

Topological shaping of vortex neutron beams using forked phase gratings

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Beams of light or matter that carry well-defined states of orbital angular momentum (OAM) are promising probes of topological and textured condensed matter systems such as magnetic skyrmions. Using spin-echo small-angle neutron scattering, a phase-sensitive technique that allows for a relatively low intensity beam, we demonstrate the production of vortex neutron beams from forked phase gratings of various topological charges. This type of interferometric verification of OAM we present here could be further extended to other platforms using probes that are similarly limited by the coherence and strength of the currently available sources.

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I. INTRODUCTION

Orbital angular momentum (OAM) beams of light or matter display unique properties that make them promising candidates as probes of condensed matter [1–3], such as *superkicks*, enhanced scattering from processes that are kinematically forbidden for beams without OAM [4–8], and preferential absorption and scattering that depend on the probe's OAM state and the target's chirality [9–13]. An OAM state is defined by its *phase singularity* $e^{i\ell\phi}$, where $\ell \in \mathbb{Z}$ is the OAM quantum number and ϕ the azimuthal angle about the direction of travel. For most probes, the OAM state is usually verified by directly imaging the amplitude node associated with this phase singularity. In this article, we present an alternative route to OAM detection that is based on an interferometric measurement of the beam's phase, which is more suitable for low-coherence, weak-intensity beams. Forked diffraction gratings (FDGs) are often utilized as a robust method of OAM generation and measurement for both light [14–17] and matter waves [18–23]. The characteristic profile of an FDG is described by $\cos \alpha$ with $\alpha = (2\pi/p)x - m\phi$, where p is the period of the grating, $m \in \mathbb{Z}$ the grating's *topological charge*, and $\phi = \arctan(y/x)$. The topological charge corresponds to the difference of the number of grooves above and below the topological defect at the grating's center as shown in Fig. 1(a).

This function is the interference pattern of a plane wave and a charge m OAM state, and so an FDG acts as a hologram, converting an incoming neutron without OAM into a conjugate pair of diffracted $\ell = \pm m$ OAM states [24–26].

Although the generation of OAM neutron beams has been reported using various techniques [27–34], it is generally difficult to unambiguously verify the production of neutron OAM [35–37], primarily due to the weak interactions of thermal and cold neutrons ($E \lesssim 25$ meV) with matter. Therefore, we used spin-echo small-angle neutron scattering (SESANS), which is a phase-sensitive technique resembling a microscopic Mach-Zehnder-like interferometer [38] as shown in Fig. 2 to detect the phase distortions produced by the FDG rather than the amplitude distortions. This method convincingly demonstrates the production of neutron OAM for several values of m while simultaneously presenting an alternative method for quantifying the OAM of other intensity-limited probes like gamma rays [39] or Helium-3 [40] for which one cannot easily perform direct imaging or coincidence counting due to experimental constraints such as detector sensitivity or source incoherence.

II. SAMPLE DESIGN AND EXPERIMENT

Our silicon FDGs were etched using electron-beam lithography at the Center for Nanophase Materials Sciences at Oak Ridge National Laboratory [41]. As our FDG grooves have a rectangular rather than sinusoidal profile, they also produce higher-order diffraction peaks that are weaker with larger OAM values. For a 50% duty cycle grating, the rectangular profile can be described by the indicator function

$$\chi(x, y) = \frac{1}{2}[1 + \text{sign}(\cos \alpha)]. \quad (1)$$

As the production of neutron OAM requires the FDG to be coherently illuminated, the size of the FDG must be on the order

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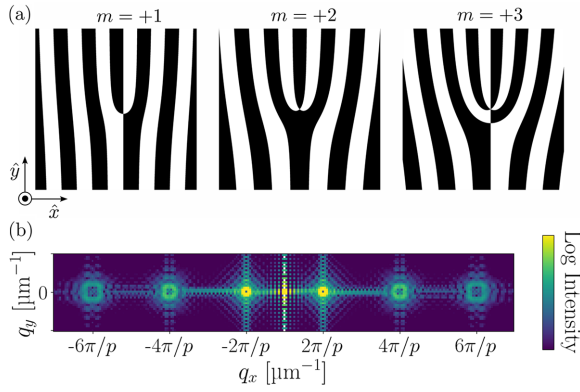


FIG. 1. (a) Examples of binary forked phase grating profiles with topological charges $m = 1, 2, 3$. (b) Plane wave simulation of the diffraction pattern produced from an $m = 1$, period $p = 2 \mu\text{m}$ forked phase grating vs transverse momentum transfer $\mathbf{q} = (q_x, q_y)$. Notice that at the usual Bragg peak locations at $\mathbf{q} = (\pm 2\pi n/p, 0)$ for $n \neq 0$ we instead observe annular “Bragg donuts,” one of the characteristic indications of an OAM state.

of the transverse *intrinsic* coherence width Δ_t of the neutron, which we define to be the average transverse size of each of the mutually incoherent neutron wave packets that form the total beam [42]. Although measurements of Δ_t are experimentally challenging, most report $\Delta_t \gtrsim 10 \mu\text{m}$. [43–47]. To enhance the signal to an observable level, the FDG motif must be repeated periodically as individual plaquettes over the area of the beam. Each of our plaquettes has a period $p = 2 \mu\text{m}$, groove depth $d \approx 4\text{--}6 \mu\text{m}$, and total size of $10 \times 10 \mu\text{m}^2$. The total etched area of the wafer is $1 \times 1 \text{ cm}^2$. As the region near the singularity is especially fragile, an additional disk was etched away near each plaquette’s center; although we do not show this additional feature in Fig. 1(a), it is included in the simulations presented below.

The phase accumulated by the neutron due to the nuclear interaction as it passes through our quasi-two-dimensional

(quasi-2D) silicon FDG is $\Phi(x, y) = -\rho_{\text{Si}}\lambda d\chi(x, y)$, where $\rho_{\text{Si}} \approx 2.07 \times 10^{-4} \text{ nm}^{-2}$ is the scattering length density of silicon and λ the neutron wavelength [41]. Assuming plane wave illumination e^{ikz} with $k = 2\pi/\lambda$, the neutron wavefunction immediately after the FDG is given by $e^{ikz}e^{i\Phi(x,y)}$, with

$$e^{i\Phi(x,y)} = f_0 + \frac{1}{2}(e^{-i\rho_{\text{Si}}\lambda d} - 1) \sum_{n=1}^{\infty} f_{nm}(\alpha), \quad (2)$$

$$f_{nm}(\alpha) = \text{sinc}(n\pi/2)[e^{in\alpha} + e^{-in\alpha}],$$

where $f_0 = (e^{-i\rho_{\text{Si}}\lambda d} + 1)/2$ is the transmission amplitude of the unscattered beam and $\text{sinc}(x) = \sin(x)/x$. From Eq. (2), we see that the angular deviations of the diffraction orders relative to the transmitted beam are $\theta \approx \pm n\lambda/p$, and each order carries an $\ell = \mp nm$ OAM state. Due to the sinc weighting of the diffraction orders, only the odd orders of n contribute, and the $n = 1$ conjugate orders dominate.

We present the full plane wave limit calculation for the diffracted neutron state in the Supplemental Material [41]. We find the far-field analytic form of the Bragg donut intensities shown in Fig. 1(b) to be

$$|f_{nm}|^2 = C_{nm} \left| (Rq')^{|nm|} {}_1F_2\left(\frac{a}{b}; \frac{-R^2 q'^2}{4}\right) \right|^2, \quad (3)$$

$$C_{nm} = \left| \frac{\pi R^2}{\lambda} \frac{(e^{-i\rho_{\text{Si}}\lambda d} - 1) \text{sinc}(n\pi/2)}{2^{|nm|+1}(2 + |nm|)(|nm|)!} e^{\mp inm\phi} \right|^2,$$

where ${}_1F_2$ is a generalized hypergeometric function with $a = (1 + |nm|/2)$ and $b = (2 + |nm|/2, 1 + |nm|)$ [48], $R > 0$ a regulator used to ensure convergence of the scattering amplitude integral, and q' the radial coordinate in reciprocal space relative to the center of the Bragg donut. The upshot of this result is that each Bragg donut for definite $|nm|$ is a linear combination of Bessel and the related Struve functions and *not* pure Laguerre-Gauss modes [49] as has previously been asserted [30]. Importantly, our calculation shows that the

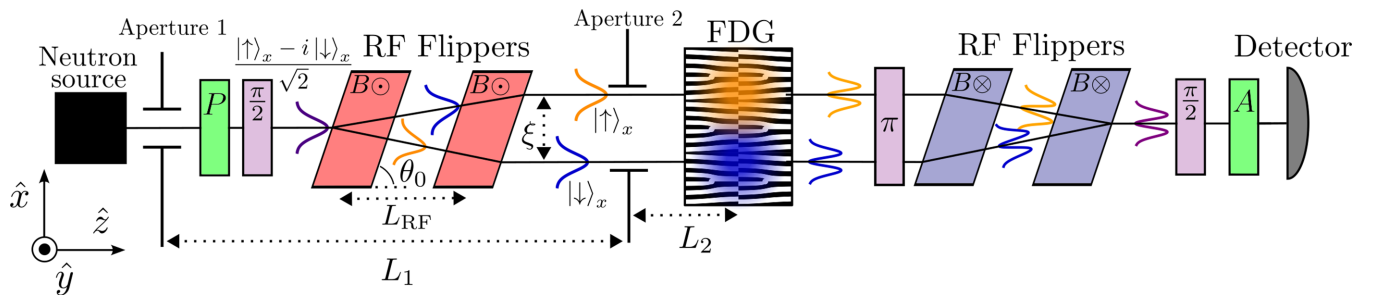


FIG. 2. Experimental setup of the SESANS measurement. The incident polarized neutron state (purple) is sheared in the $\hat{x}$ direction by the first pair of rf flippers into two spin-polarized outgoing spin-path mode-entangled states (orange and blue). Each state interacts with the FDG that imparts OAM onto that state. The relative OAM phase between the two states is measured in the usual Mach-Zehnder-type way by spatially interfering the two OAM states using a second pair of rf flippers [38]. Collimation was performed by two square apertures placed before the polarizer (Aperture 1) of size $14 \times 14 \text{ mm}^2$ and before the FDG (Aperture 2) of size $6 \times 6 \text{ mm}^2$. The distances between the two apertures and between the FDG and Aperture 2 are $L_1 = 4.82 \text{ m}$ and $L_2 = 0.09 \text{ m}$, respectively. The green components label “P” and “A” are the neutron spin polarizer and analyzer, respectively. The two $\pi/2$ -flippers start and stop the neutron precession, while the central π -flipper corrects for the magnetic inhomogeneities in the rf flippers. Additional weak guide fields are not shown. The wave packet size and separation relative to the various components are exaggerated for clarity. The FDG is used in transmission-mode for these experiments, but we have rotated it into the plane of the page for visual clarity. Additionally, the relative scale between the wave packet size, beam size, entanglement length, and plaquette size and period are adjusted, again for clarity.

radius of the Bragg donut (defined as the maximum intensity) scales *linearly* with the topological charge m for $m \geq 1$ [50], and is *independent* of Δ_t since our plane wave calculation assumes the $\Delta_t \rightarrow \infty$ limit. Similar results were found for the analogous optical near- and far-field diffraction from FDGs [51,52].

In principle, these Bragg donuts can be observed using the technique of small-angle neutron scattering (SANS) as was previously attempted [30,31], although those data were difficult to interpret due to both a large wavelength bandwidth and shallow grating grooves. Instead, we performed a SESANS measurement of our FDGs [53]. In our setup shown in Fig. 2, each neutron is initially polarized in the $\hat{y}$ direction and is then coherently split into two paths by the first radio-frequency (rf) neutron spin flipper that acts like a polarizing beam splitter [54]. These independent trajectories are made parallel by a second rf flipper. Each neutron is therefore spin-path mode-entangled [55,56] (i.e., intraparticle entangled in the spin and position degrees of freedom [57–59]). The *entanglement length* ξ is the experimentally tunable, real-space separation of the two spin states and thus the length scale probed in the sample [60]. For our setup,

$$\xi = \frac{m_n f_{\text{RF}} L_{\text{RF}} \cot \theta_0}{\pi \hbar} \lambda^2 = \xi_0 \lambda^2, \quad (4)$$

where m_n is the neutron mass, $f_{\text{RF}} = 2$ MHz the rf flipper frequency, $L_{\text{RF}} = 1.2$ m the distance between each pair of rf flippers, $\theta_0 = 40^\circ$ the rf flipper inclination angle, and $\hbar$ the reduced Planck's constant. With these parameters, the *entanglement constant* was fixed to $\xi_0 \approx 1.37 \times 10^4 \text{ nm}^{-1}$ for all measurements reported here. The neutron wavelength band used was 0.3–1.05 nm, which translates to an entanglement length range of 1.2–15 μm . After interacting with the sample, the final two rf flippers spatially recombine the neutron spin states for subsequent polarization analysis and detection. The observed SESANS polarization is hence a measure of the relative OAM phase imparted by the FDG on the two entangled neutron spin states separated by the *entanglement vector* $\xi = (\xi_x, \xi_y)$. Additionally, SESANS also has the advantage that the polarization is independent of the intrinsic neutron coherence width unlike the SANS intensity [60].

Using the phase-object approximation (POA) for the scattering amplitude, we can efficiently simulate the expected SESANS polarization signal from our FDG. Under general assumptions discussed in the Supplemental Material [41], the empty-beam-normalized polarization P/P_0 is given by the cosine Fourier transform of the total Bragg donut diffraction pattern, projected onto the *encoding direction* given by $\hat{\xi} = \xi/\xi$ [60]. In the POA, the normalized polarization can be further simplified to

$$\frac{P(\xi)}{P_0} = \frac{1}{\sigma} \text{Re} \left[\int_{\mathbb{R}^2} dx dy e^{i\Phi(x+\xi_x, y+\xi_y)} e^{-i\Phi(x, y)} \right], \quad (5)$$

where σ is the total scattering cross section, and $\text{Re}[\cdot]$ denotes the real part. The simulations shown in Fig. 3 demonstrate that the polarization strongly depends on the phase imparted by the FDG, which is determined by the grating's topological charge. Therefore, we can verify the production of OAM by comparing the measured SESANS polarization to expected polarization from a POA simulation, and in the case of good

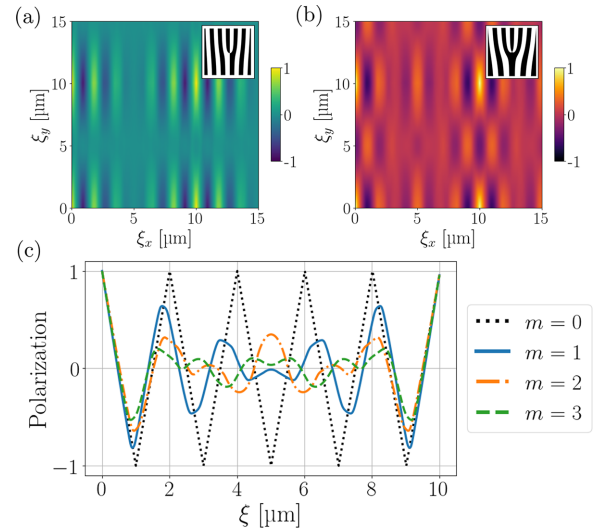


FIG. 3. Simulated SESANS polarization for a plaquette with charge (a) $m = 1$ and (b) $m = 2$, with the insets being the original FDG profile. For visual clarity, we used a constant $\lambda = 0.4$ nm incident neutron spectrum and a $10 \times 10 \mu\text{m}^2$ plaquette size with $d = 38 \mu\text{m}$ (such that $\Phi = -\pi\chi$) and $p = 2 \mu\text{m}$. Notice that the polarization is periodic in both ξ_x and ξ_y with plaquette size. These plots demonstrate that SESANS can serve as a direct probe of the sample's topological structure as the polarization signal for each m is markedly distinct. (c) Slices through the origin along $\xi_y = 0$ of the simulated SESANS polarizations for plaquettes of various charges. These particular $\xi = \xi_x$ slices of the 2D polarizations correspond to the case where the grooves in the $\hat{y}$ direction are perpendicular to the encoding direction $\hat{\xi}$. An arbitrary slice through (ξ_x, ξ_y) can be chosen by changing the angle between the sample and encoding direction.

agreement we can establish that the FDG did indeed produce the Bragg donut diffraction pattern given in Eq. (3) and displayed in Fig. 1(b). Finally, the analytical solution and POA simulations both show that each Bragg donut carries the appropriate phase singularity with $\ell = \mp nm$.

III. RESULTS AND DISCUSSION

We performed the experiment on the Larmor instrument at the ISIS Neutron and Muon Source located in the UK; see Fig. 2 for a sketch of the experimental setup. We measured the FDGs in two orientations, the first with the encoding direction perpendicular to the grooves (the perpendicular orientation) and the second with the encoding direction parallel to the grooves (the parallel orientation). With this convention, the perpendicular orientation corresponds to $\xi = \xi_x$ and the parallel orientation to $\xi = \xi_y$.

As shown in Fig. 4, there is good agreement between the data (solid traces) and the POA simulations (dashed traces). We present measurements of six different gratings individually in the perpendicular orientation, two each of charge 1, 2, and 3. The intricate pattern seen in each of the SESANS polarizations serves as the unique, identifying fingerprint of the particular OAM state. We also show the measured polarization from two charge $m = 1$ gratings in the parallel orientation in Fig. 4(b); the observed polarization in this

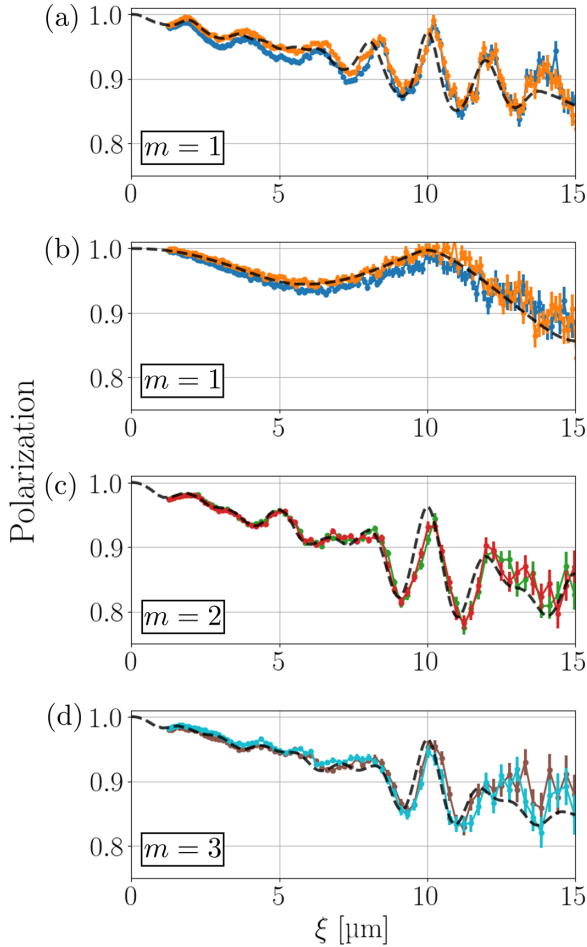


FIG. 4. Measured SESANS polarization data of two charge $m = 1$ gratings in the (a) perpendicular and (b) parallel orientation. Measured polarizations of two charge (c) $m = 2$ and (d) $m = 3$ gratings in the perpendicular orientation. In all parts, the dashed black trace corresponds to a SESANS POA simulation convolved with the instrument resolution function [41]. The two solid traces and data points in each part correspond to the measured polarizations from a particular FDG. The sloping background is an artifact of using a pulsed, time-of-flight neutron source; this can be understood by considering that both the phase ($\Phi \propto \lambda$) and the entanglement length ($\xi \propto \lambda^2$) vary with the polychromatic neutron pulse in which the wavelength is determined by the neutron's flight time [41]. Note that the distinct structures for each m seen in Fig. 3(c) are still present here but are partially obscured by this background.

orientation is relatively featureless and similar for all charges, only displaying a single broad peak at $10 \mu\text{m}$ corresponding to the interplaquette spacing. For each of the POA simulations shown in Figs. 4 and 5, the only free parameter was the depth of the grooves, which was varied between 3 and $6 \mu\text{m}$ in order to match the experimentally observed polarization contrast. Specifically, the charge 1, 2, and 3 data were best fit using a depth of 5.5 , 3.9 , and $4.2 \mu\text{m}$, respectively. These SESANS-determined depths are consistent with the depths found from SEM images presented in the Supplemental Material [41].

To enhance the polarization contrast, we also performed measurements with FDGs stacked back-to-back, with gratings

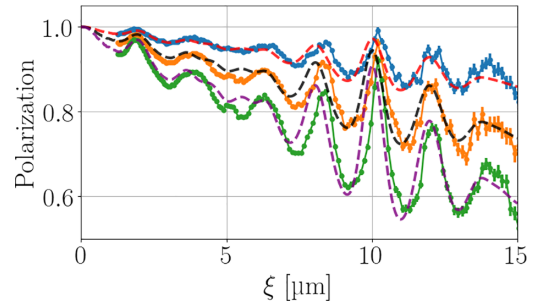


FIG. 5. Comparison of the measured SESANS polarization data of a single grating (blue), and two (orange) and three (green) stacked charge $m = 1$ gratings. The red dashed trace is the simulation result for a single charge $m = 1$ grating of depth $d = 5.5 \mu\text{m}$ reproduced from Fig. 4(a), and the black and purple dashed traces the result of a simulation with a single FDG of depths $d\sqrt{2}$ and $d\sqrt{3.5}$, respectively. For the three-grating simulation, we attribute the deviation from the expected apparent depth of $d\sqrt{3}$ to the fact that the third grating was produced by a slightly different process that most likely resulted in slightly deeper grooves [41].

separated from one another by $\sim 1 \text{ mm}$. As shown in Fig. 5, the observed polarization from the stacked gratings is the product of the single-grating data. Therefore, stacking gratings of the same charge and groove depth d results in a similar polarization to that which would be found from a single grating with an increased groove depth of $d\sqrt{n_g}$, where n_g is the number of gratings. Importantly, the individual plaquettes were not required to be placed in registry to observe this enhancement. Because etching FDGs with a depth-period aspect ratio greater than $\sim 5:1$ is challenging, stacking multiple gratings can provide a simpler alternative means to enhance the contrast.

IV. CONCLUSION

We have demonstrated the production of neutron OAM beams with forked phase gratings using the phase-sensitive, interferometric technique of SESANS. While the SESANS signature of OAM is less visually striking than the donut-shaped Bragg peaks anticipated in conventional SANS, the SESANS data are of substantially higher quality. Our results demonstrate that the scattered wavefunction is *not* a pure Laguerre–Gauss mode. Moreover, they indicate that the radius of the Bragg donuts if observable with SANS would scale *linearly* with the topological charge m , rather than with $\sqrt{m}$, as previously inferred from lower-quality SANS measurements [30,31]. We conjecture that such phase-sensitive methods may be more generally applicable to detecting the OAM of weakly scattered radiation such as neutral atoms and molecules or gamma rays. Furthermore, we have demonstrated that the signal in our experiments can be enhanced by stacking together gratings of the same charge, and so small-period forked gratings could be used to both spatially separate and enhance the intensity of neutron beams with well-defined values of OAM. We have also shown that SESANS is extremely sensitive to real-space structures, being able to discriminate between samples with similar but ultimately distinct features, and so could be used to probe topological defects in materials.

We suspect that OAM neutron beams could be effective probes of both the structure [61] and dynamics [62] of topological systems such as skyrmions [63] and other magnetically textured materials [64] that have spintronic applications [65–67]. Additionally, spin-orbit neutron beams that are entangled in their spin and OAM degrees of freedom are another promising type of OAM beam for the measurement of topological materials [68]. On the other hand, grating interferometry with FDGs has been proposed in order to both enhance the imaging resolution and serve as a direct probe of chiral structures [69,70].

Finally, we acknowledge that the exact nature of the OAM content of our neutron beam remains an open question, namely, whether this OAM is truly a single neutron or only a beam property. Elucidation of this point requires both higher-precision measurements of the intrinsic coherence width of the neutron and a more sophisticated theoretical analysis that *ab initio* incorporates the neutron's intrinsic coherence width.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [71,72].

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