Origins and Demonstrations of Electrons with Orbital Angular Momentum

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Abstract

The surprising message of the 1992 paper of Allen, Beijersbergen, Spreeuw, and Woerdman (ABSW) was that photons could exhibit orbital motion in free space, which subsequently launched advancements in optical manipulation, microscopy, quantum optics, communications, many more fields. It has recently been shown that this result also applies to quantum mechanical wavefunctions describing massive particles (matter waves). This article discusses how electron wavefunctions can be imprinted with quantized phase vortices in analogous ways to twisted light, demonstrating that charged particles with nonzero rest mass can exhibit orbital motion in free space. With ABSW as a bridge, connections are made between this recent work in electron vortex wavefunctions and much earlier works, extending a 175 year-old tradition in matter wave vortices.

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I. INTRODUCTION

The work of Allen et al. \cite{1} can be interpreted as a crucial link in a chain of thought extending to a much earlier hypothesis of a vortex nature to matter. In 1850, William Rankine presented a paper titled 'Hypothesis of Molecular Vortices' to the Royal Society of Edinburgh \cite{2}. This inspired further work by Lord Kelvin (Sir William Thomson), who supposed that atoms could be described as knots of vortices in the aether \cite{3}. J. J. Thomson (no relation to Lord Kelvin) developed this idea further by analyzing interactions between intertwined vortices of matter \cite{4}. After the discovery of the electron by J. J. Thomson himself, a finding for which he ultimately won the 1906 Nobel Prize in physics \cite{5}, he and others abandoned the vortex theory of matter. However, the general idea that particles (and electrons in particular) could have a vortex structure, and an associated angular momentum and azimuthal phase, continued to play an important role in 20th century physics and beyond. The relatively recent experimental demonstrations of free electron vortex states \cite{6,7,8} thus represent a continuation of a centuries-old idea.

Not long after J. J. Thomson’s discovery of the electron, his son, George Paget Thomson, demonstrated the wavelike nature of electrons by diffracting cathode rays from a thin crystal \cite{9}, a finding that earned him the 1937 Nobel Prize \cite{10}. The son’s discovery that electrons could behave like a delocalized fluid re-opened the door to the elder Thomson’s vortex theory of matter. Paul Dirac considered that an electron wavefunction scattering from a theoretical magnetic charge (magnetic monopole) would acquire an azimuthal phase \cite{11,12}. In order for the scattered electron wavefunction to not be multivalued, Dirac argued that the amplitude must vanish along a nodal line, and that the azimuthal phase about this singularity must be quantized, thus continuing the thoughts of J. J. Thomson, Kelvin, and Rankine. With these considerations, Dirac was able to derive a theoretical value of a magnetic monopole quantum in terms of the electron charge $e$, the speed of light, and Planck’s constant $\hbar$.

As Dirac showed, a quantum vortex is a de Broglie wavefunction that has nontrivial topology; a vortex state cannot be continuously deformed into a different vortex state without introducing another dislocation in the fluid or wave function. At the vortex core, along the nodal line, the phase of the wavefunction is undefined and the amplitude is zero. The quantum vortex can be defined by a spatial wavefunction that includes an azimuthal phase factor, $\exp(i\ell \phi)$, where integer $\ell$ is the topological charge of the spiral phase singularity,
and \( \phi \) is the polar angle about the axis of the phase singularity. From this it can be seen that such quantum vortex wave functions are orbital angular momentum eigenstates, with
\[
\hat{L}_z \psi_\ell = -i\hbar \partial_\phi \psi_\ell = \ell \hbar \psi_\ell.
\]

Quantum vortices have been observed in a variety of matter wave systems; first in superfluid helium \cite{13}, then flux vortices in type II superconductors \cite{14}, and on to Bose-Einstein condensates \cite{15}. The related concepts of wavefunction vortices, topology, and angular momentum have become central to many modern topics in condensed matter physics and many-particle physics. Several theoretical works proposed vortex solutions in free particle waves, too. Nye and Berry developed a theory of phase dislocations and singularities in general wave fields \cite{16}, and later analyzed electron ‘whirling wave’ solutions \cite{17}. Tonomura et al. \cite{18, 19} proposed the use of electron holography (electron matter wave interferometry) to detect magnetic monopoles, describing the spiral electron wavefront topology and tell-tale forked interference fringes that would result from a scattering event with a monopole. Bialynicki-Birula et al. \cite{20} predicted the presence of quantum vortex states in particle wavefunctions and theoretically studied their motion. Bliokh et al. predicted phase vortex states for free electrons \cite{21} and considered their relativistic propagation dynamics.

In 1992, Allen, Beijersbergen, Spreeuw, and Woerdman (ABSW) \cite{1} pointed out that photons could also be prepared in quantum vortex states, opening an accessible path to quantum vortices in freely propagating single-particle systems (photons). Commonly referred to as ‘optical vortex beams’, ‘twisted light’, or optical orbital angular momentum (OAM), these photon angular momentum states have enabled thousands of advances in a great many fields within optics. That similar states could also be applied to matter waves only became obvious after parallel advances in the unrelated fields of coherent electron microscopy and matter wave optics.

Recently, researchers demonstrated that freely propagating electrons can similarly be prepared in quantum vortex states \cite{6–8}. Similar to an optical vortex, the electron vortex is remarkable for its spiral phase singularity and associated quantized OAM. Yet unlike photon OAM, electrons possess both charge and rest mass. Thus, these demonstrations of free electron vortices were the first experimental observations of single, non-interacting massive particle wavefunctions occupying vortex states. The electric charge of these OAM states imparts a magnetic moment, such that electron vortices can interact with magnetic fields and quantum systems in new ways compared to photon OAM. Furthermore, the de
Broglie wavelength of electrons ($\lambda = 1.97$ pm in the case of 300 keV electrons) is much shorter than for experimentally accessible photons, and provides a means of exploring OAM effects at much smaller length scales. These unique properties of the electron vortices will be discussed here.

The outline of this paper is as follows: The key experimental components needed for observations of electron vortices - a coherent electron beam system and singular electron optical elements - will first be discussed in Section II. Experimental demonstrations of electron vortex beams with OAM will be presented in Section III followed by a model of such states and a discussion of their magnetic properties in Section IV. Finally, Section V will conclude by making a connection between this recent work and that of Suzuki and Willis, who noted that an azimuthal phase in an optical field could be generated by diffraction from forked structures [22, 23].

II. COHERENT ELECTRON OPTICS

A schematic representation of the experiments discussed here is shown in Fig. 1. The recent experimental work with electron vortex beams is enabled by two technologies: (a) improvements in high-resolution transmission electron microscopy, and (b) electron wavefront engineering using nanofabricated optics.

A. Transmission electron microscope: A coherent electron optics bench

Many of the tools necessary for experiments in coherent electron optics are provided in a modern transmission electron microscope (TEM). State-of-the-art aberration-corrected TEMs provide a bright source of coherent electrons in the field emission gun (the electron analog to a tunable laser), several magnetic lenses (the electron analog to high-precision spherical lenses) with adjustable focal lengths, several sets of independently adjustable magnetic quadrupole lenses (the electron analog to cylindrical lenses), scan coils (the electron analog to prisms), if an aberration corrector is present there are additional electrostatic and magnetostatic quadrupoles, hexapoles, and even octopoles, adjustable apertures, four multi-axis automated stages with sub-micron positioning accuracy, a Möllendstedt wire (the electron equivalent of an optical biprism), an electron energy loss spectrometer (EELS) with
Figure 1. Schematic illustration of electron vortices produced from a diffraction hologram. A spatially coherent electron wavefunction (a) illuminates a nanofabricated forked diffraction grating (b). The resulting diffracted portions of the wavefunction (c) possess quantized phase vortices. This diffraction pattern from many non-interacting electrons being diffracted is imaged in an electron microscope (d). For the experimental image adapted for (d), the undiffracted 0-order beam was blocked by a beam stop.

sub-eV accuracy, high-speed single pixel detectors (the equivalent of a photodiode), imaging detectors (CCDs with scintillators) with high quantum efficiency and high dynamic range. In addition, a modern TEM provides extremely stable control electronics, a moderately high vacuum, and an efficient airlock for introducing and removing materials from the system.

The electron beams supplied by a TEM equipped with a field emission gun (FEG) are analogous to a low-intensity laser in terms of spatial coherence. While there is no predictable phase relationship between consecutive electrons due to the stochastic nature of the emission process, the very small source size and beam-defining optics result in a high degree of transverse (spatial) coherence. While the transverse coherence width of the electron can be large (nearly as large as the width of the beam, which can vary from 0.1 nm to 1 mm), the energy spread of the beam ($\Delta E \approx 0.4\ eV$) and the accelerating voltage (typically $E = 300\ \text{keV}$) results in a longitudinal coherence length for the electrons of only approximately $2\ \mu\text{m}$. While this coherence length (and associated coherence time) is quite short compared to that of lasers, note that this coherence length is quite long compared to the electron de Broglie wavelength ($\lambda = 1.97\ \text{pm}$). The relative longitudinal coherence of electrons in the TEM,
defined as the ratio of the coherence length to the de Broglie wavelength, is on the order of $10^6$ - comparable to a typical HeNe laser. Thus, for the studies of electron OAM presented here, it is reasonable to model the electron wavefunctions as being monochromatic.

It should also be noted that, the experiments described here probe single-electron physics. To a good approximation, the electrons in the TEM beam are both (a) non-interacting and (b) prepared identically. It is only near the field emission tip that the density of electrons is large enough for mutual Coulomb interactions comes in to play, yet even here the phase-space packing density of electrons is quite low. After the electrons are accelerated to 300 keV, they are separated by an average distance on the order of 10 cm apart within the column, even when using a high beam current of 100 pA. This average spacing is too diffuse for multiple electrons to interact. The high degree of transverse spatial coherence of the beam means that electrons are prepared with nearly identical transverse wave functions. In a typical experiment, an imaging detector measures the probability distribution of an ensemble of a great many identically prepared electrons. Thus, the digital images presented here, to a reasonable approximation, represent the probability distribution of a single electron.

B. Singular Electron Optics

Conceptually, there is little difference between the physics of engineering the phase of a photon and that of a particle wavefunction. Electron vortices are prepared using a new class of singular optics for charged particles. Spiral phase plates for electrons - either material [6] or magnetic [24, 25] - can be used to directly imprint an azimuthal phase onto an electron beam. Interestingly, the magnetic spiral phase plate concept approximates the same scattering from a magnetic monopole that Dirac studied [11, 12]. As is the case of singular optics for light, spiral phase plates are challenging to fabricate, and it is quite difficult to imprint a precisely quantized spiral phase onto an electron wavefunction using these devices. Unless a precisely controlled single quanta of magnetic flux emerges from the tip of the magnetic needle, magnetic spiral phase plates will imprint a 'seam' or step in the phase along a radius away from the central vortex, resulting in fractional OAM that is not stable upon propagation [26]. Magnetic needle-based phase plates also become saturated in the external field of a magnetic lens, such as is the case when using them in conventional aperture locations within an electron microscope. However, two groups working independently are making advances
in the design, fabrication, and use of these devices [24, 25].

Diffraction holograms provide an alternative means of precisely imprinting OAM in all types of matter waves - not just electrons. Similar to gratings used in light optics [27, 28], electron vortices can be produced by diffraction from nanoscale holographic gratings featuring a fork dislocation [7, 8, 29] (Fig. 1). As in optics, the dislocation at the center of a forked grating encodes a spiral phase. This spiral phase can be revealed by Fourier-filtering the spatial components of an image of the grating, as shown in Fig. 2. The number of extra grooves present in the grating determines the topological charge (winding number) of the spiral phase. The lateral nanoscale position of lines (actually grooves) within the grating pattern determines the \textit{picometer} shift in the phase of the electron de Broglie wave. Thus, the overall size of the patterned area, the resolution with which the line features can be placed, and the spatial coherence of an array of such lines are all factors of the hologram design.

Figure 2. (a) A TEM micrograph image of a silicon nitride forked diffraction grating used to imprint spiral phase onto electron beams. The lighter lines are grooves etched partially through the silicon nitride membrane using focused ion beam (FIB) milling, and have a spatial periodicity of 76 nm. (b) The fork in the diffraction grating encodes an azimuthally varying spatial phase (represented by color scale) in the lateral registration of the grooves, revealed here by Fourier-filtering the TEM micrograph image in (a). (c) A radially-averaged plot of the phase in (b) shows that it varies smoothly and linearly as a function of the polar angle \( \phi \) and wraps continuously to \( 2\pi \). In an actual electron diffraction experiment, this spiral phase is imprinted onto the +1 diffraction order, and the opposite winding direction is imprinted onto the -1 diffraction order.
Electron diffraction holograms must have an extremely small average spatial period (lattice spacing) between lines in order to provide sufficient angular separation between diffracted electron beams. This can be achieved using modern nanofabrication techniques, such as e-beam lithography (EBL) [30, 31] or focused ion beam (FIB) milling [8, 29, 32]. In FIB milling, a beam of 30 keV Ga\(^+\) ions is focused to nanoscale dimensions onto a substrate and mills a hole into it of a prescribed depth. The Ga\(^+\) beam is then steered according to the desired holographic pattern. The substrate is this case is a thin (30 nm, 50 nm, or 100 nm) membrane of silicon nitride suspended in a silicon frame, coated with a thin layer of metal in order to make it conductive. This recipe for electron diffraction holograms was used to produce the various gratings shown in this paper. With spatial periodicities (pitch) between 50 nm and 100 nm, these gratings provide an angular separation between diffracted 300 keV electrons of 39 \(\mu\text{rad}\) to 20 \(\mu\text{rad}\), respectively, which is sufficient for use in a TEM.

Silicon nitride is suitable for nanoscale diffractive optics for electrons because it is mechanically strong, does not melt under exposure to an intense electron beam, and it is transparent to electrons which, as will be discussed, is important for use in efficient phase gratings and interferometry. We find that these structures can be used over the course of several months in a TEM. With prolonged exposure to an electron beam under non-ideal vacuum conditions, the diffraction gratings can get contaminated due to carbon deposition, which decreases their diffraction efficiency and introduces unwanted phase distortions in the beam. Plasma cleaning the gratings restores their original functionality.

A major challenge with diffractive electron optics is that only a fraction of the electron beam incident on a grating gets diffracted into a desired diffraction order. The diffraction efficiency of a grating, defined as the beam current in a desired diffraction order relative to the beam current incident on the grating, thus becomes a major concern for use in an electron microscope, where experiments are often starved for signal. Whereas our early nanofabricated forked holograms [29] served as binary masks for low-energy electrons, for the demonstration of electron vortices in [8] we developed electron-transparent gratings. The patterned silicon nitride gratings used for much of our research work by modulating the phase of an illuminating plane wave, rather than modulating the amplitude of a wave by removing parts of the beam (scattering or absorbing electrons). Several groups are now optimizing the depth and shape profile of the grooves milled into these electron-transparent membranes in order to maximize the beam current in a desired diffraction order [32, 35]. Non-relativistic
electrons propagating through the material membrane acquire a phase \( \phi = eVL/v\hbar \), where \( V \) is the mean inner potential of the membrane, \( L \) is the projected thickness, and \( v \) is the electron’s velocity. Thus, varying the thickness of the grating as a function of position allows one to modulate the phase of the electron wave. For silicon nitride patterned with FIB, we find that a thickness modulation \( \Delta L \) of 30 nm to 40 nm is enough to modulate the phase of a 300 keV electron by \( \pi \) radians. We have successfully made blazed gratings that place over 70% of the incident beam current into a diffraction order, and sinusoidal gratings that suppress the 0-order and place 30% of the incident beam current in each of the +1 and -1 diffraction orders.

Figure 3. Scanning electron microscope (SEM) image of several different singular electron holograms (circular regions) fabricated on the same silicon nitride membrane. This patterned membrane is suspended over a square window in a silicon frame. Though horizontal support structures are apparent on some of the gratings, at this image resolution the fine grooves of the gratings are not visible. This SEM image was recorded at much lower beam energies than those used in TEM. In a TEM, the membrane is transparent, such that an in-focus image barely shows any contrast where the gratings are.

While a digitally reconfigurable hologram such as a spatial light modulator (SLM) does not yet exist for electron beams, several different nanofabricated holograms can be placed on a single silicon nitride membrane (Fig. 3). Such a device can be installed as an aperture of a condenser lens in a TEM, and positioned such that only one grating at a time is illuminated.
Figure 4. (a) A TEM image of the 80 keV electrons diffracted from the grating in Fig. 2. The central order has been removed by a beam block. A color scale is used for this and subsequent images of diffraction patterns in order to make less intense features visible. (b)-(d) Magnified images of the +1, +2, and +3 diffraction orders. (e) Azimuthally averaged radial profiles of each of these diffracted beams reveals the dependence of the vortex core size on the topological charge. The dashed lines are fits derived from a Laguerre-Gaussian model of the electron wavefunctions (Equation 1).

by electrons. This provides versatility to conduct many different experiments with the same device, reducing the need to repeatedly vent the TEM column.

III. EXPERIMENTS AND DEMONSTRATIONS OF FREE ELECTRON VORTICES

A. Production of electron beams with OAM from diffraction holograms

Fig. 4 presents a TEM electron diffraction experiment using the grating shown in Fig. 2. At 80 keV ($\lambda = 4.18$ pm), electron beams diffracted from this grating had a free space angular spread of only 50 $\mu$rad. However, the strength of the TEM imaging system was employed to magnify this angular separation to observe the diffracted waves in the far field regime (Fraunhofer approximation), such that the diffraction spots could be resolved when projected onto the imaging detector.

Experimental results such as those shown in Figs. 4 indicate that low-OAM states for electron vortex beams appear to be stable. That is, despite the rest mass and charge of the electron wavefunction, these states do not appear to radiate or decay upon propagation through free space and their spiral phase is maintained. These states are stable even to higher
OAM. As shown, in Figs. 5 and 6, there appears to be no limit to the amount of OAM that can be imparted to an electron wavefunction. The electrons detected in the diffracted rings in Fig. 5 with high OAM are like Rydberg states, but without the central potential. These high OAM ring-shaped distributions do not radiate in free space and are each eigenstates of orbital angular momentum. Using focal series, we find that these distributions are self-similar upon propagation. However, as will be discussed in Section IV, the radial size of the wavefunction evolves.

Figure 5. (a) An SEM micrograph image of a nanofabricated silicon nitride diffraction hologram with 15 extra grooves for producing electron beams with high OAM. (b) The $\ell = 15$ phase topology encoded in the hologram can be revealed by Fourier-filtering the SEM micrograph image. (c) TEM images of an actual electron diffraction pattern (300 keV) show electron vortex beams with multiples of $15\hbar$ units of OAM.

B. Measuring the spiral phase of electron OAM beams

As discussed in [8], the use of electron-transparent phase gratings provides a convenient way to characterize the wavefront of the diffracted beams. Fig. 7a shows the results of a simple, single-grating electron interferometry experiment in which a wide electron beam coherently illuminates a forked grating and a large area of the transparent membrane surrounding it. The portion of the electron wavefunction that is transmitted through the unpatterned membrane serves as a reference wave, and at intermediate values of TEM defocus this overlaps the diffracted beams emanating from the grating. The interference between a helical electron wavefront and a flat wavefront is shown in Fig. 7a. The fork dislocation predicted [18, 19] and first measured [6] by Tonomura et al. is visible in the interferogram.
Figure 6. Electrons with extremely high OAM diffracted from a grating with topological charge of 200 (a) and 800 (b). A color scale has been applied to (a) to make higher orders visible. For (b), a log scale is applied to make lower intensity, higher diffraction orders with extremely high amounts of OAM visible in the image. Electrons with $4000\hbar$ are visible. Such beams are very sensitive to imperfections in the electron optics, and any small leftover astigmatism results in elliptic distortions to the beams.

A holographic reconstruction of these fringes, shown in Fig. 7b, makes the spiral phase topology of the diffracted beams quite evident.

In most ways, free electron wavefunctions possessing OAM behave just like twisted light. The dark node in the center of the electron vortex beam remains there under propagation through free space - even throughout the Rayleigh range of a beam waist [8] - as described by the Laguerre-Gaussian wavefunction developed for optics (Equation 1). The evolution of optical OAM modes in astigmatic optical systems has been explored experimentally using cylindrical lenses; e.g., [36]. Likewise, electron OAM states transform similarly upon propagation through astigmatic optical systems [37,39]. In Figure 8, electron OAM beams were propagated through a magnetic quadrupole lens, the electron-optical equivalent of a cylindrical lens. An electron vortex beam with topological charge 2 breaks up into two separate vortices. This can be used as a quick way to characterize the OAM of the electron beam [39,40]. Schattschneider et al. used this to transform electron beams modes [41].
Figure 7. Electron interferometry reveals the spiral phase topology imprinted onto electron vortex beams diffracted from the grating shown in Fig. 2. (a) An intermediate-field (far-defocus) TEM image of the grating reveals interference between the diffracted wave (circled in dashed yellow) and a reference wave directly transmitted through the surrounding transparent membrane, as discussed in [8]. The brighter concentric circular pattern on the right is the undiffracted 0-order of the grating. (b) Holographic phase reconstruction of the interferogram confirms the spiral phase of the diffracted beam.

IV. MODELING FREE ELECTRON VOR Ritches

Free electron vortex states can be visualized and formally modeled in several ways. Just as with all quantum vortices, the electron vortex wavefunction is defined by an azimuthal phase, \( \exp \left( i \ell \phi \right) \), where the integer \( \ell \) is the topological charge, or winding number, defining the spiral phase singularity. The corresponding OAM of the electron about an axis parallel to the vortex core (defined here as the optical axis in the +z direction) is equal to the topological charge times Planck’s constant, \( L_z = \ell \hbar \). Unlike quantum vortices in bound systems, however, free electron vortices have characteristic dimensions that evolve in space and time, as described by the Schrödinger equation.

A. A convenient model for paraxial electron OAM wavefunctions

A Laguerre-Gaussian beam with no radial modes \((p = 0)\) provides a convenient, simple model of the electron vortex wavefunction \( \psi_\ell \). It describes the evolving width, wavefront
Figure 8. Transformation of electron OAM states within astigmatic electron optical systems. A magnetic quadrupole was used to introduce the astigmatic perturbation to the beam. The central dark spot of a single topological charge state distorts into an edge-type phase dislocation, forming a dark line across the spot (upper left). (Right) A series of images of astigmatic higher OAM states shows that each state becomes an ellipse. The numbers in each image of this sequence denote the increasing strength of the quadrupole field. One can see that negative OAM states on the left side of the diffraction pattern are all tilted in different directions from those on the right. The evolution of each OAM state is reminiscent of a current loop precessing in space. At high enough quadrupole excitation (lower rightmost image), the OAM states seem to have inverted.

The spatial phase $\Phi_\ell(\rho, \phi, z)$ of this wavefunction is

$$\Phi_\ell(\rho, \phi, z) = k z + \ell \phi + \frac{k \rho^2}{2 R(z)} - (|\ell| + 1) \arctan(\frac{z}{z_R}) + \varphi_\ell$$

where $R(z)$ is the radius of wavefront curvature. For generality, $\varphi_\ell$ is a phase that can be introduced by interactions with external parameters that depend on the topological charge. When describing propagation through free space, $\varphi_\ell = 0$. 

$$\psi_\ell(\rho, \phi, z, t) = \frac{A_\ell}{w(z)} \left( \frac{\sqrt{2}\rho}{w(z)} \right)^{|\ell|} \exp\left( -\frac{-\rho^2}{w(z)^2} \right) e^{-i\omega t} e^{i\Phi_\ell(\rho, \phi, z)},$$

where $A_\ell$ is a normalization constant such that $A_\ell \int \psi_\ell(\rho, \phi, z) d\vec{r} = 1$, $\omega = \frac{E}{\hbar}$ is the temporal frequency related to the electron’s energy, and the characteristic transverse size of the wavefunction, $w(z)$, evolves upon propagation according to standard Gaussian optics.
Equation 1 can be used to visualize the electron vortex wavefunction as a surface of constant phase. In Figure 9(a), such a wavefront is plotted as a parametric surfaces defined by $r(\rho, z) = (\rho \cos \phi(\rho, z), \rho \sin \phi(\rho, z), z)$, where $\phi(\rho, z)$ is provided by setting $\Phi_\ell$ in Eq. 1b equal to an integer multiple of $2\pi$. In this visualization, the amplitude of the wave (the modulus of Eq. 1a) is represented by the shading and opacity of the surface. A wavefunction model of freely propagating electron OAM states, such as Equation 1, can be applied to understand the scattering of these states from other systems, or their dynamics in external fields.

![Figure 9](image)

Figure 9. Representations of an electron vortex wavepacket. (a) A Laguerre-Gaussian wavefunction serves as a model for electron de Broglie wavepackets in a focused electron vortex beam. The helical wavefront is plotted as a surface of constant phase, with an opacity is proportional to the amplitude of the wavefunction. (b) A geometric model of the beam can be derived by evaluating the momentum of the Laguerre-Gaussian function. Here, the probability current evaluated at the radius of peak amplitude of the wavefunction forms a collection of skewed straight rays. (c) In the center-of-mass frame of reference, shown here at various times during propagation through a focus, the electron vortex probability distribution is torus-shaped, with extent along $z$ determined by the spread in longitudinal momentum. The size of the donut contracts as the electron propagates through a focus, and then expands again afterwards.

A ray model of the free electron vortex OAM state above can be derived by considering the local charge current density of the wavefunction above. The charge current density associated with this wavefunction is given by $\vec{j}(\vec{r}, t) = -\frac{ie\hbar}{2mc} \left( \psi^* \vec{\nabla} \psi - \psi \vec{\nabla} \psi^* \right)$. Berry and McDonald [42] performed an identical calculation of the local momentum distribution of an optical
vortex. They found that while the energy flow lines within an optical vortex wavefunction can curl around the optical axis in the evanescent regions of a Laguerre-Gaussian beam, the flow lines become simple straight rays at the locations of peak probability in the beam. A similar interpretation of optical vortex beams was provided by [43]. A geometric model of charged particles following skewed, straight trajectories in space (Fig. 9b) offers practical benefits in visualization and simulations.

Note that in a reference frame co-propagating with the electron center-of-mass, the electron vortex probability distribution can be visualized as a toroidal cloud of charge, with a longitudinal extent determined by the energy spread along the axis(Fig. 9c). This donut-shaped electron cloud rotates, and as it does so it expands in free space (or contracts, if initially given a negative radial momentum by a lens). A simple model of the free electron vortex as a loop of charged current can serve as a useful tool to understand how these states interact with and evolve in magnetic fields, as will be discussed in the following section.

B. Free electron vortices in a magnetic field

Unlike photon OAM, the electron is a charged particle, and so the quantized OAM of the electron vortex state has an associated magnetic dipole moment. This magnetic moment is different from the intrinsic magnetic moment due to the electron’s spin. The size and direction of an electron vortex magnetic moment $\mu_\ell$ can be calculated from the charge current density using the formula

$$\vec{\mu}_\ell = \frac{1}{2} \int \vec{r} \times \vec{j} d\vec{r}.$$ 

The azimuthal term of the cross product:

$$\mu_\ell = -ie\hbar \int_0^\infty \int_0^{2\pi} \left[ \psi_\ell^*(\frac{1}{\rho} \frac{\partial \psi_\ell}{\partial \phi}) - \psi_\ell \left( \frac{1}{\rho} \frac{\partial \psi_\ell^*}{\partial \phi} \right) \right] \rho^2 d\phi d\rho$$

Inserting Eq. 1, we find that each electron vortex wavefunction possesses an orbital magnetic moment equal to $\mu_\ell = -\frac{e\hbar}{2m_e}$. This serves as another example that all magnetic phenomenon arise from the angular momentum of electrical charges.

We can generally express the magnetic moment of any electronic state as $\mu_\ell = -g\mu_B \ell$ where $\mu_B = \frac{e\hbar}{2m_e}$ is the Bohr magneton and $g$ is the gyromagnetic ratio. In deriving Eq. 2 we have shown that $g = 1$ for the specific case of a Laguerre-Gaussian electron vortex wavefunction, as is the case for well-defined orbital states in bound electron systems. Gallatin et al. [44] used this to show that because the gyromagnetic ratio $g = 1$, in an external magnetic
field the orbital axis (and orbital magnetic moment) of an electron vortex wavefunction will
precess. However, the magnetic moment of the vortex state precesses at a different rate in
the magnetic field compared to the classical orbital motion of the center-of-mass. That is,
the Larmor precession frequency of the orbital magnetic moment is half of the cyclotron
frequency.

The Hamiltonian of an electron vortex propagating in an external magnetic field is

\[ H = H_0 + U \]

where \( H_0 \) is the unperturbed Hamiltonian of the electron and \( U \) is the Zeeman interaction
energy of the electron vortex with the external field:

\[ U = -\vec{\mu}_{\ell} \cdot \vec{B} = \frac{e\hbar \ell B}{2m_e} \]

where we have assumed the magnetic field points along the orbital quantization axis of the
vortex wavefunction. Using the WKB approximation, we compute that this interaction leads
to a field-dependent phase shift of the electron wavefunction:

\[ \varphi_{\ell} = \frac{U\Delta t}{\hbar} = \frac{U\Delta z}{\hbar v} = \frac{e\ell B\Delta z}{2m_e v} = \frac{e\ell B\Delta z\lambda}{2\hbar} \]

Bliokh et al. \[45\] also derives this phase shift. The orbital magnetic moment of the electron
vortex state interacts in unique ways with an external magnetic field, for example inducing
rotations in superpositions of OAM in a longitudinal field \[45,47\], OAM precession in a
transverse field \[44\], or inducing spin-orbit couple in a nonuniform field \[48\]. Further exper-
imental research in this area is likely to provide insight into fundamental electron physics,
magnetism, and electron microscopy.

V. CONCLUSION

Applications for engineered phase singularities such as optical vortices have been de-
veloped for optical imaging, communications, quantum optics, and micromanipulation for
over two decades. The many applications of optical vortices developed for light microscopy,
such as spiral phase microscopy \[49\], are generating a surge of interest within the electron
microscopy community, with hopes that electronic analogs to these techniques might be de-
veloped \[50\]. The quantized OAM of electron vortex state couple to matter in ways that are
slightly different than photon OAM, and this holds promise for a new way to probe chirality and angular momentum at the nanoscale [51–55].

In the 1950’s, Suzuki and Willis [23] investigated diffraction from edge dislocations in crystals, demonstrating optical vortices in beams of light using diffraction gratings. To understand electron and x-ray diffraction from edge dislocations in crystals - i.e., forked periodic structures - they conducted optical experiments to simulate such electron diffraction using a Lipson diffractometer. They produced an optical mask model of crystal edge dislocations, a two-dimensional array of holes with a dislocation in the center. When they diffracted coherent light from the structure, they observed dark spots in the diffracted beams, which we now know are due to phase vortices. In [23], Willis reviews Suzuki’s theoretical model of the wavefunction giving rise to the dark spots. Indeed, Suzuki had apparently posited a spiral phase singularity, though did not apparently publish this result himself.

In 1992, Allen, Beijersbergen, Spreeuw, and Woerdman revolutionized the world of optics by showing that light can occupy quantized orbital states, spawning thousands of discoveries in the quarter century since. Recently, several groups showed that the ABSW result applies to electron wavefunctions and other matter waves, too. Thus, optical OAM is a general property of all wavefunctions propagating in free space. These recent results underline how the ABSW paper brings new life to a 175-year-old idea in physics: the topology and angular momentum of vortex structures provides a convenient model for many fundamental effects in low-energy physics.

VI. DATA ACCESSIBILITY

Raw data files consist primarily of TEM images in the DM3 format, and are available upon request to the corresponding author.

VII. AUTHORS’ CONTRIBUTIONS

BM conceived of and designed the study, performed experiments and data analysis, and drafted the manuscript. AA and JP fabricated the diffraction holograms. PE and ML carried out experiments in Figs. 2, 4, and 7, AH carried out experiments in Fig. 5 and 8, VG and TH carried out experiments in Fig. 6. All authors read and approved the manuscript.
Figure 10. Early work on optical diffraction (top row) from 2D masks is reproducible with electron diffraction (bottom row). (a) An optical mask produced by Willis (adapted from [23]) simulates a natural crystal edge dislocation, with a topological charge of 1 in the $x$ direction and 0 in the $y$ direction. (b) Light diffracted by this mask produces optical vortices (figure adapted from [23]). Here the $(0, 3)$, $(0, 4)$, and $(0, 5)$ diffraction spots corresponding to optical vortices with $\ell = 3, 4,$ and 5, respectively, have been enlarged to reveal the central dark spots characteristic of topological charge. (d) A nanofabricated 2D grating for electrons simulates an edge dislocation in a single crystal lattice plane. (d) The resulting electron diffraction pattern forms a 2D array of electron vortices. In this diffraction image (300 keV, $\lambda = 1.97$ pm), the diffraction spots were defocused in order to enlarge the diffraction spots.
VIII. COMPETING INTERESTS

I have no competing interests.

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[36] Bekshaev AY, Soskin MS, Vasnetsov MV. Transformation of higher-order optical vortices


