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Prospects for out–of–plane magnetic field measurements through interference of electron vortex modes in the TEM

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Abstract. Magnetic field mapping in transmission electron microscopy is commonplace, but all conventional methods provide only a projection of the components of the magnetic induction perpendicular to the electron trajectory. Recent experimental advances with electron vortices have shown that it is possible to map the out of plane magnetic induction in a TEM setup via interferometry with a specifically prepared electron vortex state carrying high orbital angular momentum (OAM). The method relies on the Aharonov-Bohm phase shift that the electron undergoes when going through a longitudinal field. Here we show how the same effect naturally occurs for any electron wave function, which can always be described as a superposition of OAM modes. This leads to a clear connection between the occurrence of high-OAM partial waves and the amount of azimuthal rotation in the far field angular distribution of the beam. We show that out of plane magnetic field measurement can thus be obtained with a much simpler setup consisting of a ring-like aperture with azimuthal spokes. We demonstrate the experimental setup and explore the achievable sensitivity of the magnetic field measurement.

Keywords: rotational DPC, magnetometry, 4DSTEM, TEM, electron vortex beams, Aharanov-Bohm, Zeeman shift

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1. Introduction

Electron beams are used in many characterisation techniques to explore the properties of materials, ranging from spectroscopies (e.g. Auger, energy loss spectroscopy, energy dispersive X-ray spectroscopy, ...) to diffractive methods (e.g. low energy electron diffraction, reflection high energy electron diffraction, ...). Electron optics studies the control, focusing and manipulation of electron beams and has seen tremendous progress since transmission and scanning electron microscopes were invented, with continuous improvements to the lenses and the sources, the development of new methods such as holography and new components such as aberration correctors [1].

Despite the many parallels with light optics, the different properties of electron beams mean they exhibit unique physics, yielding many interesting applications [2, 3, 4, 5, 6]. The charge makes them interact with static electromagnetic fields, and the short wavelength of fast electrons makes it possible to focus them down to sub-Ångström resolution [2, 7]. Thanks to this, electron beams in transmission electron microscopes have been used to quantitatively measure microscopic fields inside materials, i.e. the in-plane components of electrical and magnetic fields, through different techniques (electron holography, Lorentz microscopy differential phase contrast, and, more recently, center-of-mass shift detection). All these methods, regardless of the technical details, rely on the lateral forces that in-plane fields exert on the electrons inducing a deflection that can be measured either directly, or as a phase gradient. Measuring the out-of-plane component of the magnetic field, however, was elusive so far.

In the past we have studied how electron vortex beams, carrying orbital angular momentum (OAM), do interact with a magnetic field parallel to the propagation direction, resulting, depending on the exact configuration, in a variation of the kinetic OAM, or in an energy shift [8, 9]. Both effects however vanish as the electron beam leaves the magnetic field, resulting only in a remaining phase shift. More recently, Grillo *et al.* have demonstrated a holographic scheme to measure this phase shift, allowing to access the out of plane magnetic field component [10].

Here we present an alternative and simplified method based on a superposition of electron vortex beams (EVB) to bring out these phase shifts allowing to measure the out of plane magnetic field. The proposed methodology is first exposed, then tested experimentally both in a uniform magnetic field, and on a magnetic sample. We then discuss its applications, properties and perspectives.

2. Theoretical description

2.1. Electron Vortex Beams in a magnetic field

Before outlining our experimental setup, we will briefly recall a few relevant results on the interaction of electron vortex beams with magnetic fields [8, 11, 10]. Two limiting cases for an EVB interacting, in its propagation, with a longitudinal magnetic field have been derived. The first considers a uniform magnetic field, the second a magnetic flux concentrated in a single flux line passing through the center of the vortex. To study the process for a general wave function it is convenient to use the (complete) basis set of Bessel functions. The basis consists of terms in the form:

$$\psi_\ell(r, \phi, z) = j_{|\ell|}(k_\perp r)\exp[i(\ell\phi + k_z z)] \tag{1}$$

where r,ϕ, z are the cylindrical coordinates, $k_\perp$ is the transverse wave number. It is well known that the individual terms are non-diffracting and are eigenstates of forward momentum and orbital angular momentum operators with eigenvalue $p_z = \hbar k_z$ and $L_z = \ell\hbar$ [8, 12, 13, 14]. As such, they also each carry an associated magnetic moment $\boldsymbol{\mu} = -\frac{e\hbar}{2m_e}\mathbf{L} = -\ell\mu_B\hat{\mathbf{z}}$ where e, m_e and μ_B are the elementary charge, the electron's mass and the Bohr magneton. In the presence of a uniform longitudinal magnetic field $\mathbf{B} = B\hat{\mathbf{z}}$ the Bessel terms each undergo an energy shift due to the Zeeman effect equal to

$$\Delta E_\ell = -\boldsymbol{\mu}\cdot\mathbf{B} = B\ell\mu_B. \tag{2}$$

This energy shift is of the order of $\ell \times 10^{-4}\,eV$ and is extremely small compared to the kinetic energy of the electrons which is, in standard TEMs, of the order of 50 to $300\,keV$. This energy shift therefore has no significant impact on the trajectory of the electron beam. Such a small energy shift is effectively undetectable not only due to its magnitude but also because it disappears when the electron leaves the magnetic field, before it can actually be detected §. The temporary change in energy does however induce a phase shift in the electron wave which remains after leaving the field region. If the magnetic field has an extension d along the beam's propagation direction, the total phase shift accumulated can be expressed as

$$\theta_{Z,\ell} = \frac{B\ell\mu_B}{\hbar}\frac{d}{\beta c} \tag{3}$$

where c is the speed of light in vacuum and βc is the electron velocity.

The case of a single flux line carrying a finite flux (an infinitely thin solenoid) oriented along the beam's propagation direction and containing the flux ϕ, shows significant differences. Supposing the flux line coincides with the central axis of the Bessel beam terms, the circular symmetry is maintained, and the (canonical) orbital angular momentum is conserved. Nevertheless the flux line changes the kinetic OAM $\mathcal{L}_z$ by an amount equal to the magnetic flux parameter $\alpha = \frac{e\Phi}{2\pi\hbar}$ i.e. the amount of flux quanta contained in the flux line, yielding $\langle\mathcal{L}_z\rangle = \hbar(\ell - \alpha)$, changing also the radial amplitude distribution of the term [8]. If the magnetic flux is removed (e.g. if the flux line is a very thin and long solenoid where the current is then reduced to zero, or a very elongated magnetic needle which can be schematised as a flux line with two monopoles at the ends), the original kinetic OAM is restored to the same value as the canonical one, leaving us again with no directly measurable effect of the magnetic field's presence. Due to the change in kinetic OAM however, the beam will have again acquired a propagation dependent phase (this can be heuristically thought of as a difference in

§ Assuming the field is localised in the vicinity of the sample and does not include the detector.

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optical path due to the OAM change) which depends on both the topological charge ℓ and the flux parameter α [10]:

$$\theta_{AB,\ell} \approx \left(\frac{d\,\lambda}{\pi\rho^2 m_e}\right)(\ell+\alpha)^2. \tag{4}$$

were λ is the wavelength of the electron and ρ the radius of the ring of maximum intensity. In a superposition of two opposite OAM modes $\psi \propto \psi_\ell + \psi_{-\ell}$ the phase difference between the two components becomes

$$\Delta\theta_{AB,\ell} \approx \left(\frac{d\,\lambda}{\pi\rho^2 m_e}\right) 4\ell\alpha. \tag{5}$$

which shows a phase difference which is linearly proportional to ℓ.

2.2. Decomposition of wave functions

These results have a few implications for a more generic wave function. Let's suppose for simplicity that the wave function under study can be written as the product of a radial, an azimuthal and a longitudinal part $\psi(r,\phi,z) = A(r)f(\phi)\exp(-ikz)$. While this seems a strong hypothesis, it is sufficient to describe the experiments presented below. The electron wave can then easily be written as a sum of OAM components:

$$\psi_\ell^{Ref}(r,\phi) = \sum_{\ell=-\infty}^{\infty} c_\ell A(r) e^{i\ell\phi} \tag{6}$$

where the coefficients c_ℓ are calculated by integrating

$$c_\ell = \int |A(r)|^2 f(\phi) e^{(-i\ell\phi)} r\,dr\,d\phi. \tag{7}$$

As shown numerically in Figure 1a this allows to reconstruct waves with an arbitrary azimuthal profile as a superposition of OAM eigenstates.

As we have shown above, in both field configurations considered, the accumulated phase shift is proportional to the value of ℓ. When such phase shifts act on all the components of a superposition of OAM modes, this causes a uniform rotation of the whole wave function. In particular if all components are shifted by $\ell\theta_0$ the wave function is rotated by $\phi = -\theta_0$ (see Figure 1b). This carries the important consequence that we don't need to create an electron vortex beam state with a definite ℓ in order to exploit these phase shifts, a simple measurement of a rotation in the far field pattern already carries information on the magnetic fields experienced by the samples, and represents an interesting alternative to the far more elaborate interferometric scheme presented by Grillo *et al.* [10].

The higher sensitivity given by using high-ℓ vortex beams is here guaranteed by the presence of sharp edges in the aperture, which ensures that high-ℓ modes will have a significant weight $|c_\ell|^2$ in the superposition.

In the case of the uniform magnetic field, using Equation 3, we can write:

$$\theta_Z = \frac{B\mu_B}{\hbar}\frac{d}{\beta c} = \omega_L t \tag{8}$$

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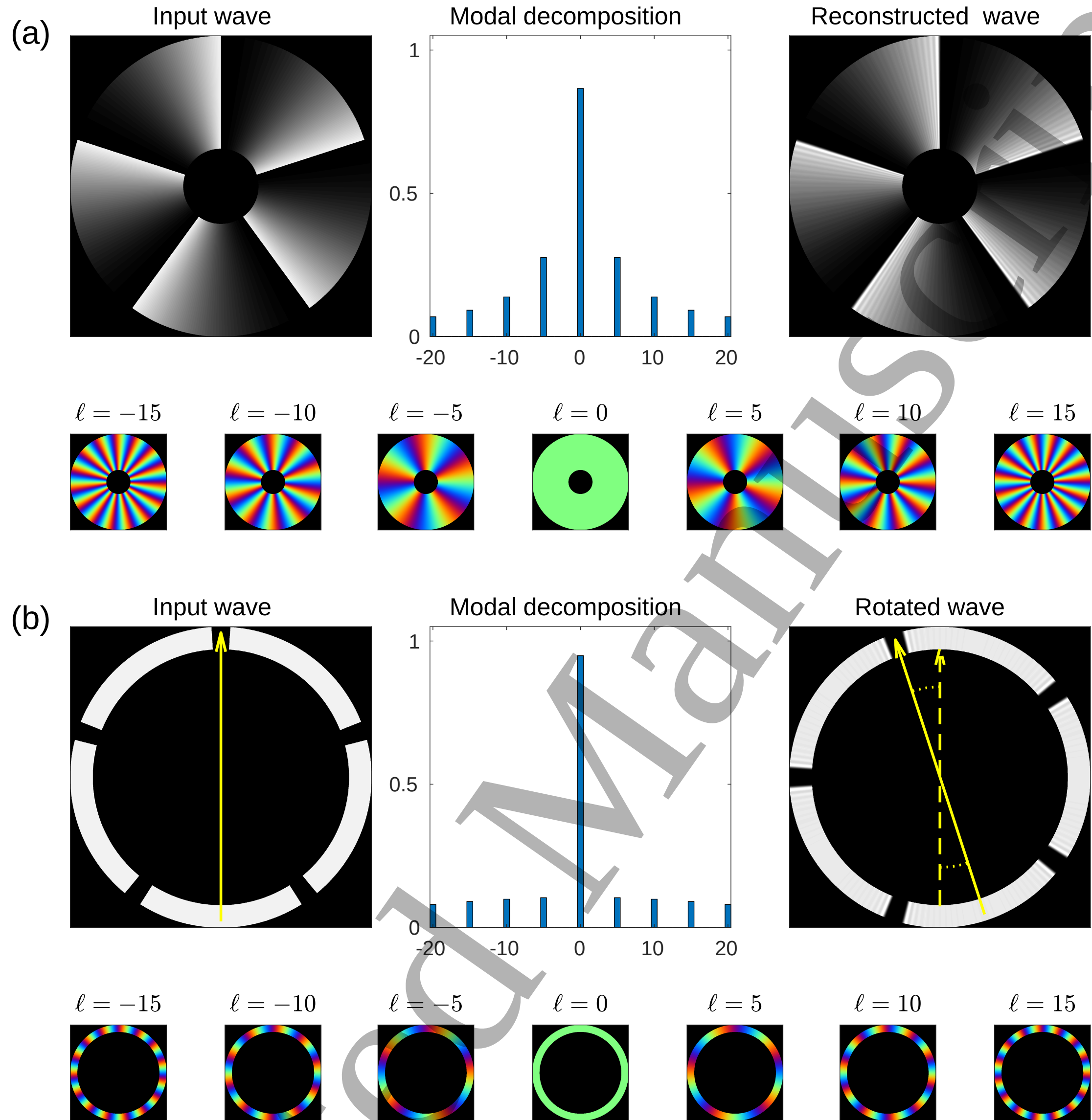

Figure 1. Decomposition of wave functions into orbital angular momentum (OAM) modes. A wave function $A(r)\Phi(\phi)$ can be decomposed into OAM components in the form $A(r)\exp(i\ell\phi)$, obtaining the coefficients c_ℓ. Panel (a) shows an example wave function (left) together with the resulting OAM spectrum $|c_\ell|$ and the reconstructed wave function. The absolute values of the coeffiecients c_ℓ are displayed here for better visibility, and not the real mode weights $|c_\ell|.^2$ which are required to follow $\sum_\ell |c_\ell|.^2 = 1$. The bottom row shows the wave functions for the lowest order relevant modes ($\ell = n \cdot 5$ with $-3 \leq n \leq 3$), with the color intensity encoding the wave amplitude and the colour encoding the phase. The successful reconstruction depends on both moduli and phases of the c_ℓ coefficients. In order to rotate the reconstructed wave by an angle θ_0 as in panel (b), each OAM component must be shifted by a different phase $\ell\theta_0$. As the example wave function chosen here has five-fold azimuthal symmetry, the only coefficients c_ℓ different form zero will be the ones for $\ell = n \cdot 5$ with n integer. In both cases we have displayed the OAM spectrum for $-20 \leq \ell \leq 20$, though we have employed for the reconstruction components covering the range $-150 \leq \ell \leq 150$.

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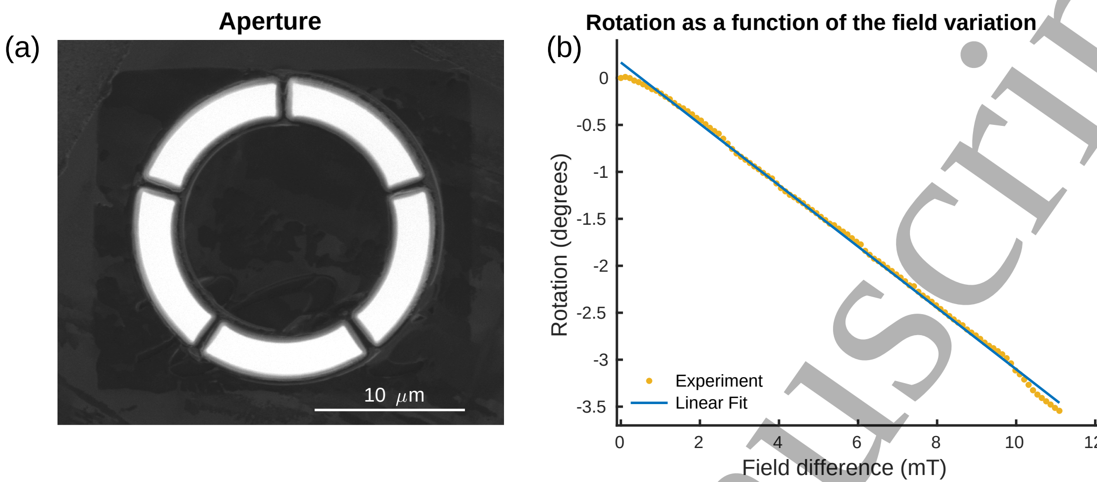

Figure 2. Rotation of an electron vortex beam superposition in a uniform magnetic field. (a) A segmented annular aperture, imaged here with an ion beam microscope, is fabricated out of a platinum film with the ion beam of the microscope. The aperture is then inserted in the illumination system of a transmission electron microscope and its image is projected on a CCD camera, after passing through the lenses of the microscope. The region within the objective lens can be thought of as a large (compared to the electron wavelength) region of uniform magnetic field. (b) In a uniform magnetic field, the different OAM components of the beam undergo an energy shift proportional to the OAM, resulting in a rotation of the whole image. Variations in the current flowing through the objective lens cause the rotation angle to vary, allowing us to measure its dependency on the field.

where $\omega_L = \frac{Be}{2m_e}$ is the well known Larmor frequency and $t = \frac{d}{\beta c}$ is the time spent in the field [15, 16]. That is to say in a uniform magnetic field any electron wave rotates, upon propagation, at exactly the Larmor precession frequency. This interesting phenomenon is counterintuitive, as one might intuitively expect such rotation to have cyclotron frequency $\omega_c = 2\omega_L$. While this fact is well known in electron microscopy the usual derivation is more cumbersome [2], while this approach offer a simpler alternative point of view.

3. Experiments

3.1. Test in Vacuum

In order to test the proposed setup experimentally, we fabricated an annular aperture with five support bar, similar to that shown in Figure 1b, by milling a 1 μm thick gold film with the focused ion beam of a FEI Helios dual beam instrument (Figure 2a). We then inserted the aperture in the illumination system of a FEI Titan^3 transmission electron microscope operating at 300 kV. Through the action of the illumination lenses the Fourier transform of the aperture transmission function is projected onto the sample

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position. For a conventional round aperture, this wave function would take the form of an Airy disc, while for a very thin annulus, it would be an approximate electron Bessel beam [17, 18]. Our current case is intermediate and can be modelled with a integral of Bessel modes beam depending on the width of the annulus. The probe shows significant rippling compared to an Airy disc, owing to the presence of the large non-transparent central part of the aperture similar to a Bessel beam, but only modest extension of the depth of focus.

Modern electron microscopes employ a twin-lens design, where the sample is immersed in the magnetic field of the lens, similarly in concept to what happens in an optical oil-immersion lens. This magnetic field has an extension of several millimeters, a very long extension compared to both the wavelength (2 pm) and the Rayleigh range of the beam. Thanks to this, we can use a simple Zeeman shift model to describe the interaction between the magnetic field and the electron beam. When the microscope is operated in diffraction mode, the projection system performs a second Fourier transform thus conjugating the detection stage of the microscope to the aperture plane, though with a magnetic field dependent rotation. Small changes in the current flowing through the lens, and thus in its magnetic field, cause this rotation to vary slightly. We therefore record images of the aperture while varying slightly the current flowing through the objective lens, then quantify this rotation through image registration methods. By using a I-B (current-field) characteristic curve, obtained by introducing a Hall probe in the sample plane, [19], we can assess the image rotation as a function of the field variation as shown in Figure 2b. The rotation, though small (the average rotation between two successive images is only 0.6 mrad), is retrieved reliably and with very low noise. We can take two approaches in estimating the sensitivity of this technique. The first one is by calculating the proportionality factor between rotation, field and distance travelled inside the magnetic field.

$$k_L = \frac{\omega_L}{\mathbf{B}\beta c} = \frac{e}{2\beta c m_e} \approx 378\,\mathrm{rad T^{-1} m^{-1}} \tag{9}$$

and appears very low for microscopic scale measurement. This implies that a field of 1 T, over the length of 1 μm gives only a rotation of about 0.4 mrad. However, given the quality of the data in Figure 2b this seems to be well within the present detection limits.

3.2. Test on magnetised nanopillars

A second and more realistic test of this approach can be done by attempting to measure the rotation induced by a magnetic sample, which we fabricated for this purpose using focused electron beam induced deposition, such as in Franken *et al*[20]. We fabricated three nanopillars with an approximate diameter of 100 nm and lengths of 0.3, 0.7 and 1.5 microns respectively, and with a separation of 3 μm, as shown in Figure 3a (further images in the Supplementary Material). Special precautions must be taken when observing this type of sample in a TEM since the high magnetic fields in the sample plane is likely to severely disrupt magnetic materials. To this end, we switched

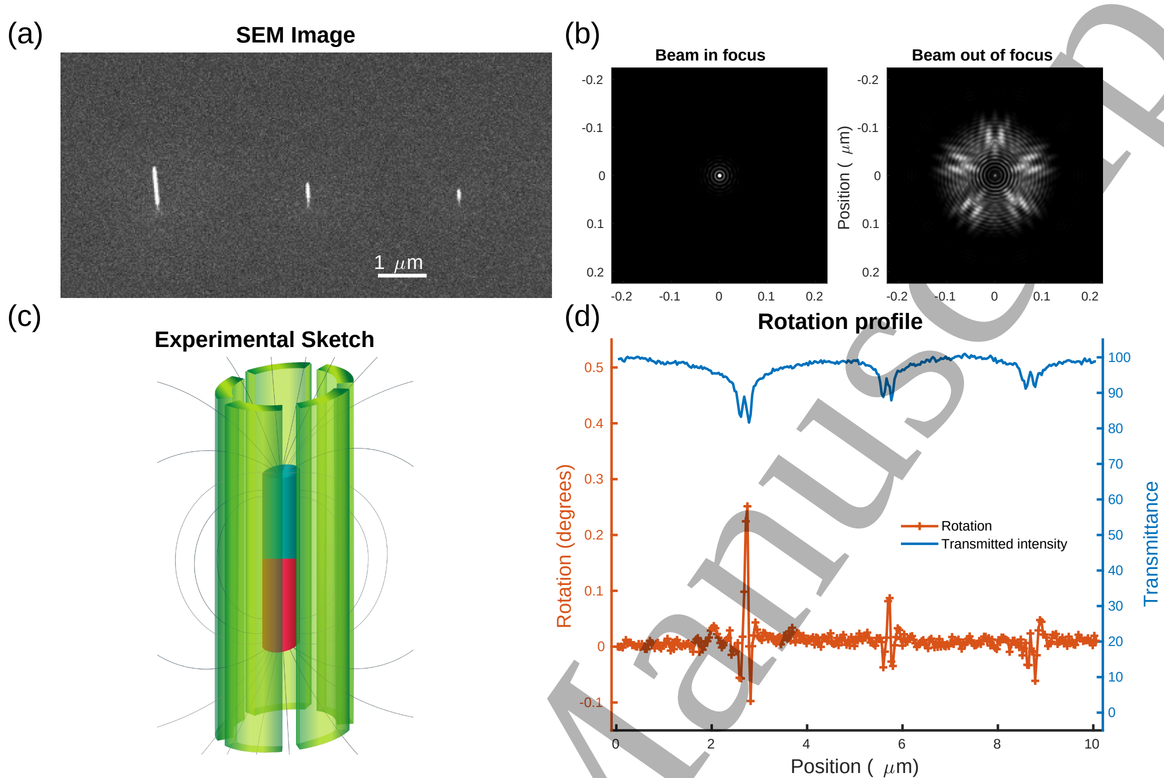

Figure 3. Rotation induced by a localised, out-of-plane, magnetic field. (a) By using electron beam induced deposition we fabricated iron nanopillars of different height. (b) We then projected over the sample the beam generated by the aperture in Figure 2a, slightly unfocused so that the intensity has a ring-like envelope and is wider than the nanopillars. (c) In this configuration, the beam is wider than the pillar and can 'encircle' it. We then scanned the beam collecting an image of the far field intensity (i.e. an image of the aperture) per each position. (d) The total transmitted intensity (in blue) allows to identify the pillars' positions, and by measuring the rotation of the images we can estimate the flux through the nanopillars, and from that the magnetic fields. The approximate value obtained is $\approx 3T$.

off the objective lens entering the 'Lorentz' mode, where the field generated by the lenses in the sample position is minimised. In this mode, since the objective lens is turned off, angles are much lower than usual and the convergence angle of the focused beam is about 140 μrad. We then tuned the illumination system so that the beam would be focused above the sample's position, and the beam would already have a more ring-like intensity distribution on the sample position as in Figure 3b. We then moved the beam in a linear fashion over the three pillars, recording the far-field intensity distribution for 300 beam positions.

By integrating the collected intensity we can compute the so-called bright-field signal (Figure 3b, in blue), corresponding to the total intensity transmitted through the sample, which appears to have three ring-like dips corresponding to the pillar's positions. The ring-like shape is expected, as it is an obvious consequence of the intensity

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distribution of the impinging defocused beam. Through the same image registration procedure used before, we can measure the rotation of the far-field intensity (Figure 3b, in red). Clear peaks can be observed in correspondence to the pillars, and their intensity appears to scale in the expected manner with the height of the pillars. When the beam is centered on the nanostructure we have circular symmetry and we can use the description outlined above to describe the Aharonov-Bohm phase shift induced in the various pairs of OAM components by the magnetic flux in the pillar, and therefore link the image rotation to the field. The estimated field is comprised between 2 and 4 Tesla, which appears high for FEBID deposited iron (bulk iron has a saturation field of $\sim 2.2\,T$, and the expected Fe content of the wire is 80% or lower), but is of the correct order of magnitude [21].

4. Conclusion

We have recalled the rotational beviour of electron vortex beam superpositions in the presence of uniform or localised longitudinal magnetic field, and we have demonstrated experimentally how this behaviour can be used as a probe for said magnetic fields both in a uniform magnetic fields and in the presence of magnetised nanopillars. This can be done by installing a special modified aperture in the illumination system of a commercial transmission electron microscope, projecting its Fourier transform on a magnetised sample, then measuring the apparent rotation of this aperture in the far field of the sample. This constitutes one of the first demostrations of a measurement of out-of-plane magnetic fields in a TEM.

The technique appears to have a sensitivity comparable to the previous experiments [10], and therefore constitutes an interesting alternative. The method we present here appears significantly easier to implement than the previous holographic approach, since there is no need for an undisturbed reference that passes outside the magnetic field region, which makes it much easier to combine with conventional methods for the measurement of in-plane fields (such as DPC/COM, as discussed below), and to straightforwardly scan the beam over the sample to repeat the measurement in different positions, and on different nanostructures. On the the other hand more work will be necessary to make the quantification more accurate and on par with the holographic approach, as the beam has a more complex shape at the sample position. Aside from this the techniques share many of the same strong points and weaknesses. Phase shifts induced by non-magnetic interactions (e.g. out-of-plane electrostatic fields) are independent of ℓ, and hence do not contribute to a rotation. The low magnitude of the phase shifts present in both techniques means however that it might be very hard to measure if the beam has to pass through a thick sample, as the strong scattering would significantly affect the diffraction pattern. The sensitivity of the holographic method could be increased by increasing the value of ℓ, while that of our proposal could be increased by optimising the aperture design, adding more thin spokes that would make the detection of rotations easier (and hence maximising the weights $|c_\ell|^2$ of

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the high$-\ell$ modes in the beam), but the weak nature of the effect, along with the Bragg scattering that would severely impact the diffraction pattern, are significant hurdles for the achievement of sub-nanometer resolutions. Coherence does not seem to be a limiting factor in modern instrumentation, and conceivably the main effect of limited lateral coherence would be a blurring of the beam in the sample position, with consequent averaging over the corresponding area. In the current regime of high energy and very small rotations, the method proposed here appears analogous to a technique currently employed in electron microscopy to measure the transverse component of static electromagnetic fields known as, depending on the variant, Center of Mass (COM) or Differential Phase Contrast (DPC). This approach relies on measuring the far field deflection of the beam due the transverse field, which allows to compute the fields through Ehrenfest's theorem. The method proposed here comprises a rotational extension of this technique, giving access to the longitudinal component, a form of *rotational DPC*. These methods could be combined obtaining a three-dimensional DPC, giving access to all three components of the magnetic field.

Methods

Aperture production

We deposited a $1\,\mu m$ thick Pt layer on top of a commercial SiN film commonly used in TEM as a sample support. The aperture was milled into this metallic layer using the focused gallium ion beam of a FEI Helios dual beam instrument. This aperture was then inserted into the illumination system of a FEI Titan^3 transmission electron microscope.

Nanopillars production

Utilising an FEI Helios G3 equipped with an $Fe_2(CO)_9$ gas injection system, iron nanopillars of the desired height were deposited on top of an electron-transparent 20 nm thick amorphous silicon nitride support film by allowing the beam to dwell on a fixed location for a controlled amount of time such as in Franken *et al*[20].

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Author Contributions

G.G. and J.V. conceived the experiment. A.B. manufactured the TEM aperture and calibrated the objective lens magnetic field. D.M. manufactured the iron nanopillars. G.G., A.B., J.V. designed the experimental set-up, and G.G. performed the TEM experiments, and analysed the data. All Authors contributed to writing the paper.

Data Availability

The detected far field patterns are available on Zenodo at the address https://doi.org/10.5281/zenodo.3232898

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