scherzer defocus setting for high spherical aberration. The particular difference between the Scherzer pass-band at a high spherical aberration and the Scherzer pass-band at small spherical aberration is the higher point resolution of the latter.

In the simulated through-focus through-thickness map for Ge [1 1 0] given in Fig. 1, it can be seen how the image contrast gradually changes from the simple channelling-determined pattern to more complicated patterns by introducing the defocus aberration. For a small specimen thickness and a defocus of $Z = -7.5$ nm, according to relation (10), phase contrast occurs, with a pass-band narrower than the extended Scherzer pass-band, or the extended pass-band with reduced delocalisation.

The above considerations on combinations of spherical aberration and defocus useful for high-resolution imaging of smallest object structures, hold true also for a negative spherical aberration and the corresponding defocus values of over-focus. With the present layout of the hexapole correction system it is possible to generate a negative spherical aberration as well, which gives the additional freedom of producing positive or negative phase contrast, i.e. “white-atom” or “black-atom” structure images.

3. Experimental

High-resolution imaging of different object structures at a vanishing, or small, delocalisation requires a set-up of the aberration correction system with a vanishing spherical aberration, and an objective lens focus setting close to Gaussian focus, i.e. $Z > 0$. Any deviation from the Gaussian focus introduces a delocalisation of $R = |Z| / \delta_{f_{\text{max}}}$. Whether it is induced by a misalignment of the defocus, or by a defocus change over the field of view due to an inclined bottom surface of the object under investigation. To date, sample preparation techniques used for materials science investigations generally lead to wedge-shaped samples with more or less inclined bottom surfaces. A similar situation is obtained in the imaging of theatorial parts of spherical particles, where particles of different diameter on e.g. a carbon film are imaged under different effective focus. Therefore, it is useful in high-resolution imaging at a low level of delocalisation to record through-focus series of images with close defocus steps in order to get the desired specimen locations in-focus. We note that even under these conditions it is, in single-crystalline specimens, not always easy to determine which of the images of such a series is the proper zero-defocus image.

We found that in practical high-resolution work, a particular advantage of the fully corrected objective lens is the possibility to take advantage of aberration-free tilts of the incident electron beam up to a few millirads. This allows to align accurately the desired specimen low-index crystallographic direction with the incident beam for optimum imaging of the atomic structure. Due to the particular mechanical problems of specimen tilting stages this is difficult to achieve using the stage tilt controls only. As a consequence the electron microscopist using a standard electron microscope is often forced to give up a desired specimen area in favour of a neighbouring one where by chance, due to sample bending, the atomic structure occurs at optimum orientation. In standard microscopes with a high spherical aberration, tilting the beam for the sake of adjustment of the proper specimen orientation would induce aberrations, in particular defocus, two-fold astigmatism, and coma [14]. In the aberration-corrected microscope such aberrations do not occur, except a simple image displacement caused by defocus.

The information limit of the Philips CM 200 FEG ST equipped with the aberration-correction system was measured using a Young’s fringe analysis. Fig. 4 displays the respective diffractograms of image pairs of amorphous tungsten deposited on a thin carbon film.
of the microscope is carried out using aberration measurements deduced from Zemlin tables [15,16].

In order to demonstrate the conditions for experimental high-resolution investigations at a vanishing spherical aberration, four examples of practical work with the aberration-corrected instrument will be presented in the following. These concern three types of sample, a multilayer structure comprising thick GaSb and thin AlSb layers, a heterostructure with a thin AlAs layer embedded in GaAs, and a polycrystalline thin film of $\text{Ba}_0.7\text{Sr}_{0.3}\text{TiO}_3$.

Fig. 5 displays images from a defocus series of a GaSb/AlSb/GaSb heterostructure oriented along the [0 0 1] zone axis. The specimen was prepared by cleaving along {1 1 0} planes, thus forming a wedge of 90° with the bottom surface inclined at 45° with respect to the electron beam. The defocus series of 20 images was taken with the first defocus at 30 nm over-focus and defocus steps of 3.6 nm towards under-focus. At Gaussian focus, $Z = 0$, with respect to the specimen edge, Fig. 5(A), the amorphous material at the specimen edge is imaged under minimum contrast conditions, and the adjacent thinnest part of the crystalline material exhibits pure amplitude contrast revealing a projection of the object structure. Image contrast of specimen locations further away from the edge is obscured more and more by the defocus-induced delocalisation due to the inclined bottom surface. The latter effect can be observed more clearly by analysing the point-spread close to the specimen edge for different images of the defocus series. Fig. 5(B)–(E) display images with growing defocus levels of $-3.6 \text{ nm}$, $-7.2 \text{ nm}$, $-14.4 \text{ nm}$ and $-28.8 \text{ nm}$, resulting in delocalisations of $0.07 \text{ nm}$, $0.13 \text{ nm}$, $0.26 \text{ nm}$ and $0.53 \text{ nm}$, that can be observed directly from the growing fringe distance of the Fresnel diffraction fringes at the specimen edge.

Fig. 6 displays the image at Gaussian focus from a series of images of a GaAs/AlAs/GaAs heterostructure oriented along the [0 0 1] zone axis. The heterostructure was grown at a low temperature of 220°C, and annealed for 30 min at 750°C, with a nominal AlAs layer thickness of 6 monolayers. The Gaussian image is free from delocalisation...
and it, therefore, permits an analysis of the local Al content by evaluating the (0 0 2) image intensity distribution, unit-cell by unit-cell. The intensity map was transformed into a profile of the Al content by averaging of individual profiles along the layer structure and by exploiting the almost linear chemical sensitivity of the (0 0 2) intensity.

Figs. 7 and 8 display images of a polycrystalline Ba$_{0.7}$Sr$_{0.3}$TiO$_3$ thin film with individual grains oriented along the [0 0 1] zone axis. The film exhibits a large number of irregular and small-angle tilt grain boundaries. Fig. 8 displays two images taken at Gaussian focus and at Z = -30 nm. The image at Gaussian defocus is free from delocalisation revealing the structure of the dislocations at the small-angle grain boundary clearly. The second image reveals nearly the same contrast pattern in the periodic parts of the specimen, caused by a defocus offset to the first image close to the Fourier image defocus period $\Delta Z = 2/(\lambda g^2)$ of the perovskite lattice. Since its defocus is off Gaussian defocus delocalisation is visible at the defect locations, e.g. the dislocation cores of the small-angle tilt boundary. The corresponding disk of confusion of the point-spread function is indicated in Fig. 8(B).

Fig. 9 displays an image at Gaussian focus of a Ba$_{0.7}$Sr$_{0.3}$TiO$_3$ thin film in the [1 1 0] zone axis orientation. The image represents a structure projection of the perovskite lattice with large dots corresponding to (BaSr) columns, weaker dots to Ti columns, and faint dots to O columns.
The Ti–O column spacing of 0.14 nm in the [1 1 0] zone axis orientation is resolved.

4. Discussion

The high-resolution investigations clearly show that operating the aberration-corrected microscope at a vanishing spherical aberration leads, in many cases, to imaging conditions with vanishing delocalisation. The prerequisite for simple, directly interpretable, images of the projected structure, however, is a precise control of the defocus levels, and of the local specimen tilt. If the investigations have to be made rapidly, e.g. for materials sensitive to beam irradiation damage, or if a precise defocus control is not possible due to an inclined bottom surface of the specimen, taking defocus series of images is a great help. More
Fig. 8. Grains and grain boundaries in a Ba$_2$Sr$_{0.7}$TiO$_3$ thin film. The defocus of the upper image is 0 nm (A); the defocus of the lower image is −30 nm (B).

Fig. 9. Ba$_2$Sr$_{0.7}$TiO$_3$ thin film in a [110] zone axis orientation. Arrows point to a Ti column and an adjacent O column.

critical to high-resolution investigations is the control of the specimen tilt. The precise adjustment of the incident electron beam with respect to the specimen orientation turned out to be of great advantage in all our practical work. Furthermore, while it is sometimes possible for simple, lattice-matched and unstrained object structures, e.g. the GaSb/AlSb and GaAs/AlAs multilayers investigated in this work, to align the specimen properly for the whole field of view, one frequently encounters cases where lattice defects, boundaries and interfaces induce local strain fields leading to a change of lattice orientation over the field of view. This was the case also in the different grains in the polycrystalline Ba$_{0.7}$Sr$_{0.3}$TiO$_3$ thin film investigated here. It turned out during practical work
that using the beam-tilt control to compensate for small local specimen misalignments, high-resolution information could be readily obtained from large areas which in conventional instruments would have produced very non-uniform contrast. The necessary small beam adjustments could be performed rapidly in the imaging mode, without the need to switch to diffraction mode. We note that recording the beam-tilt control settings provides a means to measure crystal tilts, e.g. across boundaries, at high precision on an atomic level.

Acknowledgements

The authors are grateful to Dr. Max. Haider, CEOS GmbH, Heidelberg, for his kind and continuous support. The project on aberration correction of a transmission electron microscope was funded by the Volkswagen Stiftung. The project on improving the electrical stability of the corrector power supplies was funded by the Deutsche Forschungsgemeinschaft.

References