Picometer Transmission Electron Microscopy

K. W. Urban*

Institute for Solid State Research and Ernst Ruska Center for Microscopy and Spectroscopy with Electrons, Research Center Jülich, 52425 Jülich, Germany
*k.urban@fz-juelich.de

The science and technology of materials has to rely increasingly on the understanding of structures and physical properties down to the atomic level. In particular this holds for nanoscience and nanotechnology. The key to this is the recent introduction of aberration-corrected electron optics opening up the way to genuine atomic-resolution electron microscopy. This permits to study atomic structures at an unprecedented precision in the order of a few picometers and to solve materials problems directly on the atomic level [1].

Over decades high-resolution techniques like (crossed) lattice-fringe imaging were applied successfully in materials science [2]. Being based on conventional electron optics these techniques have rarely been able to offer genuine atomic resolution. Rather they supplied collective information such as local lattice symmetry or lattice plane distances. Today's optical technology allows to define atomic resolution in a very stringent way: For a given substance all atomic species are accessible, the behaviour of individual atomic-columns is observable, the occupancy as well as individual lateral atom-column shifts can be measured. These are new qualities going far beyond what has been possible before in conventional instruments.

It is quite unfortunate for the field that outside the electron optics community it is generally believed that electron microscopy is nothing else than a continuation of light optics with radiation of lower wavelength. This is combined with the view that image formation is based on some sort of electron absorption phenomenon revealing the position of the atoms. Although entirely wrong this view is, superficially regarded, corroborated by the fact that, in general, only single images are published in the literature selected in such a way that their contents fits to the reader's intuition. This has created the impression that electron microscopy is a "snap-shot technique". In addition, the impression that transmission electron microscopy in atomic dimensions is an "incoherent" technique providing us with "images" understandable in a straightforward way is supported by the claim frequently made in electron microscopic work that the thin-sample projected-potential approximation holds. On this basis the reconstructed wave function is taken as a projection of the actual sample structure. Although this may hold true in selected favourable cases it is quite easy to show that this approximation rarely holds for realistic materials science investigations. In any case it is by far not a sufficient basis for the ultra-precision investigations possible in today's aberration-corrected electron optical instruments [3].

Since studies in atomic dimensions are exploiting quantum mechanics the term "image" looses its classical meaning and intuitive image interpretation is inadequate. As a consequence, for picometre electron microscopy, the non-linear electron-optical imaging process has to be inverted numerically [4,5]. In general the starting point is an interferometric operation mode of the instrument where series of acquisitions are employed to calculate the electron wave function at the exit plane of the specimen. The second step is a numerical procedure based on iterative solutions of the Dirac equa-

tion for relativistic electrons for a model of the atom arrangement improved step by step until it is able to fit the measured experimental wave function. Since neither the specimen thickness nor the direction of incidence of the electron wave can be measured directly in atomic dimensions these imaging parameters have to be taken as variables together with the coordinates for many tens of atomic positions which have to be adjusted in the fitting procedure. We note the following points representing characteristic features of these numerical procedures inherently involved in ultra-high precision transmission electron microscopy:

1) In the past it was considered sufficient to obtain in the described calculations a qualitative fit of the intensity distribution at the position of high-nuclear charge atoms. This is by far not enough. Not only the relative height of all the intensity maxima has to be predicted correctly but also the diffuse grey-scale background which provides us with a very sensitive measure for small, otherwise undetectable specimen tilts or small residual aberrations which have to be corrected in order to arrive at a model precise enough for picometer-range measurements. Recently, after an explanation was found for the Stobbs factor problem absolute intensity fits are within reach opening up additional options for ultra-high resolution studies [6].

2) In samples of realistic thicknesses there is not only *phase contrast* to be considered but also *amplitude contrast*. In the past this type of contrast has rarely been considered in high-resolution studies. Today we know that it is largely responsible for the contrast under NCSI (negative spherical aberration imaging) conditions. The origin of amplitude contrast is electron diffraction channelling, and it is enhancing the intensity at the atomic positions. In order to benefit from it the phase contrast has to be inverted (so called negative phase contrast) so that the two contrast types are additive (rather than subtractive as in the conventional case). The substantial contrast improvement is an essential pre-condition for picometer-precision requiring high signal-to-noise ratios.

Recent examples of picometer electron microscopy are the measurement of the width of ferroelectric-domain walls in $PbZr_{0.2}Ti_{0.8}O_3$ (PZT) [7], the measurements of the coupling of elastic strain fields to polarization in PZT/$SrTiO_3$ epitactic systems [8] and of oxygen-octahedron tilt and polarization in $LaAlO_3$/$SrTiO_3$ interfaces [9]. Here atom shifts in the order of a few picometers could be measured.

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