Revitalizing uncorrected electron microscopes by a low-cost, plug-and-play spherical aberration corrector using a sculpted thin film

Lu, P.[1], Shiloh, R.[2], Remez, R.[2], Jin, L.[1], Lereah, Y.[2], Tavabi, A.H.[1], Dunin-Borkowski, R.E.[1] and Arie, A.[2]

[1] Forschungszentrum Jülich, Germany, [2] Tel Aviv University, Israel

The development of the transmission electron microscope (TEM) heralded a new age in the characterization of the atomic structure of matter. However, for a long time its spatial resolution was limited to approximately 100 times the electron wavelength, as a result of the unavoidable positive spherical aberration of electron lenses, as described by Otto Scherzer in a seminal paper in 1936 [1]. His theorem is based on a number of constraints, including the fact that electromagnetic fields generated by round lenses are axially symmetric and static and that the neighborhood of the optical axis is charge-free. He subsequently made several proposals for correcting spherical aberration by relaxing these constraints [2]. It was only two decades ago that Haider et al. [3] and Krivanek et al. [4] finally succeeded in experimentally correcting spherical aberration in the imaging and probe-forming lenses of electron microscopes, respectively, by using non-rotationally-symmetric multipole lenses. However, such aberration correctors require major changes to an electron column, additional stable power supplies and water cooling, precise manufacture, accurate alignment and dedicated operation protocols. Thus, the only practical way to improve the resolution of an uncorrected TEM is to replace it altogether, at considerable cost, by a new aberration-corrected microscope.

An alternative approach involves imparting a structured phase shift to the electron beam by means of changes in the mean inner potential and thickness of a material. The mean inner potential is related to the charge density distribution in a material [5], and can therefore be used to violate Scherzer's charge-free constraint [2]. Although extensive research has been carried out in this direction, attempts to fabricate phase-shifting thin films that act as spherical aberration correctors were never successfully translated into practice. Here, we describe how the primary spherical aberration of the probe-forming optics in a scanning transmission electron microscope (STEM) can now be compensated by using a refractive thickness-modulated thin film [6]. We fabricated a thin film corrector using focused ion beam milling with nanometer precision and installed it in the Condenser 2 aperture plane in a 300 kV uncorrected FEI microscope with a known spherical aberration coefficient C_s of 2.7 mm (Figs 1a-b). High-angle annular dark-field (HAADF) STEM images of crystalline Si <110> showed a clear improvement in spatial resolution when using the corrector, allowing 136-pm-separated dumbbells to be resolved (Figs 1c-h). Through-focus series of Ronchigrams collected from $SrTiO_3$ <100> both without (Fig. 2a) and with (Fig. 2b) the thin film corrector demonstrate an enhancement of the aberration-free angle from ~7.4 to ~12 mrad, in accordance with its intended purpose of phase modulation and spherical aberration correction.

In summary, a thin film corrector can be implemented as an immediate low-cost upgrade to existing uncorrected electron microscopes as a plug-and-play tool without re-engineering the electron column or complicated operation protocols. The principle could be extended to the correction of additional aberrations of electron lenses.

References

[1] O. Scherzer, Zeitschrift Für Phys. 101, 593 (1936).
[2] O. Scherzer, J. Appl. Phys. 20, 20 (1949).
[3] M. Haider, H. Rose, S. Uhlemann, E. Schwan, B. Kabius, and K. Urban, Ultramicroscopy 75, 53 (1998).
[4] O. L. Krivanek, N. Dellby, and A. R. Lupini, Ultramicroscopy 78, 1 (1999).
[5] M. Beleggia, L. C. Gontard, and R. E. Dunin-Borkowski, J. Phys. D. Appl. Phys. 49, 294003 (2016).
[6] R. Shiloh*, R. Remez*, P.-H. Lu*, L. Jin, Y. Lereah, A. H. Tavabi, R. E. Dunin-Borkowski, A. Arie, arXiv:1705.05232 (2017).
[7] This work was supported by the German Research Foundation (German-Israeli Project cooperation Grant) and the Israel Science Foundation (Grant No. 1310/13). The authors are grateful to Les J. Allen and Juri Barthel for fruitful discussions.

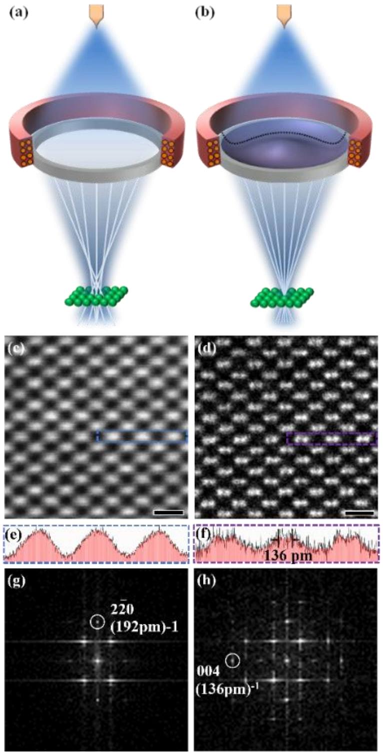

Figure 1. (a-b) Schematic diagrams of the setup for spherical aberration correction using a thin film corrector. (c-h) HAADF-STEM images and their Fourier power spectra of a Si <110> crystal imaged (c-g) without and (d-h) using a thin film corrector. Si <110> dumbbells with 136 pm separation can only be resolved after correction. The scale bars in (c) and (d) are 500 pm.

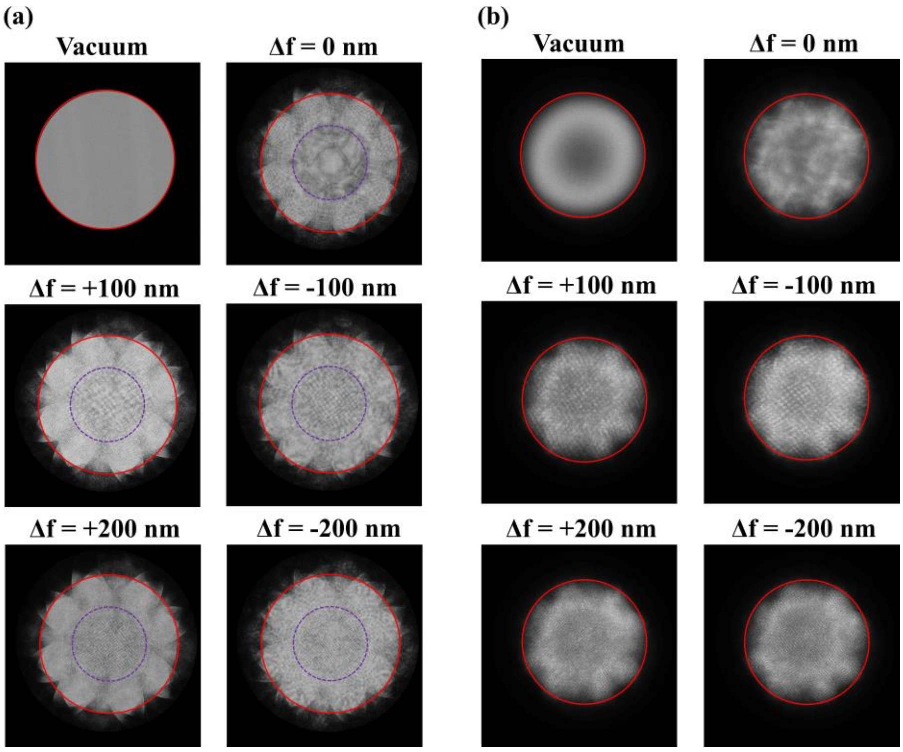

Figure 2. Through-focus series of Ronchigrams recorded from a $SrTiO_3$ <100> crystal (a) without a corrector at a convergence semi-angle of 14 mrad (red circles) and (b) using a thin film corrector at a convergence semi-angle of 12 mrad (red circles). The aberration-free angle indicated by the non-distorted lattice is (a) ~7.4 mrad (purple circles) without the corrector and (b) the full aperture range, i.e., ~12 mrad, when using the thin film corrector.