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Prospects for versatile phase manipulation in the TEM: Beyond aberration correction

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ABSTRACT

In this paper we explore the desirability of a transmission electron microscope in which the phase of the electron wave can be freely controlled. We discuss different existing methods to manipulate the phase of the electron wave and their limitations. We show how with the help of current techniques the electron wave can already be crafted into specific classes of waves each having their own peculiar properties. Assuming a versatile phase modulation device is feasible, we explore possible benefits and methods that could come into existence borrowing from light optics where the so-called spatial light modulators provide programmable phase plates for quite some time now. We demonstrate that a fully controllable phase plate building on Harald Rose's legacy in aberration correction and electron optics in general would open an exciting field of research and applications.

1. Introduction

When Harald Rose started to work on the implementation of the spherical aberration (C_s) corrector he was trying to overcome the intrinsic limits of the simple circularly symmetric electron optical lenses. Scherzer himself had realized that using multipolar lenses could have allowed to break free of the restrictions imposed by the Scherzer theorem [1–3]. However, due to the formidable challenges posed by its realization, this concept was only successfully implemented 50 years later by the Haider–Rose–Urban project [4–6]. The goal of the C_s corrector however, despite all its flexibility, is to "flatten" the phase imposed by the optical systems as much as possible in order to make the smallest possible probe (in STEM) and to collect and interpret the highest possible frequencies (in TEM). This has, of course, had a groundbreaking impact on the field, pushing the resolution of both TEM and STEM beyond the angstrom limit, and making it possible to reach the very high current densities that have allowed analytical techniques to reach atomic resolution.

We would like to peek into the territory of phase manipulation. Optical microscopy techniques have greatly benefited from the invention of the spatial light modulator, a device which allows to tune a light wave in both phase and amplitude. A corresponding "Spatial Electron Modulator" however is far from being available to the TEM community. In this paper we will review the methods currently available for the manipulation of the electron wave, and outline possible applications.

2. Review: the state of the art

It is not yet possible to freely and arbitrarily tune the wave function of an electron beam. However there are a number of techniques which allow this, to some degree. Many of these have already proven useful, despite all their limitations, in order to expand the flexibility of the TEM or in the developing field of singular electron optics.

2.1. Electromagnetic fields in free space

An electron encountering a region of space containing an electric potential acquires a phase shift equal to

$$\Delta\Phi_{\mathrm{E}} = \frac{\pi}{\lambda E_0}\int_{\Gamma} eV(\mathbf{r})\, dl. \tag{1}$$

Indeed electrostatic optical elements are widespread in lower energy applications, ranging from scanning electron microscopes, low energy or photoemission electron microscopes to Mott detectors, and electrostatic aberration correctors have been devised as well in the form of electron mirrors. While impractical for the

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manipulation of high energy electrons, tetrode mirror aberration correctors have been demonstrated to successfully correct spherical and chromatic aberrations in LEEM-PEEMs and SEMs [7–13].

Similarly, magnetic vector potentials also influence the phase of electron waves. When passing through a region of space with non-zero magnetic vector potential, an electron acquires an additional Aharonov–Bohm phase of

$$\Delta\Phi_{AB} = \frac{e}{\hbar}\int_{\Gamma} \mathbf{A}\cdot d\mathbf{l}. \tag{2}$$

This phase is acquired even when the magnetic field $\mathbf{B} = \nabla \times \mathbf{A}$ is zero. Magnetic lenses commonly employed in the TEM can also be explained in the light of this effect, as can multipolar aberration correctors.

The multipolar corrector aims to determine the phase plate impressed on the beam by manipulating the magnetic field by the boundary conditions at the ends of the magnetic poles. This is fundamentally possible because fields in free space are described by analytical functions, and as such an entire region of space can be entirely described by the properties of its boundary. While this is certainly true, the level of accuracy and stability required for this type of manipulation to be effective is extremely high, and was one of the factors that made the realization of aberration correctors so challenging.

The phase of an electron probe can also be altered by interacting with light. When a charged particle is immersed in a strongly varying electromagnetic field as found e.g. in laser cavities it is subjected to a ponderomotive force. For an electron passing through the focal point of a laser beam, perpendicular to the electron propagation, Müller et al. [14] calculated the phase shift to be

$$\Delta\Phi_{P} \propto \frac{P\lambda}{2\beta\gamma}, \tag{3}$$

where P and λ are the local energy density and wavelength of the laser, $\beta = v/c$ and $\gamma = (1-\beta^2)^{-1/2}$, the Lorentz factors for the relativistic electron. By focusing the laser in the back focal plane of the objective lens a Zernike phase plate can in principle be obtained [15–17]. In this case the unscattered wave component is phase shifted by the strongly varying light intensity in the focal point of the laser while the scattered wave is left unchanged. Efforts are being made to implement this technique [18]. Note that also here, the phase plate is defined by the standing optical wave which is determined by its boundary conditions (the cavity) making it far from trivial to produce a fully versatile phase plate this way.

2.2. Phase manipulation using aberration correctors

The development and commercialization of aberration correction for the minimization of the Seidel aberrations has been absolutely fundamental in enabling reliable, consistent access to sub-Angstrom resolution in modern electron microscopes. The electric and magnetic multipoles (quad/octopole or hexapole systems) of an aberration corrector can be individually addressed and tuned to counteract both the intrinsic spherical (and recently chromatic) aberration and the remaining parasitic aberrations. Typically, for an electron beam to be considered "corrected", there should be a variation of no more than $\pi/4$ in the phase, within a given opening angle. This leads to a good approximation to a plane wavefront, which can be focused to produce a small diffraction limited probe for STEM, or an aberration-free lens which can correctly transfer high frequencies in TEM.

While manipulating the phase of the wavefronts using aberration correctors to closely approximate the ideal plane wave is very well suited to normal TEM and STEM studies, with the degrees of freedom given by the free-lens mode of an aberration corrected microscope, the electron wave can be designed and optimized for the experiment at hand, with structures available beyond simple plane waves, or Airy disc probes.

The aberration function in Saxton notation is

$$\begin{aligned}\chi = \frac{2\pi}{\lambda}[&\theta A_0 \cos(\phi-\phi_{11}) \\ &+\frac{1}{2}\theta^2\{A_1\cos(2(\phi-\phi_{22}))+C_1\} \\ &+\frac{1}{3}\theta^3\{A_2\cos(3(\phi-\phi_{33}))+B_2\cos(\phi-\phi_{31})\} \\ &+\frac{1}{4}\theta^4\{A_3\cos(4(\phi-\phi_{44}))+S_3\cos(2(\phi-\phi_{42})) \\ &+C_3\}+\cdots]\end{aligned} \tag{4}$$

where χ is the phase shift of the wave front at (θ, ϕ).

The possibilities offered by the free tuning of the aberrations have been studied previously for optimizing phase contrast in atomic resolution HRTEM. Lentzen et al. have described how to combine defocus, third and fifth order spherical aberrations in order to obtain positive or negative Zernike-type phase contrast by employing a controlled amount of positive or negative spherical aberration [19,20]. In particular, the negative spherical aberration imaging (NCSI) technique has shown a remarkable enhancement of the contrast for light elements such as oxygen or nitrogen, allowing their direct imaging, while previously their position had been only accessible by performing exit-wave reconstruction [21–23].

A further example of probe restructuring using aberration correctors was demonstrated recently by Clark et al. [24], wherein free manipulation of the aberrations was combined with an annular aperture to produce a high intensity vortex beam.

Thus, it can be seen that by selecting a single θ value with the annular aperture, and minimizing all aberrations other than the A_i (where A_0 is beam tilt and $A_1 \cdots A_n$ are the $i+1$-fold astigmatisms), we can create a phase linearly increasing with azimuthal angle ($\chi \approx m\phi$) typical of a so-called vortex beam using the appropriate weights for the cosine series. This phase structure, and the resulting

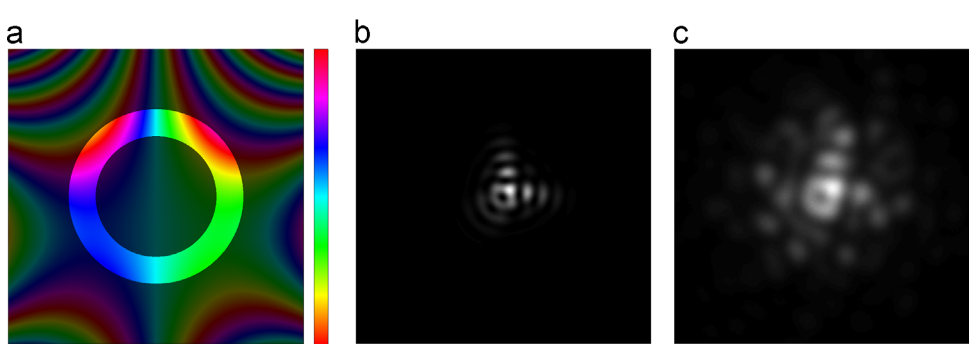

Fig. 1. Creation of an electron vortex through aberration manipulation. A_1 and A_2 were optimized. Higher orders of astigmatism were beyond experimental limits. The annulus has an average radius of 7 mrad. Both intensity images represent 2 nm by 2 nm. (a) Phase map imposed by the A_i to produce a vortex phase, with overlaid annular aperture. (b) Resulting simulated intensity pattern in the sample plane. (c) Experimental intensity pattern in the sample plane, obtained in an FEI Titan[3] at 300 kV.