Vortex beams for atomic resolution dichroism

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Abstract

Vortex beams carrying orbital angular momentum have been produced recently with electron microscopy by interfering an incident electron beam with a grid containing dislocations. Here, we present an analytical derivation of vortex wave functions in reciprocal and real space. We outline their mathematical and physical properties and describe the conditions under which vortex beams can be used in scanning transmission microscopy to measure magnetic properties of materials at the atomic scale.

Keywords

electron vortex, orbital angular momentum, STEM, EELS, dichroism, ion vortex

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It is now possible, using state-of-the-art electron microscopes, to obtain direct chemical identification of individual atoms based on image intensity [1] together with spectroscopic fine-structure information [2]. However, it has not so far been feasible to measure magnetic properties of individual atoms or single atomic columns. One way to achieve atomic-scale magnetic information within an electron microscope would be to polarize the electron beam such that each electron carries a specific orbital angular momentum (OAM).

Vortex photon beams carrying an OAM have been known (and widely used) for almost 20 years in optical physics [3,4]. Recently, it has been shown that vortex beams with desired OAM can also be produced in a transmission electron microscope (TEM) [5,6,7]. Specifically, by interfering an incident electron beam with a grid containing dislocations, Verbeeck \textit{et al.} [6] showed that electron vortex beams can be generated to measure magnetic circular dichroism with a spatial resolution of 250 nm using electron-energy loss spectroscopy (EELS) in a TEM. Their work also implies that by using a scanning (S)TEM (see Fig. 1), it may be possible to achieve the goal of atomically resolved measurements of magnetic properties.

In this letter, we derive an analytical description in reciprocal and real space of vortex beams carrying an OAM by evaluating the diffracted pattern formed by a plane wave that has passed through a grid containing dislocations. We find that a vortex beam can be expressed by generalized hypergeometric functions. It has an O-ring shape and contains a phase singularity in real and reciprocal space. Our calculations show that electron vortex probes, produced with current state-of-the-art aberration-corrected electron microscopes, achieve a spatial resolution in imaging and spectroscopy that is \(\simeq 10\) times worse than that for electron probes without OAM. We find that an alternative route to achieve atomic resolution is to either accelerate the electrons to 2 MV (or above) or to use ion beams (He\textsuperscript{+}) rather than electrons.

An analytical expression of an electron (or ion) vortex wave function carrying an OAM can be obtained by calculating the diffraction pattern when a plane wave goes through a grid containing fork dislocations. This is equivalent to obtaining the
Fourier transform of the mathematical function that describes such a grid. Mathematically, an ideal grid with \( l \) fork dislocations can be defined as 
\[
\cos(l\phi - \arccos \phi),
\]
where \( a \) is the grid spacing and \((r,\phi)\) are cylindrical coordinates in real space. Thus, the diffraction pattern is obtained by evaluating the following integral:
\[
\frac{1}{2} \left( e^{il\phi - iar\cos \phi} + e^{-il\phi + iar\cos \phi} + e^{-il\phi + ikr} + e^{il\phi - ikr} \right) dr,
\]
where \( k \) is the vector position in reciprocal space. Note that Eq. (1) contains two terms. Each term can be interpreted as a single transmitted wave function carrying an OAM of \( \pm lh \), and the center of propagation in reciprocal space which is located at \( k_x = \pm 1/a, k_y = 0 \). The diffraction pattern when a plane wave is transmitted through a real physical (binary) grid \( l \) dislocations follows Bragg’s law and is composed of a diffracted beam (without OAM) centered at \( k = 0 \) plus a set of \( n \) pairs of diffracted beams centered at \( k_x = \pm n/\alpha, k_y = 0 \) with an OAM equal to \( \pm nlh \), where \( n \) is just the diffraction order number. The phase and amplitude of each pair of diffracted electron (ion) beams of order \( n \) from the binary grid can be described by those of an ideal grid (1), where \( l \) is replaced by \( nl \), multiplied by the function \( \frac{1}{2} \sin (n\pi/2) \) [4]. Therefore, we can write a general vortex wave function \( \Lambda_l(k) \) carrying an OAM of \( lh \) as
\[
\Lambda_l(k) = e^{il\phi - iar\cos \phi + ikr} dr.
\]
The integral in Eq. (2) for a circular grid of radius \( R \) is
\[
\Lambda_l(k) = e^{il\theta} \frac{1}{K^2} \int_0^{KR} r J_l(\tau) d\tau,
\]
where \( J_l \) is the Bessel function of the first kind with order \( l \) and \( \theta \) the angular coordinate in reciprocal space (see the appendix for details). Eq. (3) defines a vortex wave function in reciprocal space.

Figure 2 shows the calculated intensity, phase and amplitude profile of a vortex wave \( \Lambda_1(k) \), \( \Lambda_1(k) \) presents an O-ring shape and a phase singularity at \( k = 0 \), as can be observed in Fig. 2. The \( k \) values where \( \Lambda_1(k) = 0 \) (labeled as \( s_i \) in Fig. 2) depend entirely on

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1 For instance, see p. 158 in Ref. [4]. The definition of an ideal grid is given in the context that the transmitted wave function consists only of two vortex beams with opposite OAM as is shown in Eq. (1) and discussed in the main text.
the radius $R$, as expected from the diffraction theory. Larger values of $R$ result in smaller values of $s_l$ and vice versa. This means, from a calculation perspective, that the magnification of an electron vortex at the objective aperture plane can be controlled by choosing an appropriate value of $R$.

The analytical expression for a single electron (ion) vortex wave in real space is obtained by applying an inverse Fourier transform to $\Lambda_l(k)$.

Therefore, a vortex carrying an OAM $lh$ can be expressed in real space as

$$\Lambda_l(r) = \int e^{-i k r - i \chi(k)} \Lambda_l(k) \, dk. \quad (4)$$

In Eq. (4), we have added the term $i \chi(k)$ in the exponential in order to describe the change of phase that an electron (ion) undergoes due to imperfections (aberrations) of the objective lenses. $\chi(k)$ is known as the aberration function [9]. Eq. (4) contains all the physics for a general description of a vortex in real space. However, for the investigation purpose of this study, which is to find how small an electron (ion) vortex can be made in real space, we set $\chi(k)$ to zero without loss of generality in our calculations. The last approximation is allowed because of the development in the last few years of aberration-correction optics [10]. When aberrations can be neglected up to values $k \leq k_{max}$, an electron vortex in real space is

$$\Lambda_l(r) = e^{i \phi} \int_0^{k_{max}} \frac{J_l(kr)}{k} \int_0^{kR} \pi \Lambda_j(\tau) \, d\tau \, dk. \quad (5)$$

With the definition of vortex beams in real space given by Eq. (5), we can proceed to investigate the electron optical conditions at which the electron vortexes reach their smallest physical size. This can be done by calculating the intensity profile $|\Lambda_l(r)|^2$. For all our calculations, we define $k_{max} = \alpha/\lambda$, where $\alpha$ is the half angle of the objective aperture ($\sim$30 mrad in state-of-the-art aberration-corrected electron microscopes) and $\lambda$ the wavelength of the electron.

First, we examine how an electron probe changes in size depending on the number of diffracted rings in $\Lambda_l(k)$ that are allowed within the objective probe-forming aperture (in other words, the relative magnification between the aperture in the diffraction plane and the objective aperture). Figure 3a shows the electron vortex probe intensity with an OAM of $h$ as a function of the number of diffracted rings, $s_l$, allowed in the objective aperture. The probe was calculated for the case of an electron microscope operating at 100 kV. We find that the best optical condition to obtain the smallest electron probe is when only the first diffracted ring ($s_1$) is allowed through the objective aperture. Choosing a value smaller than $s_1$ for $\Lambda_l(k)$ results only in effectively decreasing the total current of the electron probe without reducing significantly the electron confinement radius $R_c$ (defined as the position where the maximum intensity occurs) or the full-width at half-maximum on the Gaussian-like profile node $\Delta r$. This is to be expected, since the spread of an electron vortex in reciprocal space necessarily implies a localization of the electron probe in real space. We find the same behavior for electron vortexes with higher OAM.

We next calculate how the increase in the OAM of the electron vortexes affects their confinement in real space. We find that under optimum optical

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\[ \Lambda_l(r) = e^{i \phi} \int_0^{k_{max}} \frac{J_l(kr)}{k} \int_0^{kR} \pi \Lambda_j(\tau) \, d\tau \, dk. \quad (5) \]

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Footnote: In order to derive $\Lambda_l(r)$ from Eq. (4), we have used the same approach outlined in the appendix for $\Lambda_l(k)$, in particular, the integral with respect to $k$. In the definition of $\Lambda_l(r)$, we have left out phase shifts proportional to $\ell$ and other integration constants.
conditions (defined earlier), $\Delta R$ for electron vortexes with different OAMs increases slightly with the OAM (Fig. 3b). We also find that their confinement radius, $R_c$, increases linearly and in quasi-discrete values with the OAM. The last result is due to the quantization of angular momentum and can be understood in classical terms by the expression that relates angular momentum $L$, with the linear momentum $p$ and the radius of rotation $r$ of a particle, i.e. $L = r \times p$. Since the linear (transferred) momentum is the same for all the electron vortexes (i.e. the electron vortexes have the same kinetic energy and are formed with the same objective aperture), the orbit in which the electrons rotate necessarily needs to increase to satisfy the conservation of angular momentum. The smallest probe for an electron vortex at 100 kV (OAM = $\hbar$) has an $R_c = 0.370$ nm and a $\Delta R = 0.404$ nm. If images or EEL spectra were going to be taken using that electron probe, then the structural and spectroscopy information obtained would have a spatial resolution $d$ of 1.14 nm, where $d = 2R_c + \Delta R$. To put the last result into perspective, under the same optical conditions, an electron probe without the OAM is about 10 times smaller, which allows the acquisition of images and spectra with atomic resolution.

Increasing the acceleration voltage to 300 kV produces an electron vortex probe with an $R_c = 0.200$ nm and a $\Delta R = 0.215$ nm, resulting in $d = 0.615$ nm (see Fig. 4a). Increasing $\alpha$ to 40 mrad gives $R_c = 0.150$ nm, $\Delta R = 0.162$ nm and $d = 0.462$ nm. Even in the unrealistic case that $\alpha$ could be increased up to 60 mrad, we find that electron vortex beams would not be small enough to provide imaging and spectroscopy at the atomic scale. If the electrons are accelerated to 2 MV, then the vortex probe shrinks to values $R_c = 0.050$ nm, $\Delta R = 0.055$ nm and $d = 0.155$ nm (Fig. 4a), achieving the goal of atomic resolution.

Finally, we calculate the probe size for an STEM employing a beam of helium ions. We find that in a proof of principle He$^+$ microscope operating...
without aberration-corrected lenses (40 kV, δ = 0.1 mrad) [11], the probe size has an \( R_c = 2.030 \) nm, \( \Delta_R = 2.237 \) nm and \( d = 6.297 \) nm (Fig. 4b). In an aberration-corrected He microscope allowing a value \( \delta = 4 \) mrad, the He vortex probe is similar to that of a 2 MV electron vortex probe (i.e. \( R_c = 0.050 \) nm, \( \Delta_R = 0.057 \) nm and \( d = 0.157 \) nm).

In conclusion, we derive an analytical description in reciprocal and real space of vortex beams carrying the OAM. We find that in current aberration-corrected STEM, vortex probes can be confined to a diameter of 0.6 nm, which allows for the mapping of magnetic properties at the sub-nanometer level. But in order to achieve atomic resolution, our calculations indicate that either electrons have to be accelerated to 2 MV (or above) or the microscope needs to use ion beams (He\(^+\)) instead of electrons.

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**Appendix**

The integral shown in Eq. (2) can be evaluated by expanding \( k \cdot r \) as \( kr(\cos \phi \cos \theta + \sin \phi \sin \theta) \), where \( \phi \) and \( \theta \) are the angular coordinates in real and reciprocal space, respectively. For simplification purposes, it is also convenient to define \( \xi = r(-a + k \cos \theta) \) and \( \eta = kr \sin \theta \). The terms in the exponential \( i \xi \cos \phi \) and \( i \eta \sin \phi \) then can be replaced by using the definition of Bessel functions through a Laurent series to obtain the following expression:

\[
\Lambda_l(\mathbf{k}) = \sum_{n,m=-\infty}^{\infty} i^n \int_0^R r J_n(\xi) J_m(\eta) dr \int_0^{2\pi} e^{i(l+n+m)\phi} d\phi.
\]

(A.1)

The integral with respect to \( \phi \) vanishes for all values of \( n \) and \( m \) that do not satisfy the condition \( l = -(n + m) \). Thus, the only remaining terms after integrating with respect to \( \phi \) are

\[
2\pi \sum_{n=0}^{\infty} i^n \left[ (-1)^{n+1} J_n(\xi)J_{n+l}(\eta) + i J_{n+l}(\xi)J_n(\eta) \right] + 2\pi \sum_{n,m=1}^{\infty} i^n (-1)^m J_n(\xi)J_m(\eta) \delta_{l,m+n}.
\]

(A.2)

Eq. (A.2) can be simplified even further by performing the following analysis. First, the amplitude of the final solution must preserve an axial symmetry. By setting \( \xi \) or \( \eta \) to zero, the only term that survives in Eq. (A.2) is \( 2\pi (-1)^l J_l(\eta) \) or \( 2\pi i^l J_l(\xi) \), respectively. The previous result means that the amplitude and phase of Eq. (A.2) can simply be expressed as \( 2\pi J_l(\kappa) \) and \( e^{il\phi} \), respectively, where \( \kappa^2 = \xi^2 + \eta^2 \) and \( \nu \) is a linear function of \( l \) and the angle \( \theta \), i.e. \( \nu = l(\theta + \text{constant}) \). The value of the constant is obtained by noting that for \( \theta = \pi/2 \) (\( \xi = 0 \)), \( e^{i\nu} = (-1)^l \) and for \( \theta = 0 \) (\( \eta = 0 \)), \( e^{i\nu} = i^l \). This means that \( \nu = \theta + l\pi/2 \). Performing the change of variable of \( \tau = \kappa r \), Eq. (A.1) can be reduced to

\[
\Lambda_l(\mathbf{k}) = e^{il\phi} \frac{1}{k^2} \int_0^R r J_l(\tau) d\tau.
\]

(A.3)

Note that in Eq. (A.3), the variable \( a \) does not appear. Nevertheless, \( a \) can be recovered by defining \( k^2 = (k_x + 1/a)^2 + k_y^2 \), where the sign of \( a \) depends on the sign of \( l \). In the definition of \( \Lambda_l(\mathbf{k}) \), we left out the phase \( l\pi/2 \) and the constant \( 2\pi \). \( \Lambda_l(\mathbf{k}) \) has some interesting mathematical and physical properties which we will briefly discuss next for all values of \( l \), with the exception of \( l = 0 \), since \( \Lambda_0(\mathbf{k}) \) becomes the well-known solution for the Fraunhofer diffraction of a circular aperture.

(i) \( \Lambda_l(\mathbf{k} = 0) = 0 \). When \( R \to \infty \), \( \Lambda_l(\mathbf{k}) \) is constrained around the vicinity of \( k = 0 \) but it never reaches \( k = 0 \).

(ii) \( \Lambda_l(\mathbf{k}) \) by definition carries angular momentum \( l\hbar \). This statement can easily be verified by applying the momentum operator \( \hat{L}_z = -i\hbar \partial_\theta \) to \( \Lambda_l(\mathbf{k}) \).

(iii) \( \langle \Lambda_l(\mathbf{k}) | \Lambda_l(\mathbf{k}) \rangle \propto \delta_{l,l'}. \) Note as well that vortex beams always come in pairs as illustrated in Eq. (1).
The former statement is another way of expressing the law of conservation of angular momentum (i.e. the total angular momentum before the plane wave goes through the dislocation grid is zero).

\( (iv) \) \( \Lambda_l(k) \) vanishes when \( k \to \infty \).

\( (v) \) \( \Lambda_l(k) \) is not normalized. The integral with respect to \( r \) results in a generalized hypergeometric function of the form \( \mathcal{F}_0(; ; z) \) that needs to be calculated with the help of a computer for each value of \( l \) and \( R \). For the sake of simplicity and mathematical aesthetics, we decided to define \( \Lambda_l(k) \) as a non-normalized wave function.

References


