

Seeing Atoms at Sub-Ångstrom Resolution with Aberration-Corrected TEM

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Resolution is the ability to determine if a feature in an image represents two objects rather than one. Rayleigh's resolution criterion [1], an accepted standard in optics, was derived as a means for judging when two sources of light (stars) were distinguishable from a single source. In microscopy, resolution is the ability to determine if detail in an image represents distinct (separated) objects. In high-resolution TEM, these objects are atoms. Resolution of $|d|$ is achieved when atoms separated by a (projected) distance $|d|$ can be perceived as separate objects. Presence of the $1/|d|$ frequency in the TEM image spectrum is not sufficient to demonstrate a corresponding resolution of $|d|$ [2]. The standard test of TEM image resolution should be the separation of atom peaks in the image [3].

In the high-resolution TEM, the sample is oriented along a relatively low-index zone axis where the atom columns to be imaged are well separated in projection, ideally by more than the microscope resolution. The incident electron beam interacts strongly with the crystal, forming multiple diffracted beams. These diffracted beams are brought together by the objective lens so that they can interfere to create an image. ***TEM images are able to depict the projected atom columns because they are interference patterns of the directly transmitted beam with beams diffracted from the specimen.*** By imaging under well-established conditions that have been understood and utilized for decades, phase-contrast HRTEM images can be obtained in which either the intensity minima or maxima correspond to the true column positions of the projected crystal lattice.

Structural information from the specimen is encoded in the phase of the scattered electron waves [4]. Although the electron phase is not an observable (it is not gauge invariant [5]), phase differences can be measured by interference experiments. A direct way is by electron holography [6], but the usual method is to image at the "optimum" or "extended Scherzer" defocus [7]. Then objective lens phase shifts allow interference of the scattered electron waves exiting the specimen to turn the relative phases of the waves into image peaks mapping the atom positions (at the resolution of the microscope). This result has been verified by theory [8], simulation [9], and countless experiments. The native, or Scherzer, resolution limit of the uncorrected high-resolution transmission electron microscope is $d_x = 0.64C_s^{1/4}\lambda^{3/4}$ where C_s is the spherical aberration coefficient and λ the electron wavelength [7,10]. Increasing the accelerating voltage (and the cost) improves resolution by reducing electron wavelength. Objective pole-piece saturation keeps the product of C_s and λ (almost) constant with increasing voltage, so that a wavelength of $\lambda(\text{Å})$ will allow a resolution $d(\text{Å})$ of 12 times $\sqrt{\lambda}$. Mid-voltage TEMs can achieve 1.9Å resolution at 200 kV (1.7Å at 300kV), but require aberration correction (C_s reduction) to reach sub-Ångstrom levels [11]. C_s can be reduced directly (hardware correction [12]), or by using several images (with phase changes known from objective lens defocus) to compute the phase of the electron wave exiting the specimen (software correction [13]).

Hardware and software correction have each produced sub-Ångstrom images [10,14], and allowed the imaging of light atoms such as oxygen [15,16]. The LBNL One-Ångstrom Microscope (OÅM) combines a modified CM300FEG/UT TEM with FEI focal-series reconstruction (TrueImage™) software by Coene and Thust [17,18] to achieve sub-Å resolution to 0.78Å [19]. Modifications include hardware correction of 3-fold astigmatism to 0.68Å and information limit extension to 0.78Å [10]. The OÅM can image atoms as light (small) as nitrogen [20], carbon [10], and lithium [21].

Focal-series reconstruction (FSR) is more than just C_s correction. Its compensation of imperfect objective lens transfer provides improvement over any single image. Single images of diamond and silicon (Fig. 1) show carbon atoms clearly separated by 0.89Å and (at the information limit of the OÅM) silicon atoms separated by 0.78Å. C_s -corrected OÅM images (Fig. 2), assembled from 20-member focal series, are "cleaner", due to lack of second-order contributions. Note that second-order components can be removed from single images by subtracting a minimum-contrast image [22] (at zero-defocus for the case of zero C_s), thus extending the interpretable specimen thickness [23] as for focal-series reconstruction. Comparison of figures 1 and 2 shows that TEM images are able to depict atom positions just as well as do FSR images, provided both have the same resolution [24].

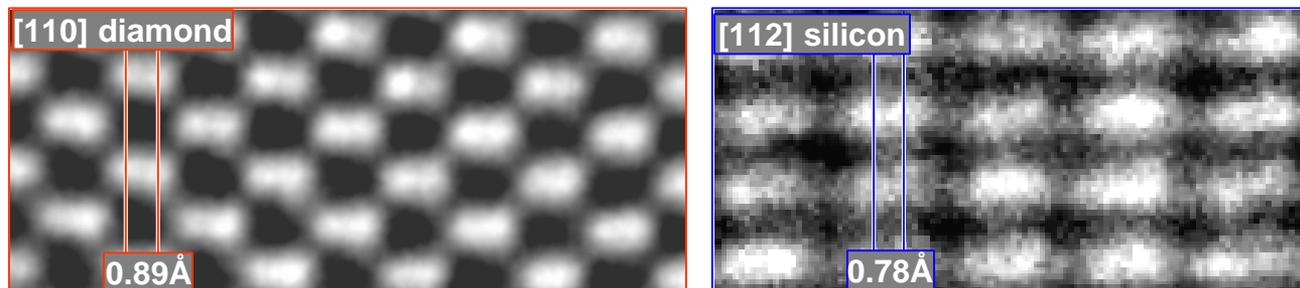


FIG. 1. Single-shot images obtained close to alpha-null defocus [10], shown at magnifications of 50 million times. Left image shows 0.89Å carbon atom spacing in [110] diamond [10]. Right image shows 0.78Å silicon atom spacing in [112] silicon at the OAM resolution limit [19].

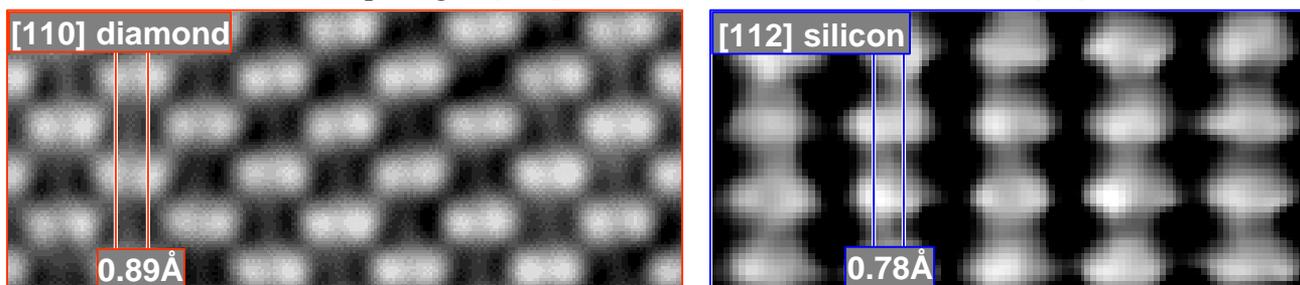


FIG. 2. OAM images, aberration-corrected by reconstruction from focal-series of 20 images, shown at 50 million times magnification. Left image shows 0.89Å carbon atom spacing in [110] diamond [10]. Right image shows 0.78Å silicon atom spacing in [112] silicon. Focal-series-reconstructed images appear “cleaner” than single-shot images due to lack of “second-order interferences” [2].

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