Electron Beam Aberration Correction Using Optical Near Fields

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The interaction between free electrons and optical near fields is attracting increasing attention as a way to manipulate the electron wave function in space, time, and energy. Relying on currently attainable experimental capabilities, we design optical near-field plates to imprint a lateral phase on the electron wave function that can largely correct spherical aberration without the involvement of electric or magnetic lenses in the electron optics, and further generate on-demand lateral focal spot profiles. Our work introduces a disruptive and powerful approach toward aberration correction based on light-electron interactions that could lead to compact and versatile time-resolved free-electron microscopy and spectroscopy.

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The development and widespread use of spatial light modulators have revolutionized optics by enabling an increasing degree of control over light beam propagation. Likewise, the extension of this concept to electron optics could provide the means for controlling the electron wave function and its interactions with atomic-scale samples. Electron microscopes already reach precise spatial and temporal control over the amplitude and phase of the wave function of beam electrons employed as sample probes. Over the last decades, costly and sophisticated arrangements of magnetostatic and electrostatic lenses have been engineered to eliminate electron-optics aberrations [1,2], making it possible to focus electron beams with subangstrom precision in state-of-the-art scanning transmission electron microscopes. These capabilities are crucial for atomic-scale imaging and spectroscopy [3,4].

Parallel efforts have led to the development of amplitude and phase reconstruction techniques such as ptychography [5] or electron tomography and holography [6], which have proved useful in both imaging low-contrast samples [7,8] and acquiring additional information on the sample, such as electric or magnetic field distributions [9–11]. An alternative approach has consisted in preparing electron beams with on-demand focal spot phase and intensity distributions designed to introduce phase contrast and selectively interact with targeted types of excitations such as plasmons of specific multipolar symmetry [12] or chiral modes and materials magnetic properties [13,14]. Such phase-shaped electron beams can be obtained through diffraction by a static phase plate [12,15–17] or ingenious use of lens aberrations [18,19]. A recent work also demonstrates programmable electron phase plates based on arrays of electrically biased transmission elements [20].

The interaction of free electrons with optical near fields in illuminated nanostructures opens exciting possibilities as a further mechanism to control the electron wave function.

This phenomenon has been exploited to develop the so-called photon-induced near-field electron microscopy [21], which has been the subject of intense experimental [21–40] and theoretical [41–48] efforts. By synchronizing the arrival of ultrashort electron and laser pulses near the sample, the former can undergo stimulated absorption or emission of up to hundreds of photons [39,40]. This technique has been predicted to imprint optical phase on the lateral electron wave function [45], which has been demonstrated to generate vortex electron beams via photon-to-electron angular momentum transfer [37]. The synergetic combination of spatial light modulators and ultrafast electron microscopy constitutes a powerful platform for the control of free-electron wave functions, including the possibility of compensating beam aberrations and shaping the focal spot. While other alternatives to traditional electron-optics components have been suggested, such as using nanofabricated structures or thin films for wave front shaping and correction [49,50], our target is to introduce a more versatile and tunable solution.

In this Letter, we theoretically demonstrate the correction of spherical aberration in an electron beam upon transmission through an illuminated thin film, where light-electron phase transfer compensates for the undesired deviation of the transverse electron wave function from a spherical wave front, thereby resulting in nearly unaberrated focusing down to subangstrom focal spots. The proposed implementation of this type of photonic aberration corrector (PAC) in an electron microscope is schematically depicted in Fig. 1, where the new element is placed after the condenser along the electron optical path in order to imprint the required phase on the electron wave function to compensate for the aberration produced by subsequent electron-optics focusing elements. The PAC consists of an optically opaque electron-transparent thin film (e.g., a metal film deposited on a Si₃N₄ membrane, as\
already used in photon-induced near-field electron microscopy studies [25,35], but we discuss additional options in Sec. S3 of the Supplemental Material [51] on which a lateral optical pattern is projected with diffraction-limited spatial resolution. Electron interaction with semi-infinite light fields [35] in this film then produces energy sidebands in the transmitted electrons that can be optimized to accommodate ~1/3 of the electrons in the first sideband (i.e., electrons that have gained one photon energy). A monochromator inserted right after the PAC removes the rest of the energy sidebands before entering an electron-optics module for focusing at the sample. Aberration correction is thus performed through the PAC phase plate, which represents an alternative to traditional aberration correctors. This type of design inherits the flexibility of light patterning through spatial light modulators, here demonstrated for aberration correction, but also enabling arbitrary shaping of the electron focal spot.

**Electron beam propagation through the electron microscope.**—We represent fast electrons by their space- and time-dependent wave function ψz(R)e−iE0t, where we consider monochromatic electrons of energy E0 that depend on transverse coordinates R = (x, y) at each propagation plane determined by z. Free propagation from z′ to z is described by the expression [44,52]

\[
ψ_z(R) = \int \int \frac{d^2Qd^2R'}{(2π)^2} e^{i(Q \cdot (R-R') + q_z(z-z'))}ψ_{z'}(R'),
\]

where the outer integrals extend over transverse wave vectors Q, qz = \sqrt{q_0^2 - Q^2} is the longitudinal wave vector, \(\hbar q_0 = m_\gamma v\) and v are the average electron momentum and velocity vectors, respectively, \(\gamma = 1/\sqrt{1-v^2/c^2}\) is the Lorentz factor, \(m_\gamma\) is the electron rest mass, and c is the speed of light in vacuum. In what follows, we focus on electron beams of well-defined chirality, characterized by an azimuthal orbital quantum number \(m\), such that the wave function takes the form \(ψ_z(R) = ψ_{z'}(R)e^{im\phi}\), where \((R, \phi)\) are polar coordinates. Additionally, electrons in microscopes are collimated and therefore safely described in the paraxial approximation, for which \(q_z \approx q_0 - Q^2/2q_0\). These considerations allow us to carry out some of the above integrals to find [51]

\[
ψ_z(R) = (F^m_{\chi, z'} \cdot ψ_{z'})|_R \\
≡ (−i)^{m+1} q_{z-z'}^m e^{iq_{z-z'}(z-z')} \\
× \int_0^\infty R'dR'J_m(q_{z-z'}R')e^{i\xi_{z-z'}(R^2+R'^2)/2}ψ_{z'}(R'),
\]

where \(ξ_{z-z'} = q_0(z-z')\), \(J_m\) is a Bessel function, and we implicitly define the free-propagation operator \(F^m\) using matrix notation with a dot standing for integration over the radial coordinate \(R\).

**FIG. 1.** Proposed experimental arrangement incorporating a photonic aberration corrector (PAC) to mitigate electron spherical aberration through an electron optical phase plate. The PAC module (light orange frame) is placed just before the electron-optics focusing module (electromagnetic lenses inside the light blue frame) at a distance \(z_{L, in} - z_{XO}\) from the crossover point. A coherent electron wave is prepared by condenser lenses placed after an electron gun. Both the transmitted beam and the light wave prepared by a spatial light modulator are restricted by a circular aperture of radius \(R_{max}\).

Transmission through the microscope sketched in Fig. 1 results in an electron wave function at the sample given by

\[
ψ_{\chi, sample} = F^m_{\chi, sample - z_{L, out}} \cdot T_L \cdot F^m_{z_{L, in} - z_{PAC}} \cdot T_{PAC} \cdot ψ_{\chi, PAC},
\]

where \(ψ_{\chi, PAC}\) represents the electron incident on the plane of the corrector at \(z_{PAC}\), while \(T_{PAC}\) and \(T_L\) account for transmission through the PAC and electron lenses (orange and blue frames in Fig. 1, respectively). For a thin lens, the latter is well described by [53,54]

\[
T_L|_{RR} = δ(R - R')e^{i[q_0\Theta/2]} e^{-i[q_0\Theta/2]} Θ(R_{max} - R),
\]

where \(f\) is the focal distance, a pupil blocks propagation above a radial distance \(R_{max}\), and the phase \(χ(\Theta)\) accounts for aberrations in the lenses as a function of exit angle \(θ = R/(z_{sample} - z_{L, out})\). Here, we concentrate on spherical aberration, so we express \(χ(\Theta) = C_q q_0 \Theta^4/4\) in terms of the (length) coefficient \(C_q\) [52,55]. For simplicity, in what follows we consider a spherical wave \(ψ_{\chi, PAC}(R) ∝ e^{i[q_0\Theta/2]}(z_{PAC} - z_{XO})\) with \(m = 0\) emerging from a perfect point-like crossover that can be produced by another set of condenser lenses placed after the electron source and
profile of subangstrom width treated in Fig. 2(c). We plot the normalized beam electron density $|\psi_{\text{sample}}|^2$ [Eq. (2)] obtained through (a) aberration-free electron optics; (b) electron optics introducing spherical aberration with $C_3 = 1$ mm; and (c) same as (b) including a PAC module. We consider 60-keV electrons, a focal distance $f = 1$ mm, $z_L - z_{so} = 40 f$, and an aperture $R_{\text{max}} = 30 \mu m$. The corrected beam profile in (c) is calculated by using a realistic spatial dependence of the coupling parameter $\beta$ as a function of radial distance $R$ in the PAC [panel (e), obtained from Eqs. (6) and (8) with $\lambda_0 = 500$ nm light wavelength], which differs from the ideal $\beta$ that is needed to perfectly correct the aberration [panel (d), Eq. (7)].

preceding the PAC. In practice, the condenser lenses together with collimating apertures provide lateral coherence of the electron beam over the aperture restricted by $R_{\text{max}}$, as needed for a correct performance of the PAC. We note that the PAC can also correct for aberrations introduced by the preceding lenses, but for demonstration purposes, here we only consider the electron-optics aberration produced by the objective lens placed after the PAC. Additionally, we take the PAC to coincide with the near and far sides of the optical lenses at the virtual plane $z = z_L$.

In the absence of aberrations ($\chi = 0$), $\psi_{\text{sample}}$ is focused at a position $z = z_{\text{sample}}$ determined by the lens formula $1/(z_{\text{sample}} - z_L) + 1/(z_L - z_{so}) = 1/f$ [51]. This is illustrated in Fig. 2(a) for 60-keV electrons (~5 pm wavelength) with $z_L - z_{so} = 40 f$ (implying $z_{so} - z_L \approx f$), $f = 1$ mm, and $R_{\text{max}} = 30 \mu m$. The focal spot is limited by diffraction at the aperture, yielding a $\psi_{\text{sample}} \sim J_1 (R/\Delta)$ transverse profile of subangstrom width $\sim \Delta = f/q_0 R_{\text{max}} = 0.26 \AA$. In contrast, a typical spherical aberration corresponding to $C_3 = 1$ mm produces a substantially broadened and shifted focus [Fig. 2(b)], accompanied by satellite foci along the optical axis.

**Electron optical phase plate.**—We intend to cancel the aberration phase $\chi$ by imprinting an additional phase on the electron wave function through the interaction with an optical near field [35,44,45]. For this purpose, we consider an optically opaque electron-transparent film subject to external illumination, a configuration that has been demonstrated to produce large coupling to the electrons [35]. We assume that inelastic scattering due to interaction with the film material can be neglected, while the additional phase acquired through this interaction should only contribute as a position-independent overall factor. The condition of weak inelastic scattering is met by thin metal films with thickness $<10$ nm, well below the electron inelastic mean free path [51,56]. Alternatively, a thin dielectric membrane can be used together with more intense optical illumination, as we demonstrate in Sec. S3 of the Supplemental Material [51].

The transmitted electron wave function consists of sidebands of energies $E_0 + \hbar \omega_0$ separated from the incident energy by multiples $\ell$ ($<0$ for loss, $>0$ for gain) of the photon energy $\hbar \omega_0$. In the nonrecoil approximation, the wave function associated with each transmitted sideband $\ell$ consists of the incident wave function times a multiplicative factor accounted for by the operator $[35,46]$

$$T_{\text{PAC}}|_{RR'} = \delta(R - R') J_\ell (2|\beta|) e^{i(\arg(-\beta)} ,$$

where the coupling coefficient

$$\beta(R) = \frac{e}{\hbar \omega_0} \int_{-\infty}^{\infty} dz E_z(R, z) e^{-i\omega_0 z/v}$$

captures the electron-light interaction through the (along-the-beam) $E_z$ component of the optical electric-field amplitude, which bears a dependence on transverse coordinates $R$ that can be controlled through a spatial light modulator. We first consider axially symmetric illumination [implying $m = 0$ in Eq. (2)] and express the incident optical field $E_z = \int k_\parallel dk_\parallel f(0, k_\parallel R) e^{i k_\parallel z - i \omega_0 t}$ as a combination of cylindrical Bessel waves with in- and out-of-plane wave vector components $k_\parallel$ and $k_z = \sqrt{k_0^2 - k_\parallel^2}$, respectively, limited by the free-space light wave vector $k_0 = \omega_0/c$. Upon insertion of this field into Eq. (5), we find
\[ \beta(R) = \int_0^{k_0} k_\parallel dk_\parallel J_0(k_\parallel R) \beta_{k_\parallel}, \] (6)

where the coefficient \( \beta_{k_\parallel} \) is proportional to \( \alpha_{k_\parallel} \) and also includes light components reflected at the film [35]; we stress that \( \beta_{k_\parallel} \) can therefore be controlled through the applied angular light profile \( \alpha_{k_\parallel} \).

**Design of the PAC field profile.**—We now design the PAC based on an electron optical phase plate in which \( \ell' = 1 \) is selected, while other sidebands (\( \ell' \neq 1 \)) are filtered out by a monochromator (Fig. 1), using for example a Wien filter [1] (easily capable of separating peaks with an energy difference \( \hbar\omega \sim 1 \text{ eV} \)). The aberration phase \( \chi \) introduced through \( T_L \) [Eq. (3)] can then be eliminated from Eq. (2) by setting

\[ \arg \{-\beta(R)\} = -\chi(\theta) \] (7)

in \( T_{\text{PAC}} \) [Eq. (4)], where \( R = \theta(z_{\text{sample}} - z_L) \). We can maximize the current by imposing \( |\beta| = \beta_0 \approx 0.92 \), which yields an absolute maximum fraction of the \( \ell' = 1 \) sideband

\[ J_1^2(2\beta_0) \approx 34\% \] [51]. The PAC then involves a reduction in electron current by a factor of \( \sim 2/3 \), which, together with beam losses due to interaction with the PAC material, represents a drawback compared to traditional solutions for aberration correction in which most of the beam current is transmitted. The spatial dependence of \( \beta = -\beta_0 e^{-i\chi} \) required to produce perfect aberration correction and maximum \( \ell' = 1 \) current is presented in Fig. 2(d) according to Eq. (7) for \( E_0 = 60 \text{ keV} \) and \( f = C_3 = 1 \text{ mm} \). However, optical diffraction at the used finite light wavelength \( \lambda_0 = 2\pi/k_0 \) limits the profile of \( \beta \) that can be achieved in practice using far-field illumination. We find a nearly optimum realistic profile by setting \( \beta = -\beta_0 e^{-i\chi} \) in Eq. (6) and approximately inverting this equation to yield

\[ \beta_{k_\parallel} = -\beta_0 \int_0^{R_{\text{max}}} RdRJ_0(k_\parallel R)e^{-i\chi(R/(z_{\text{sample}} - z_L))] \] (8)

(this inversion is only exact in the \( k_0 R_{\text{max}} \gg 1 \) limit). The coupling coefficient obtained by inserting Eq. (8) back into Eq. (6) is plotted in Fig. 2(e) for a photon wavelength \( \lambda_0 = 500 \text{ nm} \), which resembles the perfect-correction coefficient of Fig. 2(d) up to \( R \sim 20 \mu\text{m} \), but deviates substantially from that target value at larger \( R \) (i.e., where the phase \( \chi \) exhibits rapid variations over a distance \( \sim \lambda_0 \)). Although the resulting corrected beam profile plotted in Fig. 2(c) is not perfect, it still provides an impressive improvement in electron focusing compared to the aberrated spot shown in Fig. 2(b) [51]. We note that the degree of correction increases when \( \lambda_0 \) is made smaller relative to \( R_{\text{max}} \), as we show in Fig. 3, which further predicts a remarkable aberration compensation using blue light. We also verified that aberration correction by the PAC does not introduce any undesired beam tails (see Fig. S5 in the Supplemental Material [51]).

The PAC can be fed using light with definite chirality \( m \) [i.e., \( E_z(R, z) = E_z(R, z)e^{im\phi} \)], which directly translates through Eqs. (4) and (5) into \( T_{\text{PAC}} \propto e^{im\phi} \), still described by Eqs. (6) and (8) with \( J_0 \) substituted by \( J_m \). Results for the transverse profile of the electron focus using chiral PACs with \( m = 1 \) and 3 are compared with the \( m = 0 \) profile in Fig. 4, revealing the formation of donuts...
associated with an electron wave function of $e^{im\varphi}$ azimuthal symmetry. More complex profiles are possible, which should be reachable using a spatial light modulator to project light on the PAC. For example, Fig. 4(d) shows the result obtained by projecting a symmetric combination of $m = 1$ and $m = -1$ light.

Conclusion.—In summary, we propose the use of electron optical phase plates as a way of tailoring the amplitude and phase of the electron-transverse wave function in an electron beam. Specifically, we theoretically demonstrate correction of spherical aberration without the involvement of complex electron-optics elements. This concept can be straightforwardly applied to eliminate any undesired distortions introduced by electron optics in both standard and ultrafast electron microscopes. Although we apply analytical methods to produce a proof-of-principle design, improvement in the capabilities of such phase plates could be gained through machine learning, which should enable the design of more complex electron spot shapes or correction of a general combination of aberrations following the example of light optics [57]. Ultimately, iterative improvement of the PAC could be attained through a feedback loop involving measurement of the electron spot and modification of the projected light profile. Additionally, temporal manipulation of the imprinted optical phase offers interesting possibilities for the exploration of sample dynamics through time-varying electron spot profiles. The versatility and compactness of electron optical phase plates hold potential for active control of electron wave functions beyond the present application in aberration correction.

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[1] P. Hawkes and J. Spence, Springer Handbook of Microscopy (Springer Nature, Switzerland, AG, 2019).
[2] M. Haider, H. Rose, S. Uhlemann, E. Schwan, B. Kabius, and K. Urban, Ultramicroscopy 75, 53 (1998).
[3] P.E. Batson, N. Dellby, and O.L. Krivanek, Nature (London) 418, 617 (2002).
[4] D. A. Muller, L. Fitting Kourkoutis, M. Murfitt, J. H. Song, H. Y. Hwang, J. Silcox, N. Dellby, and O.L. Krivanek, Science 319, 1073 (2008).
[5] Y. Jiang, Z. Chen, Y. Han, P. Deb, H. Gao, S. Xie, P. Purohit, M. W. Tate, J. Park, S. M. Gruner et al., Nature (London) 559, 343 (2018).
[6] P.A. Midgley and R.E. Dunin-Borkowski, Nat. Mater. 8, 271 (2009).
[7] H.-W. Fink, H. Schmid, E. Ermantraut, and T. Schulz, J. Opt. Soc. Am. A 14, 2168 (1997).
[8] F. S. Yasin, T. R. Harvey, J. J. Chess, J. S. Pierce, C. Ophus, P. Ercius, and B. J. McMorran, Nano Lett. 18, 7118 (2018).
[9] M. R. McCartney, N. Agarwal, S. Chung, D. A. Cullen, M.-G. Han, K. He, L. Li, H. Wang, L. Zhou, and D. J. Smith, Ultramicroscopy 110, 375 (2010).
[10] O. Nicoletti, F. de la Peña, R. K. Leary, D. J. Holland, C. Ducati, and P.A. Midgley, Nature (London) 502, 80 (2013).
[11] K. Shibata, A. Kovács, N. S. Kiselev, N. Kanazawa, R. E. Dunin-Borkowski, and Y. Tokura, Phys. Rev. Lett. 118, 087202 (2017).
[12] G. Guzzinati, A. Beche, H. Lourenco-Martins, J. Martin, M. Kociak, and J. Verbeeck, Nat. Commun. 8, 14999 (2017).
[13] S.M. Lloyd, M. Babiker, G. Thirunavukkarasu, and J. Yuan, Rev. Mod. Phys. 89, 035004 (2017).
[14] J. Rusz and S. Bhowmik, Phys. Rev. Lett. 111, 105504 (2013).
[15] J. Verbeeck, H. Tian, and P. Schattschneider, Nature (London) 467, 301 (2010).
[16] B.J. McMorran, A. Agrawal, I.M. Anderson, A. A. Herzing, H. J. Lezec, J. J. McClelland, and J. Unguris, Science 331, 192 (2011).
[17] R. Shiloh, Y. Lereah, Y. Lilach, and A. Arie, Ultramicroscopy 144, 26 (2014).
[18] P. Schattschneider, M. Stöger-Pollach, and J. Verbeeck, Phys. Rev. Lett. 109, 084801 (2012).
[19] L. Clark, A. Béché, G. Guzzinati, A. Lubk, M. Mazilu, R. Van Boxem, and J. Verbeeck, Phys. Rev. Lett. 111, 064801 (2013).
[20] J. Verbeeck, A. Béché, K. Müller-Caspary, G. Guzzinati, M. A. Luong, and M. Den Hertog, Ultramicroscopy 190, 58 (2018).
[21] B. Barwick, D.J. Flannigan, and A.H. Zewail, Nature (London) 462, 902 (2009).
[22] F.O. Kirchner, A. Gliserin, F. Krausz, and P. Baum, Nat. Photonics 8, 52 (2014).
[23] L. Piazza, T.T.A. Lummen, E. Quiñonez, Y. Murooka, B. Reed, B. Barwick, and F. Carbone, Nat. Commun. 6, 6407 (2015).
[24] A. Feist, K.E. Echternkamp, J. Schauss, S.V. Yalunin, S. Schäfer, and C. Ropers, Nature (London) 521, 200 (2015).
[25] T.T.A. Lummen, R.J. Lamb, G. Berruto, T. LaGrange, L.D. Negro, F.J. García de Abajo, D. McGrouther, B. Barwick, and F. Carbone, Nat. Commun. 7, 13156 (2016).
[26] K.E. Echternkamp, A. Feist, S. Schäfer, and C. Ropers, Nat. Phys. 12, 1000 (2016).
[27] C. Kealhofer, W. Schneider, D. Ehberger, A. Ryabov, F. Krausz, and P. Baum, Science 352, 429 (2016).
[28] A. Ryabov and P. Baum, Science 353, 374 (2016).
[29] G.M. Vanacore, A.W.P. Fitzpatrick, and A.H. Zewail, Nano Today 11, 228 (2016).
[30] Y. Morimoto and P. Baum, Nat. Phys. 14, 252 (2018).
[31] M. Kozák, J. McNeur, K.J. Leedle, H. Deng, N. Schönenberger, A. Ruehl, I. Hartl, J. S. Harris, R. L. Byer, and P. Hommelhoff, Nat. Commun. 8, 14342 (2017).
[32] A. Feist, N. Bach, T.D.N. Rubiano da Silva, M. Mäller, K.E. Priebel, T. Domräse, J.G. Gatzmann, S. Rost, J. Schauss, S. Strauch et al., Ultramicroscopy 176, 63 (2017).
K. E. Priebe, C. Rathje, S. V. Yalunin, T. Hohage, A. Feist, S. Schäfer, and C. Ropers, Nat. Photonics 11, 793 (2017).

E. Pomarico, I. Madan, G. Berruto, G. M. Vanacore, K. Wang, I. Kaminer, F. J. García de Abajo, and F. Carbone, ACS Photonics 5, 759 (2018).

G. M. Vanacore, I. Madan, G. Berruto, E. Pomarico, R. J. Lamb, D. McGrouther, I. Kaminer, B. Barwick, F. J. García de Abajo, and F. Carbone, Nat. Commun. 9, 2694 (2018).

Y. Morimoto and P. Baum, Phys. Rev. A 97, 033815 (2018).

G. M. Vanacore, G. Berruto, I. Madan, E. Pomarico, P. Biagioni, R. J. Lamb, D. McGrouther, O. Reinhardt, I. Kaminer, B. Barwick et al., Nat. Mater. 18, 573 (2019).

K. Wang, R. Dahan, M. Shentcis, Y. Kauffmann, S. Tsesses, and I. Kaminer, Nature (London) 582, 50 (2020).

S. Nehemia, R. Dahan, M. Shentcis, O. Reinhardt, Y. Adiv, K. Wang, O. Beer, Y. Kurman, X. Shi, M. H. Lynch, and I. Kaminer, arXiv:1909.00757.

O. Kfir, H. Lourenço-Martins, G. Storeck, M. Sivis, T. R. Harvey, T. J. Kippenberg, A. Feist, and C. Ropers, Nature (London) 582, 46 (2020).

F. J. García de Abajo, A. Asenjo Garcia, and M. Kociak, Nano Lett. 10, 1859 (2010).

S. T. Park, M. Lin, and A. H. Zewail, New J. Phys. 12, 123028 (2010).

S. T. Park and A. H. Zewail, J. Phys. Chem. A 116, 11128 (2012).

F. J. García de Abajo, B. Barwick, and F. Carbone, Phys. Rev. B 94, 041404(R) (2016).

W. Cai, O. Reinhardt, I. Kaminer, and F. J. García de Abajo, Phys. Rev. B 98, 045424 (2018).

V. Di Giulio, M. Kociak, and F. J. García de Abajo, Optica 6, 1524 (2019).

O. Kfir, Phys. Rev. Lett. 123, 103602 (2019).

O. Reinhardt, C. Mechel, M. Lynch, and I. Kaminer, arXiv:1907.10281.

V. Grillo, A. H. Tavabi, E. Yucelen, P.-H. Lu, F. Venturi, H. Larocque, L. Jin, A. Savenko, G. C. Gazzadi, R. Balboni et al., Opt. Express 25, 21851 (2017).

R. Shiloh, R. Remez, P.-H. Lu, L. Jin, Y. Lereah, A. H. Tavabi, R. E. Dunin-Borkowski, and A. Arie, Ultramicroscopy 189, 46 (2018).

See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.125.030801 for a derivation of Eq. (1), a calculation of the coupling coefficient β for thin films, additional elements of the simulation with the PAC and the electron lenses at the same virtual plane, further details on the PAC, and a comparison of electron spot profiles with and without aberration correction.

L. J. Allen, M. P. Oxley, and D. Paganin, Phys. Rev. Lett. 87, 123902 (2001).

P. Hawkes, B. Lencová, and M. Lenc, J. Microsc. 179, 145 (1995).

R. F. Egerton, Physical Principles of Electron Microscopy: An Introduction to TEM, SEM, and AEM (Springer, New York, 2005).

D. M. Paganin, T. C. Petersen, and M. A. Beltran, Phys. Rev. A 97, 023835 (2018).

H. Shinotsuka, S. Tanuma, C. J. Powell, and D. R. Penn, Surf. Interface Anal. 47, 871 (2015).

O. Albert, L. Sherman, G. Mourou, T. B. Norris, and G. Vdovin, Opt. Lett. 25, 52 (2000).
S1. DERIVATION OF EQ. (1) IN THE MAIN TEXT

Because a monochromatic electron wave function $\psi_z(R)$ satisfies the Helmholtz equation $(\nabla^2_R + \partial_{zz} - q_0^2)\psi_z(R) = 0$ for fixed electron wave number $q_0$, its propagation from a transverse plane $z'$ to $z$ can be realized through the integral

$$\psi_z(R) = \int \frac{d^2 Q}{(2\pi)^2} e^{iQ\cdot R + q_0(z-z')} \int d^2 R' e^{-iQ\cdot R'} \psi_{z'}(R'),$$

(S1)

where we use the notation $R = (x, y)$, the $R'$ integral produces the Fourier transform along the transverse directions, each component of wave vector $(Q, q_z)$ with $q_z = \sqrt{q_0^2 - Q^2 + i0^+}$ and $\text{Im} \{q_z\} > 0$ evolves as a plane wave, and the $Q$ integral yields the inverse Fourier transform after propagation. We note that evanescent waves with $Q > q_0$ die away in the $q_0(z - z') \gg 1$ limit, so only $Q < q_0$ contributes to the integral in Eq. (S1) for propagation along a distance $z - z'$ spanning many electron wavelengths. Additionally, in the paraxial approximation for well collimated beams, only small components $Q \ll q_0$ contribute to Eq. (S1), so we can approximate $q_z \approx q_0 - Q^2/2q_0$. Using this expression and considering wave functions with an azimuthal dependence given by $\psi_z(R) = \psi_z(R)e^{im\varphi_R}$ in terms of a well-defined angular momentum number $m$, we can perform the integrals over $\varphi_R$ and $\varphi_Q$ in Eq. (S1) using the identity (Eq. (9.1.21) of Ref. [1])

$$\int_0^{2\pi} d\varphi e^{\pm iz \cos \varphi} e^{im\varphi} = 2\pi \delta_{m0} J_m(z).$$

We find

$$\psi_z(R) = e^{iq_0(z-z')} \int_0^{\infty} QdQ J_m(QR) e^{-iQ^2(z-z')/2q_0} \int_0^{\infty} R'dR' J_m(QR') \psi_{z'}(R')$$

$$= (-i)^{m+1} q_0 \int_0^{\infty} e^{iQ_0(z-z')} \int R'dR' \exp \left[ \frac{iQ_0(R^2 + R'^2)}{2(z - z')} \right] J_m \left( \frac{q_0 RR'}{z - z'} \right) \psi_{z'}(R'),$$

where the second line, which coincides with Eq. (1) in the main text, is obtained after exchanging the order of integration and applying the Weber second exponential integral (Sec. 13.31 of Ref. [2])

$$\int_0^{\infty} d\theta J_m(\theta x) J_m(\theta x') e^{-i\theta^2/2} = (-i)^{m+1} e^{(x^2 + x'^2)/2} J_m(xx').$$

S2. CALCULATION OF FOCAL SPOTS

For simplicity, we assume the near and far sides of the electron-optics lens system to coincide with the PAC in a virtual plane at $zL$. In practice, this allows us to set $L_{\text{in}} = L_{\text{out}} = zL$ in Eq. (2), which noticing that $F_{\Delta z}$ becomes the identity operator when $\Delta z = 0$, reduces to $\psi_{z,\text{sample}} = F_f^* \cdot T_{\text{L}} \cdot T_{\text{PAC}} \cdot \psi_{z,\text{PAC}}$, where $f' = z_{\text{sample}} - zL$. Inserting in this equation the definitions provided in the main text for the different factors and considering illumination

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of the PAC with a chiral light field $E_z(R, z) \propto e^{im\varphi_R}$, we find the expression (with $\ell = 1$)

$$
\psi_z(R) \propto e^{im\varphi_R + q_0 z + q_0 R^2 / 2f} \int_{R_{\min}}^{R_{\max}} R'dR' \left( \frac{q_0 RR'}{z - z_l} \right) J_\ell(2|\beta(R')|) + \text{arg}[-\beta(R')]
\times e^{i(q_0 R^2 / 2)[1/(z - z_m) + 1/(z_L - z_\infty) - 1/f]}
$$

for the electron wave function in the sample region, where $\chi(R/f') = C_3 q_0 R^4 / 4 f'^4$ and we consider points separated by a small distance from the focal point $(R, z) = (0, z_{\text{sample}})$ compared with $f'$, where $z_{\text{sample}}$ is defined by the condition that the argument of the last exponential vanishes, leading to the lens equation $1/(z_{\text{sample}} - z_L) + 1/(z_L - z_\infty) = 1/f$.

**S3. COUPLING COEFFICIENT $\beta$ FOR A THIN FILM**

We calculate the coupling coefficient $\beta$ using Eq. (5) in the main text combined with the out-of-plane light electric field $E_z$ in an illuminated film. Considering p-polarized light of frequency $\omega_0$ and amplitude $E_0$ impinging from the $z < 0$ region with angle $\theta$ relative to the $z$ direction on a self-standing homogeneous thin film of thickness $d$ and local dielectric function $\epsilon$, confined within the surfaces $z = 0$ and $z = d$, we find

$$
E_z = E_0 \sin \theta \times \begin{cases} 
    e^{ik_z z} + r_p e^{-ik_z z}, & \text{for } z \leq 0 \\
    A e^{ik_z^l z} + B e^{-ik_z^l z}, & \text{for } 0 < z < d \\
    t_p e^{ik_z(z - d)}, & \text{for } d \leq z
\end{cases}
$$

where

$$
r_p = r_p^0 \left[ 1 - \frac{(r_p^0)^2(k'_z/k_z)c^2k'_z d}{1 - (r_p^0)^2 e^{2ik'_z d}} \right],
$$

$$
t_p = \frac{(r_p^0)^2(k'_z/k_z)c^2k'_z d}{1 - (r_p^0)^2 e^{2ik'_z d}},
$$

$$
A = \frac{1}{\sqrt{\epsilon}} \frac{r_p^0}{1 - (r_p^0)^2 e^{2ik'_z d}},
$$

$$
B = \frac{1}{\sqrt{\epsilon}} \frac{(r_p^0)^2 e^{2ik'_z d}}{1 - (r_p^0)^2 e^{2ik'_z d}}
$$

are expressed in terms of the Fresnel reflection coefficients $r_p = (e k_z - k'_z)/(e k_z + k'_z)$ and $t_p^0 = 2 \sqrt{\epsilon} k_z/(e k_z + k'_z)$. Here, $k_z = k_0 \cos \theta$, $k'_z = k_0 \sqrt{\epsilon - \sin^2 \theta}$, and $k_0 = \omega_0 / c$. Equation (5) in the main text can now be readily integrated to yield

$$
\beta = \frac{i e E_0 \sin \theta}{\hbar \omega_0} \left[ \frac{1}{\omega_0/v - k_z} + \frac{r_p}{\omega_0/v + k_z} - \frac{t_p e^{-i\omega_0 d/v}}{\omega_0/v - k_z} + \frac{A e^{(-i\omega_0/v + k'_z) d} - 1}{\omega_0/v - k'_z} + \frac{B e^{(-i\omega_0/v + k'_z) d} - 1}{\omega_0/v + k'_z} \right].
$$

(S2)

In the perfect-electric-conductor (PEC) limit for the film material, we have $r_p = r_p^0 = 1$ and $t_p = A = B = 0$, so the coupling coefficient reduces to

$$
\beta_{\text{PEC}} = \frac{2i e v E_0 \sin \theta}{\hbar \omega_0^2} \left[ \frac{1}{1 - (v/c)^2 \cos^2 \theta} \right]
$$

According to this expression, the light field required to achieve the optimum PAC operation regime ($\beta = \beta_0 \approx 0.92$) for 60-keV electrons ($v/c \approx 0.45$) in the PEC limit at $\theta = 45^\circ$ incidence with a light wavelength $\lambda_0 = 500$ nm is $E_0 \approx 4 \times 10^7$ V/m.

A first indication that films made of a real metal (e.g., aluminum) or even a dielectric (e.g., Si$_3$N$_4$, $\epsilon \sim 4$) can perform well to couple light and electrons is provided by examining their reflection coefficient $|r_p|$, which is shown in Fig. S1 for three different film thickness in the $d = 5$-15 nm range. In particular, the metal reaches high values well above 0.5 over a wide range of light wavelengths and incidence angles, while the dielectric yields more moderate but still substantial reflectivities.

In Fig. S2, we show that the PEC limit is nearly approached by aluminum films of thickness $d \sim 15$ nm. Interestingly, thin dielectric films such as Si$_3$N$_4$, which are often used as supporting TEM membranes, can also produce the optimum $\beta$ by increasing the light intensity approximately one order of magnitude with respect to aluminum, which should be possible because optical losses are then much lower. Importantly, the use of dielectric films composed of light elements would significantly reduce inelastic scattering of the electron beam, which would be favorable for the design of PACs.
FIG. S1: Wavelength and angle dependence of the p-polarization Fresnel reflection coefficient $|r_p|$ for thin aluminum (top row) and dielectric (bottom row) films. We describe the optical response of aluminum as a function of light frequency $\omega_0$ through the Drude dielectric function $\epsilon = 1 - \omega_p^2/\left(\omega_0^2 + i\omega\gamma\right)$ with $\hbar\omega_p = 15.1$ eV and $\hbar\gamma = 0.15$ eV. The dielectric film is assumed to have a constant $\epsilon = 4$. We consider three different film thicknesses, as indicated at the top of each column.

FIG. S2: Thickness dependence of the light field amplitude required to produce $\beta = \beta_0 \approx 0.92$ on 60-keV electrons ($v/c \approx 0.45$), as calculated from Eq. (S2) for aluminum and $\epsilon = 4$ dielectric films at a wavelength $\lambda_0 = 500$ nm. We use the same dielectric function for aluminum as in Fig. S1. Results for a perfect mirror are shown for comparison.

S4. ADDITIONAL FIGURES

We show in Fig. S3 further details of the electron optical phase plate used for the PAC in the main text. In Fig. S4 we present longitudinal and transverse cuts of the spots plotted in Fig. 2 of the main text to facilitate their comparison. Fig. S5 is similar to the inset of Fig. 3 in the main text, but showing the ratio of full widths at 20% of the intensity maximum instead of the half maximum.

[1] M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1972).
[2] G. N. Watson, *A Treatise on the Theory of Bessel Functions* (Cambridge, Cambridge University Press, 1944).
FIG. S3: Details of the electron-light coupling in the optical phase plate used for the PAC element proposed in this paper. (a) Interaction with the optical field $E_z$ produces inelastic sidebands in the spectrum of the transmitted electrons, separated by multiples of the photon energy $\hbar \omega_0$ with respect to the incident electron energy $E_0$. (b) A monochromator is used to retain electrons only within the first sideband $\ell = 1$. The transmitted wave function consists of the incident wave function times a factor $J_1(2|\beta|)/(\beta)$, where the coupling parameter $\beta$ scales linearly with $E_z$ and therefore varies with lateral position along the film. The field amplitude is adjusted to take values in the region to the left of the vertical orange line $|\beta| < \beta_0 \approx 0.92$ below the maximum first-sideband transmission intensity $J_1^2(2\beta_0) \approx 34\%$.

FIG. S4: Profiles showing the electron probability $|\psi_{\text{sample}}|^2$ along transverse (a) and longitudinal (b) cuts across the focal spot maximum for a nonaberrated beam and aberrated beams under the conditions of Fig. 2 in the main text. The profiles are normalized to their respective maxima. The lower $x$ scale in (a) is normalized such that the unaberrated curve is universal. The lower $z$ scale in (b) is normalized using $\Delta z = 1/[4\pi/(q_0 R_{\text{max}}^2) + 1/f - 1/(z_{\text{sample}} - z_{\text{out}})] - 1/[1/f - 1/(z_{\text{sample}} - z_{\text{out}})]$. The upper horizontal scales in both (a) and (b) are obtained for $R_{\text{max}} = 30 \mu$m.

FIG. S5: Full width of the beam profile at 20% maximum intensity divided by the corresponding value obtained for an unaberrated beam. This plot shows the same behavior as the ratios of FWHM in the inset of Fig. 3 of the main text, thus confirming that the aberration correction with the PAC does not introduce parasitic tails.