Supplementary information: Aberration-corrected transmission electron microscopy with Zernike phase plates

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Figure S1: (a) Sketch of employed objective aperture lamella with inserted SEM image of mounted grid carrying an amorphous carbon thin film used as Zernike PP. (b) Enlarged SEM image of single hole of the grid with hole for Zernike PP in center. Scale bar is 50\,\mu m.
SI1. Calculation of a PCTF with PPs

The PCTFs considering partial spatial coherence and a realistic phase-shift distribution of the PPs is calculated in dependence of the spatial frequency (u) by the following equation:

$$PCTF(u) = \sum_{\delta\xi} B_{\delta\xi} \exp \left[ -\frac{(\delta\xi_x^2 + \delta\xi_y^2)}{2\Xi^2} \right] \sum_{\delta Z} \frac{B_{\delta Z}}{2\pi\Delta} \exp \left[ -\frac{\delta Z^2}{2\Delta^2} \right] \cdot \sin \left[ \chi(u, Z + \delta Z, \delta\xi) \right] \cdot E_S(u, Z + \delta Z) \cdot A_{PP}(u, \delta\xi) \quad (1)$$

where the first sum (partial spatial coherence in combination with a PP) goes over the (x,y) position $\delta\xi$ in the ZOB with width $\Xi$ (identical in x and y direction) and with $B_{\delta\xi}$ the normalization factor in order that $\sum_{\delta\xi} B_{\delta\xi} \exp \left[ -\frac{\delta\xi_x^2 + \delta\xi_y^2}{2\Xi^2} \right] = 1$. The second sum goes over the defocus variation $\delta Z$ and calculates the effects of partial temporal coherence. $B_{\delta Z}$ is the normalization factor for the second sum in order that $\sum_{\delta Z} B_{\delta Z}/(2\pi\Delta) \cdot \exp \left[ -\frac{\delta Z^2}{2\Delta^2} \right] = 1$ with the focal spread $\Delta$. The wave aberration function $\chi(u, Z + \delta Z, \delta\xi)$ is given by:

$$\chi(u, Z + \delta Z, \delta\xi) = \pi(Z + \delta Z)\lambda u^2 + \frac{\pi}{2} C_S \lambda^3 u^4 + \phi_{PP}(u, \delta\xi) + ... \quad (2)$$

with the electron wavelength $\lambda$, defocus $Z$, constant of spherical aberration $C_S$ and the phase-shift distribution of the PP $\phi_{PP}$, which is calculated in dependence of $u$ for every position in the ZOB $\delta\xi$. The envelope considering spatial coherence with the semi-convergence angle $\alpha$ is given by

$$E_S(\vec{u}) = \exp \left[ -\frac{\alpha^2}{4\lambda^2} \left( \frac{\partial \chi}{\partial \vec{u}} \right)^2 \right] \quad (3)$$

We neglected the dependence of $E_S$ on the position within the ZOB $\delta\xi$ (corresponding to different incident angles) as for the studied defocus values, $E_S$ is negligible in comparison with the effect of temporal coherence.
SI2. Beam convergence

Figure S2: (a,b) Two TEM images of an amorphous C thin film acquired with Zernike phase plate (5 µm hole radius) at different coherent convergence angles of (a) 0.07 and (b) 0.85 mrad. (c) Comparison of power spectra at different coherent convergence angles reveals similar appearance of Thon rings. Zernike PP hole edge is indicated by dashed green circle. (d) Rotationally averaged line profiles of power spectra show an increasing intensity and a minor change at the position of the PP edge marked by the black vertical line. Scale bar is (a,b) 7 nm and (c) 0.6 nm⁻¹.
### Table S1: Calibration of zero-order beam size in back focal plane (coinciding with PP plane) for different illuminated areas for a 50 µm Condenser (C2) aperture ranging from last parallel setting (624 nm) to coherent converging illumination. Data is obtained by acquisition of diffraction patterns without specimen inserted and measuring the size of the ZOB. The size for parallel illumination is probably overestimated due to the high current density and a saturation of the camera.

<table>
<thead>
<tr>
<th>Illuminated area / nm</th>
<th>ZOB size / nm⁻¹</th>
<th>ZOB size / mrad</th>
<th>ZOB size / nm in BFP</th>
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<tr>
<td>624 (parallel)</td>
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<td>0.07</td>
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<td>0.29</td>
<td>514</td>
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<tr>
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<tr>
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<td>1.41</td>
<td>2540</td>
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</table>
SI3. Stability of unheated Zernike PP

Figure S3: (a) Four power spectra of TEM images of a very thin amorphous C thin film acquired before (0s) and after insertion of the (unheated) Zernike PP thin film with no hole in the center. The strong buildup of the charge/phase shift is already evident in the first power spectra after insertion (0.24s) and further increases during the series. (b) TEM image at the end of the series shows strong contrast. (c) Using the Thon-ring positions (see d) of each power spectrum allows to detect the spatial distribution of the charge/phase shift build up on the PP. The negative phase shift increases in amplitude and also in width. (d) Plot of rotationally averaged profiles of the power spectra shows the strong change of the phase-contrast transfer, especially at lower spatial frequencies. Scale bars are (a) 0.3 nm$^{-1}$ and (b) 20 nm.
Figure S4: (a-c) Three high-resolution aberration-corrected TEM images of Fe$_3$O$_4$ nanoparticles acquired with Zernike phase plate (3.5 µm hole radius) without changing any parameter. Time passed after initial image is indicated. Image appearance is stable for over one minute, which is proven by the (d) line profiles taken along the arrow indicated in (a). Scale bar is 6 nm and 1 nm$^{-1}$ for inset power spectrum in (a).
Figure S5: (a,b) Two high-resolution aberration-corrected TEM images of a MoS$_2$ nanoparticle acquired with Zernike phase plate (4 µm hole radius) without changing any parameter. (c) Comparison of power spectra reveals similar appearance. (d) Line profiles taken along the arrow indicated in (a) shows similar contrast and indicates stable imaging conditions. Scale bar is 5 nm and power spectra have a size of 20 nm$^{-1}$ x 20 nm$^{-1}$. 
Figure S6: (a,c) Two HRTEM images of a graphene sample acquired at 80 kV (a) with and (c) without a Zernike phase plate (5 µm hole radius) and (b,d) corresponding power spectra. The charge build up under the reflections of the graphene manifests itself in a distortion of the image and the corresponding power spectra. Scale bars are (a,c) 3 nm and (b,d) 2 nm⁻¹.
SI4. Comparison of HRTEM images with and without PP

Figure S7: Aberration-corrected TEM image of aC film with Fe$_3$O$_4$ nanoparticles and corresponding power spectra acquired (a) with and (b) without Zernike phase plate (radius of 4 μm) without changing any additional parameter. Defocus is measured to (a) 266 nm and (b) 296 nm, phase shift is (a) 0.45π. Mean image intensity is (a) 197.8 and (b) 200.3. Scale bars are 5 nm and power spectra/fitted spectra are displayed up to 5 nm$^{-1}$ and 2.86 nm$^{-1}$, respectively.
Figure S8: Aberration-corrected TEM image of identical sample region of an aC film with Fe$_{3.4}$O$_4$ nanoparticles and corresponding power spectra acquired at 80 kV (a,b) with and (c,d) without Zernike phase plate (radius of 6 µm). Mean image intensity is (a) 58.4 and (b) 61.4. Scale bars are 4 nm and power spectra are displayed up to 5.6 nm$^{-1}$. 