Electron Vortex Beams with High Quanta of Orbital Angular Momentum

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Abstract

Electron beams with helical wavefronts carrying orbital angular momentum are expected to provide new capabilities for electron microscopy and other applications. We use nanofabricated diffraction holograms in an electron microscope to produce multiple electron vortex beams with well-defined topological charge. Beams carrying up to 100 quanta, $\hbar$, of orbital angular momentum per electron are observed. We describe how the electrons can exhibit such orbital motion in free space absent of any confining potential or external field, and discuss how these beams can be applied to improved electron microscopy of magnetic and biological specimens.

The discovery that optical vortices – light beams with a spiral phase singularity at the center of helical wavefronts – carry orbital angular momentum (OAM) (*1*) has led to appreciable advances in optical microscopy (*2, 3*), astronomy (*4*), micromanipulation (*5*), quantum state manipulation (*6, 7*), and other diverse applications (*8*). In an effort to extend these applications to other types of beams, we demonstrate production of well-separated electron vortex beams with large quantized angular momentum. Electron vortices provide novel opportunities in electron microscopy.

An optical vortex beam can be characterized by the thread pitch angle of the screw-shaped wavefronts. This angle is related to the topological charge, *m*, describing the magnitude of the phase singularity at the center of the vortex. The wavefunctions of all such optical vortices include an azimuthal phase factor $e^{im\phi}$. When this phase factor is imprinted onto a conventional Gaussian beam, described in cylindrical coordinates as

$$G(\rho,z) \propto \frac{1}{w(z)} \exp\left[-i\left(kz + \frac{k\rho^2}{2R(z)} - \zeta(z)\right)\right] \exp\left(\frac{-\rho^2}{w(z)^2}\right),$$

where $w(z)$ is the beam width, $R(z)$ is the radius of wavefront curvature, and $\zeta(z)$ is the Guoy phase, it forms a particularly simple kind of optical vortex called a Laguerre-Gaussian (LG) beam, described by the wavefunction

$$\psi_{LG}(\rho,\phi,z) = G(\rho,z)\left(\frac{\sqrt{2}\rho}{w(z)}\right)^{|m|} \exp\left[i\left(m\phi + |m|\zeta(z)\right)\right].$$

For $m \neq 0$, complete destructive interference at the vortex core yields an intensity node along the entire optical axis of the vortex, such that LG beams are hollow and form characteristic ring-shaped (annular) intensity distributions when projected on a planar detector. An application of the angular momentum operator $\hat{L}_z = -i\hbar\partial_\phi$ to this wavefunction shows that the optical vortex carries a quantized projection of OAM onto the optical axis, such that $L_z = m\hbar$. The optical vortex is an orbital eigenstate for the individual particles that make up the beam (*6*).

Massive particles, such as free electrons, can also occupy such vortex states. The similarity between the Schrödinger equation describing the evolution of free particle wavefunctions and the Helmholtz equation describing the propagation of light suggests that matter waves can be manipulated and applied in similar ways to light waves. The fact that LG beams are stable paraxial solutions of both the Helmholtz equation and the Schrödinger equation led to predictions (*9, 10*) that free electron vortices were not only physically realizable but they could also be produced and applied in analogous ways to optical vortices. The first demonstration of electron vortex beams used a nanoscale spiral phase plate (*11*), formed by three pieces of thin graphite. However, it is very difficult to generate smooth helical wavefronts using this approach, and it was noted that the resulting electron vortices had non-integer topological charge, such that the electrons occupied mixed quantized orbital states (*12*). Microscale gratings were recently used to create electron vortex beams with topological charge $m = 1$ (*13*).

We use nanoscale diffraction holograms to produce free electrons with large angular momentum in pure quantized orbital states (Fig 1). Our work is based on earlier demonstrations using nanofabricated gratings for coherent electron interferometry (*14-16*), which led to a proposal to apply similar techniques to generate electron vortices (*10*). This approach, also adopted by (*13*), uses a grating mask with a fork dislocation to holographically imprint a phase vortex onto a diffracted beam (*17, 18*). Advantages of this technique over spiral phase plates are that the diffracted beam automatically possesses integer topological charge, regardless of the wavelength, and it can be scaled to generate vortices with large quanta of OAM by including a higher order fork dislocation. Transmission of a beam with wavelength λ through a binary hologram with slit spacing *d*, the grating period, and a fork dislocation defined by *b* extra half slits produces multiple diffracted vortex beams. The n^{th} diffraction order has distinct topological charge $m = nb$, such that negative diffraction orders propagating to one side of the central order beam have quantized OAM that is antiparallel to the propagation direction, and vice versa for positive diffraction orders. The diffracted beams propagate at discrete angles $\alpha = \lambda/d$ relative to one another, so for applications requiring isolated electron vortex beams it is necessary to use holograms with sufficiently small grating periods.

With a view towards future applications requiring high quality, isolated electron vortex beams, we have placed a particular emphasis on fabricating diffraction holograms with nanoscale feature sizes. Using a focused ion beam (FIB) to mill silicon nitride membranes (*19*), we made multiple transmission holograms with 50 nm, 75 nm, and 100 nm grating periods over 5 μm diameter circular areas, featuring fork dislocations that encode various amounts of topological charge, from $b = 1$ to b = 25 (Figs. 2A and 2B). These holograms have grating periods that are an order of magnitude smaller than those demonstrated in (*13*), which provides a correspondingly larger separation angle between beams. The smaller feature size also allows higher topological charge to be encoded within the same finite aperture area, enabling us to demonstrate electron beams with OAM up to $100\hbar$.

The electron vortices are produced by diffraction from the holograms in a transmission electron microscope (TEM) operating at 300 keV (*19*). Images of the diffraction patterns (Figs. 2, 3) show the ring-shaped projections of electron vortex beams onto the detector plane. Electron vortex beams with distinct topological charge are selectively generated in particular diffraction orders from different holograms. In Figure 3, one can discern the 4^{th} diffraction order produced by a grating with dislocation number $b = 25$ (Fig. 2B). The corresponding topological charge in this beam is *m* = 100, and it is composed of individual electrons that each carry 100 $\hbar$ quanta of OAM. Electrons that have such high OAM are localized to a thin annulus around a large hollow core.

The free space evolution of a hollow core in a wavefunction can be used to determine the existence of a vortex. A wavefunction with OAM has zero amplitude at the vortex core due to the repulsive centrifugal barrier. This is manifested in an LG beam by a central node that persists through the focus and increases in diameter as a function of distance from the beam waist. On the other hand, an initially ring-shaped wavefunction without OAM, such as that formed with an annular beam-defining aperture, will spread radially both inwards and outwards, such that the hole at the center of the wave becomes smaller and eventually disappears. We measured the hollow structure of the electron vortex beam by focusing it in space and then imaging the beam at multiple transverse planes relative to the beam waist (Fig. 4A)(*19*). The dark core can be observed throughout the Rayleigh range of each diffracted electron beam. Intensity line profiles through the first diffraction order, an *m* = 15 beam, show that the dark vortex core increases in diameter with distance from the focus (Fig. 4B). A quantitative analysis shows that over this range the vortex core diameter increases away from the beam waist by a factor of 2.8±0.4 (Fig. S1), which indicates that the diffracted electron beams possess angular momentum. An interference experiment between the electron vortex beam and a plane wave confirms that this OAM is due to a helical phase (Fig. S2).

These results demonstrate that electrons can be prepared in quantized orbital states with large OAM, in free space devoid of any central potential, electromagnetic field (*20*), or medium that confines the orbits. The electron vortex state is nonradiative in free space, as it must be, as emission of a photon would violate the simultaneous conservation of energy and linear momentum. Unlike a classical vortex, this orbital motion cannot be attributed to the collective behavior of many electrons in the beam; at the low beam currents of this experiment, the separation between individual electrons along the optical axis is several orders of magnitude larger than the longitudinal extent of each wavepacket (*21*). Such high-OAM electron vortex states can exist at rest, too, because unlike in a beam of photons one can produce, using a decelerating electric field, a reference frame in which the forward motion of the electron vortex is zero along the optical axis.

The electron vortex is understandable in terms of semiclassical physics, provided the correct model is used for how these states evolve in time. In the reference frame where the electron vortex has no forward motion, the wavefunction is uniformly delocalized in a ring. The radius of this ring-shaped vortex wavefunction may change with time, but the centroid of the wavefunction is stationary. Thus, there is no acceleration associated with the electron's center of mass and so no external force is necessary to preserve or define the orbital motion. A picture of this wavefunction evolving in space is provided by the simulation of a focused LG beam (Fig. 4C). The helical wavefronts of the beam are slightly tilted both azimuthally and radially, such that their locus of peak amplitude sweeps out a hyperboloid surface describable as a family of straight lines (Fig. 4D). Thus, the electron vortex can be modeled simply as a coherent superposition of classical straight trajectories that are slightly skewed and offset from the optical axis of the vortex (*22*).

Electron vortex beams are expected to provide new capabilities for electron energy loss spectroscopy (EELS) in a TEM (*23*). Computed scattering cross sections between electron vortices and atoms show that it is possible for the electron vortex to transfer both quantized OAM and energy to the atoms. This transfer could be used to induce atomic transitions that were previously inaccessible in conventional EELS, such that an OAM-dependent signal could provide new chemical, crystallographic, optical, electronic, and magnetic information about a sample. This finding was recently confirmed experimentally (*13*). We are particularly interested in developing this technique for magnetic imaging, which will require the use of well-separated electron vortex beams.

Electron vortices can also enable spiral phase microscopy (*2, 24*) in a TEM, enhancing the visibility of edges in samples with low absorption contrast, such as unstained biological specimens, without sacrificing spatial resolution. Computer-generated holograms have been used to implement spiral phase measurements in transmission light microscopes (*24*). Nanofabricated holograms could be implemented in a similar way in a TEM, provided that the hologram provides sufficient angular separation between vortex beams. This technique could provide some advantages over emerging TEM phase plate technologies (*25, 26*), since the spiral phase provides additional information (*2*). This improved phase contrast will impact TEM imaging of electron-transparent samples, such as biological specimens, macromolecules, carbon nanotubes, and polymers.(*27*)

References and Notes

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20. While the beams in this experiment do traverse magnetic fields in the lenses, the fields are confined to very small intervals relative to the entire path of the electrons.

21. Assuming a 1 nA, 300 keV electron beam with 0.3 eV energy spread, the longitudinal extent of a electron's wavepacket along the optical axis is about 3 μm, whereas the average separation distance between consecutive electrons is 4 cm.

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Figure 1. Representation of the formation of electron vortices. (A) A spatially coherent plane wave of electrons illuminates (B) a nanofabricated hologram and (C) diffracts into multiple electron vortex beams which are then (D) imaged using a CCD. The depictions of the vortex beams shown at (C) simulate the wavefronts, and not the trajectories, of the electrons. The beam cross section displayed at (D) is a measured diffracted electron intensity distribution. For simplicity, the TEM imaging optics used to demagnify the diffraction pattern are not shown in this diagram.

Figure 2. Nanofabricated gratings with fork dislocations (top row) used to create electron beams with quantized phase vortices (bottom row). A spatially coherent beam of 300 keV electrons transmitted through (**A**) a 75 nm period grating with $b = 1$ and (**B**) a 100 nm period grating with $b = 25$ (dislocations magnified in insets) forms diffraction patterns (**C**) and (**D**), respectively. To preserve the structural integrity of the high order hologram shown in **(B)**, the area around the central dislocation has been masked off. Each ring-shaped spot in **(C)** and **(D)** is the transverse intensity profile of an electron vortex beam. The topological charge is indicated below each diffracted beam. The diffracted electron intensity distributions are shown in false color to make the higher diffraction orders more evident.

Figure 3. Electron vortices with large OAM. The false color image shows transverse intensity profiles of electron vortex beams produced using the hologram shown in Fig. 2C. The labels indicate the associated values of L_z possessed by each electron in an order. An electron vortex with $L_z = +100\hbar$ ($m = 100$) is evident with a different color scale applied to the the right side of the image containing the 4^{th} and part of the 3^{rd} diffraction orders.

Figure 4. Evolution of electron vortex beams going through focus. **(A)** A focal series of images shows multiple diffracted electron vortex beams going through a beam waist. The beams are produced using a grating with a *b* = 15 dislocation. The slanted elongation of the intense central beam near the beam waist is an artifact resulting from saturation of the CCD camera. **(B)** Line profiles though the 1^{st} diffraction order (indicated by the dashed green line in **A**) clearly show the presence of a persistent central intensity node that expands from the beam waist, indicating a phase vortex and therefore OAM. **(C)** A simulation of the wavefronts and transverse amplitude of an $m = 5$ Laguerre-Gaussian beam going through focus illustrates the helical structure of the wavefronts. **(D)** A geometric (ray) optics model reveals that the electron vortex beam can be understood as a superposition of semiclassical straight line trajectories.