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Letter to the Editor: Image Formation in the High-Resolution Transmission Electron Microscope

A recent article in these pages compares STEM images with an image obtained with the One-Ångstrom Microscope (OÅM) at Lawrence Berkeley National Laboratory (LBNL). Although the experimental work is of excellent quality, Diebold et al. (2003) offer an incorrect explanation of the image formation process in the high-resolution transmission electron microscope. It is important that this misinterpretation be corrected before it comes to be accepted as factual by other scientists who are not expert in the field of high-resolution transmission electron microscopy.

The article correctly describes the two stages of HRTEM image formation: 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, it goes on to make the incorrect 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 publication's emphasis of this 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 so *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 (e.g., Smith, 1997). Any high-resolution TEM operated under well-established conditions (conditions that have been understood and utilized for decades) will produce phase-contrast images in which intensity peaks correspond to the atomic 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 (e.g., Cowley, 1975). Although the electron phase is not observable, phase differences can be measured with interference experiments. One direct way is by electron holography (e.g., Lichte, 1991), but the usual method is to image the specimen at the Scherzer (1949) defocus or the "optimum" extended-Scherzer defocus (O'Keefe, 1992). At these values of 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 (e.g., Cowley & Iijima, 1972; Cowley, 1975), by simulation (e.g., O'Keefe et al., 1978), and in countless experimental observations (e.g., Hofmann & Ernst 1994; Smith, 1997).

Of course, it is true that a misfocused TEM can be made to depict atom positions incorrectly, but the same is true for many optical instruments. 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.

Unfamiliarity with images at ultrafine resolution (e.g., the OÅM image in Figs. 6b and 10a of Diebold et al., 2003) could make lower resolution images seem incorrect by comparison. Because the resolution of the OÅM[1] 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 resolution and mistake it as a special property of the reconstruction process. This possibility is suggested by the following statement in Diebold et al. (2003): "HRTEM 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."

[1]The OÅM project was conceived to produce images at ultrafine resolution (O'Keefe, 1993) and was implemented using a Philips CM300FEG/UT with hardware modifications designed to correct objective lens threefold astigmatism and extend information transfer to below 1 Å (O'Keefe et al., 2001*a*). Instead of imaging atom peaks by extracting the spatial distribution of the relative phase from the electron wave by direct interference (as in a HRTEM at optimum focus), the OÅM uses FEI focal-series reconstruction software (Coene et al., 1996; Thust et al., 1996) to derive the relative electron phase from a series of images. The result of this focal-series reconstruction is to produce the spatial distribution of relative phase with peaks that correspond to the atom positions. The OÅM is capable of achieving resolutions down to 0.078 nm (O'Keefe et al., 2001*b*) and of imaging columns of atoms as light as lithium (Shao-Horn et al., 2003).

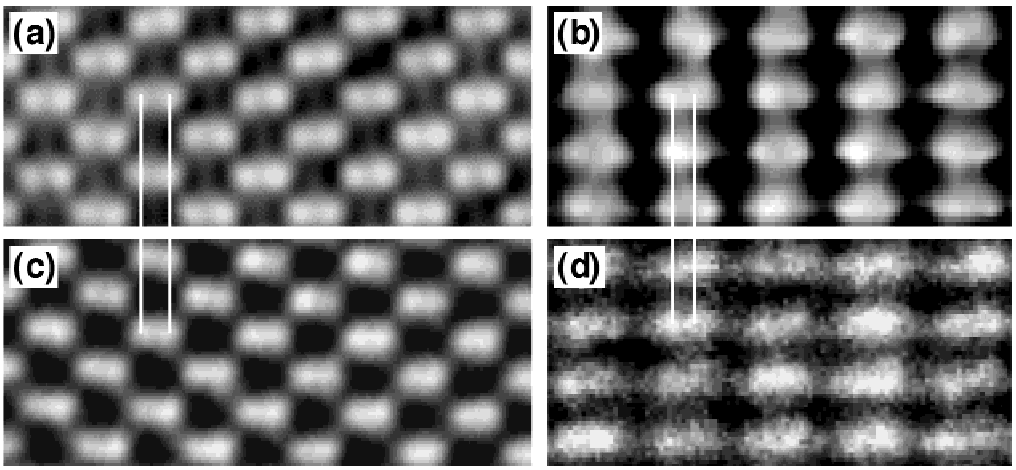

Figure 1. Comparison of reconstructed and direct images. Reconstructed (**a**, **b**) and direct (**c**, **d**) images show atom positions for [110] diamond (**a**, **c**) and [112] silicon (**b**, **d**). OÅM images are C_S-corrected by reconstruction from 20-member focal series. Direct images are obtained at alpha-null defocus (O'Keefe et al., 2001*a*). Diamond images reveal 0.089-nm carbon atom spacing (O'Keefe et al., 2001*a*). Silicon images show 0.078-nm atom spacing (marked) at the OÅM resolution limit (O'Keefe et al., 2001*b*). Images are shown at the same magnification of 39 million times; the diamond images are 1.78 nm across and the silicon 1.57 nm.

The statement appears to confuse the effects of improved resolution and focal-series reconstruction. Any high-resolution TEM "can produce direct images of the crystal structures." Such images will be limited to the resolution of the particular TEM and achieve sub-Ångstrom resolution only if the TEM can reach sub-Ångstrom resolution. Resolution may be limited by spherical aberration or by chromatic aberration (O'Keefe, 1992) if spherical aberration is corrected. Correction may be made by hardware or by software, such as focal-series reconstruction software. 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 just the established 0.078-nm resolution of the OÅM (O'Keefe et al., 2001*b*) and will be different for other TEMs. For example, the original investigation that led to the OÅM project produced images with 0.138-nm resolution from a JEOL ARM-1000 using a simple linear focal-series reconstruction (Wenk et al., 1992).

It is 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. OÅM reconstructions show the same atom peaks as equivalent OÅM direct images taken with the correct objective lens phase changes. This agreement has been demonstrated for carbon atoms (Fig. 1a,c) separated by 0.089 nm in [110] diamond images (O'Keefe et al., 2001*a*). Further, the positions of atom peaks in [112] silicon images are the same in OÅM reconstructed and direct images (Fig. 1b,d). Correspondence of direct and reconstructed atom peak positions is confirmed down to the OÅM information limit of 0.078 nm (O'Keefe et al., 2001*b*). This is $\sqrt{3}$ times better resolution than is required for the OÅM [110] silicon atom image of Diebold et al. (2003).

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 variation in the phase that carries the information on atom positions or make them visible directly by interference. Improved 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 chosen to extract these positions from the phase of the electron wave. Any (C_S-corrected) HRTEM operated under the correct conditions and with the same resolution would show an image with the same atom positions.

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Authors' Response

The main purpose of the article by A.C. Diebold and coworkers (2003) is to propose a robust method for determination of gate oxide thickness. O'Keefe objects to a statement in this paper that "*Lattice images do NOT depict the projected atom columns; instead, they are interference patterns of the directly transmitted beam with diffracted beams.*"

We agree with O'Keefe's statement that "TEM images ARE able to depict the projected atom positions." However, as we elaborate in our article immediately following the statement quoted above, images showing reverse contrast are intrinsic to the phase contrast method. It is precisely the fact that a large number of very different phase contrast images may be obtained from any one specimen that makes such images directly interpretable, only if a detailed study of simulated images as a function of image and specimen variables is performed and the imaging conditions are carefully chosen. The strength of phase contrast imaging is high contrast and sensitivity to structural details.

O'Keefe's objection is based upon the specific case of Scherzer imaging in a conventional electron microscope where the information limit is close to the Scherzer point resolution and if thin areas are imaged. A simulation of this situation is shown in Figure 1. We assume that the identical location of contrast minima and atom column positions is what he refers to by claiming, "intensity peaks correspond to the atomic positions of the projected crystal lattice."

However, at a particular specimen thickness, within the same image, both contrast maxima and minima can mark atom positions even if lattice images of thin samples are recorded at Scherzer defocus as shown in Figure 2. Furthermore, it is even *desirable* to record a large number of very different lattice images at other focus values (Lichte, 1991). Lichte defocus, for example, minimizes delocalization but

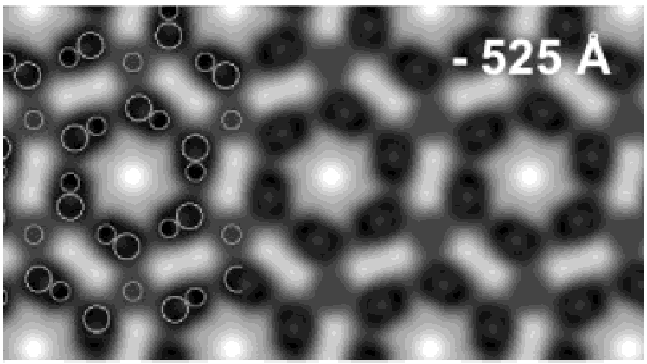

Figure 1. Simulated lattice image of a thin Si_3N_4 [0001] crystal. The crystal structure is inserted. Large circles: Si. Small circles: N. Open circles enable a better comparison of intensity with column positions. Scherzer defocus (800 kV). A LaB_6 cathode is assumed. Note that contrast minima relate to atom positions.

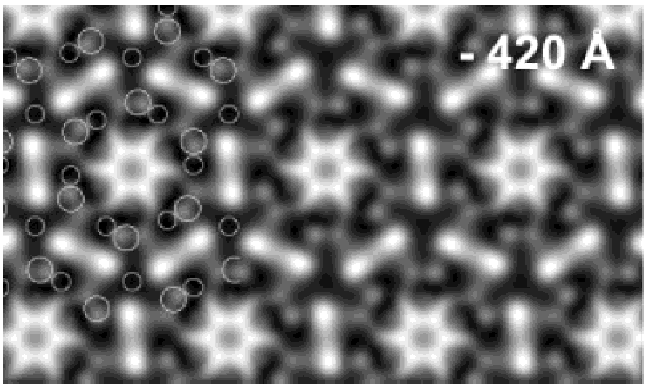

Figure 2. Same sample as Figure 1 with Scherzer defocus (300 kV). A field emission cathode is assumed. Note that contrast minima relate to nitrogen atom positions whereas contrast maxima relate to silicon columns.

adds complexity to the image patterns. Nevertheless, it is the most suitable focus setting for the recording of focus series for the purpose of reconstructing electron exit waves (Coene et al., 1996). A varying image pattern is precisely the reason that such images are only interpretable if a detailed study of simulated images as a function of imaging, and specimen variables, is performed, unless electron exit waves are properly reconstructed.

None of this should be surprising or controversial. For instance, further discussion of this Si_3N_4 example can be found in chapter 7.2 of Kirkland's (1998) book, including a demonstration of contrast reversal at Scherzer defocus, even with a Scherzer aperture, when the sample thickness changes from 10 to 15 nm (for a 200-kV beam). He notes that "the sign of the contrast will change periodically with defocus for a given thickness and also periodically with thickness for a given defocus, making image interpretation very difficult. Image simulation is one means of sorting out what is going on in the image." Exit wave reconstruction is another.

The analogy of photographing trees in a light optical camera that O'Keefe uses is, in our opinion, not applicable here, as a camera does not form a phase contrast image like a TEM, but rather an incoherent image. An out-of-focus image disappears. It is not possible to obtain an image of trees with inverted contrast.

In summary, we entirely agree that "TEM images actually do show the positions of projected atom columns *under the proper conditions*" (emphasis added). The point is that knowing those proper conditions requires knowledge of the atom species and positions, which is precisely the information we are trying to determine from the image in the first place. Although results from NCEM's OÅM were shown in our article, the focus of our article was neither to "report in these pages data obtained with the One-Ångstrom Microscope" nor to provide current figures for "the resolution of the OÅM". Also, the discussion of OÅM sub-Ångstrom resolution and its first demonstration in 2000 by the diamond [110] phase image (Kisielowski, et al., 2000) republished in O'Keefe's letter is not germane to our article. Rather, the goal of our article was to provide a practical, robust method for determining the thickness of ultrathin gate dielectrics on silicon substrates. In our article, we emphasize that both methods (Z-contrast imaging and HRTEM) provide correct, reliable values for gate dielectric film thicknesses under the correct experimental conditions.

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