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Picometer Transmission Electron Microscopy

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In recent years aberration-corrected transmission electron microscopy has demonstrated that the new research opportunities offered by this technique are meeting the growing demand of materials science for atomic-scale characterization of materials [1]. On the other hand, electron microscopy is currently still in a process of liberating itself of the restrictions imposed on its work by the limited optical qualities of the instruments of the pre-aberration correction era [2].

With certain exceptions most of this earlier work, in spite of the fact that the images looked like atomic, was in reality not really atomically resolving. Due to aberration-induced contrast delocalization a given atomic column did also contribute intensity to the positions of neighboring columns in the image. As a consequence atomic concentrations and lateral shifts on the scale of an individual atomic column could not be measured reliably. In most cases the images showed merely the *structure* of a sample, this means a collective *non-local* property. Today things have changed; genuine atomic resolution has become available allowing to measure individual atomic shifts and concentrations. However, in order to do so the investigations have to become more quantitative, much beyond the level which appeared to be sufficient in pre-aberration correction times. But it is worth making the effort: Aberration-corrected electron microscopy allows us to measure atomic positions and lateral shifts with close to picometer precision. This changes the electron microscope from a predominantly structure-oriented tool into a physical measurement instrument.

The goal of atomic-resolution work is to measure the set of individual atom positions in a sample $\boldsymbol{X}=\{\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \dots\}$. For this we employ an electron wave field $\boldsymbol{\psi}_0$ transmitted through the specimen. This wave field interacts with the atom potential $\boldsymbol{V}(\boldsymbol{r})$ and thereby it is modified inside the sample where it is described by a superposition of Bloch states. At the exit surface of the specimen the wave field is described by the exit-plane wave function $\boldsymbol{\psi}_\mathrm{e}$. This exit-plane wave function is the object of the electron optics. The individual partial waves, in a Fourier representation of $\boldsymbol{\psi}_\mathrm{e}$, characterized by their amplitudes, spatial frequencies $\boldsymbol{g}$ and wave vector directions are modified by the electron optics. Neglecting absorption the effect of the electron optics is treated as a modification in phase of the complex wave function described by the aberration function $\chi(\boldsymbol{g})$. The image intensity distribution is then given by the Poynting vector $\boldsymbol{I}(\boldsymbol{r}_\mathrm{i})$ which for fast electrons can be approximated as to be proportional to the absolute square of $\boldsymbol{\psi}_\mathrm{i}$, the aberration-modified wave function in the image plane. It is well known that in general high-resolution images obtained in the transmission electron microscope cannot be understood by intuitive interpretation. It is therefore common practice to perform what is called "image simulation" in order to get access to the information content of the images by comparing computed intensity distributions with experimental images. However, in order to exploit the full potential of aberration-corrected transmission electron microscopy the precision of image calculations has to be improved substantially.

What is essential the inversion of the non-linear imaging process. Given a set of experimental images these have first of all to be corrected for the nonlinear transfer characteristics of the CCD ca-

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mera employed for recording the images. It could recently been shown that it is primarily the detector modulation-transfer function that has a dramatic effect on the experimental intensity vs. spatial frequency characteristics in the images [4]. Subsequently a "backward" calculation has to be carried out to retrieve the exit-plane wave function. Against a widespread assumption $\boldsymbol{\psi}_e$ does in general not supply us with a direct representation of $\boldsymbol{X}$. Neither a mapping of the real nor of the imaginary part of $\boldsymbol{\psi}_e$ gives us unambiguously the correct atomic sites. Depending on the sample thickness, the contrast varies dramatically and in a complex fashion which cannot be treated intuitively. Furthermore the $\boldsymbol{\psi}_e$ obtained is the wave function of the "real" case. This means that a tilted specimen yields as a result a "tilted" wave function, and the amplitude and phase distribution are those of the "real" case, i.e. they depend on the actual specimen thickness. Of course the set of wanted coordinates $\boldsymbol{X}$ must be universal and therefore independent of these rather arbitrary imaging circumstances. Therefore, as long as the imaging parameters are not precisely known, the reconstructed wave function is not yet sufficient for picometer microscopy. This means that the backward calculation has to be continued. As usual this is done by choosing a first-guess model, use tabulated (independent atom) potential coefficients employed to solve the relativistically corrected Schrödinger equation for the wave function. Since neither the sample thickness nor the beam tilt is in general known with sufficient accuracy, these have to be introduced as free parameters in an iterative fit of the model in order to reach an optimum match between calculated and experimental exit-plane wave function. Only after a self-consistent set of atomic coordinates and imaging parameters is obtained we can consider the problem solved.

The inversion of the imaging process as it was just described looks rather canonical. Nevertheless, concerning picometer precision electron microscopy there are important points which have to receive much greater attention than this was standard in the past. First of all, it is quite likely that the original set of images is affected by small residual aberration values. These have to be corrected in the backward calculation. Furthermore, for realistic sample thicknesses unavoidable specimen tilts in the order of 1 mrad are resulting in projection-related distortions of the atomic images affecting the precision of the measurements. On the other hand, provided that the contrast behavior of the diffuse background in the micrographs is taken into account the effect of even 0.5 mrad tilts can be detected in the image calculations and thus be taken care of.
We point out that it is not the employed measurement procedures but the extremely small dimension of the effects to be measured which are making the high demands described. This has been exploited e.g. in perovskitic oxides to investigate the behavior of individual dipoles in polarization-domain walls [3,5]. On the other hand, in addition to picometer-precision of lateral atom-position measurements aberration-corrected transmission electron microscopy combined with numerical inversion of the imaging process is able to reveal chemical bonding effects and to offer not only lateral but also atomic depth resolution, as has recently been shown in graphene [6,7].

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