Aberration-corrected atomic-resolution transmission electron microscopy

M. Lentzen[1]

1. Institute of Solid State Research and Ernst Ruska Centre for Microscopy, Research Centre Jülich, 52425 Jülich, Germany

m.lentzen@fz-juelich.de
Keywords: high-resolution transmission electron microscopy, aberration correction, sub-angstrom resolution, atomic resolution, contrast theory

The past decade of transmission electron microscopy has witnessed a dramatic gain in instrumental resolution towards the sub-angstrom scale. The gain was achieved chiefly through improvements in mechanical, electrical, and thermal stability of the instruments. An equally important dramatic gain in contrast and interpretability of recorded image intensities was achieved through aberration correction, in particular of the strong spherical aberration of the objective lens. In sum, today atomic resolution with high contrast has been achieved, and it is to be expected that with the aid of the next generation of improved instruments the very limits of resolution and contrast in transmission electron microscopy will be explored.

Historically two routes were followed to implement aberration correction in transmission electron microscopy: firstly, hardware or lens aberration correction, based on Scherzer's idea of using multipole lenses for the compensation of aberrations [1]; secondly, aberration correction of the reconstructed electron wave function, based on Gabor's idea of in-line and off-axis holography [2]. Today both routes have come to a successful end with the realisation of the double-hexapole lens aberration corrector [3], and its incarnations in various commercial instruments, and with the realisation of through-focus series wave function reconstruction [4, 5] and biprism off-axis electron holography [5].

More had to be done in order to make the instrumental improvements applicable to materials science investigations: contrast theory had to be extended in order to match the requirements for sub-angstrom structural investigations, and instrumental aberrations had to be precisely measured and controlled.

The contrast theory used today has been extended by following Scherzer's idea of employing phase contrast in the electron microscope [6]: lens defocus and variable spherical aberration are combined such that the Scherzer passband extends towards the information limit; both aberrations are further tuned in order to result in a small contrast delocalisation [7]. The contrast of thin objects could be strongly amplified by discarding the positive phase contrast mechanism, forced by the fixed positive spherical aberration of the objective lens, and by using the negative phase contrast mechanism instead [8]. This is possible today by over-compensating the fixed positive spherical aberration of the objective lens with a lens aberration corrector, resulting in a small negative spherical aberration, which is then combined with a small over-focus of the objective lens. It could be shown that the contrast amplification for negative phase contrast is based on the transmission of linear and nonlinear contributions to the image intensity with the same sign [8].

Even for thicker crystalline objects, which deviate significantly from the weak phase object approximation due to dynamical electron scattering, the image contrast can be optimised in an aberration-corrected instrument through special settings for lens defocus and spherical aberration [9]. The key idea is compensation of the phase of the quantum-mechanical column states, which grows with increasing object thickness. For column phases

G. Kothleitner, M. Leisch (Eds.): MC2009, Vol. 1: Instrumentation and Methodology, DOI: 10.3217/978-3-85125-062-6-003,

between $\pi/2$ and π negative phase contrast through a negative spherical aberration combined with an over-focus is favourable; for column phases exceeding π, however, it is traditional positive phase contrast through a positive spherical aberration combined with an under-focus.

An outlook for the next generation of aberration-corrected instruments, aiming at an information limit of 0.05 nm, shows that further progress can be made in optimising high-resolution contrast. Today aberrations up to the third-order are optimised; in future lay-outs contrast may be enhanced by including the spherical aberration of the fifth order [10]. This will be particularly helpful to fill the transfer gap at small spatial frequencies, which has grown in today's aberration-corrected instruments with improving information limits, relative to the small gap in the olden days of high-resolution transmission electron microscopy.

The high contrast achieved already with today's aberration-corrected instruments is a basic requirement for two goals in materials science investigations: firstly, the accurate measurement of atom column positions in order to detect minute structural changes and, secondly, the accurate measurement of the chemical occupancy of atomic columns. Experimental results support that aberration-corrected transmission electron microscopy has contributed to a break-through in materials science: the oxygen sub-lattice of oxides has been imaged with atomic resolution [11]; the concentration of light elements was measured, and oxygen deficiencies were detected quantitatively [12]; minute column displacements have been measured in 90° grain boundaries of YBCO [13]; the structure of ferroelectric domain walls in PZT has been determined with atomic precision [14], and the appertaining macroscopic polarisation could be measured, based on the microscopic measurements.

With the aid of the future generation of aberration-corrected instruments the accuracy for the measurement of column positions will improve: estimates predict an accuracy of 1 pm, in comparison with an accuracy of 2 pm for a present-day 300-kV instrument or an accuracy of 4 pm for the 200-kV aberration-corrected prototype, the CM 200 installed at Ernst Ruska Centre Jülich [13].

Approaching the target of 0.05-nm information limit, however, will be most likely be accompanied by the observation of limits to structure resolution not induced by the instrument but by the quantum-mechanical scattering process. For crystalline materials the modulation of the exit plane wave transferred by the microscope is governed by quantum-mechanical channelling states, which have a finite width in the range from 0.05 nm to 0.07 nm, depending on the scattering power of the column [10]. A limit to the peak column contrast observable in recorded image intensities will be limited by the decrease of Rutherford scattering with increasing scattering angles.

1. O. Scherzer, Optik **2** (1947) p114.
2. D. Gabor, Proc. Roy. Soc. A **197** (1949) p454.
3. Haider et al., Nature **392** (1998) p768.
4. A. Thust et al., Ultramicroscopy **64** (1996) p211.
5. W. Coene et al., Phys. Rev. Lett. **69** (1992) p3743.
6. H. Lichte, Ultramicroscopy **20** (1986) p293.
7. O. Scherzer, J. Appl. Phys. **20** (1949) p20.
8. M. Lentzen et al., Ultramicroscopy **92** (2002) p233.
9. C.L. Jia et al., Microsc. Microanal. **10** (2004) p174.
10. M. Lentzen, Ultramicroscopy **99** (2004) p211.
11. M. Lentzen, Microsc. Microanal. **14** (2008) p16.
12. C.L. Jia et al., Science **299** (2003) p870.
13. C.L. Jia and K. Urban, Science **303** (2004) p2001.
14. L. Houben et al., Ultramicroscopy **106** (2006) p200.
15. C.L. Jia et al., Nature Mat. **7** (2008) p57.

G. Kothleitner, M. Leisch (Eds.): MC2009, Vol. 1: Instrumentation and Methodology,
DOI: 10.3217/978-3-85125-062-6-003,