Title
Image formation in the high-resolution transmission electron microscope

Author
O'Keefe, Michael A.

Publication Date
2004-03-04

Image Formation in the High-Resolution Transmission Electron Microscope

Michael A. O'Keefe
Materials Sciences Division, LBNL 2-200, 1 Cyclotron Road, Berkeley, CA 94720, USA

Letter to the Editor:

Image Formation in the High-Resolution Transmission Electron Microscope

In the course of reporting in these pages results obtained with the One-Ångstrom Microscope (OÅM) at Lawrence Berkeley National Laboratory, A. C. Diebold and his co-authors offer an incorrect interpretation of the image formation process in the high-resolution electron microscope (Diebold et al., 2003). It is important to rectify this problem before the misinterpretation comes to be accepted as factual by scientists who are not expert in the field of high-resolution transmission electron microscopy.

The authors correctly describe the two stages of the image formation process: interaction of the incident electron beam with the crystal to form multiple diffracted beams, followed by interference of two or more of the diffracted beams to form a "lattice" image. However, the authors go on to make the extraordinary statement that "*Lattice images do NOT depict the projected atom columns; instead, they are interference patterns of the directly transmitted beam with diffracted beams*." The authors' determination to emphasize their misstatement with italic and uppercase text makes its inaccuracy particularly regrettable. In fact, TEM images ARE able to depict the projected atom columns. And they are able to do this BECAUSE they are interference patterns of the directly transmitted beam with beams diffracted from the specimen.

Materials scientists have come to rely on the fact that high-resolution transmission electron microscopes are able to produce micrographs that are images of atoms, or atom columns, or unresolved groups of atoms. Any high-resolution TEM, operated under well-established conditions (conditions that have been understood and utilized for decades), produces phase-contrast images in which intensity peaks correspond to the true column positions of the projected crystal lattice.

In the high-resolution transmission electron microscope, structural information from the specimen is encoded in the spatial distribution of the phase of the scattered electron waves (Cowley, 1975). Although the electron phase is not observable (it is not gauge invariant), phase differences can be measured with interference experiments. A direct way is by electron holography (Lichte, 1991), but the usual method is to image the specimen at the "optimum" or "extended" Scherzer (1949) defocus. At this focus, the objective lens shifts the phase of the scattered electron wave exiting the specimen such that interference causes the relative phase of the wave to form image peaks that map the atom positions at the resolution of the microscope. This result has been verified many times by theory (Cowley & Iijima, 1972), simulation (O'Keefe et al., 1978), and countless experiments.

Of course it is true that a misfocused TEM will depict atom positions incorrectly, but the same is true of any optical instrument. No one makes the statement that camera images "do NOT depict the positions of trees" merely because it is possible to photograph a forest with the camera misfocused sufficiently to produce false "tree images" by overlap of blurred representations of the real trees.

It may be that the authors were led to an incorrect interpretation of the nature of TEM images by the exceptional resolution of the One-Ångstrom Microscope (OÅM), e.g., the OÅM image shown in their Figures 6b and 10a. The OÅM project was designed to extend the resolution of a mid-voltage TEM to the sub-Ångstrom region using hardware enhancements and focal-series reconstruction of the electron wave at the specimen exit surface (O'Keefe, 1993). The OÅM is a Philips CM300FEG/UT modified to correct objective lens three-fold astigmatism and extend information transfer to below one Ångstrom (O'Keefe et al., 2001a). It is capable of achieving resolutions down to 0.078 nm (O'Keefe et al., 2001b), and of imaging columns of atoms as light as lithium (Shao-Horn et al., 2003). Instead of imaging atom peaks by extracting the spatial distribution of the relative phase from the electron wave by direct interference (as in a TEM at optimum defocus), the OÅM uses software to extract the relative electron phase from a series of images and display it with atom positions appearing as peaks in the spatial distribution (Coene et al., 1996; Thust et al., 1996).

Because the OÅM's resolution far exceeds the 0.17 nm limit of a typical 300-keV TEM, it is possible to misinterpret the image improvement produced by the OÅM's extended resolution as some perceived property of the reconstruction process. This possibility is suggested by the authors' statement "HR-TEM combined with focal series reconstruction can produce direct images of the crystal structures with sub-Ångstrom resolution down to about 0.08 nm, because the phase of the electron exit wave marks the position of the projected atomic columns and the resolution is improved." The statement is ambiguous and appears to confuse the effects of improved resolution and focal series reconstruction. Any high-resolution TEM "can produce direct images of … crystal structures". Such images will be limited to the resolution of the particular TEM, and achieve sub-Ångstrom resolution only if the TEM has sufficient resolution. Resolution may be limited by spherical aberration, or by the microscope information limit if spherical aberration is corrected (O'Keefe, 1992). Correction may be made by hardware, or by software such as focal-series reconstruction. In addition, there is nothing particularly special about 0.08 nm, and focal-series reconstruction will not automatically produce this resolution. The figure of 0.08 nm is merely the resolution of the OÅM (O'Keefe et al., 2001b) and will be different for other TEMs. For example, the original investigation that led to the OÅM project produced images with 0.138 nm resolutions from a JEOL ARM-1000 using focal-series reconstruction (Wenk et al., 1992).

It is indeed true that "the (relative) phase of the electron exit wave marks (displays) the position of the projected atomic columns" in the focal series reconstruction. However, it is just as true for any high-resolution TEM image taken under the correct imaging conditions. Theory predicts that images obtained either directly or with focal-series reconstruction will show the same peak positions corresponding to the same atom positions, provided only that both images are obtained under the correct conditions and possess the same resolution. Reconstructions and equivalent direct images, taken with the correct objective lens phase changes, show identical atom peaks (Fig 1a, c) for carbon atoms separated by 0.089 nm in [110] diamond (O'Keefe et al., 2001a). Even at the OÅM information limit of 0.078 nm (O'Keefe et al., 2001b), positions of atom peaks in [112] silicon match in reconstructed and direct images (Fig 1b,d); this is √3 times better resolution than required for the [110] silicon atom image used by the authors. These OÅM experiments confirm the correspondence of direct and reconstructed atom peak positions.

It cannot be emphasized too strongly that high-resolution TEM images actually do show the positions of projected atom columns under the proper conditions. This is true whether we reconstruct the spatial fluctuations in the phase that carries the information on atom positions, or make them visible directly by interference. Improvement in the quality of atom position information in OÅM images is due to the OÅM's improved resolution, not to the fact that focal-series reconstruction is the method used to extract these positions from the electron wave phase.

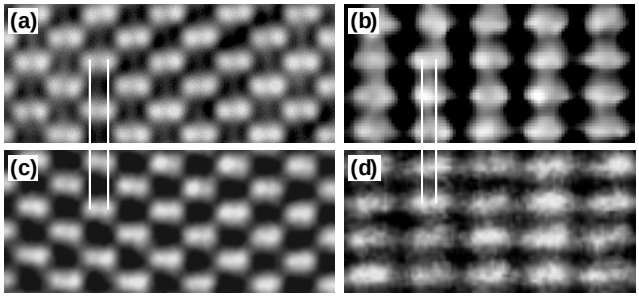

Figure 1. Comparison of reconstructed and direct images. Reconstructed (a, b) and direct (c, d) images show atom positions for [110] diamond (a, c) and [112] silicon (b, d). OÅM images are aberration-corrected by reconstruction from 20-member focal-series. Direct images are obtained at alpha-null defocus (O'Keefe et al., 2001a). Diamond images reveal 0.089 nm carbon atom spacing (O'Keefe et al., 2001a). Silicon images show 0.078 nm atom spacing (marked) at the OÅM resolution limit (O'Keefe et al., 2001b). Magnification is 25 million times.

Michael A. O'Keefe
Materials Sciences Division, LBNL 2-200
1 Cyclotron Road, Berkeley, CA 94720, USA
Principal Investigator of the LBNL One-Ångstrom Microscope Project, 1992-2002
maok@LBL.gov

COENE, W.M.J., THUST, A., OP DE BEECK, M. & VAN DYCK, D. (1996). Maximum-likelihood method for focus-variation image reconstruction in high resolution transmission electron microscopy. *Ultramicroscopy* **64**, 109-135.

COWLEY, J. M. & IIJIMA, S. (1972) Electron microscope image contrast for thin crystals. *Z. Naturforsch.* **27a**, 445-451.

COWLEY, J.M. (1975). *Diffraction Physics*. North Holland / American Elsevier.

DIEBOLD, A.C., FORAN, B., KISIELOWSKI, C., MULLER, D.A., PENNYCOOK, S.J., PRINCIPE, E. & STEMMER, S. (2003). Thin dielectric film thickness determination by advanced transmission electron microscopy. *Microscopy & Microanalysis* **9**, 493-508.

LICHTE, H. (1991). Optimum focus for taking electron holograms. *Ultramicroscopy* **38,** 13-22.

O'KEEFE, M.A., BUSECK, P.R. & IIJIMA, S. (1978). Computed crystal structure images for high resolution electron microscopy. *Nature* **274,** 322-324.

O'KEEFE, M.A. (1992). 'Resolution' in high-resolution electron microscopy. *Ultramicroscopy* **47**, 282-297.

O'KEEFE, M.A. (1993). Using coherent illumination to extend HRTEM resolution: why we need a FEG-TEM for HREM. *LBL Symposium on Microstructures of Materials*, edited by K. Krishnan (Berkeley: San Francisco Press), pp. 121-126.

O'KEEFE, M.A., HETHERINGTON, C.J.D., WANG, Y.C., NELSON, E.C., TURNER, J.H., KISIELOWSKI, C., MALM, J.-O., MUELLER, R., RINGNALDA, J., PAN, M., & THUST, A. (2001a). Sub-Ångstrom high-resolution transmission electron microscopy at 300keV. *Ultramicroscopy* **89**, 4: 215-241.

O'KEEFE, M.A., NELSON, E.C., WANG, Y.C. & THUST, A. (2001b). Sub-Ångstrom resolution of atomistic structures below 0.8Å. *Philosophical Magazine B* **81,** 11: 1861-1878.

SHAO-HORN, Y., CROGUENNEC, L., DELMAS, C., NELSON, E.C. & O'KEEFE, M.A. (2003). Atomic resolution of lithium ions in $LiCoO_2$ battery material. *Nature Materials* **2**, 464-467.

THUST, A., COENE, W.M.J., OP DE BEECK, M. & VAN DYCK, D. (1996). Focal-series reconstruction in HRTEM: simulation studies on non-periodic objects. *Ultramicroscopy* **64**, 211-230.

SCHERZER, O. (1949). The theoretical resolution limit of the electron microscope. *J. Applied Physics*, **20**, 20-29.

WENK, H.-R., DOWNING, K.H., MEISHENG, HU & O'KEEFE, M.A. (1992). 3D structure determination from electron-microscope images: electron crystallography of staurolite. *Acta Cryst.* A**48,** 700-716.

Work supported by the Director, Office of Science, Office of Basic Energy Sciences, Materials Science Division, US Department of Energy, contract DE-AC03-76SF00098. Electron microscopy was carried out using the OÅM at the NCEM, LBNL. Thanks are due to Drs. Charles E. Lyman and Lawrence F. Allard for critical reading of this letter.