New insight into the information carried by electrons

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SUMMARY

Since the development of electron holography, it has been recognised that the transmission electron microscope is not only an extremely powerful microscope, but also a versatile electron-optical bench. New developments in phase retrieval and phase tailoring have opened the way to further access to degrees of freedom of free electrons, like orbital angular momentum. Recent advances in orbital angular momentum sorting have added a new observation domain to the usual space and momentum, opening perspectives to a new kind of microscopy.

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Dating back to early 1900s, the need to overcome the resolution limit of light microscopes pushed the research towards finding other kinds of probes capable of unveiling the finer details of micrometric structures. The wavelength of light was one of the limiting factors for resolution. It was at that time that, inspired by the electron diffraction experiments of 1927, Knoll and Ruska in 1932 proposed and developed an instrument, the electron microscope (Knoll and Ruska, 1932), capable of overcoming this limit. Despite the inconvenience of working in vacuum, using high voltage and having limited access to the sample and to the electron-optical elements, the electron microscope established itself as a promising alternative to the light microscope. This has been a huge revolution: being the wavelength of high-energy electrons approximately four orders of magnitude smaller than that of visible light, the possibilities were manifold. Ruska was later awarded the Nobel Prize in Physics for this discovery.

However, the wavelength of the electron was not the limiting factor for the electron microscope resolution. There were other major factors such as thermal, mechanical, and electromagnetic instabilities, and aberrations. It was then in 1948 when the Nobel prize awardee Dennis Gabor developed electron holography (Gabor, 1948) to improve the resolution of the electron microscope. He proposed this new microscopic principle to circumvent spherical aberration correction, which was too challenging at that time and found successful application only in late 1990s (Haider *et al.*, 1998). However, holography didn't have the highest impact concerning resolution. This discovery led also to something different: the unlocking of the phase information of the electron beam thanks to its interferometric character and to Fourier optics.

Holography and phase manipulation found successful application both in light and electron optics. Optical benches offer ease of access to the various optical elements, and phase-varying optical elements, the spatial light modulators (Sampsell, 1990), allow for a precise, local, programmable wavefront phase shifting. With electrons, technological requirements are more demanding and these operations are somewhat more difficult. However, the highly coherent electron source and the flexibility of the transmission electron microscope (TEM) prove its versatility as an electron-optical bench for experiments involving the phase information of the electron beam.

Phase manipulation techniques origin from the computational version of Gabor's holography. In fact, the resulting pattern of a holographic process can be used to shift the phase of a reference wavefront and to retrieve the original beam of interest encoded in the holographic process. This way, wavefronts of different intensity and phase structures can be generated.

An interesting example is the generation of wavefronts with spiralling phase, which confers them a given orbital angular momentum (OAM) value (Yao and Padgett, 2011). The application of this concept to electrons is particularly valuable as, due to their electrical charge, they are capable of interacting with magnetic fields. Early examples of this kind of electron beams are the works of the groups of Tonomura (Uchida and Tonomura, 2010) Verbeeck (Verbeeck *et al.*, 2010) and McMorran (McMorran *et al.*, 2011) in 2010s. These works unlocked a new degree of freedom of electrons, and paved the way for its applications in different techniques. One example is making electromagnetic chiral dichroism (Schattschneider *et al.*, 2006) conveniently feasible in a TEM (Verbeeck *et al.*, 2010), so to obtain information on the spin and orbital magnetic moment of atoms (Edström *et al.*, 2016).

Once found the way to endow electron beams with OAM, the need for reading this information became relevant. The main issue is that this information is mainly encoded in the phase of the electron wavefront, which is not directly measurable. One way of tackling this challenge is to apply phase retrieval techniques that could reveal the phase structure of the beam (Hue *et al.*, 2010; Lubk *et al.*, 2013), which properly manifests in the diffraction space and then requires dedicated experimental techniques (Venturi *et al.*, 2017).

There are however other more direct ways of measuring the OAM of an electron beam. One example (Guzzinati *et al.*, 2014) uses hard masks that allow for checking if the beam has a particular OAM value. One other example (Saitoh *et al.*, 2013) uses fork-gratings, which have the property of adding or subtracting OAM to the beam, eventually leading to a beam with no OAM then revealing its former OAM value. Conversely, one recent application (Grillo *et al.*, 2017) inspired by an optics work (Berkhout *et al.*, 2010) allows, by using two holographic masks, to directly measure and sort the OAM spectrum of the electron beam. In general, few electron beams have a well-defined OAM value, but rather they are normally characterised by a superposition of OAM states. In particular, if their OAM content is the result of an actual interaction, like one with a magnetic object, they normally have a full OAM spectrum with different intensity coefficient for each OAM value. In principle, this OAM-sorting technique would allow for the measurement of the magnetisation of a sample with very few electrons, as the observation takes place directly in the OAM space. With technological improvements that can enhance the efficiency of this device, it would be possible to obtain magnetic and structural information from very small and beam-sensitive samples ranging from molecular magnets to magnetotactic bacteria.

The recently developed phase manipulation techniques, along with the improved TEM characteristics, offer new ways to encode information in the electron wavefunction and to retrieve information from it. Some applications have been explored and hopefully many more are yet to come, conveying us towards exciting years for transmission electron microscopy, beyond high resolution.

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