

Three-Dimensional Determination of the Coordinates of Individual Atoms in Materials

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In 1959, Richard Feynman challenged the electron microscopy community to locate the positions of individual atoms in substances (1). Over the last fifty-five years, significant advances have been made in electron microscopy. With the development of aberration-corrected electron optics (2,3), two-dimensional transmission electron microscopy (TEM) imaging has reached sub-0.5 Å resolution (4). Using scanning TEM (STEM) (5-6) and a three-dimensional (3D) image reconstruction method known as equal slope tomography (EST) (8,9), electron tomography has achieved 2.4 Å resolution and was applied to image the 3D core structure of edge and screw dislocations at atomic resolution (10,11). More recently, TEM has been used to determine the 3D atomic structure of gold nanoparticles by averaging 939 particles (12). Notwithstanding these important developments, however, Feynman's 1959 challenge – 3D localization of the positions of atoms in a substance without using averaging or a priori knowledge of sample crystallinity – remains elusive. Here, we report the determination of the 3D positions of 3,769 individual atoms in a tungsten needle sample with a precision of ~19 picometers and the identification of a point defect inside the sample in three dimensions (13). Our method consists of acquisition of a tilt series with an aberration-corrected STEM, 3D EST reconstructions of the tilt series, tracing individual atoms from the reconstructions, and 3D atomic model refinement, in which the crystallinity of the sample is not assumed. By comparing the positions of individual atoms with a body-centred-cubic (bcc) crystal lattice, we determine the atomic displacement field and the full strain tensor with a 3D resolution of ~1 nm³ and a precision of ~10⁻³, which are further verified by density functional theory calculations and molecular dynamics simulations (13). The ability to precisely localize the 3D positions of individual atoms in materials without assuming crystallinity, measure point defects, and determine the 3D atomic displacement field and the full strain tensor is expected to find important applications in both fundamental and applied sciences. [14]

References:

- [1] Feynman, R. P. in Feynman and Computation (ed. Hey, J. G.) 63–76 (Perseus Press, Cambridge, Massachusetts, 1999).
- [2] Haider, M., *et al.* *Nature* **392**, 768–769 (1998).
- [3] Batson, P. E., Dellby, N. & Krivanek, O. L. *Nature* **418**, 617–620 (2002).
- [4] Erni, R. *et al.* *Phys. Rev. Lett.* **102**, 096101(2009).
- [5] Midgley, P. A. & Weyland, M. STEM Tomography. In *Scanning Transmission Electron Microscopy: Imaging and Analysis*. ed. Pennycook, S. J. & Nellist, P. D. pp. 353–392 (Springer, 2011).
- [6] LeBeau, J. M. *et al.* *Phys. Rev. Lett.* **100**, 206101 (2008)
- [7] D. A. Muller. *Nature Materials* **8**, 263–270 (2009).
- [8] Miao, J., Förster, F. & Levi, O. *Phys. Rev. B* **72**, 052103 (2005).
- [9] Zhao, Y. *et al.* *Proc. Natl. Acad. Sci. USA* **109**, 18290–18294 (2012).
- [10] Scott, M. C. *et al.* *Nature* **483**, 444–447 (2012).
- [11] Chen, C.-C. *et al.* *Nature* **496**, 74–77 (2013).
- [12] Azubel, M. *et al.* *Science* **345**, 909–912 (2014).
- [13] Xu, R. *et al.* *Nature Mater.* **14**, 1099–1103 (2015).
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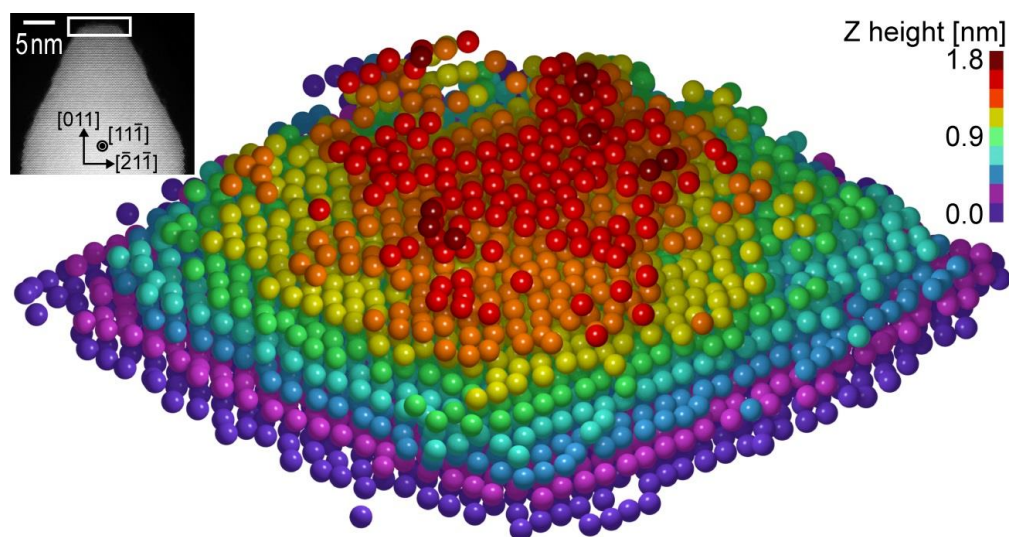


Figure 1. 3D positions of individual atoms in a tungsten needle sample revealed by electron tomography. The experiment was conducted using an aberration-corrected STEM. A tilt series of 62 projections was acquired from the sample by rotating it around the [011] axis. The inset shows a representative projection at 0°. After post-processing, the apex of the sample (labelled with a rectangle in the inset) was reconstructed by the EST method. The 3D positions of individual atoms were then traced from the reconstructions and refined using the 62 experimental projections. The 3D atomic model of the sample consists of 9 atomic layers along the [011] direction, labelled with crimson (dark red), red, orange, yellow, green, cyan, blue, magenta and purple from layers 1 to 9, respectively. (Ref. 13)