



Sculpturing the electron wave function using nanoscale phase masks



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ABSTRACT

Electron beams are extensively used in lithography, microscopy, material studies and electronic chip inspection. Today, beams are mainly shaped using magnetic or electric forces, enabling only simple shaping tasks such as focusing or scanning. Recently, binary amplitude gratings achieved complex shapes. These, however, generate multiple diffraction orders, hence the desired shape, appearing only in one order, retains little of the beam energy. Here we demonstrate a method in electron-optics for arbitrarily shaping electron beams into a single desired shape, by precise patterning of a thin-membrane. It is conceptually similar to shaping light beams using refractive or diffractive glass elements such as lenses or holograms – rather than applying electromagnetic forces, the beam is controlled by spatially modulating its wavefront. Our method allows for nearly-maximal energy transference to the designed shape, and may avoid physical damage and charging effects that are the scorn of commonly-used (e.g. Zernike and Hilbert) phase-plates. The experimental demonstrations presented here – on-axis Hermite–Gauss and Laguerre–Gauss (vortex) beams, and computer-generated holograms – are a first example of nearly-arbitrary manipulation of electron beams. Our results herald exciting prospects for microscopic material studies, enables electron lithography with fixed sample and beam and high resolution electronic chip inspection by structured electron illumination.

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1. Introduction

Today, spatial manipulation of electron beams is achieved mainly using electrostatic and magnetic fields. Altering the spatial profile of the beam may be proven useful in many fields incorporating phase microscopy [1,2], electron holography [3–6], and electron–matter interactions [7]. With the advance of nano-fabrication technology, unique possibilities have opened for fundamental research in electron optics. Nano-scale wrought thin films, using a focused ion-beam (FIB) for example, can be fashioned as phase- or amplitude-masks to create holograms that may be utilized in material science and in fabrication of microelectronic circuits.

A number of groups have proposed analogies between free-electron and light optics. Specifically, it has been suggested that under certain approximations feasible in a standard transmission electron microscope (TEM), the Klein–Gordon equation describing the dynamics of free electrons may be replaced by the paraxial Helmholtz wave propagation equation, which is used in light optics [8,9]. One of the first realizations in this direction was already made in 1998 – a Fresnel lens in AlF_3 film was fabricated

by electron beam nano-lithography [10]. More recently, Uchida and Tonomura measured vortex electron beams [11], having a helical phase front structure and carrying orbital angular momentum [12], Verbeeck et al. [13] and McMorran et al. [14] fabricated a binary mask for the generation of off-axis vortex beams and Voloch-Bloch et al. [15] experimented with the generation of electron Airy beams that preserve their shape and propagate in free space along curved parabolic trajectories [16]. These shaped electron beams open exciting possibilities in electron microscopy. For example, it was shown that vortex beams can be used to characterize the magnetic state of ferromagnetic materials [7,13], whereas Airy beams can be used for realization of a special type of electron interferometer [15]. However, the experimental demonstrations of these analogies have mainly relied upon favorable natural deformations in an observed specimen [11] or, in the case of holographic projections [13–15] reconstruction in the first diffraction order using binary amplitude-based masks.

Here, we utilize widespread light-optics methods in holography and diffractive-optics to design, fabricate and experimentally measure images produced by phase-based masks using TEM. Holograms produced in this way benefit from potentially maximal energy efficiency, contrast-resolution and flexibility in their usage, marking them as the next standard in electron beam shaping.

In our experiment, the electron beams are shaped by patterning thin Silicon-Nitride (SiN) membranes using focused ion beam

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(FIB) milling. These membranes, ranging from 5 nm to 150 nm in thickness, are a popular choice due to their low scattering and mechanical robustness. Much like light waves passing through glass and acquiring a phase-shift dependent on the material's refractive index, an electron passing through a SiN membrane will similarly accumulate a phase factor directly related to the thickness of the interacting material according to [8]

$$\varphi = \frac{2\pi}{\lambda}(n-1)t = \frac{2\pi eU_i}{\lambda E} \frac{E_0 + E}{2E_0 + E} t \quad (1)$$

where λ is the electron's wavelength, $E_0 = m_0c^2$ and $E = eU$ are the electron's rest and kinetic energy, respectively, where the kinetic energy is given by the acceleration voltage U and the electron's charge e . The most interesting quantities are the thickness t and the material's mean inner potential U_i , which comprises the electron-optical refractive index n . A relatively thin film is sufficient, e.g. for a 200 keV electron, the required thickness to generate a π -phase shift is 42 nm. The scattering through this film is fairly low, hence this can be considered a nearly pure phase plate for our purposes.

Computer-generated holograms (CGH) are an invaluable tool in optics. This concept conventionally allows designing a slide that stores the amplitude and phase of a wavefront, which may later be reconstructed by illuminating the hologram with a reference beam, and observing the result in the diffraction plane. CGH design may be classified as off-axis (first and higher-order) or on-axis (zero-order), the latter potentially enjoying the full conversion of energy to the desired shape.

Phase plates [17], which may be thought of as the simplest manifestations of phase-only CGHs, are generally used in electron microscopy for phase contrast [2] imaging, as first suggested by Zernike in 1942 [1]. An example of such a phase plate is the Hilbert phase plate, which imparts a π -phase shift between two halves of the impinging electron beam, thus generating an approximation to the Hermite–Gauss (HG) 01 or 10 mode (which are solutions of the paraxial wave equation).

2. On-axis phase-plates

In our first experiment, we studied the ability to generate such basic beams – the HG11-like as well as Laguerre–Gauss LG01-like (vortex) beams, by imparting a π phase-shift to opposite quadrants of a disc and a continuous spiraling slope covering 2π radians, respectively. We fabricated an additional periodic (Bragg) grating; the distance between the measured diffraction orders and knowledge of the grating's period Λ yields the effective propagation distance L to the diffraction plane according to $L = \Lambda x / 2\lambda$, where $\lambda \approx 2.5$ pm is the electron's wavelength at 200 keV, which is used as a metric for the measurements. The fabrication process included a coating of 10 nm of gold or 5 nm of titanium on one side of 50 nm or 100 nm SiN membranes, respectively, to reduce charging effects while in observation under the microscope. Titanium was expected to reduce electron scattering over gold. The designs were milled into the membranes from the uncoated side, using a Raith IonLine FIB with a 35 kV gallium beam, currents ranging between 18 pA and 24 pA.

Calibration of the FIB for selective-depth milling may be done as follows. Depending on the desired accuracy, a number of pixels are milled (we found $1 \mu m^2$ squares convenient), each with a linearly-increasing beam dwell-time. Once milling has begun, the beam's dwell-time is directly proportional to the milled depth and as such, the pixel where the 100 nm membrane is breached indicates the associated beam dwell-time. Thus, appropriate scaling may be applied to the dwell-times of a CGH's pixels, as was

done in the second part of this article. While our CGHs were designed to operate correctly for 200 keV electrons, it may be shown by direct substitution in Eq. (1) that a 1 nm error in milling depth or 1 kV error in acceleration voltage yield a 2.3% change in phase shift, φ . This allows correcting for systematic depth errors in the microscope by slight changes in acceleration voltage.

In order to observe the evolution of the wave-function in maximum magnification we used a 10 μm - or 30 μm -diameter objective aperture in accordance with the diameter of the hologram. This aperture is essential to obtain viable diffraction pattern. A non-modulated area of the membrane is imaged in LAD (diffraction) mode, where we found a distance of 180 m most convenient. Making sure we are in eucentric focus (objective lens at 6%), we focus the spot to a minimum – in this way, the beam impinging on the sample is approximately collimated. The collimation condition does not have to be strictly met to generate the holograms, but it makes possible the correct assessment of effective distances using the Bragg grating diffraction pattern. Lastly, we used the “Free lens control” software to set the magnification to the maximum possible, and brought the hologram to view by mechanically controlling the stage. Then, the evolution of the beam may be observed from image to diffraction plane and beyond, by changing the diffraction lens only. Fig. 1 depicts these results: in (a), the beam's shape is measured passing through the membrane with no additional modulation, where its focusing and defocusing is marked by the 4σ -diameter, σ being the standard deviation of the intensity profile. In (b), the HG11-like mode shows shape invariance for an effective distance of nearly 150 m, while the existence of a spiral phase front is evident from the enduring dark centre in (c). The effective distances are calculated from the measurements of the ± 1 diffraction orders generated by the 445 nm-period Bragg grating, as exemplified in (d), where z denotes the relative effective distance the beam traverses in proximity to this plane. Microscopic images of the masks we used are recorded in (e and g). In Appendix A we mathematically show that the phase plate in (F) yields a dominant mode, HG11, as clearly observed in (b).

It is most important to distinguish these phase-plates from previous results [13,14,18] which may be obtained with popular holographic binary schemes [19]; our beams have nearly all the energy of the impinging beam transformed to the single desired shape, on-axis. This is a major advantage with respect to the binary holograms that generate multiple diffraction orders at different angles, of which only the pattern in the first (positive or negative) diffraction order is usually desired. A second, auspicious argument must be made: commonly used phase-plate methods, such as the Zernike and Hilbert phase plates, are usually placed in the back focal-plane of the objective lens and thus suffer high-intensity electron bombardment that may cause physical damage and charging effects [20]. Conversely, our phase plates are placed in other apertures, where the beam is collimated and low in intensity, thus reducing or possibly avoiding such difficulties all together.

If the appropriate conditions are available, the vortex beam may be shrunk down to nanometer size to study, for example, electron–matter interactions such as orbital angular momentum transfer to and from the internal electron states [7]. The HG11 and similar phase-plates may be used, for instance, in conjunction with the application of phase-contrast microscopy [17] to study nearly transparent objects, e.g. biological samples [21]. The two examples given here show the ability to generate, with patterned phase masks, different electron wave functions that are solutions of the paraxial wave equation. The same technique may be utilized to generate other beams that satisfy this equation [22], e.g. higher order Hermite–Gauss and Laguerre–Gauss beams, Bessel Airy and parabolic beams, etc.

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