ELECTRON MICROSCOPY

A new spin on electron beams

Ideas about angular momentum that have been borrowed from optics could allow the magnetic and spin structures of materials to be studied on atomic scales with electron vortex beams.

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We are used to electrons occupying states with quantized spin and orbital angular momenta in atoms, but existing methods for generating high-energy electron beams are very inefficient at either producing or transferring states with well-defined spin and/or orbital angular momentum. This failure stems from the dominance of the direct Coulomb interaction over the spin–spin and spin–orbit interactions for high-energy electrons. As a consequence, even though electron beams can be focused to subatomic dimensions and used to measure the local composition and bonding of a material, they cannot reveal magnetic and spin information on comparable length scales. However, recent work by groups in Japan[1] and Europe[2] points the way to filling this gap by showing that it might be possible to measure and manipulate orbital angular momentum on the atomic scale with electron microscopes.

The key to creating an electron beam with quantized orbital angular momentum relies on techniques that were developed by John Nye and Michael Berry in 1974 to study dislocations in optical fields[3]. For example, if the pattern of the wave fronts resembles the pitchfork shape shown in Fig. 1a, then a dislocation core or vortex appears at the apex of the pitchfork, with the probability current (which is the energy flux or Poynting vector in optics) circulating about a point of indeterminate phase and zero amplitude. For electrons, the probability current associated with the vortex possesses an intrinsic orbital angular momentum and an associated magnetic moment that is determined by the shape of the wave packet[4].

The number of wave fronts added or removed at the core is an integer, l, and this results in the orbital angular momentum being quantized and equal to $l\hbar$, where $\hbar$ is Planck's constant, h, divided by 2π, and the integer l is known as the topological charge of the vortex. The optics community has long exploited optical vortex beams to transport orbital angular momentum independent of its polarization (or spin) state in applications such as optical traps and quantum information.

The first demonstration of orbital angular momentum in an electron microscope, reported by Masaya Uchida and Akira Tonomura of RIKEN earlier this year[1], relied on a stack of thin films of graphite naturally (and fortuitously) forming a spiral phase plate. They produced an electron beam with a helical phase and confirmed the presence of a vortex with $l = 1$ by interfering the vortex beam with a reference plane wave to form an electron hologram. This groundbreaking work was a proof of concept rather than a general tool, however, because there were problems avoiding contamination of the beam and maintaining precise spatial control of the pattern.

A few months later, Jo Verbeeck and He Tian of the University of Antwerp, and Peter Schattschneider of the Vienna University of Technology, overcame these problems by, essentially, running the RIKEN experiment in reverse[2]. Verbeeck and co-workers generated a hologram by digitally interfering a vortex beam with a reference plane wave in a computer. Then they carved a binary version of the hologram into a thin platinum foil using a focused ion beam. Finally, by illuminating the hologram with a coherent electron beam, they demonstrated that vortex beams with $l = \pm 1$ were produced in the far-field diffraction plane (Fig. 1a); beams

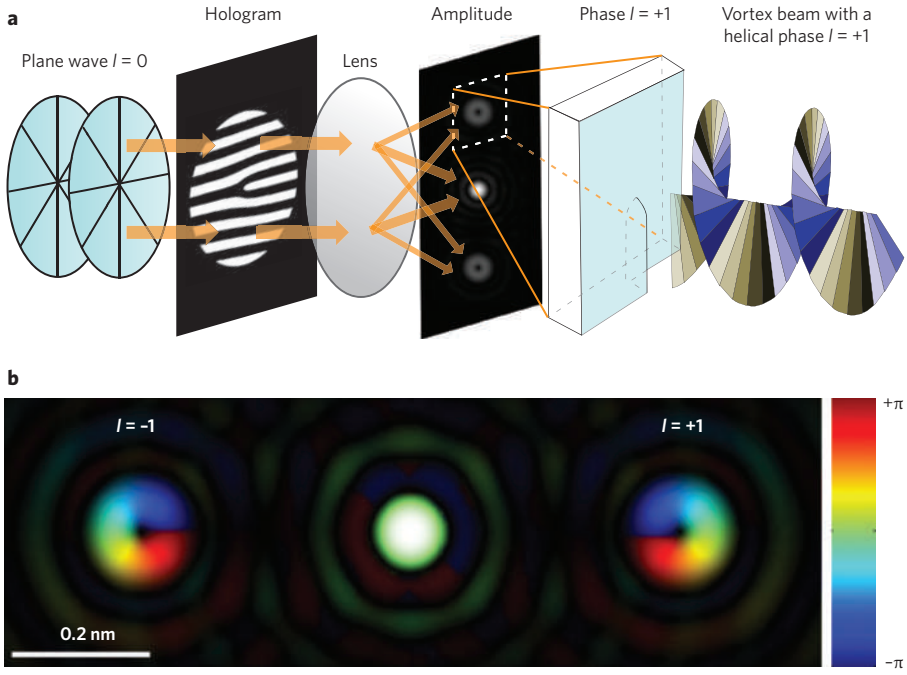

Figure 1 | Adding a twist to electron beams. **a**, Schematic showing a plane wave of electrons (left) being converted to a vortex beam (right) by a pitchfork hologram mask placed in the front focal plane of the electron lens. The image plane (to the right of the lens) contains the Fourier transform of the hologram, with two vortex side beams (thin orange arrows) spinning in opposite directions (and with opposite orbital angular momentum), and also a plane wave beam without any angular momentum. The dark centre of each side beam is the vortex; the strength (and sign) of the vortex is proportional to the orbital angular momentum. The discontinuity in the phase of the upper beam is also shown. **b**, Simulations of an atomic-scale vortex beam produced by a hologram mask (such as that in **a**) and a 300 keV electron beam typical of an aberration-corrected transmission electron microscope. The wave amplitude is indicated by the image intensity, and the phase by the colour bar. Note the phase discontinuities from $-\pi$ to $+\pi$ (blue to red) point in opposite directions for the $l = -1$ (left) and the $l = +1$ (right) vortices.

with higher-order topological charges were also observed.

The 'reverse' application has many practical advantages: the masks are easy to fabricate and are stable under a high-energy electron beam, and beams with arbitrary topological charges can be designed. Moreover, simulations that we have performed suggest that it should be possible to demagnify the beam down to atomic dimensions in an aberration-corrected scanning transmission electron microscope by placing the mask in the pre-specimen diffraction plane (Fig. 1b). (Verbeeck and co-workers produce a vortex beam in the post-specimen diffraction plane by placing the mask in the image plane where an aperture port was available.) This would make it possible to translate, rotate and excite individual atoms and molecules with an electron beam that was just an atom wide and carried a given amount of orbital angular momentum.

The Antwerp–Vienna team also demonstrated that hologram masks can be used in combination with electron energy loss spectroscopy (EELS) to study the handedness (or chirality) of systems. X-ray absorption spectroscopy with circularly polarized radiation is widely used for such experiments today, and although similar experiments have been performed with electrons, they have involved restricted and highly inefficient scattering geometries. Using hologram masks to generate and analyse beam states with well-defined orbital angular momentum provides a new and more efficient way of studying chiral systems.

By comparing the left- and right-handed side bands in the EELS signal from an iron sample, Verbeeck and co-workers were able to measure the spin state of ferromagnetic iron for a region with a diameter of about 250 nm, showing an improvement of a factor of ten over previous electron measurements in discrimination between angular momentum states. It should already be possible to image individual magnetic nanoparticles with this approach. Furthermore, by running an aberration-corrected scanning transmission electron microscope in a confocal geometry in combination with a hologram mask, it might be possible to image the magnetic moments of three-dimensional transition-metal elements on individual atomic columns. ❐

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DNA SEQUENCING

Detecting methylation with force

An atomic force microscope with antibodies attached to its tip can be used to determine methylation patterns in individual DNA strands by making hundreds of force spectroscopy measurements.

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Cytosine is one of the four bases of DNA and cytosine methylation — the addition of a methyl group (CH_3) to this base — is important in many cellular processes[1,2]. There is evidence that human cancers exhibit alterations in DNA methylation patterns[3], which is why researchers are looking for new ways to overcome the technical challenges involved in the accurate detection of such patterns[4]. Writing in *Nature Nanotechnology*, Peter Hinterdorfer of the Johannes Kepler University of Linz and colleagues in Austria and the UK now report a simple and direct approach to detecting methylation patterns in DNA using single-molecule force spectroscopy[5].

Force spectroscopy measures the relationship between molecular length and tension, and has been used to examine the nanomechanics and mechanochemistry of DNA, sugars and proteins[6–8]. This technique has also proved unique in its ability to capture information about dynamic processes such as the rupture of antigen–antibody complexes[9]. To analyse DNA methylation patterns, Hinterdorfer and colleagues linked methylated single-stranded DNA molecules to a glass substrate and attached an antibody that is specific towards 5-methylcytosine (Fig. 1a) to the tip of an atomic force microscope (AFM). The antibody was attached to the tip by an Fc (crystallizable fragment) arm, and had two free Fab (antigen-binding fragment) arms, giving it a Y shape (Fig. 1a). Bringing the microscope tip into contact with the DNA resulted in the formation of two complexes between the two Fab arms of the antibody and two 5-methylcytosines on the DNA. Because single-stranded DNA is very flexible, the two Fab arms, which are spaced a few nanometres apart, can bind two 5-methylcytosines, which are separated by tens of nanometres along the DNA contour.

By stretching the DNA–antibody complex in the AFM, the researchers captured two characteristic force peaks in single-molecule force–extension curves, which they associated with consecutive rupture events between the antibody arms and two methylcytosines (Fig. 1a). Because contacts between both Fab arms and various methylcytosines form randomly, performing these measurements repeatedly allowed the Fab arms to contact all possible pairs of methylated bases. The methylation patterns could then be derived by analysing the separations between force peaks.

The first DNA oligomer examined by the team contained five equally spaced methylcytosines. There were three unmodified bases between neighbouring methylcytosines, giving the nearest neighbour spacing equivalent to the length of four nucleotides. That means that there are 4 possible pairs in which the methylcytosines are separated by 4 nucleotide lengths, 3 pairs separated by 8 nucleotide lengths, 2 pairs that are separated by 12 nucleotide lengths and 1 pair separated by 16 nucleotide lengths (inset to Fig. 1b). Hundreds of force spectroscopy measurements on this DNA sample yielded a relatively complex distribution of the spacing between two rupture events (Fig. 1b). Notably, when the distribution was fitted with numerous Gaussian functions, four maxima at 4, 8, 12, and 16 nucleotides were identified. Moreover, as expected, the relative amplitudes of these maxima were 4:3:2:1.