

Single-Grating Interferometry in Scanning Transmission Electron Microscopy

by

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DISSERTATION ABSTRACT

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Title: Single-Grating Interferometry in Scanning Transmission Electron Microscopy

Electron microscopy is a critical tool for understanding the natural world atom-by-atom. Scanning Transmission Electron Microscopy (STEM) rasters a focused electron probe across the sample and detects the amount of scattered current, quickly generating an interpretable image of the sample amplitude. Light elements and weakly-scattering samples, however, appear invisible to STEM. For these samples, which include many biological and 2D materials, we need phase contrast imaging. Ptychography, the main such technique in electron microscopy today, struggles to recover low-spatial-frequency information like sample thickness, especially at low dose conditions, and only recovers a relative phase measurement. Here, we continue development of an alternative phase contrast technique called STEM holography (STEMH), which uses a single pre-specimen diffraction grating as a beamsplitter to form an N-beam electron interferometer. STEMH can recover absolute and linear phase contrast, especially at the low spatial frequencies existing techniques fail at. However, its multiple-beam illumination of the sample currently prevents realizing quantitative atomic resolution interferometry in the TEM. We investigate how to improve our diffractive beamsplitter by studying one-dimensional phase grating theory, and show that current technology cannot achieve ideal two-beam performance. We perform STEMH on nanomagnetic samples and demonstrate its flaws in sub-nanometer resolution images of silver nanocubes, gold nanoparticles, and a few-atomic-layer sheet of GaN. Finally, we integrate

ptychography within STEMH, showing initial simulations of an algorithmic solution to the multiple-beams problem.

This dissertation includes previously published and unpublished co-authored material.

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CHAPTER I

INTRODUCTION

As an imaging technique, the ultimate goal of electron microscopy is to quantitatively measure the amplitude and phase of nanoscale samples at the highest possible resolution. After decades of technological development, commercially available transmission electron microscopes (TEMs) are now regularly capable of capturing the sample amplitude. This in itself is a marvel. The TEM consists of a gun accelerating free electrons to relativistic speeds of $0.3 - 0.8c$ and a series of electromagnetic lenses with individually tunable focal lengths all within a high vacuum system that ensures electrons can proceed from the gun and through the sample. Hans Busch first conceived how to use solenoids as electromagnetic lenses in 1926 [1], with Ernst Ruska and Max Knoll first developing a transmission electron microscope in the 1930s and achieving about 7 micron resolution by 1938 [2, 3]. In 1936, Max Scherzer demonstrated TEM electromagnetic lenses inherently suffer serious spherical and chromatic aberrations that limit resolution to roughly 2 Å, just above the typical distance between atoms in a lattice [4]. It took until the 1990s to implement aberration correctors that eliminated the inherent spherical aberrations and compensated for all non-chromatic aberrations up to fourth order [5]. This unlocked 0.5 Å resolution, also called sub-angstrom or atomic resolution in the 2000s [6, 7].

Consequently, generating amplitude contrast at atomic resolution in transmission electron microscopy is largely a solved problem. Phase contrast is not. We need phase contrast to detect samples too weak to meaningfully perturb the trajectory of an incident electron, namely biological samples and magnetic and electric potentials. Interferometry is an easy answer to supply, but

difficult to implement in TEMs due to their one-way experimental geometry and limited beamsplitter technology. Historically, the leading phase contrast TEM measurement technique was electron holography, which uses a positively-charged wire post-specimen to curl the incident electron plane wave around itself and interfere the part of the electron wavefront that passed through the sample with the part that passed through vacuum. First demonstrated by Mollenstedt and Duker in 1955 [8], electron holography was a specialized but important technique for pushing advancements in TEM resolution following a proposal by Gabor [9] for numerical aberration correction [10, 11, 12, 13, 14]. However, in the last decade, holography has fallen far out of fashion. Unlocked by new direct electron detector technology [15], ptychography is now the main phase contrast imaging technique in electron microscopy. Using scattering information from neighboring diffraction measurements of the sample to solve the sample phase [16, 17], electron ptychography has set new resolution records while becoming the new workhorse of atomic-resolution materials science [18, 19, 20].

Despite its success, ptychography struggles to acquire low-spatial-frequency information and does not acquire absolute phase measurements [21, 22, 23], especially at the low electron doses necessary to prevent knock-on damage in the fragile phase-only specimens we need phase contrast imaging for [24]. Non-iterative ptychographic techniques lose all contrast at low spatial frequencies [21, 22] while iterative ptychography methods require significant computational resources to converge in the same regime [23]. In practice, this leads to slight [24] or no [22] phase contrast at zero spatial frequency. Electron (biprism) holography, as an interferometric technique, can capture the absolute phase of the sample at all spatial frequencies [25], but suffers from near-field imaging effects while having

stringent source coherence requirements [26]. Attempts to correct these issues included adding multiple biprisms to the TEM column, which requires much more complicated customization of the TEM [27], or doing beamsplitting prior to the sample, which ran up against the technological constraints of the time [28, 29, 30].

The McMorran group first realized a more Mach-Zehnder-like setup that, instead of illuminating the sample with a plane wave and interfering the wave post-specimen, split the beam into multiple probes focused at the sample plane in 2018 [21]. Called Scanning Transmission Electron Microscopy Holography (STEMH), this technique uses a custom nanofabricated diffraction grating as a beamsplitter instead of an electron biprism. Fig. 1 depicts this experimental geometry. Using a diffraction grating significantly relaxes the source coherence requirements while eliminating the near-field interference effects that plague biprism holography. As an interferometric technique, STEMH is able to acquire the absolute phase contrast at low spatial frequencies that ptychography cannot. Once paired with advanced direct electron detectors, our group was able to demonstrate sub-nanometer resolution phase contrast imaging with STEMH [31].

This dissertation attempts to address the two flaws with STEMH that prevent its adoption as a complementary phase contrast imaging technique to ptychography and TEM holography. Both issues follow from our beamsplitter. For ideal interferometry, the STEMH beamsplitter would produce exactly two beams: a probe and a reference, but our diffraction grating beamsplitters produce between five and nine non-negligible diffraction orders. The first hurdle is ghost images. Fig. 2 shows a STEMH measurement of a silver nanocube with an edge length of 100 nm. Ideally, STEMH would record the image of a projected square. Instead, each beam projects a copy of the sample, so the final image contains multiple copies of

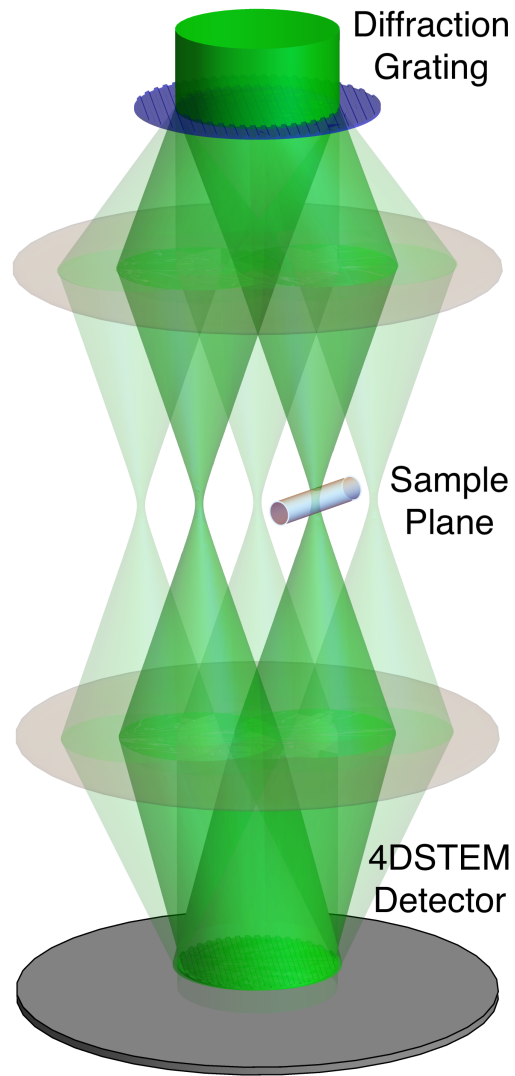


Figure 1. A schematic of the scanning transmission electron microscopy holography (STEMH) experimental geometry. The electron beam is incident on a nanofabricated diffraction grating, which splits the beams into multiple probes which are focused at the sample plane. The interference of these various paths is imaged by a 4DSTEM direct electron detector.

the sample stacked on top of each other. While one can reasonably easily discern the multiple copies in Fig. 2a, which measures the total counts at each beam stop in the scan, it is extremely difficult to interpret the phase reconstruction (the true STEMH measurement) in Fig. 2b.

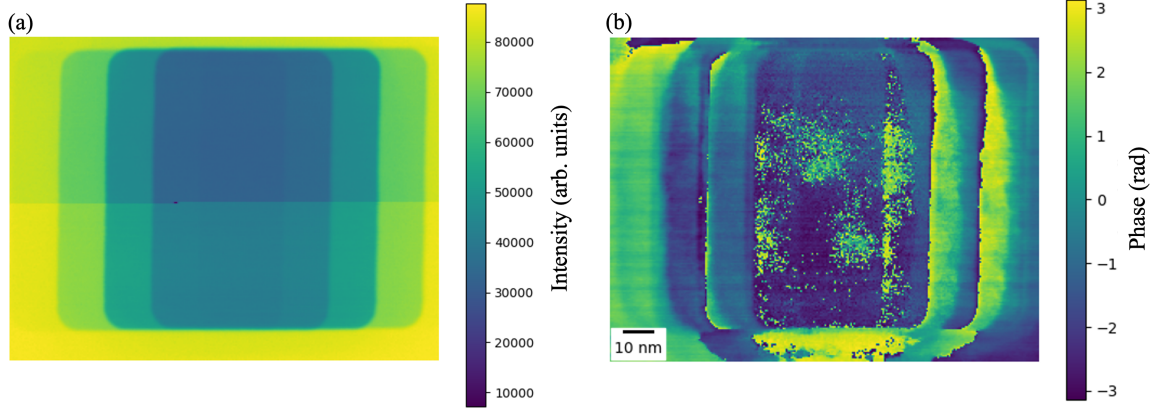


Figure 2. A Scanning Transmission Electron Microscopy Holography (STEMH) measurement of a silver nanocube 100 nm in length. (a) The measured intensity. (b) The measured phase. The scale bar applies identically to both images.

The second hurdle is the current qualitative nature of the phase measurements. Consider a toy one-dimensional STEMH model with only three probes (the 0th and ± 1 diffraction orders of the grating) where only the +1st order interacts with the sample. Let the probe separation be x_0 , the specimen phase be ϕ , and the relative amplitude of the n th probe be c_n . Then the electron wavefunction after the sample ψ is

$$\psi(x) = c_{-1}\delta(x + x_0) + c_0\delta(x) + c_{+1}e^{i\phi}\delta(x - x_0) \quad (1.1)$$

The detector records the modulus squared of the Fourier transformed post-specimen wavefunction $|FT[\psi(x)]|^2 = |\psi(w)|^2$

$$|\psi(w)|^2 = \sum_i |c_i|^2 + 2c_{-1}c_0 \cos(wx_0) + 2c_0c_{+1} \cos(wx_0 - \phi) + 2c_{-1}c_{+1} \cos(2wx - \phi), \quad (1.2)$$

a linear combination of waves phase shifted by the delay of the first order probe passing through the sample—classic interference fringes. The first step of the STEMH reconstruction algorithm is to take a FFT of the fringes, which yields

$$\begin{aligned} \sum_i |c_i|^2 \delta(x) + (c_{-1}c_0 + c_0c_{+1}e^{-i\phi})\delta(x - x_0) + (c_{-1}c_0 + c_0c_{+1}e^{i\phi})\delta(x + x_0) \\ + c_{-1}c_{+1}e^{-i\phi}\delta(x - 2x_0) + c_{-1}c_{+1}e^{i\phi}\delta(x + 2x_0) \end{aligned} \quad (1.3)$$

All interference from neighboring probes (the pairs (-1,0) and (0, +1)) is sent to the first peak at $x = x_0$. The Fourier transform cannot isolate the -1st and 0th probe interference from the 0th and +1st probe interference. We instead can only measure a phase $\arg(c_{-1}c_0 + c_0c_{+1}e^{-i\phi})$, which is decidedly not ϕ . Admittedly, in this toy example, we could get the sample phase via the phase of the second order peak. However, in real experiments with more probes, this peak would contain similarly polluting interference between, for example, the 0th and +2nd order probes, as interference of probes n orders apart is all sent to the n th peak.

Chapter II of this dissertation investigates one-dimensional phase diffraction grating theory in an attempt to understand how we could better design our beamsplitters to limit the number of probes incident on the sample. The main takeaway for STEM holography is that the theoretically optimal two-beam diffraction grating places only 81% of incident electrons into the desired beam paths, leaving far too many electrons in higher orders for direct quantitative measurements. This Chapter also provides novel developments in the scalar one-

dimensional phase grating theory, including the first new phase grating profiles with exact analytical formulas for their full diffraction since the 1990s.

Chapter III dives into the experimental considerations necessary to perform STEMH. We walk through grating fabrication, microscope alignment, the base-level theory, and the reconstruction process. We demonstrate experimental measurements of nanomagnetic stripe domains in Fe/Gd multilayers, Au and Ag nanoparticles, and few-atomic-layer GaN.

Chapter IV considers the integration of iterative ptychography with STEM holography. We discuss the particular algorithmic changes necessary to tailor the ptychographic post-processing to the STEMH geometry, then demonstrate simulations using the combination to remove ghost images from the final phase reconstruction image.

We conclude with future directions that could expand upon the great potential STEM holography has as a complementary absolute phase contrast measurement technique at all spatial frequencies.

CHAPTER II

NEW ANALYTICAL ONE-DIMENSIONAL PHASE GRATINGS IN SCALAR DIFFRACTION THEORY

This chapter is adapted from a similarly titled article accepted for publication by the Journal of the Optical Society of America A, cowritten with Kyle Cole, Devon Ball, and Benjamin McMorran. I developed the idea for the project, derived the theory and its analysis, and wrote the manuscript. KC fabricated the example diffraction gratings. DB and I acquired the TEM diffraction data from the gratings. DB and KC acquired the atomic force microscopy scans of the grating profiles. KC and BM provided edits to the original text.

This chapter describes my investigation into one-dimensional phase grating theory. While the gratings we nanofabricate are two-dimensional, their phase profiles are uniform along one axis; as a result, we get one line of individual diffraction peaks. In our group, the norm was to fabricate a binary square wave (a Ronchi ruling) with a π phase shift. While trying to understanding what other phase grating profiles could serve as better beamsplitters for STEM holography, I found a hole in the literature: there simply are not that many phase grating profiles with known analytical diffraction efficiencies in scalar (Kirchoff or Fourier) diffraction theory. The reason for this is two-fold. One, application moved beyond scalar diffraction. Near-field effects can only be neglected when the smallest grating feature size is much larger than the wavelength of the incident light. This approximation does not hold, for example, in the vast majority of gratings illuminated by light in the ultraviolet to infrared regimes. Two, the math is moderately complicated, as using scalar diffraction theory to model a phase grating with phase profile $\phi(x)$ requires Fourier transforming $\exp[i\phi(x)]$. However, the time

is ripe to re-explore this regime. X-ray, electron, neutron, and other massive quanta have much smaller wavelengths than the gratings used to manipulate them, and are increasingly being exploited as experimental probes. These optical subfields could use the grating profiles I describe in this chapter.

2.1 Introduction

Many mature modern technologies, including optical communications, spectroscopy [32], and intraocular lenses [33], employ diffraction gratings as versatile wavelength selectors or beamsplitters. Several sophisticated tools, such as rigorous coupled wave analysis (RCWA) [34], the Fourier modal method (FMM) [35], and finite difference time domain (FDTD) methods [36], model full vectorial solutions to Maxwell's equations for a given grating geometry. Such rigor is necessary to derive physical predictions when the smallest grating feature size s is roughly the same order of magnitude as the incident light wavelength λ [37, 38, 39]. However, in burgeoning subfields like x-ray [40], electron [41], and neutron optics [42], the wavelengths in question range from the edge of modern fabrication limits (nanometer scale; soft x-rays, ultracold neutrons), to the size of an atom (Angstrom scale; tender x-rays, cold neutrons), and beyond (sub-Angstrom scale; hard x-rays and gamma rays, hot neutrons, relativistic free electrons). Here, we can safely use the much simpler scalar diffraction theory.

Also called Fraunhofer, or far-field, diffraction in Fourier optics, or Raman-Nath diffraction in acousto-optics, scalar diffraction theory describes the complex-valued output of a plane wave incident on some object, when the resulting wavefunction is viewed functionally infinitely far from said object. This requires either measuring the final wavefunction some transverse distance L away from the object such that $s \ll L$, or at the back focal plane of a lens placed behind

the object. Describing the object by a transfer function $T(x, y)$, the far-field wavefunction is the Fourier transform of the transfer function

$$\Psi(p, q) = \frac{|\psi_0|}{f\lambda} \iint_{-\infty}^{\infty} T(x, y) e^{-i2\pi(px+qy)} dx dy, \quad (2.1)$$

where $p = \frac{x'}{f\lambda}, q = \frac{y'}{f\lambda}$ are reciprocal space coordinates with units of inverse length m^{-1} , f is the lens focal length, $|\psi_0|$ is the intensity of the plane wave incident on the grating, and x', y' are Cartesian coordinates in the back focal plane. We measure $|\Psi(p, q)|^2$. This familiar and deeply studied formalism significantly reduces the mathematical and computational overhead for grating design.

Gratings are often categorized as either transmission or phase gratings depending on whether their beam-shaping affects the amplitude or phase of incident light respectively. Young's double slit is an example of the former type, but many applications use the latter, as most light incident on a phase grating is kept in the optical system. In many physical systems both the amplitude and phase of the wavefront are perturbed, but researchers tend to only discuss pure amplitude or pure phase gratings. This is because we have limited mathematical tools to analytically describe diffraction from gratings with any phase modulation. Transmission gratings are easy to model; the diffraction from a grating with pure amplitude modulation $T(x, y) = A(x, y)$ is described by the Fourier transform $FT[A(x, y)]$. However, describing the effect of any phase modulation $\phi(x, y)$ requires computing $FT[A(x, y)e^{i\phi(x, y)}]$. The variation residing inside a complex exponential vastly increases the difficulty of finding a closed-form description of the grating diffraction (although the computational benefits remain).

This challenge is reflected in the paucity of one-dimensional pure phase gratings ($A(x) = 1$) with known analytical formulas for their diffraction. There

exist three main profiles in the literature [43, 44, 45, 46]: rectangular-, triangular-, and sine-wave gratings. The binary grating or Ronchi ruling is a rectangular grating with a 50/50 duty cycle, while the sawtooth wave and blazed gratings are specific forms of triangular-wave gratings. The simplicity of these structures allows for straightforward derivations of analytical formulas for the relative intensity of each and every diffraction order. However, further examples of such one-dimensional thickness profiles are scarce at best. Some combinations and remixes of these profiles have computable diffraction patterns. The Dammann grating, a concatenation of N rectangular blocks of up to N different heights and widths, was introduced in 1971 [47] and had a closed-form equation for all diffraction efficiencies first published in 1977 [48]. This prompted a flurry of follow-up work as this new foundation for innovation led to efficient beamsplitters [49, 50]. Similarly, one can interleave two blazed gratings of different slopes and heights [51, 52]. In general, the derivation of pure phase grating diffraction from any polygonal structure should be an algebraic exercise, but we are unaware of further work investigating such profiles.

Continuous phase profiles with known analytical diffraction efficiencies in every order are a different story entirely. The lone examples we are aware of are “tilted-sine” [53] and “sine-cycloid” [54] profiles, whose stated, but not derived, analytical diffraction efficiency formulas appear correct. Otherwise, analytical work has found the fundamental limits for optimal beamsplitting [55, 56], determines the profile for optimal diffraction into a particular set of diffraction orders [57, 58, 59], or describes the diffraction of an arbitrary thickness profile via a combination of infinite products and sums [60].

Yes, one can brute-force a machine-learning or other computational search method to find a thickness profile whose diffraction is optimized for a desired application [61, 62, 63, 64, 65, 66]. However, the result often requires fine sub-wavelength features beyond current fabrication techniques for small-wavelength optics. The same problem occurs for metasurfaces, large 2D planes composed of many distinct subwavelength thick scatterers, or meta-atoms [67]. Nor are Dammann gratings immune. The smallest feature size on a Dammann grating decreases with the number of desired equal-intensity diffraction orders. The best Dammann seven-beamsplitter (due to symmetry, the third-simplest case) has a minimum feature size two orders of magnitude smaller than the grating period [48]. Alternative approaches avoid the difficulty of analyzing 1D phase gratings and use known profiles while: encoding more general functions on Dammann gratings [68], stacking two 1D gratings [69, 70], extending 1D profiles to 2D [71, 72, 73, 74], or randomizing grating parameters [75, 76, 77]. It is our belief that the field has skipped past some viable options remaining in the 1D domain, and that this regime deserves a second look.

We note some recent work finding a closed-form expression for 1D binary gratings with simultaneous amplitude and phase modulation [78] and exploring how the periodicity of 1D Dammann gratings can be altered to selectively increase the intensity of diffraction orders that are multiples of any integer n [79].

Here, we provide closed-form descriptions of diffraction patterns for three novel thickness profiles: full- and half-wave rectified sine waves and a periodic extension of the parabola. We consider both amplitude and phase modulation. These are only slight variations on existing basic one-dimensional patterns and should be compatible with current fabrication techniques. We

demonstrate measured electron diffraction from nanofabricated full- and half-wave rectified gratings. Computationally, analytical solutions can be evaluated very rapidly within existing simulation software, outpacing a numerically integrated determination of one's diffraction pattern, thus accelerating the development pipeline. As for optical applications, all three gratings can produce three equal intensity beams, i.e., have triplicate beamsplitter capabilities, when using pure phase modulation. The half-wave rectified grating has four such distinct phase delays, two of which are so close as to create a very robust region of essentially three-beam performance. For a half-wave rectified grating made of BK7 glass (index of refraction 1.518) diffracting light in air from a He:Ne laser (632.8 nm wavelength), any depth between 3.13 and 3.28 microns creates three diffraction orders within, at worst, 1.26% diffraction efficiency from each other. This and one further half-wave triplicate pattern have smaller relative parasitic orders than similar patterns for the sinusoidal grating and optimum triplicating beamsplitter [57].

In addition, we reinvestigate two trailblazing optimal pure phase gratings to, for the first time, provide analytical representations of their diffraction efficiencies in all diffraction orders. The works of [57] and [58] use the calculus of variations to determine the pure phase profiles that maximize the diffraction in the 0th and ± 1 st, and 0th and +1st, diffraction orders respectively. Both original papers also derive analytical formulas for the optimized diffraction order efficiencies, but not for the infinite number of other diffraction orders produced. These results are different from the above, and are consequently discussed separately, because 1. we did not independently derive the phase profiles and 2. we do not expand the theory to include loss.

2.2 Theory

Consider a unit-amplitude plane wave of wavelength λ normally incident on a diffraction grating described by a complex-valued transfer function $T(x)$ with period P . Scalar diffraction theory tells us the resulting wavefunction measured at a plane functionally infinitely far from the object will be composed a series of outgoing beams, or diffraction orders. The relative intensity of these orders, indexed by an integer m , depend on the Fourier coefficients c_m of the transfer function

$$c_m = \frac{1}{P} \int_0^P T(x) e^{-i\frac{2\pi}{P}mx} dx. \quad (2.2)$$

The measurable intensity of each order $\eta_m = c_m^* c_m = |c_m|^2$ is called the diffraction efficiency. For a more rigorous treatment of how diffraction orders arise from the scalar theory, see [44, 45]. The diffraction grating transfer function depends on the diffraction thickness profile $t(x)$ according to

$$T(x) = e^{i\tilde{\Phi}t(x)} = e^{(-\gamma+i\Phi)t(x)} \quad (2.3)$$

where $\tilde{\Phi} = \Phi + i\gamma$ describes the phase modulation and beam attenuation per unit length as the incident plane wave traverses the grating. The phase modulation per unit length depends on the quanta and the physics of the quanta-grating interaction. For photons, $\Phi = k\Delta n$, where k is the wavenumber $\frac{2\pi}{\lambda}$ and Δn is the difference in refractive index between the grating and surrounding medium. For electrons, $\Phi = \sigma V_{MIP}$, where σ is the acceleration-voltage-dependent interaction parameter and V_{MIP} is the mean inner potential of the grating material [80]. For neutrons, $\Phi = \rho b_c \lambda$, where ρ is the number density of scatterers in the material and

b_c is the coherent scattering length [81]. γ is a material-dependent parameter with units of inverse length. If d is the maximum depth of the thickness profile $t(x)$, we can often write the Fourier coefficients as functions of $\tilde{\phi} = \tilde{\Phi}d$. In general, the Fourier coefficients are not periodic in the maximum phase shift $\Phi d \in [0, \infty)$.

If the thickness profile is odd ($t(x) \equiv t_o(x)$ if $t(-x) = -t(x)$), Euler's identity breaks the integrand of c_m into even and odd parts.

$$e^{i(\tilde{\Phi}t_o(x) - \frac{2\pi}{P}mx)} = \cos\left(\tilde{\Phi}t_o(x) - \frac{2\pi}{P}mx\right) + i \sin\left(\tilde{\Phi}t_o(x) - \frac{2\pi}{P}mx\right)$$

After changing variables by $\frac{2\pi}{P}x \rightarrow x$, the symmetric integrals of even and odd functions about the origin force $c_{m,o}$, the diffraction coefficients for an odd phase profile, to be

$$c_{m,o} = \frac{1}{\pi} \int_0^\pi \cos\left(\tilde{\Phi}t_o\left(\frac{P}{2\pi}x\right) - mx\right) dx \quad (2.4)$$

$$= \frac{1}{\pi} \int_0^\pi \left[\cos\left(\tilde{\Phi}t_o\left(\frac{P}{2\pi}x\right)\right) \cos(mx) + \sin\left(\tilde{\Phi}t_o\left(\frac{P}{2\pi}x\right)\right) \sin(mx) \right] dx \quad (2.5)$$

Note $c_{m,o}$ is purely real, so its phase can only be 0 or π . It will be advantageous to use either form of $c_{m,o}$.

Scalar diffraction theory is typically invalid in more standard photon-optical regimes. It does not include polarization dependence and consequently struggles to accurately predict the far-field distribution of light when the ratio of the grating feature size to the incident wavelength $\frac{s}{\lambda} > 0.1$. This is not a hard-and-fast rule, as the exact ratio where the error becomes non-negligible depends on the grating duty cycle [82], and can be partially addressed with slight variations to the scalar theory [83]. However, in small-wavelength optics, by definition, this is not a concern. The more likely failure of this model is in the large ϕ regime. We are approximating

the modulation of the incident beam throughout the grating (along the optical axis z) as occurring in one infinitesimally-thin plane. Using a large quantity of phase-shifting material to generate a large phase shift may break this approximation and introduce near-field scattering effects not considered by this theory.

We can extend the general scalar diffractive theory to consider thickness variations $t(x)$ interspersed by regions of no beam modulation. For a numeric study of sinusoidal and Gaussian phase gratings with a similar concept, see [84]. We define the transfer function as

$$T(x) = \begin{cases} e^{i\tilde{\Phi}t(x)}, & 0 \leq x \leq fP \\ 1, & fP < x < P \end{cases} \quad (2.6)$$

where the thickness variations occur over an active region of length fP , where P remains the total period and $f \in [0, 1]$ is the duty cycle. If we are considering such arbitrary duty cycle gratings, we use a bar over the Fourier coefficient $\bar{c}_m$. Using Eq. (2.2), the diffraction of these gratings is described by

$$\bar{c}_m(f, \tilde{\Phi}) = \frac{1}{P} \int_0^{fP} e^{i\tilde{\Phi}t(x)} e^{-i\frac{2\pi}{P}mx} dx + \frac{1}{P} \int_{fP}^P e^{-i\frac{2\pi}{P}mx} dx. \quad (2.7)$$

To simplify this expression, first consider the Fourier expansion d_n of our active region transfer function $\exp[i\tilde{\Phi}t(x)]$, which has a period fP :

$$d_n = \frac{1}{fP} \int_0^{fP} e^{i\tilde{\Phi}t(x)} e^{-i\frac{2\pi}{fP}nx} dx. \quad (2.8)$$

Up to a factor of f , we can interpret the first integral of Eq. (2.7) as the mf th Fourier coefficient of $\exp[i\tilde{\Phi}t(x)]$, special only in that $mf \in \mathbb{R}$. The second term in

Eq. (2.7) is an elementary integral that requires a separate case if $m = 0$. Together, the Fourier coefficients for arbitrary duty cycle gratings can be written as

$$\bar{c}_m(f, \tilde{\Phi}) = \begin{cases} f (d_{mf} - e^{-imf\pi} \text{sinc}(mf\pi)), & m \neq 0 \\ 1 - f + fd_0, & m = 0 \end{cases}. \quad (2.9)$$

Beware since $mf \in \mathbb{R}$, blindly substituting the coefficient for the normal ($f = 1$) grating into Eq. (2.9) can generate incorrect predictions. The boundary conditions $\bar{c}_m(0, \tilde{\Phi}) = \bar{c}_m(f, 0) = \delta_{m,0}$ are good sanity checks, since either $f = 0$ or $\tilde{\Phi} = 0$ eliminates any beam modulation. In other words, the “grating” is simply free space, and we would expect to see all intensity to remain along the optical axis, i.e., in the 0th order.

2.3 Absolute Sinusoid

2.3.1 Full-Wave Rectified Gratings. The absolute sinusoid has a period P half the period of the sinusoid it derives from.

$$t^{\text{f.r.}}(x) = A \left| \sin \frac{2\pi}{2P} x \right| \quad (2.10)$$

Fig. 3a depicts this thickness profile. In alternating current electronics, this function results from a full-wave rectifier. The diffraction pattern for this thickness profile depends on

$$c_m^{\text{f.r.}} = \frac{1}{P} \int_0^P e^{i\tilde{\Phi}A \sin(\frac{\pi}{P}x)} e^{-i\frac{2\pi}{P}mx} dx. \quad (2.11)$$

Splitting this into real and imaginary parts, using $\tilde{\phi} = \tilde{\Phi}A$, and making the substitution $u = \frac{\pi}{P}x$, we get

$$c_m^{\text{f.r.}} = \frac{1}{\pi} \int_0^\pi \cos(2mu - \tilde{\phi} \sin u) - \frac{i}{\pi} \int_0^\pi \sin(2mu - \tilde{\phi} \sin u) du \quad (2.12)$$

These are Anger $\mathbf{J}_n$ and Weber $\mathbf{E}_n$ functions of order $n = 2m$, however, we do not need to fully resort to more obscure special functions, as for any integer n , $\mathbf{J}_n(z)$ reduces to the Bessel function $J_n(z)$ [85]. Thus, the full-wave rectified sine wave phase grating has Fourier coefficients

$$c_m^{\text{f.r.}} = J_{2m}(\tilde{\phi}) - i\mathbf{E}_{2m}(\tilde{\phi}). \quad (2.13)$$

If we have a pure phase grating ($\tilde{\phi} = \phi$), the diffraction pattern is

$$\eta_m^{\text{f.r.}} = |c_m^{\text{f.r.}}|^2 = J_{2m}^2(\phi) + \mathbf{E}_{2m}^2(\phi). \quad (2.14)$$

Both $J_{-m}(z) = (-1)^m J_m(z)$ and $\mathbf{E}_{-m}(z) = (-1)^m \mathbf{E}_m(z)$, so $c_m^{\text{f.r.}} = c_{-m}^{\text{f.r.}}$. Both Anger and Weber functions are built-in to Mathematica and available in Python via the package mpmath [86, 87]. Fig 3b plots the first five unique diffraction efficiencies for a pure phase grating. Unsurprisingly, the diffraction efficiency formula for the full-wave rectified sinusoidal phase grating has some resemblance to the diffraction efficiency for the sinusoidal phase grating $\eta_m^{\text{sin}}(\phi) = J_m^2(\phi)$. This latter function enjoys the coincidence that the diffraction order m is identical to the order of the Bessel function. This is not the case for the full-wave rectified diffraction efficiency, where the orders of the special functions are twice the diffraction order.

2.3.2 Half-Wave Rectified Gratings. Again taking inspiration from electronics, we move onto the half-wave rectified sine wave. Here, the grating period matches the period of the sine wave used to describe the non-zero portion of the thickness profile

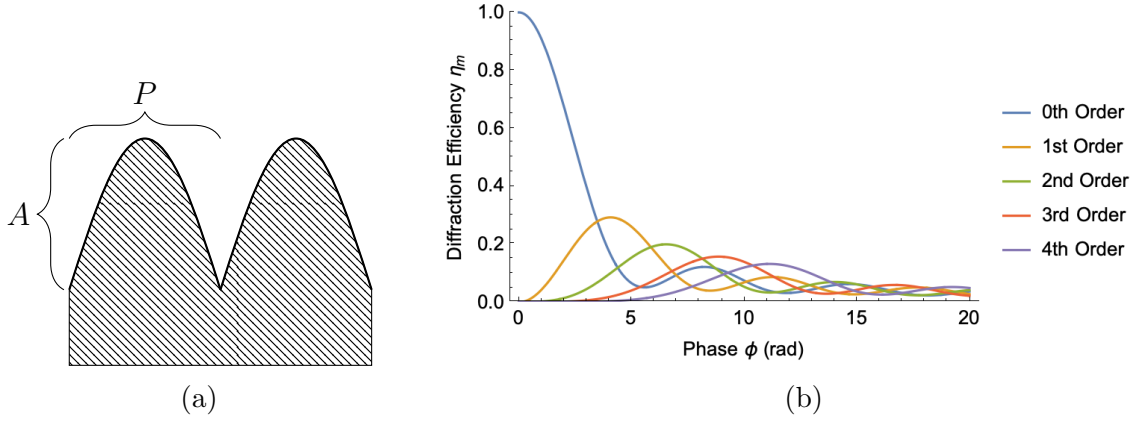


Figure 3. (a) A schematic of the full-wave rectified phase grating with (b) its diffraction efficiency in the first five orders with increased phase delay ϕ and no amplitude loss.

$$t^{\text{h.r.}}(x) = \begin{cases} A \sin \frac{2\pi}{P} x, & 0 \leq x \leq \frac{P}{2} \\ 0, & \frac{P}{2} \leq x < P \end{cases}. \quad (2.15)$$

Fig. 4a presents a schematic of the phase profile. Again using $\tilde{\phi} = \tilde{\Phi}A$, a very similar integral to Eq. (2.11) appears here:

$$c_m^{\text{h.r.}} = \frac{1}{P} \int_0^{\frac{P}{2}} e^{i\tilde{\phi} \sin(\frac{2\pi}{P}x)} e^{-i\frac{2\pi}{P}mx} dx + \frac{1}{P} \int_{\frac{P}{2}}^P e^{-i\frac{2\pi}{P}mx} dx \quad (2.16)$$

$$= \frac{1}{2} \mathbf{J}_m(\tilde{\phi}) - \frac{i}{2} \mathbf{E}_m(\tilde{\phi}) + \frac{1}{2\pi} \int_{\pi}^{2\pi} e^{-imv} dv. \quad (2.17)$$

This second integral is $\frac{1}{2}i^m \text{sinc}\left(\frac{m\pi}{2}\right)$. Assuming a pure phase grating, the ultimate diffraction efficiencies are

$$\eta_m^{\text{h.r.}} = \frac{1}{4} \left[J_m^2(\phi) + \mathbf{E}_m^2(\phi) + \text{sinc}^2\left(\frac{m\pi}{2}\right) \right] + \frac{1}{2} \text{sinc}\left(\frac{m\pi}{2}\right) \times \begin{cases} J_m(\phi), & m \equiv 0 \pmod{4} \\ -\mathbf{E}_m(\phi), & m \equiv 1 \pmod{4} \\ -J_m(\phi), & m \equiv 2 \pmod{4} \\ \mathbf{E}_m(\phi), & m \equiv 3 \pmod{4} \end{cases} \quad (2.18)$$

Fig. 4b plots the first five unique diffraction efficiencies for a pure phase grating. Note for $\Phi A > 16.88$ rad, $\eta_0^{\text{h.r.}} > \eta_{\pm 1}^{\text{h.r.}}$. The modulation-less space has contributed a phase independent, low- m -localized term $\frac{1}{4} \text{sinc}^2\left(\frac{m\pi}{2}\right)$ to the diffraction efficiency. 90.5% of the total sinc function contribution to the diffraction pattern intensity is in the $m = 0$ and $m = \pm 1$ orders. This creates a less interesting, although perhaps more useful, diffraction pattern which is generally dominated by the lower diffraction orders.

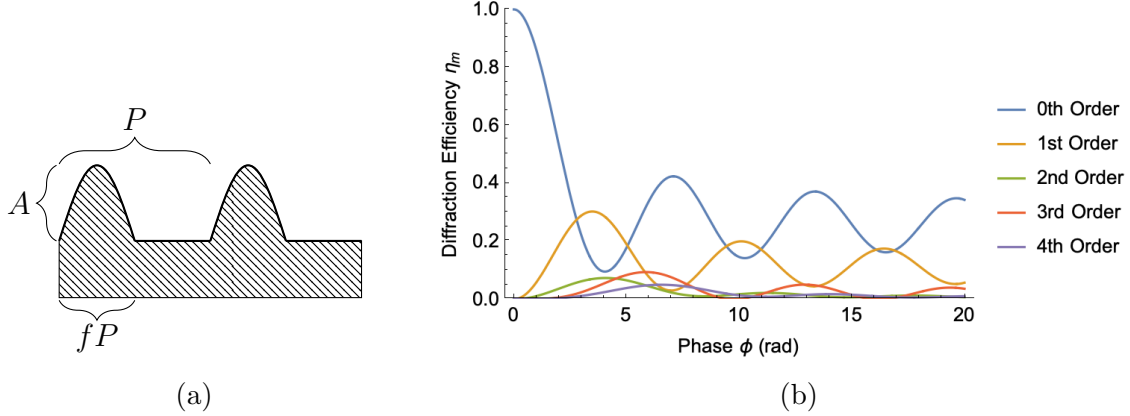


Figure 4. (a) A schematic of the half-wave rectified phase grating with (b) its diffraction efficiency in the first five orders with increased phase delay ϕ and no amplitude modulation.

2.3.3 Arbitrary Duty Cycle. We now generalize the full- and half-wave rectified gratings ($f = 1$ and $f = 1/2$ respectively) to absolute

sinusoid gratings with any duty cycle. We must insert the absolute sinusoid Fourier coefficients c_m without simplifying $\mathbf{J}_m \rightarrow J_m$, since here we have a non-integral order $d_{mf}(\tilde{\phi}) = \mathbf{J}_{2mf}(\tilde{\phi}) - i\mathbf{E}_{2mf}(\tilde{\phi})$. The resulting m th order Fourier coefficients are

$$\bar{c}_m^{\text{abs}}(f, \tilde{\phi}) = \begin{cases} f \left(\mathbf{J}_{2mf}(\tilde{\phi}) - i\mathbf{E}_{2mf}(\tilde{\phi}) - e^{-imf\pi} \text{sinc}(mf\pi) \right), & m \neq 0 \\ 1 - f + fJ_0(\tilde{\phi}) - if\mathbf{E}_0(\tilde{\phi}), & m = 0 \end{cases}. \quad (2.19)$$

This function retains the property $\eta_m^{\text{abs}} = \eta_{-m}^{\text{abs}}$. We explore this further in Appendix A.1. Figs. 5a and b show how the first four unique diffraction orders vary with both f and ϕ for a lossless grating. Note how each order contains bands with m maxima over $0 < f \leq 1$ due to the sinc term.

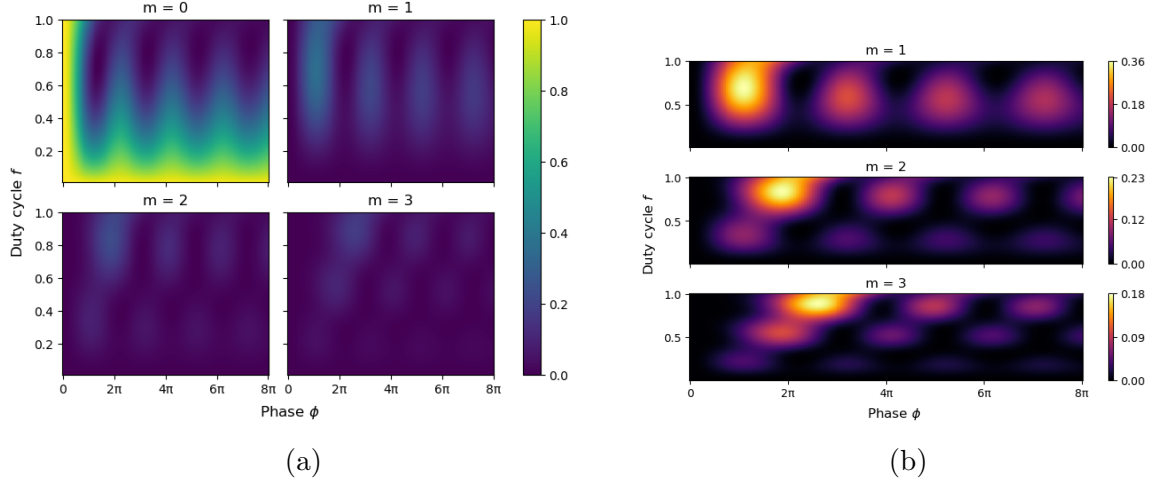


Figure 5. Diffraction efficiencies of the first four orders for the absolute sinusoid pure phase grating with arbitrary duty cycle as a function of phase delay ϕ and duty cycle f . (a) The zeroth through third diffraction efficiencies are plotted on the same color scale to compare their relative intensities. (b) The first through third diffraction efficiencies are plotted with individual color scales to highlight the qualitative features of each diffraction order as a function of ϕ and f .

2.4 Sine Wave

We note an identical method determines the diffraction of a standard sinusoidal phase grating without applying the Jacobi-Anger expansion, then Fourier

transforming the resulting infinite series, as done in [45]. It also allows us to treat the sinusoidal grating as a mixed amplitude and phase grating.

$$c_m^{\text{sin}} = \frac{1}{P} \int_0^P e^{i\tilde{\phi} \sin(\frac{2\pi}{P}x)} e^{-i\frac{2\pi}{P}mx} dx \quad (2.20)$$

$$= \frac{1}{2\pi} \int_0^{2\pi} \cos(\tilde{\phi} \sin v - mv) dv - \frac{i}{2\pi} \int_0^{2\pi} \sin(\tilde{\phi} \sin v - mv) dv \quad (2.21)$$

$$= \frac{1}{\pi} \int_0^\pi \cos(\tilde{\phi} \sin v - mv) dv = J_m(\tilde{\phi}) \quad (2.22)$$

where $v = \frac{2\pi}{P}x$, with the last line following from the parities of $\sin x$ and $\cos x$.

2.5 Parabolic Gratings

Our final thickness profile is the parabola, considered first in [88] and fabricated in [89], which we define over $x \in [-\frac{P}{2}, \frac{P}{2}]$ to eliminate unnecessary constant phases in c_m .

$$t^{\text{para}}(x) = \frac{4A}{P^2}x^2, \quad x \in \left[-\frac{P}{2}, \frac{P}{2}\right] \quad (2.23)$$

Fig. 6a depicts the periodic extension of the parabola. Its Fourier coefficients, after the substitution $u = \frac{2}{P}x$, are

$$c_m^{\text{para}} = \frac{1}{P} \int_{-\frac{P}{2}}^{\frac{P}{2}} e^{i\tilde{\phi}A\frac{4x^2}{P^2}} e^{-i\frac{2\pi}{P}mx} dx = \frac{1}{2} \int_{-1}^1 e^{i\tilde{\phi}u^2} e^{-i\pi mu} du \quad (2.24)$$

A more general form of this integral is Eq. 7.4.32 of [85]:

$$\int e^{-(ax^2+2bx+c)} dx = \frac{1}{2} \sqrt{\frac{\pi}{a}} e^{\frac{b^2-ac}{a}} \text{erf}\left(\sqrt{a}x + \frac{b}{\sqrt{a}}\right) + \text{const.}, \quad (2.25)$$

which leads to the m th order parabolic grating Fourier coefficients

$$c_m^{\text{para}} = (-1)^{1/4} e^{-\frac{im^2\pi^2}{4\tilde{\phi}}} \frac{\sqrt{\pi}}{4\sqrt{\tilde{\phi}}} \left(\text{erf} \left[e^{i\frac{\pi}{4}} \frac{-2\tilde{\phi} + m\pi}{2\sqrt{\tilde{\phi}}} \right] - \text{erf} \left[e^{i\frac{\pi}{4}} \frac{2\tilde{\phi} + m\pi}{2\sqrt{\tilde{\phi}}} \right] \right). \quad (2.26)$$

The first two factors drop out in the diffraction efficiency $\eta_m = c_m^* c_m$ and the complex conjugate $[\text{erf}(z)]^* = \text{erf}(z^*)$ so

$$\eta_m^{\text{para}} = \frac{\pi}{16\tilde{\phi}} \left(\text{erf} \left[e^{i\frac{\pi}{4}} \frac{-2\tilde{\phi} + m\pi}{2\sqrt{\tilde{\phi}}} \right] - \text{erf} \left[e^{i\frac{\pi}{4}} \frac{2\tilde{\phi} + m\pi}{2\sqrt{\tilde{\phi}}} \right] \right) \times \left(\text{erf} \left[e^{-i\frac{\pi}{4}} \frac{-2\tilde{\phi} + m\pi}{2\sqrt{\tilde{\phi}}} \right] - \text{erf} \left[e^{-i\frac{\pi}{4}} \frac{2\tilde{\phi} + m\pi}{2\sqrt{\tilde{\phi}}} \right] \right). \quad (2.27)$$

Fig. 6b plots these diffraction efficiencies up to $m = 4$.

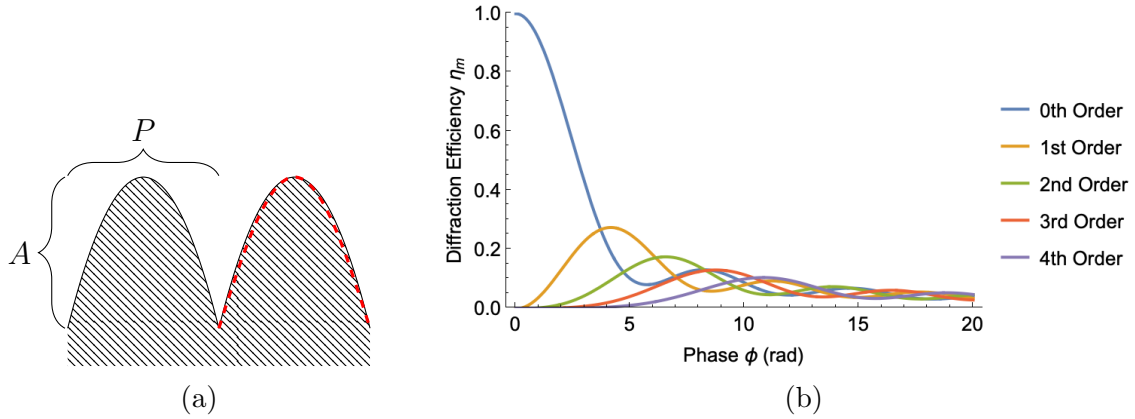


Figure 6. (a) A cartoon of the parabolic phase grating with (b) its diffraction efficiency in the first five orders with increased phase delay ϕ and no amplitude modulation. A dashed red sinusoidal profile is overlaid on the right parabola in (a) for comparison.

2.6 Applications

With all (lossy) gratings presented, Fig. 7 compares the strength of their zeroth and first orders relative to each other and the standard binary and sinusoidal phase gratings. Since we are comparing performance against known pure phase gratings, we do not consider any amplitude modulation in this section. The three

novel grating designs provide an intermediate amount of intensity in the zeroth and first diffraction order intensities relative to the two standard grating profiles. The low-order intensity for these novel profiles also decreases slower than the standard profiles with increased phase shift, which should ease the fabrication of desired diffraction efficiencies.

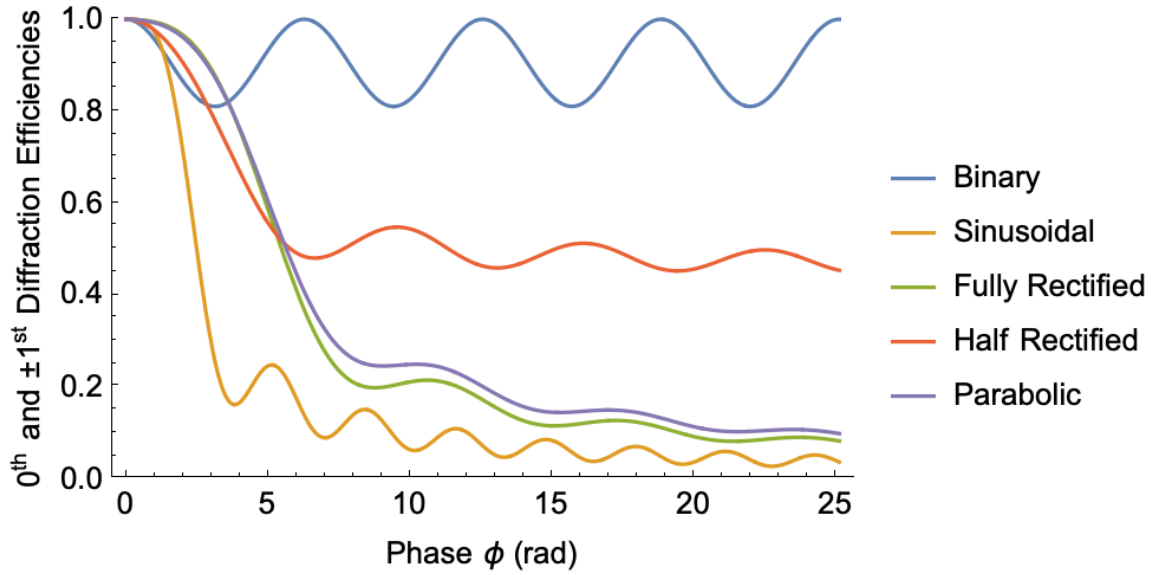


Figure 7. The sum of the 0th and $\pm 1^{\text{st}}$ diffraction efficiencies for the standard binary and sinusoidal thickness profiles and the half- and full-wave rectified and parabolic gratings.

These gratings, especially the half-wave rectified profile, could serve well as triplicators, or beamsplitters producing three equal intensity beams in the 0th and $\pm 1^{\text{st}}$ orders. Table 1 lists the absolute intensities of the seven lowest diffraction orders for the typical sinusoid, full-wave rectified, and parabolic gratings, in addition to the mathematically optimal triplicate beamsplitter design first found by [57]. The full-wave and parabolic gratings do not compare too favorably to existing designs: they have slightly less efficiency in the 0th and ± 1 orders, and more parasitic efficiency in the ± 2 and ± 3 orders. Fig. 8a shows the three-beam

splitting of the full-wave rectified grating. The advantage of the full-wave and parabolic grating is that the slower drop-off (smaller derivative $|\frac{d}{d\phi}[\eta_0 + \eta_{-1} + \eta_{+1}]|$) means it should be easier to actually fabricate the full-wave rectified triplicator than the sinusoidal or Gori triplicators.

m	Gori <i>et al.</i> (%)	Sinusoidal (%)	Full-Wave (%)	Parabolic (%)
0	30.9	30.0	27.9	26.8
± 1	30.9	30.0	27.9	26.8
± 2	1.9	4.7	6.8	7.5
± 3	1.4	0.3	1.1	1.6

Table 1. Triplicate beamsplitter diffraction efficiencies for the ideal three-beamsplitter, and the sinusoidal, full-wave rectified, and parabolic gratings.

There are exactly five locations where the half-wave rectified grating has equivalent intensity in the 0th and ± 1 orders, all visible in Fig. 4b. One, at $\phi = 5.045$ rad, contains too much light in the ± 2 and ± 3 orders as to be useful. Two are so close (16.09 and 16.88 rad) as to mark a region of near-three beam output. The maximum difference of the zeroth and first orders for $16 \text{ rad} \leq \phi \leq 17 \text{ rad}$ is only 1.26%. Table 2 compiles the first four diffraction orders for viable triplicate beamsplitting from the half-wave rectified grating. Fig. 8b-d shows the diffraction patterns for the distinct three-beam patterns from this thickness profile.

m	2.785 rad (%)	7.435 rad (%)	16.085 rad (%)
0	27.4	18.2	17.1
± 1	27.4	18.2	17.1
± 2	5.7	1.3	0.9
± 3	1.9	0.2	0.0

Table 2. Triplicate beamsplitter diffraction efficiencies for the half-wave rectified grating for three different phase delays ϕ .

The latter two examples in Table 2 compare quite favorably to extant triplicate beamsplitters. While the diffraction patterns of Figs. 6c and 6d (the

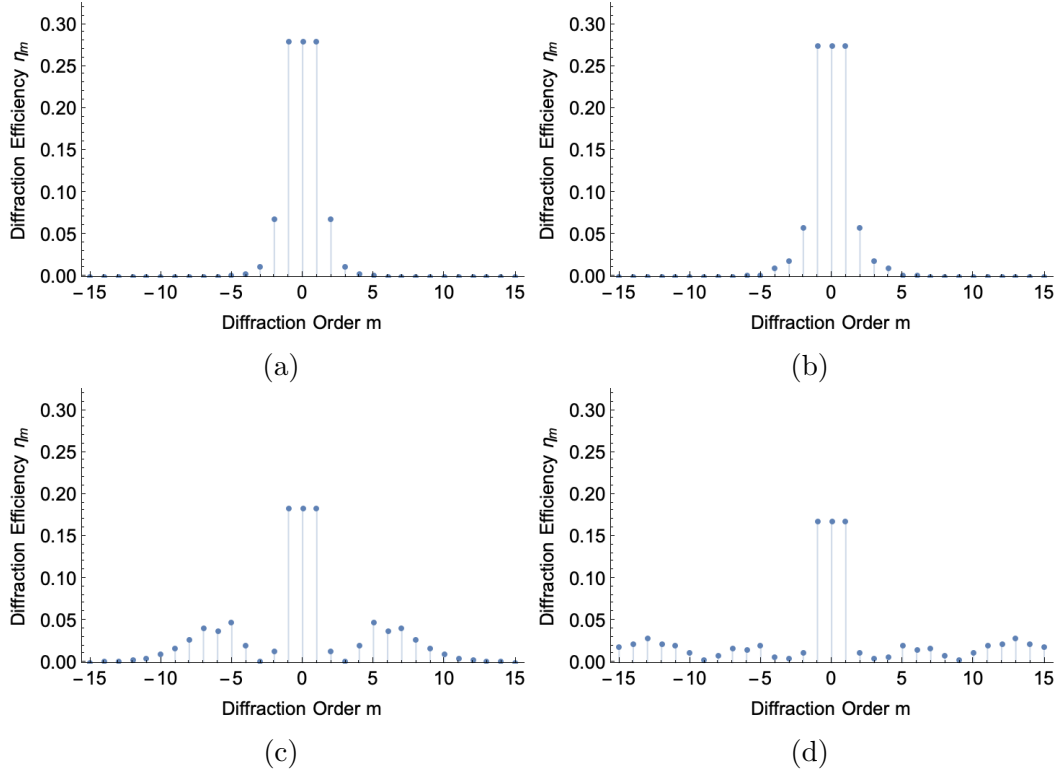


Figure 8. 1×3 beamsplitter diffraction patterns. (a) is full-wave rectified phase grating diffraction while (b-d) is half-wave rectified phase grating diffraction. (a). Full-wave diffraction with $\phi = 3.482$ rad. (b). Half-wave diffraction with $\phi = 2.785$ rad, (c). Half-wave diffraction with $\phi = 9.435$ rad, and (d). Half-wave diffraction with $\phi = 16.88$ rad.

right two columns of Table 2) have, in general, lower absolute diffraction efficiencies in the 0th and ± 1 orders, their adjacent orders are less bright. For larger phase shifts, the half-wave rectified grating diffracts more overall energy to higher orders, minimizing the efficiency of orders $m = 2$ and $m = 3$. This limits the amount of parasitic light near the region of interest (the 0th and ± 1 st orders). We can quantify this by comparing the ratios of the zeroth to second order diffraction efficiency $\frac{\eta_0}{\eta_2}$ for each grating. These are 16.3 for the Gori grating and 6.4 for the sinusoidal grating, and 14.0 and 19.0 for the $\phi = 7.435$ rad and $\phi = 16.085$ rad half-wave rectified beamsplitting cases respectively. Including the third order diffraction efficiency in the denominator, the ratio $\frac{\eta_0}{\eta_2 + \eta_3}$ is 9.4, 6.0, 12.1, and 19.0 for the Gori, sinusoidal, and two half-rectified gratings respectively. It may be fruitful to test the Gori or blazed gratings with 50% duty cycle, or $f = 1/2$, to minimize undesired intensities $\eta_{\pm 2}$ and $\eta_{\pm 3}$.

2.7 Experimental Results

To benchmark our theory, we fabricated full- and half-wave rectified gratings with focused ion beam milling (FIB) and measured their electron diffraction in a transmission electron microscope (TEM). Parabolic gratings were not fabricated because of their similarity to the full-wave rectified pattern. Gratings were milled on 100 nm thick silicon nitride membrane windows purchased from SPI Supplies. The etched backside of the membrane window is metalized with an adhesion layer of 2 nm of Ti and a charge alleviation layer of 10 nm of Pt deposited via e-beam evaporation. The uncoated membrane side is then milled with a FEI Helios 600i Ga FIB-SEM at an ion beam current of 87 pA. This corresponds to an approximate beam diameter of 50 nm. Diffraction was measured in a FEI Titan 80-200 TEM/STEM at 200 keV in Low Mag mode (real space magnification of 1550x)

and with a Large Angle Diffraction length of 810 m on a Gatan Rio Camera. The designed grating period was 488 nm, much larger than the 2.51 pm wavelength of the 200 keV electron. The depth profile of the diffraction gratings were determined by atomic force microscopy on a Bruker Dimension FastScan AFM.

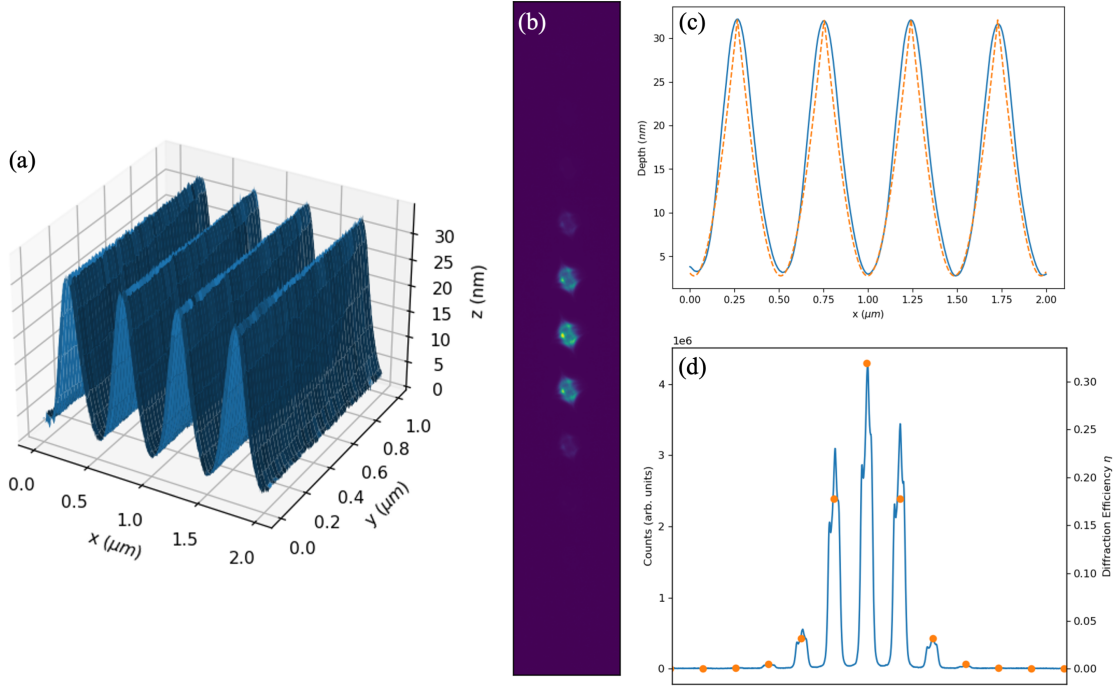


Figure 9. Experimental demonstration of full-wave rectified phase grating. (a) An AFM measurement of several periods of a nanofabricated full-wave grating. (b) The recorded electron diffraction of the full-wave grating. (c) The average depth profile along the x axis of (a). (d) Comparison of measured electron diffraction (line) and theoretically predicted diffraction efficiencies (dots).

Figs. 9a and 10a plot AFM measurements of several periods of the full- and half-wave gratings, respectively. The average peak-to-peak depths of the full- and half-wave gratings were 29 and 25 nm. Figs. 9b and 10b plot the measured electron diffraction of the full- and half-wave gratings, respectively. Structure within the diffraction spots is set by microscope aberrations and the aperture selecting the diffraction grating. The beam structure is propagated identically to each diffraction

order. To highlight the 1D nature of the experiment, we plot the measured average depth profile along the grating periodicities in Figs. 9c and 10c. Note that a sine-wave grating would be composed of alternating concave-up and concave-down grooves, whereas the full-wave rectified (absolute value of sine-wave) profile in Fig. 9c is composed solely of concave-up grooves.

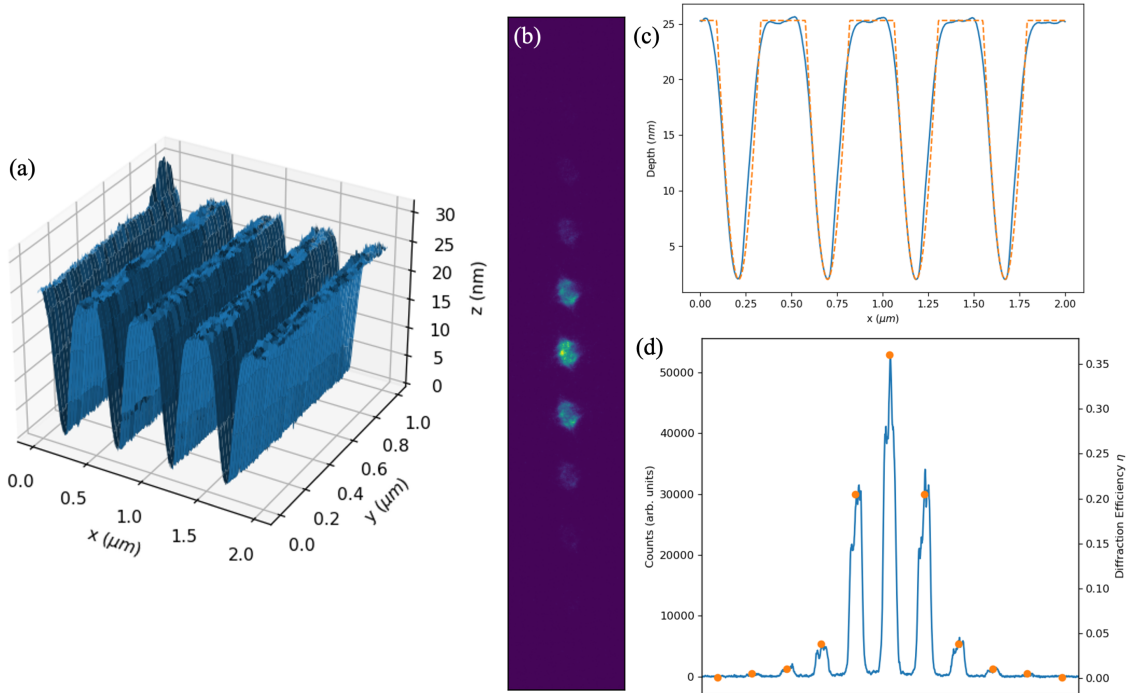


Figure 10. Experimental demonstration of half-wave rectified phase grating. (a) An AFM measurement of several periods of a nanofabricated half-wave grating. (b) The recorded electron diffraction of the half-wave grating. (c) The average depth profile along the x axis of (a). (d) Comparison of measured electron diffraction (line) and theoretically predicted diffraction efficiencies (dots).

We compare theory with experiment in Figs. 9d and 10d. We plot a one-dimensional trace of the measured diffraction overlaid with our theoretical prediction for each discrete diffraction order intensity. The trace is generated by summing the rows of the diffraction measurements in Figs. 9b and 10b. The theoretical predictions use Eqs. 2.13 and 2.16 with the measured average depths

(plotted in Figs. 9(c) and 10(c)) and $\tilde{\Phi} = \sigma V_{MIP} + i\gamma$ with $\sigma(200 \text{ keV}) = 0.0073 \text{ rad}/(\text{V} \cdot \text{nm})$, $\gamma = 0.008 \text{ nm}^{-1}$ [80], and $V_{MIP} = 13.4 \text{ V}$. A mean inner potential of 16.7 V has been measured for Si_3N_4 [90], however, the silicon nitride membranes we buy have a substantively different but unspecified stoichiometry [91]. We have indirectly determined the 13.4 V value by measuring the depth of silicon nitride necessary to impose a π phase shift and minimize the 0th order diffraction efficiency of a binary phase grating. These parameters correspond to maximum phase delays of 2.84 and 2.45 rad for the full- and half-wave rectified gratings. The theory is able to accurately predict the results of the simultaneous amplitude and phase modulation, with the exception of the ± 1 st diffraction orders, especially for the full-wave rectified grating. We attribute this disagreement to how nanofabrication rounds out the discontinuity where neighboring periods of the absolute sinusoid meet. In Fig. 9c, we see the theoretical cusps at the top of the figure, when milled, become broader smoothed edges. The same discontinuity is not so abrupt for the half-wave rectified grating, leading to a better match between the milled and theoretical profiles of the half-wave rectified grating in Fig. 10c, hence the better agreement for that design.

2.8 Optimum Triplicator

We now shift to the pure phase gratings optimized by the calculus of variations. [57] derived the phase profile which maximizes the intensity in the 0th and ± 1 st diffraction orders

$$\phi_3(x, \alpha) = \arctan(\alpha \sin x), \quad (2.28)$$

where $\alpha \in [0, \infty)$ is a parameter that tunes the relative intensity of the 0th and ± 1 diffraction orders. They then derived analytical representations of the diffraction

coefficients of interest c_0 and $c_{\pm 1}$ and determined they were equivalent in magnitude at $\alpha = 2.65718\dots$. This gives the optimum triplicator, the pure phase grating which directs the most light equally into the 0th and ± 1 st diffraction orders. Romero and Dickey later rederived a shifted version of the same profile $\arctan(\alpha \cos x)$ but did not reconsider the diffraction [55]. Fig. 11 shows examples of this phase profile for varying values of α . Their corresponding diffraction patterns are shown in the first row of Fig. 12.

Inserting the odd $\phi_3(x)$ inside Eq. (2.5), and using the identities

$$\cos(\arctan(f(x))) = \frac{1}{\sqrt{1+f(x)^2}} \quad (2.29)$$

$$\sin(\arctan(f(x))) = \frac{f(x)}{\sqrt{1+f(x)^2}}, \quad (2.30)$$

we get

$$c_m = \frac{1}{\pi} \int_0^\pi [\cos(\arctan(\alpha \sin x)) \cos(mx) + \sin(\arctan(\alpha \sin x)) \sin(mx)] dx \quad (2.31)$$

$$= \frac{1}{\pi} \int_0^\pi \frac{\cos(mx) + \alpha \sin(x) \sin(mx)}{\sqrt{1 + \alpha^2 \sin^2 x}} dx \quad (2.32)$$

$$= \frac{1}{\pi} \int_0^\pi \frac{\cos(mx) + \frac{\alpha}{2}(\cos((m+1)x) - \cos((m-1)x))}{\sqrt{1 + \alpha^2 \sin^2 x}} dx. \quad (2.33)$$

c_m is now composed of three very similar integrals. For all m , at least one is equal to zero: over the interval $x \in [0, \pi]$, $(1 + \alpha^2 \sin^2 x)^{-1/2}$ is even and the parity of $\cos nx$ matches the parity of n , so $\cos(nx)(1 + \alpha^2 \sin^2 x)^{-1/2}$ is odd over this interval for odd n . Consequently, for m even, the only nonzero integral is the first term

$$c_{m,e} = \frac{2}{\pi} \int_0^{\frac{\pi}{2}} \frac{\cos mx}{\sqrt{1 + \alpha^2 \sin^2 x}} dx. \quad (2.34)$$

For odd m , $m \pm 1$ is even, so we can write the Fourier coefficient $c_{m,o}$ in terms of the neighboring even coefficients $\frac{a}{2}(c_{m+1} - c_{m-1})$. The cosine in the numerator of Eq. (2.34) absorbs the sign of m , so $c_{m,e} = c_{-m,e}$, which forces $c_{-m,o} = -c_{m,o}$:

$$c_{-m,o} = \frac{a}{2}(c_{-m+1} - c_{-m-1}) = \frac{a}{2}(c_{-(m-1)} - c_{-(m+1)}) = \frac{a}{2}(c_{m-1} - c_{m+1}). \quad (2.35)$$

Therefore, for the rest of this section, we assume $m \geq 0$.

To compute the integral of Eq. (2.34), we expand $\cos mx$ into powers of $\cos x$ with Chebyshev polynomials of the first kind $T_m(\cos x)$. Defining the coefficient of the j th term in the n th Chebyshev polynomial of the first kind as $T_{n,j}$, $\cos mx = \sum_{k=0}^{m/2} T_{m,2k} \cos^{2k} x$. Our integral is now

$$\frac{2}{\pi} \sum_{k=0}^{m/2} T_{m,2k} \int_0^{\pi/2} \frac{\cos^{2k} x}{\sqrt{1 + \alpha^2 \sin^2 x}} dx. \quad (2.36)$$

This integral is Eq. (2) in Section 3.671 of [92], so

$$c_{m,e} = \frac{1}{\pi} \sum_{k=0}^{m/2} T_{m,2k} B\left(k + \frac{1}{2}, \frac{1}{2}\right) {}_2F_1\left(\frac{1}{2}, \frac{1}{2}; k + 1; -\alpha^2\right), \quad (2.37)$$

where $B(x, y) = \frac{\Gamma(x)\Gamma(y)}{\Gamma(x+y)}$ is the beta function and ${}_2F_1$ is the ordinary hypergeometric function. See Appendix A.2 for more details on the Chebyshev expansion and explicit definitions of $T_{n,j}$ and hypergeometric functions ${}_pF_q$. Altogether,

$$c_m = \begin{cases} \frac{1}{\pi} \sum_{k=0}^{m/2} T_{m,2k} B\left(k + \frac{1}{2}, \frac{1}{2}\right) {}_2F_1\left(\frac{1}{2}, \frac{1}{2}; k + 1; -\alpha^2\right), & m \text{ even} \\ \frac{a}{2}(c_{m+1} - c_{m-1}), & m \text{ odd} \end{cases}. \quad (2.38)$$

While this looks quite different from the combination of elliptic integrals in [57], these hypergeometric functions are sums of $K(z)$ and $E(z)$, the complete elliptic integrals of the first and second kinds, each weighted by polynomials in α .

This is guaranteed by the recurrence relation [93]:

$$\begin{aligned} \frac{(\frac{3}{2} - c)^2 z}{(c-1)(c-2)} {}_2F_1\left(\frac{1}{2}, \frac{1}{2}; c; z\right) \\ = (2z-1) {}_2F_1\left(\frac{1}{2}, \frac{1}{2}; c-1; z\right) + (1-z) {}_2F_1\left(\frac{1}{2}, \frac{1}{2}; c-2; z\right) \end{aligned} \quad (2.39)$$

which will always allow us to reduce ${}_2F_1\left(\frac{1}{2}, \frac{1}{2}; k+1; -\alpha^2\right)$ into a nonlinear combination of ${}_2F_1\left(\frac{1}{2}, \frac{1}{2}; 1; -\alpha^2\right) = \frac{2}{\pi}K(-\alpha^2)$ and ${}_2F_1\left(\frac{1}{2}, \frac{1}{2}; 2; -\alpha^2\right) = \frac{4}{\pi\alpha^2}((\alpha^2+1)K(-\alpha^2) - E(-\alpha^2))$. Some of the lowest order diffraction coefficients, written in terms of the elliptic integrals, are

$$c_0 = \frac{2}{\pi}K(-\alpha^2) \quad (2.40)$$

$$c_1 = \frac{2}{\pi\alpha} (K(-\alpha^2) - E(-\alpha^2)) \quad (2.41)$$

$$c_2 = \frac{2}{\pi\alpha^2} ((\alpha^2+2)K(-\alpha^2) - 2E(-\alpha^2)) \quad (2.42)$$

$$c_3 = \frac{2}{3\pi\alpha^3} ((5\alpha^2+8)K(-\alpha^2) - (\alpha^2+8)E(-\alpha^2)). \quad (2.43)$$

Note c_0 and c_1 here match Eq. (17) of [57].

A comment on the phases of c_0 and $c_{\pm 1}$: an even form of this diffraction profile, $\arctan(\alpha \cos x)$, was derived in [55]. An intuitive argument was provided therein for why the phases $(\arg(c_{-1}), \arg(c_0), \arg(c_{+1})) = (\frac{\pi}{2}, 0, \frac{\pi}{2})$, based on an argument first presented by Gori in [94]. A proof of this statement is provided in the Appendix of [95] and measured in [96]. However, the original diffraction profile used here from [57] is odd. In their Eq. (6), they set $c_{-1} = -c_1$, so unless $c_{\pm 1} = 0$, the phases of $c_{\pm 1}$ cannot be equal. Diffraction from the odd profile has the same amplitude as from the even one, but the phases are instead $(\arg(c_{-1}), \arg(c_0), \arg(c_{+1})) = (\pi, 0, 0)$.

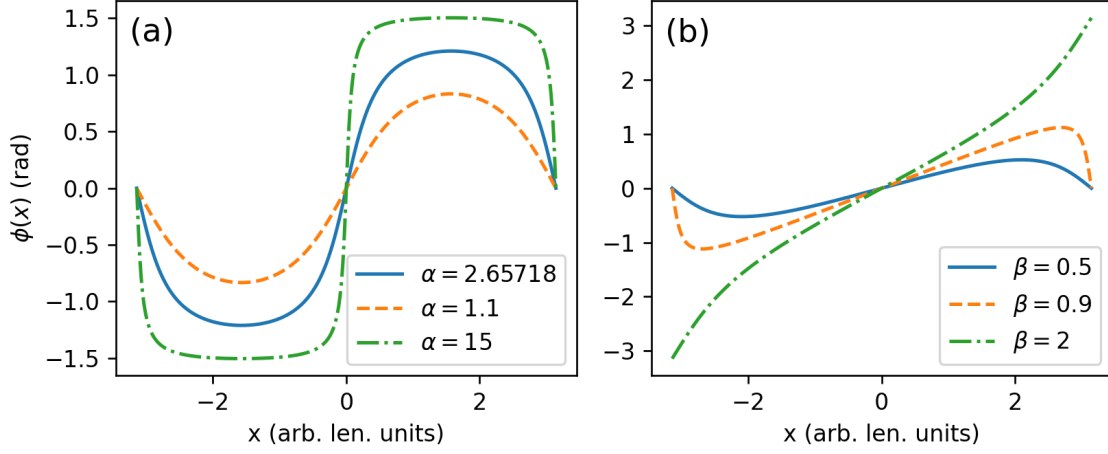


Figure 11. Profiles of the optimum triplicator and variable duplicator. (a) shows phase profiles of the optimum triplicator for three choices of parameter α : 2.65718, 1.1, and 15. (b) depicts phase profiles of the variable duplicator for three choices of parameter β : 0.5, 0.9, and 2.

2.9 Optimal Duplicator

[58] consider the problem of diffracting light into the 0th and +1st orders with the prescribed ratio $\eta_0 = r\eta_1$. Their derived phase profile is

$$\phi_2(x, \beta) = \arctan \left[\frac{\beta \sin(x)}{1 + \beta \cos(x)} \right]. \quad (2.44)$$

Fig. 11 shows this phase profile for three values of β . Their diffraction patterns are shown in the second row of Fig. 12. The ratio r as a function of β is

$$r = \frac{1}{\beta^2} \frac{(1 + \beta)E(\gamma) - (1 - \beta)K(\gamma)}{(1 + \beta)E(\gamma) + (1 - \beta)K(\gamma)} \quad (2.45)$$

where $\gamma = -\frac{4\beta}{(\beta-1)^2}$.

Plugging $\phi_2(x)$ into Eq. (2.5) gives

$$c_m = \frac{1}{\pi} \int_0^\pi \cos(\phi_2(x)) \cos mxdx + \frac{1}{\pi} \int_0^\pi \sin(\phi_2(x)) \sin mxdx \quad (2.46)$$

The arctangent argument is more complicated here, but following the same procedure as above, while simultaneously pulling a factor of $(1 + \beta \cos x)^{-1}$ out of the denominator, turns the integrand into

$$\frac{(1 + \beta \cos x) \cos mx}{\sqrt{1 + \beta^2 + 2\beta \cos x}} + \frac{\beta \sin x \sin mx}{\sqrt{1 + \beta^2 + 2\beta \cos x}}. \quad (2.47)$$

The last two terms can be simplified with a angle-sum trigonometric identity so

$$c_m = \frac{1}{\pi} \int_0^\pi \frac{\cos mx}{\sqrt{1 + \beta^2 + 2\beta \cos x}} + \beta \frac{\cos(m-1)x}{\sqrt{1 + \beta^2 + 2\beta \cos x}} dx. \quad (2.48)$$

Thus, the diffraction efficiencies of the variable beamsplitter are

$$c_m = I_m(\beta) + \beta I_{m-1}(\beta), \quad I_m(\beta) = \frac{1}{\pi} \int_0^\pi \frac{\cos mx}{\sqrt{1 + \beta^2 + 2\beta \cos x}} dx, \quad \beta \neq 1. \quad (2.49)$$

Substituting $m = 0$ and $m = 1$ produces Eqs. (21) and (22) of [58]. $I_m(1)$ does diverge, but simplifying $\phi_2(x, \beta = 1) = \frac{1}{2}x$, $x \in (-\pi, \pi)$ before plugging ϕ_2 into Eq. (2.2) gives the proper diffraction coefficients. I_m is symmetric in m , or $I_m(\beta) = I_{-m}(\beta)$, so for the rest of the section, we assume $m \geq 0$.

Mathematica is able to calculate I_m [86], in which a hypergeometric function does again appear.

$$I_m = \frac{1}{|\beta - 1| \Gamma(1 - m) \Gamma(m + 1)} {}_3F_2 \left(\frac{1}{2}, \frac{1}{2}, 1; 1 - m, 1 + m; -\frac{4\beta}{(\beta - 1)^2} \right). \quad (2.50)$$

This can be reduced to the more standard hypergeometric function ${}_2F_1$:

$$I_m = \frac{\Gamma(m + 1/2)}{\sqrt{\pi} m! |\beta - 1|} \left(\frac{-\beta}{(\beta - 1)^2} \right)^m {}_2F_1 \left(m + \frac{1}{2}, m + \frac{1}{2}; 2m + 1; -\frac{4\beta}{(\beta - 1)^2} \right) \quad (2.51)$$

In computation, it is useful to use $|m|$ (and $|m - 1|$ for I_{m-1}) for all instances of m in this formula. Like above, the hypergeometric function concisely writes a polynomial weighting of elliptic integrals of the first and second kind. The more complicated argument $\gamma = -\frac{4\beta}{(\beta-1)^2}$ arises from building the denominator of I_m

$$\begin{aligned} K\left(-\frac{4\beta}{(\beta-1)^2}\right) &= \frac{|\beta-1|}{2} \int_0^\pi \frac{1}{\sqrt{1+\beta^2+2\beta\cos\theta}} d\theta \\ E\left(-\frac{4\beta}{(\beta-1)^2}\right) &= \frac{1}{2|\beta-1|} \int_0^\pi \frac{1+\beta^2+2\beta\cos\theta}{\sqrt{1+\beta^2+2\beta\cos\theta}} d\theta. \end{aligned}$$

The remaining components of Eqs. (2.50) and (2.51) balance the terms in the numerator such that the sum of $K(\gamma)$ and $E(\gamma)$ has a numerator of $\cos mx$.

The full explicit forms of the lower order diffraction efficiencies for $\beta \neq 1$ are

$$c_{-2} = \frac{1}{15\pi\beta^2|\beta-1|} [(\beta-1)^2(8\beta^4-3\beta^2-2)E(\gamma) + (-8\beta^6+3\beta^4+3\beta^2+2)K(\gamma)] \quad (2.52)$$

$$c_{-1} = \frac{1}{3\pi\beta|\beta-1|} [-(2\beta^2-1)(\beta-1)^2E(\gamma) + (2\beta^4-\beta^2-1)K(\gamma)] \quad (2.53)$$

$$c_0 = \frac{\text{sgn}(\beta-1)}{\pi} [(\beta-1)E(\gamma) - (\beta+1)K(\gamma)] \quad (2.54)$$

$$c_1 = \frac{\text{sgn}(\beta-1)}{\pi\beta} [(\beta-1)E(\gamma) + (\beta+1)K(\gamma)] \quad (2.55)$$

$$c_2 = \frac{1}{3\pi\beta^2|\beta-1|} [(\beta^2-2)(\beta-1)^2E(\gamma) - (\beta^4+\beta^2-2)K(\gamma)] \quad (2.56)$$

$$c_3 = \frac{1}{15\pi\beta^3|\beta-1|} [-(\beta-1)^2(2\beta^4+3\beta^2-8)E(\gamma) + (2\beta^6+3\beta^4+3\beta^2-8)K(\gamma)] \quad (2.57)$$

When $\beta = 1$,

$$c_m(\beta = 1) = \frac{1}{2\pi} \int_{-\pi}^\pi e^{i(m-1/2)x} dx = -\frac{2\cos(mx)}{(2m-1)\pi} = (-1)^{m+1} \frac{2}{(2m-1)\pi}. \quad (2.58)$$

It is noted in [58] that $|c_0(\beta)|^2 = |c_1(\beta^{-1})|^2$. In fact, this identity generalizes to all diffraction coefficients, not just diffraction efficiencies: $c_n(\beta) = c_{1-n}(\beta^{-1})$.

Taking $\beta \rightarrow \beta^{-1}$ in I_m ,

$$I_m(\beta^{-1}) = \frac{1}{\pi} \int_0^\pi \frac{\cos mx}{\sqrt{1 + \beta^{-2} + 2\beta^{-1} \cos x}} = \frac{\beta}{\pi} \int_0^\pi \frac{\cos mx}{\sqrt{\beta^2 + 1 + 2\beta \cos x}} = \beta I_m(\beta). \quad (2.59)$$

Then

$$c_{1-n}(\beta^{-1}) = I_{1-n}(\beta^{-1}) + \beta^{-1} I_{1-n-1}(\beta^{-1}) = \beta I_{n-1} + I_n = c_n(\beta). \quad (2.60)$$

The Fourier coefficients (amplitude and phase), not just the diffraction efficiencies (amplitude only), have this symmetry in β and m .

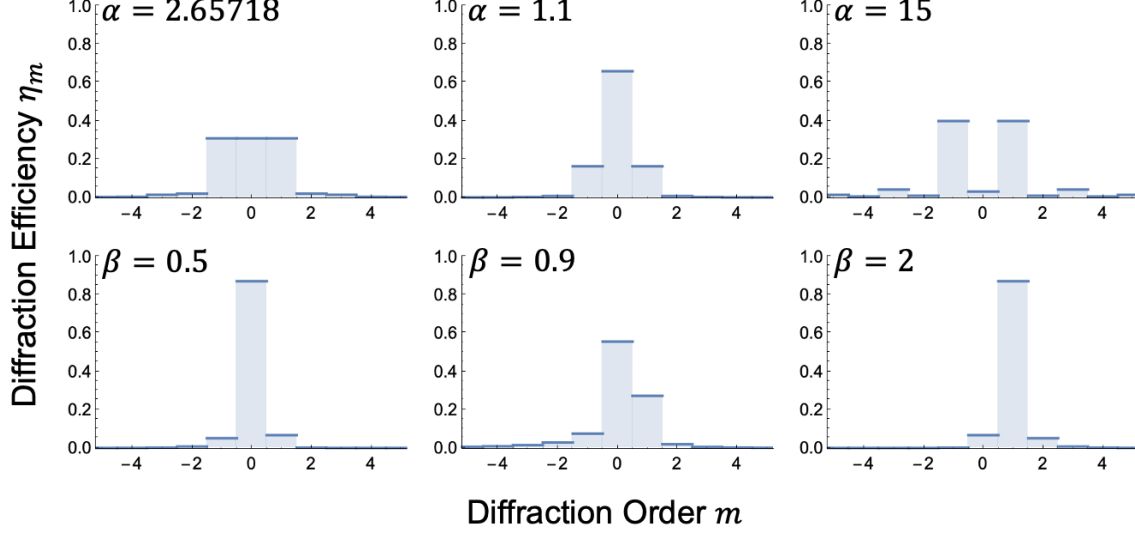


Figure 12. Analytically calculated diffraction efficiencies of the six phase profiles presented in Fig. 11. The first and second rows show diffraction from the optimal triplicator and optimal duplicator, respectively.

2.10 Design Considerations

Conventional grating profiles, including the full- and half-wave and parabolic profiles we considered before, are typically defined in terms of their maximum depth d or amplitude A . The optimal triplicator and duplicator are different. The total phase delay, critical for fabrication, is only implicitly defined by α and β . Using elementary calculus, we can find the values of $\phi_3(x)$ and $\phi_2(x)$ at their extrema to calculate the peak-to-peak amplitude $A_3(\alpha)$ and $A_2(\beta)$ of both gratings.

$$A_3(\alpha) = 2 \arctan \alpha \quad (2.61)$$

$$A_2(\beta) = \begin{cases} 2 \arctan \left(\frac{\beta}{\sqrt{1-\beta^2}} \right), & 0 \leq \beta \leq 1 \\ 2\pi, & \beta \geq 1 \end{cases} \quad (2.62)$$

We plot these amplitudes in Fig 13. The amplitude of both gratings is roughly linear for $\alpha, \beta < \frac{1}{2}$. The required maximum phase delay for the true optimum triplicator is $2 \arctan(2.65718) = 2.4217$ radians. For large α , the amplitude A_2 asymptotes to π . The variable duplicator instead rapidly grows to an amplitude of π for $\beta = 1$ before discontinuously jumping to a constant phase delay of 2π .

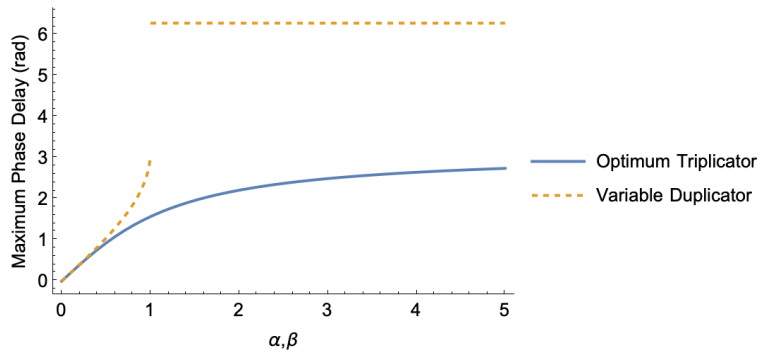


Figure 13. Peak-to-peak amplitude of optimal triplicator and duplicator phase profiles as a function of α and β .

What of the effect of increasing α and β on the diffraction patterns themselves? Shifting the optimum triplicator parameter α from 0 up to ∞ is essentially performing a smooth mapping from a horizontal line to a square wave of peak-to-peak amplitude π , as depicted in Fig. 13. As such, the diffraction pattern transitions from 100% efficiency in the 0th order to the binary phase grating pattern with diffraction efficiencies

$$\text{sinc}^2\left(\frac{m\pi}{2}\right) = \begin{cases} \frac{4}{m^2\pi^2}, & m \text{ odd} \\ 0, & m \text{ even} \end{cases}. \quad (2.63)$$

For low values of α , even coefficients grow in intensity, but are forced to zero in the binary grating limit, so even-order triplicator diffraction efficiencies first increase to a global maximum, then decrease monotonically. The lone exception is the 0th order efficiency η_0 , which is purely monotonically decreasing from $\alpha = 0$. Odd triplicator orders instead increase monotonically from 0 to their limit of the corresponding binary grating order intensity.

The optical response of the triplicator is dominated by the optimized orders, yet some non-negligible diffraction orders remain. There are six diffraction orders which have over 1% efficiency at some value of α . They are the ± 2 nd, ± 3 rd, and ± 5 th order diffraction orders and are depicted in Fig. 14a. Recall this diffraction pattern is symmetric in m , so diffraction at negative orders exist with the same intensity. The ± 3 rd and ± 5 th orders asymptote to $\frac{4}{9\pi^2} = 4.503...\%$ and $\frac{4}{25\pi^2} = 1.621...\%$ while the ± 2 nd diffraction order peaks at $2.002...\%$ for $\alpha = 3.564...$ before falling away to zero. To better understand how non-negligible these diffraction orders are, we include a shaded region in Fig. 14a whose boundary is 10% of the brightest diffraction order for a given α . In other words, the unshaded

portion of the plot contains diffraction efficiencies a lower order of magnitude than the largest diffraction efficiency. The ± 3 rd orders are the only ones to reach this shaded regime, and only for high α .

Of all the grating designs considered in this chapter, the variable duplicator is the most likely to be useful for STEM holography, as producing two beams of equal intensity would be ideal for interferometry. However, the optimal duplicator directs only 81.06% of incident light into the 0th and +1st orders ($|c_0(\beta = 1)|^2 = |c_{+1}(\beta = 1)|^2 = \frac{4}{\pi^2} \approx 0.4053$). Coincidentally, this is the exact same amount of light the binary phase grating directs into the ± 1 st orders. Phase gratings simply are unable to direct the vast majority of light into two equal beams without a significant portion leaching into neighboring diffraction orders.

These neighboring parasitic orders have one global maximum in the vicinity of $\beta = 1$: negative duplicator orders have their maxima between $0 < \beta < 1$, and positive orders have their maxima for $\beta > 1$. By the dint of $c_n(\beta) = c_{1-n}(\beta^{-1})$, each order has a near-mirror that, together, form an angel-wing-like pair visible in Fig. 14b. Duplicator orders with $-3 \leq m \leq 4$ have efficiencies above 1% for some β , but because either η_0 or η_1 rapidly grow large, only η_{-1} or η_{+2} are the same order of magnitude as the brightest diffraction order, and only for a small region around $\beta = 1$.

2.11 Conclusion

We have presented five one-dimensional phase grating thickness profiles—the half-wave and full-wave rectified sine waves, the periodic extension of the parabola, and the optimal triplicator and duplicator—which have an analytical efficiency for every diffraction order. To our knowledge, these are the first such gratings since the 1990s. These have immediate application as triplicate beamsplitters, but more

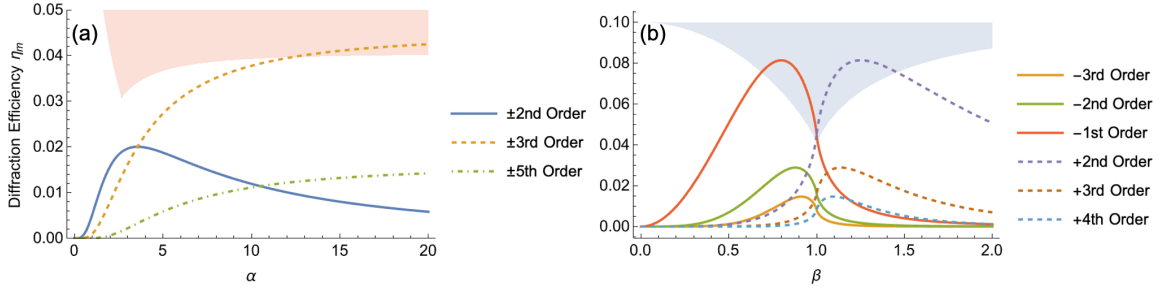


Figure 14. Parasitic diffraction orders with over 1% intensity for some value of the parameters α or β . (a) plots the three parasitic orders for the optimum triplicator and (b) plots the six for the variable duplicator. The boundary of the shaded regions is one-tenth of the largest diffraction efficiency for a given α or β .

importantly, we hope these patterns spark further analytical investigation into the basic 1D phase grating scalar theory.

CHAPTER III

CONVENTIONAL SCANNING TRANSMISSION ELECTRON MICROSCOPY HOLOGRAPHY (STEMH)

The magnetic STEMH section of this chapter is adapted from an unpublished manuscript cowritten by Andrew Ducharme and William Parker. The nanomagnetic Fe/Gd sample was prepared by Sergio Montoya and Eric Fullerton at the University of California, San Diego. Fehmi Yasin, William Parker, and Andrew Ducharme collected the magnetic STEMH data. WP collected the Lorentz TEM data. Frederik Otto at the Technical University of Berlin fabricated the GaN lamella in Fig. 25. The rest of the chapter is original work.

3.1 Introduction

Electron interferometry is identical in principle to photon interferometry, but often lags decades or even centuries behind in implementation due to the lack of electron optical elements. The picometer-scale wavelength of fast electrons and their strong coupling to electromagnetic fields, make the reward for overcoming this technical challenge—atomic-resolution interferometry—very enticing. Without mirrors or beamsplitters, the first realization of an electron interferometer used three copper crystals to divide the amplitude of the electron beam [97]. However, the historically predominant technique of this type is electron holography [98, 99], where an electrostatically charged wire, or electron biprism, splits a parallel electron beam in a transmission electron microscope (TEM) post-specimen and creates an interference pattern at the detector. Electron holography has been used to measure the absolute thickness of a sample [25], map dopants and the electrostatic potentials in semiconductors and p-n junctions [100, 101], and observe nanomagnetic phenomena such as skyrmions [102], magnetic stripe domains [103],

and even the magnetic field of individual atomic lattice planes [14]. Despite its success, TEM holography has been hampered by the need for an electron beam spatially coherent at roughly the length scale of the beamsplitter, in this case, the biprism wire width, which is on the order of individual microns. This requires more expensive field-emission electron sources [99, 104]. The biprism additionally causes Fresnel diffraction that introduces imaging artifacts in the phase reconstruction. This can be addressed by adding one or more biprisms throughout the microscope column, albeit with increasing experimental complexity and microscope customization.

In this chapter, we discuss an alternative realization of electron holography which instead splits the beam into distinct focused probes at the sample plane by inserting a nanofabricated diffraction grating into the electron beam prior to the sample. These probes are scanned in a raster pattern across the sample using Scanning Transmission Electron Microscopy, or STEM. At each stop in the two-dimensional raster, we record a two-dimensional image of the interference of the diffraction of the various probes in the far field of the sample. We can analyze each interference pattern to extract a phase map of the scanned sample plane in post-processing. This technique, first demonstrated in 2018 by the McMorran group [21, 31], is called Scanning Transmission Electron Microscopy Holography (STEMH). Fig. 15 shows a schematic of this experimental geometry. STEMH capitalizes on the better high-resolution performance of STEM over TEM while lowering the coherence requirements on the source by using a grating beamsplitter instead of a biprism.

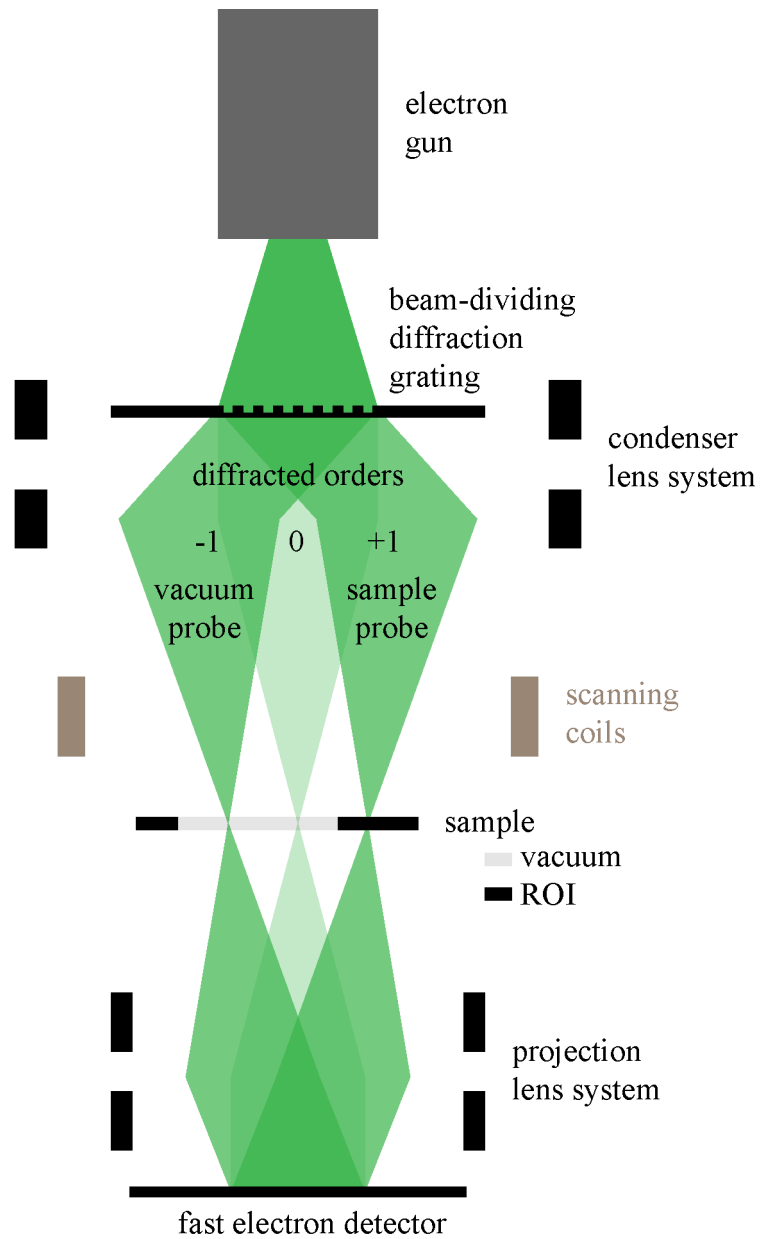


Figure 15. Schematic of the STEMH setup. A beam-dividing diffraction grating is placed into the second condenser aperture of the microscope, splitting the beam into multiple diffraction orders. Its depth profile is designed so that the majority of beam intensity is split between the +1 and -1 orders, which pass through the sample and vacuum, respectively. The projection system then recombines the probes onto the detector, forming an image of the grating modulated by the relative phase acquired by the sample probe as it passes through the sample.

3.2 Experimental Considerations

The distinctive piece of STEMH hardware is the diffraction grating. We use focused ion beam (FIB) milling to nanofabricate phase gratings in off-the-shelf silicon nitride membrane windows typically used as electron-transparent sample substrates [91]. Fig. 16a shows an example of a grating-covered SiN membrane ready to be slotted into a TEM. We coat the backside of such a membrane with an adhesion layer of Ti and charge alleviation layer of Pt via e-beam evaporation. The uncoated top side is then milled with a Ga^+ beam FIB-SEM at very low ion beam currents (typically between 7-25 pA) to prevent the splitting of the SiN membrane. We flow XeF_2 over the active area to accelerate milling and limit Ga implantation. Since we cannot rotate the diffraction pattern or the gratings in the TEM, we fabricate four copies of each grating at relative rotations of 0° , 45° , 90° , and 135° . Fig. 16b shows a micrograph of an individual phase grating.

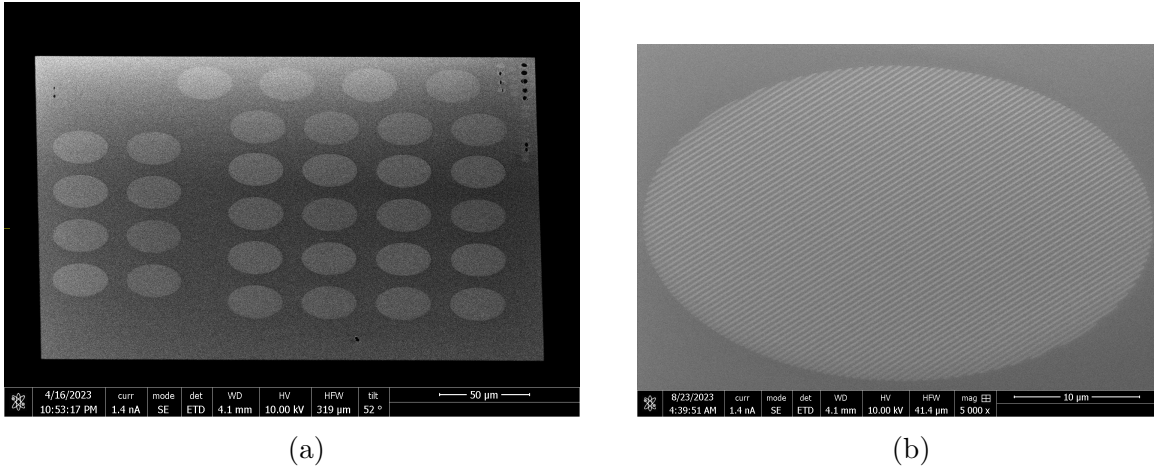


Figure 16. Scanning electron microscope micrographs of nanofabricated diffraction grating beamsplitters for STEMH. (a) A silicon nitride membrane full of gratings of slightly varying periods and mill depths to place in the condenser aperture of a TEM. (b) A single nanofabricated grating with period of 500 nm with grooves oriented at a 45° angle.

As detailed in the last chapter, the relative intensity of the probes is determined by the shape of the milled grating profile and the maximum mill depth. We use a square-wave profile with a maximum phase shift of π radians and period between 100-500 nm. The SiN maximum phase shift $\phi = \sigma V_{\text{MIP}} d$, where σ (units of radians per volt-nanometer) is an acceleration-voltage-dependent coefficient called the interaction parameter, V_{MIP} (units of volts) is the mean inner potential of the grating material, and d is the maximum mill depth (units of nanometers). The interaction parameter

$$\sigma(E) = \frac{2\pi e}{\lambda(E)E} \frac{E + m_e c^2}{E + 2m_e c^2} \quad (3.1)$$

where e is the electron charge, c is the speed of light in vacuum, h is Planck's constant, m_e is the electron rest mass, $\lambda(E)$ is the relativistic electron wavelength, and E is the accelerating voltage of the TEM. Stoichiometric SiN, Si_3N_4 , has a measured mean inner potential of 16.7 V [90], but commercial SiN membranes use a different stoichiometry that lessens the mechanical stress in the membrane and the chances of the membrane breaking [91]. In our experience with SiN membranes from vendors Norcada, Silson, and SPI, the mean inner potential for this low-stress stoichiometry is 20% lower than the measured Si_3N_4 value. Table 3 compiles the necessary mill depth to achieve a π phase shift with low-stress SiN.

Accelerating Voltage (keV)	σ (rad / V·nm)	Mill Depth (nm)
80	0.0101	23.2
200	0.0073	32.1
300	0.0065	36.1

Table 3. Necessary mill depth in low-stress SiN to impose a π phase shift on a transiting electron at varying accelerating voltages.

Assuming the SiN is a pure phase object, this phase grating creates several discrete probes at the sample plane with relative intensities

$$|c_m|^2 = \text{sinc}^2\left(\frac{m\pi}{2}\right) = \begin{cases} \frac{4}{m^2\pi^2}, & m \text{ odd} \\ 0, & m \text{ even} \end{cases}. \quad (3.2)$$

The π phase shift minimizes the even m order intensities, so the operative beam separation is the distance between the ± 1 orders, which is exactly twice the distance between neighboring orders. However, the SiN does scatter away or absorb some incident electrons, ruining the pure phase object assumption. Our group has previously measured the electron loss per nanometer of low-stress SiN as $\gamma = 0.008 \text{ nm}^{-1}$ [80]. The consequence of this amplitude-scattering can be seen in Fig. 17. The 0th order beam is greatly reduced in intensity, but in general, even orders still exist. Concerningly, the ± 2 and ± 3 orders are the same order of magnitude as the ± 1 st orders, which will lead to prominent ghost images.

We now turn to the electron optics. We perform STEMH in three-condenser TEMs. Commercial TEMs are either two-condenser or three-condenser systems. In either, the first condenser lens (the C1 lens) is always occupied by capturing the electrons coming off the electron source and focusing them to a point, while in normal STEM (single probe) operation, the second condenser lens (the C2 lens) forms the electron probe. In a two-condenser system, the size of the probe is set by the C2 aperture, while the three-condenser system can tune the probe size and dose using the C2 and C3 lenses in coordination. STEMH is far better suited for a three-condenser system not for the additional lens, but for the additional C3 aperture. The grating-filled SiN membrane is placed in the C2 aperture slot. Without an aperture in that plane, the electron microscope illuminates multiple gratings simultaneously. Fig. 18a demonstrates what occurs without a blocking C3 aperture. A standard circular aperture in the C3 aperture slot can be maneuvered

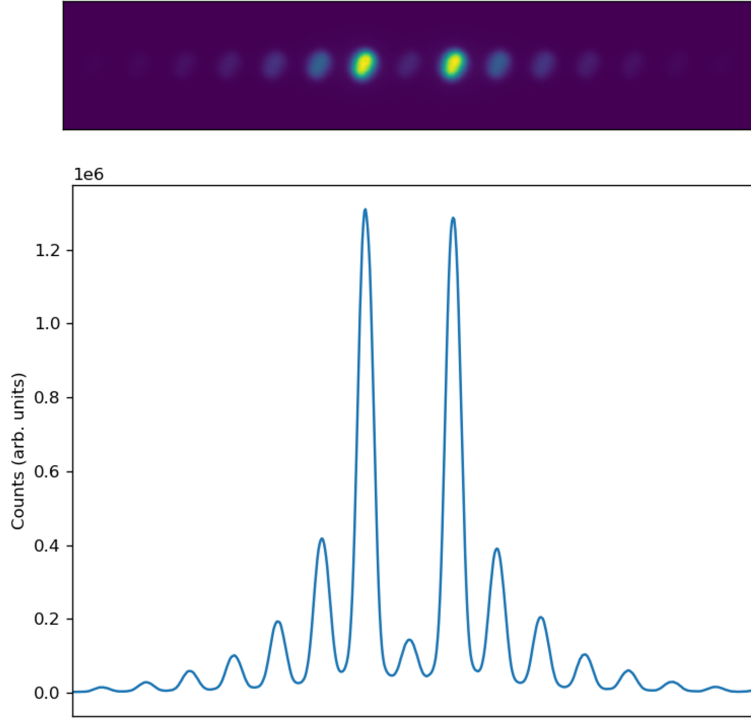


Figure 17. Measured diffraction from a nanofabricated diffractive beamsplitter for STEMH. The top image is the full diffraction pattern, with the total intensity summed along the vertical axis on the bottom.

to select an individual grating from the SiN membrane and prepare the microscope for STEMH operation. Fig. 18b shows a single grating selected by the C3 aperture.

Technically, we could perform STEMH on a two-condenser TEM using a custom membrane-aperture combination that can fit within the C2 aperture slot. With the three-condenser setup, we can mill several dozen gratings of varying parameters on a single off-the-shelf SiN membrane. With the two-condenser setup, we must weld a custom aperture to a custom SiN membrane, with the total number of gratings limited to the number of holes in the custom aperture. It is both easier and provides more flexibility to use a three-condenser system.

To align the microscope for STEMH, we first perform the standard STEM alignment procedure, including tuning the aberration corrector, if the microscope

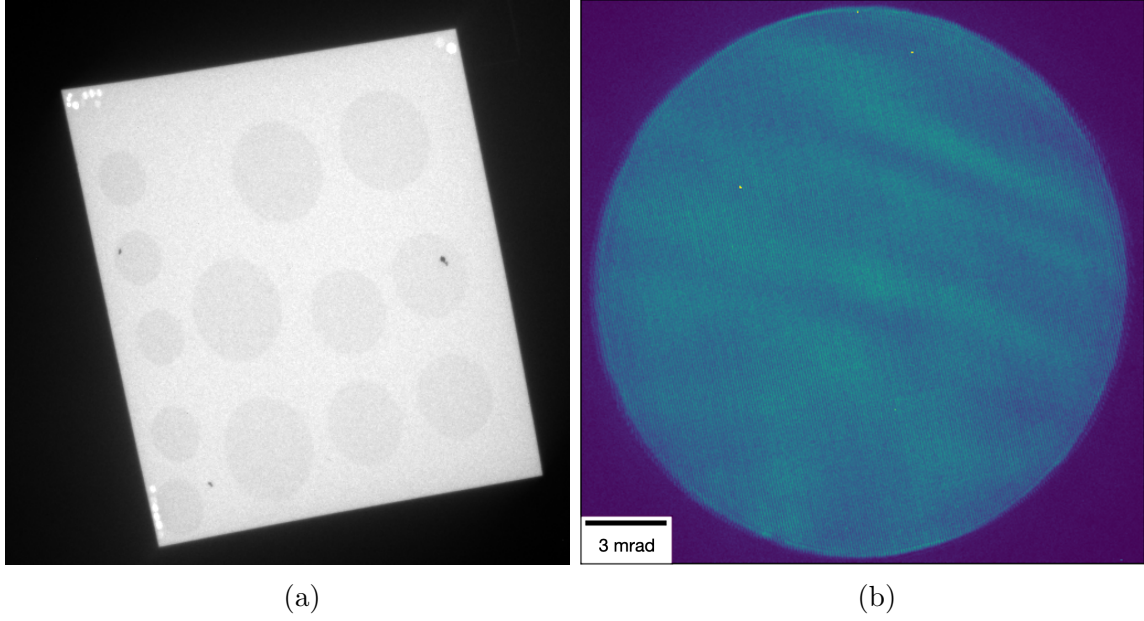


Figure 18. Examples of diffraction gratings in the electron microscope column for STEMH. (a) A projected image of the entire silicon nitride membrane unblocked by the C3 aperture. (b) A single nanofabricated grating selected by the C3 aperture. The grating grooves run roughly vertically.

being used has one. Then, we insert the membrane of gratings and the C3 aperture. We first align the C3 aperture, then a grating, with the optical axis. For sub-nanometer resolution, we then need to perform precise manual tuning of the defocus and A1 and B2 aberrations added by the grating and C3 aperture. These can be diagnosed by analyzing the ronchigram. The ronchigram is the diffraction of a convergent beam transmitted through an amorphous sample and contains features that directly correspond to particular aberrations [105]. We currently believe aberrations are added to the optical system mainly by the C3 aperture, not the SiN membrane. To test this, we captured images of the ronchigram with varying C2 and C3 apertures in an aligned aberration-corrected microscope. Due to the aberration correction, we start with a featureless ronchigram with the 70 micron diameter C2 and 2000 micron diameter C3 apertures inserted. In Fig. 19a,

we show the ronchigram after switching to a 70 micron diameter C2 aperture and 30 micron C3 aperture. There now exists some structure in the ronchigram, hence aberrations have been added to the optical system. In Fig. 19b, we replace the C2 aperture with a grating while keeping the C3 aperture fixed. Although we have lost some signal scattered away by the SiN membrane, the structure within the ronchigram remains similar. Finally, in Fig. 19c, we keep the grating fixed and change to a 20 micron C3 aperture. Again, we see the structure of the ronchigram has changed. Large shifts in the structure of the ronchigram came from changing the C3 aperture, not by inserting the grating. These additional aberrations could potentially be removed by performing the aberration correction procedure with the desired C3 aperture for STEMH already inserted and aligned with the optical axis.

To earn our claim of absolute phase measurement, we must set our field of view near a vacuum-sample interface. In the idealized two-beam STEMH model, we pass a reference beam through vacuum, acquiring no phase, and a probe beam through sample, acquiring the phase of the sample at that scan location. The phase difference between the two probes is then simply the sample phase. (Similar conditions exist if the reference and probe beams are incident on a constant thickness substrate so both beams acquire an approximately identical phase shift, which cancels out.) For this to be roughly true, we are limited to scanning sample regions within a distance equivalent to the beam separation from said interface. This is very constraining for high-resolution STEMH, where the small probe size (large convergence semi-angle α) can correspond to beam separations in the range of 5-25 nm.

Simultaneously, however, STEMH is surprisingly robust once our original reference probe enters the sample. Fig. 20 is a STEMH measurement of a whole

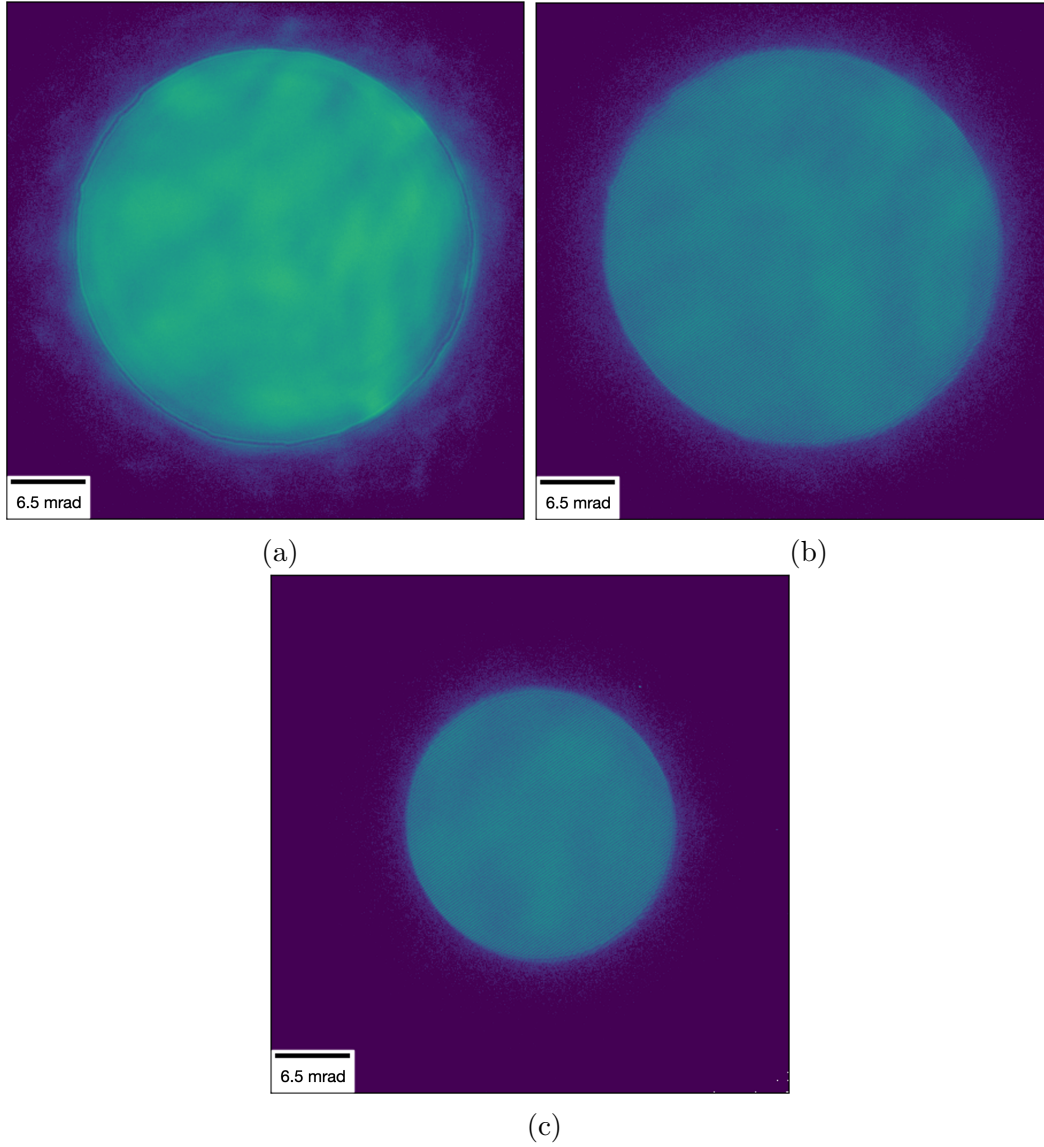


Figure 19. Observing changes in aberrations while varying C2 and C3 apertures. (a) The ronchigram generated with a 70 micron diameter C2 aperture and 30 micron C3 aperture. (b) The ronchigram generated with a grating in C2 and 30 micron C3 aperture. (c) The ronchigram generated with a grating in C2 and 20 micron C3 aperture.

Ag nanocube with edge length 100 nm with beam separation of 10 nm. We could reasonably expect the measured phase at the center of the cube, with several probes passing through the sample, to simply be noise. Instead the phase reconstruction stabilizes to a near-constant value at the center, matching the constant thickness of the sample. There is usable information in the multiple probes. Thus, when setting up a STEMH measurement, we find a vacuum-sample interface and consider the field of view relative to the beam separation, but often do not totally restrict our scans to only the first region in which an electron beam penetrates the sample. As a result, we see ghost images in our reconstructions.

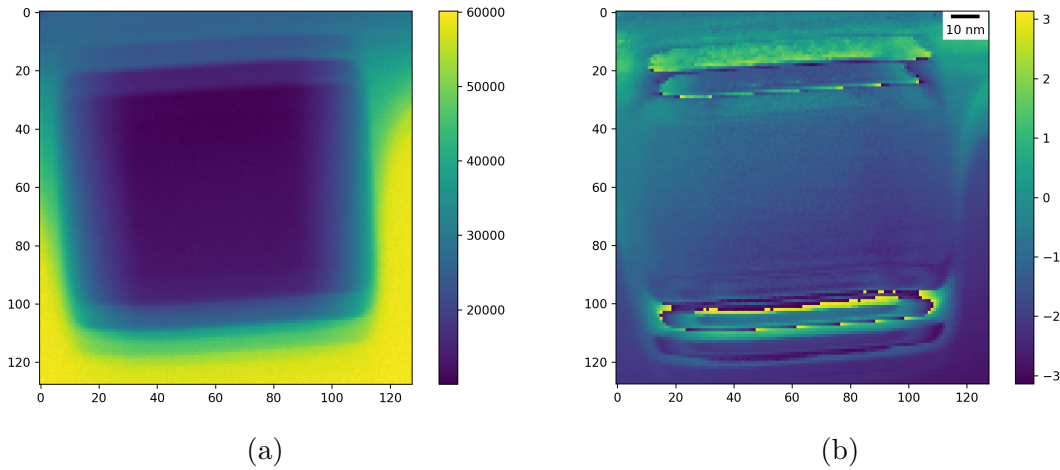


Figure 20. A Scanning Transmission Electron Microscopy Holography (STEMH) measurement of a silver nanocube 100 nm in length. (a) The measured intensity. (b) The reconstructed phase. The scale bar applies identically to both images.

The next consideration is the detector. STEMH requires 4DSTEM detectors, which capture two-dimensional diffraction images at every point in a two-dimensional beam scan (2D scan plus a 2D image equals 4D). Standard STEM measurements record a single value of transmitted current at each beam stop in the scan. This is incompatible with the necessity of capturing the two-dimensional STEMH interference pattern at each beam stop. Called a “universal detector,”

4DSTEM is a very powerful and increasingly common imaging methodology in electron microscopy [106], but is not yet ubiquitous in TEM labs. TEM holography does not need this more expensive detector, a point in its favor. Moreover, STEMH is a pixel-heavy 4DSTEM technique, because we need to sample the interference signal at a rate above the Nyquist frequency, i.e., have at least two pixels devoted to capturing an individual interference fringe. This leads to individual STEMH datasets being tens of gigabytes in size, which take a non-negligible amount of time to transfer between data storage facilities and postprocess.

3.3 Theory and Reconstruction

Before discussing how we translate the STEMH measurement in a phase contrast image, let us consider the basic theory. The TEM illuminates a grating in the C2 aperture with a plane wave and focuses its diffraction at the sample plane as a finite set of symmetric beams $n \in P = \{0, \pm 1, \pm 2, \dots, \pm N\}$. The wavefunction incident on the sample is

$$\psi_i(\mathbf{x}) = \sum_{n=-N}^N c_n a_n(\mathbf{x} - n\mathbf{x}_0). \quad (3.3)$$

Individual beams are uniformly displaced from its neighbors by $\mathbf{x}_0 = \frac{f\lambda}{d}\hat{x}$ where λ is the electron de Broglie wavelength, d is the grating pitch or spatial frequency, and f is the focal length of the probe-forming (STEM) optics. Here, we will assume the beams are approximately delta functions. This is a physically unjustifiable assumption, but allows us to understand the source of the STEMH interference pattern and a key barrier to quantitative STEM holography. For a treatment that considers probe structure, see [21]. Under the Dirac delta assumption, the incident wavefunction is

$$\psi_i(\mathbf{x}) = \sum_n c_n \delta(\mathbf{x} - n\mathbf{x}_0) \quad (3.4)$$

with complex diffraction coefficients $c_n = |c_n|e^{i\theta_n}$. The specimen in the sample plane is described by its transmission or transfer function $t(\mathbf{x}) = A(\mathbf{x})e^{i\phi(\mathbf{x})}$. We make the weak phase approximation that the sample is thin and roughly electron transparent so $A(\mathbf{x}) \approx 1$. Our beams interact with the transfer function, producing $\psi(\mathbf{x}) = t(\mathbf{x})\psi_i(\mathbf{x})$ after the sample. The microscope propagates the wavefunction to the far field, where it can be described as

$$FT[\psi(\mathbf{x})] = \psi(\mathbf{k}) = \int \sum_n c_n \delta(\mathbf{x} - n\mathbf{x}_0) t(\mathbf{x}) e^{-2\pi i \mathbf{k} \cdot \mathbf{x}} d\mathbf{x} \quad (3.5)$$

$$= \sum_n c_n t(\mathbf{x} + n\mathbf{x}_0) e^{-2\pi i \mathbf{k} \cdot (\mathbf{x} + n\mathbf{x}_0)}. \quad (3.6)$$

The 4DSTEM detector placed at the far field plane measures $I(\mathbf{k}) = |\psi(\mathbf{k})|^2$.

Our goal is to get a general form of this intensity for any combination of probes interacting with the sample. We write the transfer function as

$$t(\mathbf{x} + n\mathbf{x}_0) = \left\{ \begin{array}{ll} 1 = e^{i0}, & \text{nth probe off sample} \\ e^{i\phi(\mathbf{x} + n\mathbf{x}_0)}, & \text{nth probe on sample} \end{array} \right\} \equiv e^{i\Phi(\mathbf{x} + n\mathbf{x}_0)} \quad (3.7)$$

so

$$\begin{aligned} I(\mathbf{k}) &= \left| \sum_n c_n t(\mathbf{x} + n\mathbf{x}_0) e^{-2\pi i \mathbf{k} \cdot (\mathbf{x} + n\mathbf{x}_0)} \right|^2 \\ &= \left| \sum_n |c_n| e^{-2\pi i \mathbf{k} \cdot (\mathbf{x} + n\mathbf{x}_0) + i\Phi(\mathbf{x} + n\mathbf{x}_0) + i\theta_n} \right|^2 \end{aligned}$$

We can much more succinctly write the phase of each term as ϕ_n . The product in $I(\mathbf{k})$ is of the form

$$\left(\sum_j |c_j| e^{-i\phi_j} \right) \left(\sum_k |c_k| e^{i\phi_k} \right). \quad (3.8)$$

Consider the product of terms such that j or k is one of m or n .

$$\begin{aligned} & (|c_m| e^{-i\phi_n} + |c_n| e^{-i\phi_m}) (|c_m| e^{i\phi_n} + |c_n| e^{i\phi_m}) \\ &= |c_m|^2 + |c_n|^2 + |c_m c_n| e^{i(\phi_m - \phi_n)} + |c_m c_n| e^{-i(\phi_m - \phi_n)} \\ &= |c_m|^2 + |c_n|^2 + 2|c_m c_n| \cos(\phi_m - \phi_n) \end{aligned} \quad (3.9)$$

There exist one set of terms where the complex exponentials cancel and one set of terms that form a cosine wave. This is the standard interference pattern we get from a (two-beam) Mach-Zehnder interferometer. In the STEMH setup, we have to consider the contribution from every possible combination of integers $(m, n) \in P$, creating the interference pattern

$$\begin{aligned} I(\mathbf{k}) = \sum_n |c_n|^2 + 2 \sum_{\substack{(m,n) \in P \\ m \neq n}} |c_m c_n| \cos[-2\pi \mathbf{k} \cdot (\mathbf{x} + m\mathbf{x}_0) + \Phi(\mathbf{x} + m\mathbf{x}_0) + \theta_m \\ + 2\pi \mathbf{k} \cdot (\mathbf{x} + n\mathbf{x}_0) - \Phi(\mathbf{x} + n\mathbf{x}_0) - \theta_n] \end{aligned} \quad (3.10)$$

The explicit dependence of $\mathbf{x}$ drops out, leaving our final recorded STEM intensity, composed of interference fringes of a frequency an integer multiple of the probe separation:

$$\begin{aligned}
I(\mathbf{k}) = \sum_n |c_n|^2 + 2 \sum_{\substack{(m,n) \in P \\ m \neq n}} |c_m c_n| \cos[2\pi(n-m)\mathbf{k} \cdot \mathbf{x}_0] \\
+ \Phi(\mathbf{x} + m\mathbf{x}_0) - \Phi(\mathbf{x} + n\mathbf{x}_0) + \theta_m - \theta_n].
\end{aligned} \tag{3.11}$$

This N -beam interference pattern is equivalent to the superposition of every possible two-beam Mach-Zehnder interferometer within the N -beam interferometer. In general, we do not consider the possibility that the electron beams have an inherent phase θ_n , and assume that the phase difference $\theta_m - \theta_n = 0$. This is likely incorrect [96], and introduces a constant artifact across the scan. Future work should investigate the effect on reconstructed STEMH phases.

The wave structure of our interference pattern naturally suggests the use of a Fourier transform. Each wave of a discrete frequency maps to a particular point in Fourier space, creating a series of localized peaks

$$\begin{aligned}
FT[I(\mathbf{k})](\mathbf{x}') = \sum_n |c_n|^2 \delta(\mathbf{x}') + \sum_{\substack{(m,n) \in P \\ m \neq n}} |c_m c_n| [e^{i(\Phi(\mathbf{x}+m\mathbf{x}_0)-\Phi(\mathbf{x}+n\mathbf{x}_0))} \delta(\mathbf{x}' - (n-m)\mathbf{x}_0) \\
+ e^{-i(\Phi(\mathbf{x}+m\mathbf{x}_0)-\Phi(\mathbf{x}+n\mathbf{x}_0))} \delta(\mathbf{x}' + (n-m)\mathbf{x}_0)].
\end{aligned} \tag{3.12}$$

The phases of the peaks in the Fourier transform correspond to phase differences across the sample plane.

Eq. 3.12 forms the basis of the STEMH reconstruction algorithm. We take a measured interference pattern like the one depicted in Fig. 21a and compute its fast Fourier transform (FFT). This generates a 2D complex-valued array containing a line of peaks whose phases are related to the phase of the sample at various points across the sample plane. Fig. 21b depicts the amplitude of such a generated

array. We can select the pixel(s) containing a peak in the FFT, as in Fig. 21c, and calculate the peak phase to report as the reconstructed phase. In a two-beam (or approximate two-beam) setup with one reference beam passing through vacuum and acquiring no phase ($\Phi(\mathbf{x} + n\mathbf{x}_0) = 0$) and one probe beam passing through the sample and acquiring phase $\Phi(\mathbf{x} + m\mathbf{x}_0)$, the reconstructed phase would be exactly $\Phi(\mathbf{x} + m\mathbf{x}_0)$.

The catch is that we cannot engineer pure two-beam performance from phase gratings, so we will always have additional interferences muddying the phase signal. From Eq. (3.12), we see the $(n - m)$ th peak contains the sum of all the interferences of any pair of probes $n - m$ orders apart. For example, if $n \in \{-1, 0, +1\}$, Eq. (3.12) reduces to

$$\begin{aligned} \sum_n |c_n|^2 \delta(\mathbf{x}') &+ |c_{-1}c_{+1}| e^{i\Delta\Phi(-1,1)} \delta(\mathbf{x}' - 2\mathbf{x}_0) + |c_{-1}c_{+1}| e^{-i\Delta\Phi(-1,1)} \delta(\mathbf{x}' + 2\mathbf{x}_0) \\ &+ |c_{-1}c_0| e^{i\Delta\Phi(-1,0)} \delta(\mathbf{x}' - \mathbf{x}_0) + |c_0c_{-1}| e^{-i\Delta\Phi(-1,0)} \delta(\mathbf{x}' + \mathbf{x}_0) \\ &+ |c_0c_{+1}| e^{i\Delta\Phi(0,+1)} \delta(\mathbf{x}' - \mathbf{x}_0) + |c_{+1}c_0| e^{-i\Delta\Phi(0,+1)} \delta(\mathbf{x}' + \mathbf{x}_0) \end{aligned} \quad (3.13)$$

where we define the sample plane phase difference $\Delta\Phi(m, n) \equiv \Phi(\mathbf{x} + m\mathbf{x}_0) - \Phi(\mathbf{x} + n\mathbf{x}_0)$. The complex value of the first order peak is $|c_{-1}c_0| e^{i\Delta\Phi(-1,0)} + |c_0c_{+1}| e^{i\Delta\Phi(0,+1)}$, and its phase is

$$\frac{c_{-1} \sin \Delta\Phi(-1, 0) + c_{+1} \sin \Delta\Phi(0, +1)}{c_{-1} \cos \Delta\Phi(-1, 0) + c_{+1} \cos \Delta\Phi(0, +1)}, \quad (3.14)$$

a far cry from simply $\Delta\Phi(0, +1)$.

3.4 Magnetic STEMH

One apt STEMH application is the imaging of nanomagnetic phenomena. Existing TEM-based phase contrast magnetic imaging methods include differential

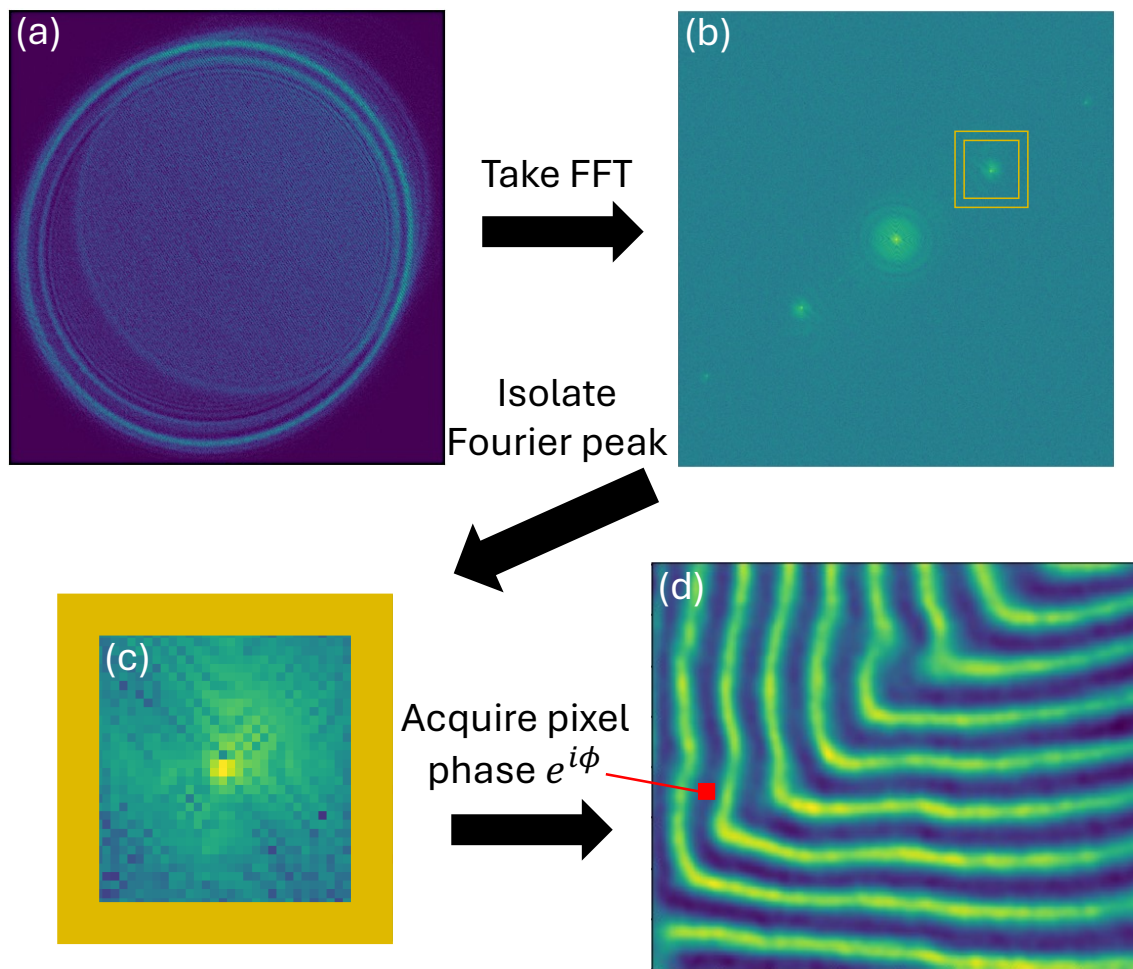


Figure 21. Reconstruction algorithm for each interference pattern captured during the 4DSTEM scan. The interference pattern (a), an individual frame in the 4DSTEM raster, is fast Fourier transformed to produce (b). The interference of the order 0 and +1 probes due to the material phase is contained at peaks closest to the center. We isolate this peak in (c) and acquire the phase of the complex-valued peak. This phase is the value plotted in a particular pixel in (d).

phase contrast (DPC) and Lorentz TEM (LTEM). STEMH has the advantage of directly measuring the Aharonov-Bohm phase of the sample, while DPC measures the gradient of the phase and LTEM forms contrast proportional to the 2D Laplacian of the phase. Additionally, to generate contrast in LTEM, we must defocus the microscope, which inherently limits the resolution. STEMH does not suffer this requirement.

We used STEMH to image stripe domains in a Fe/Gd magnetic multilayer thin film. This experiment was performed on the Transmission Electron Aberration-corrected Microscope (TEAM) I instrument at the National Center for Electron Microscopy (NCEM) at 300 keV operation using the Gatan K3 IS direct electron detector. We used Low Mag STEM mode, where the objective lens of the microscope is disabled and the aberration corrector is not used. Deactivating the objective lens is necessary for imaging magnetic specimens, as its roughly 1 T magnetic field would otherwise overwhelm the magnetic behavior of the specimen. Unfortunately, this greatly reduces the achievable resolution. However, this has the unique benefit for STEMH of greatly expanding the beam separation (hence the field of view) from tens of nanometers to, for this experiment, 2.4 microns. This large beam separation moots the issue of ghost images; since the field of view is smaller than the beam separation, we only have one image copy in the field of view.

Fe/Gd multilayer thin films are of substantial interest due to their ability to host magnetic skyrmions within a tunable range of temperatures and applied fields. 120 layers of alternating Fe and Gd were sputter deposited onto a perforated SiN membrane, with both seed and capping layers of Pt to prevent oxidation. The sample has an out-of-plane easy axis, with stripe or labyrinthian magnetic domains at remanence, separated by domain walls with a strong Bloch component.

Depending on the orientation of neighboring domain walls and the sequence of magnetic field applied prior to imaging, the magnetic domain widths are between 100-200 nm. The SiN substrate is perforated by a grid of 30 micron diameter holes, leaving ample room for the vacuum region probes to pass through.

The probe passing through the sample acquires a phase shift

$$\begin{aligned}\phi(\mathbf{r}_\perp) &= C_E \int_L V(\mathbf{r}_\perp, z) dz - \frac{\pi}{\Phi_0} \int_L \mathbf{A}(\mathbf{r}_\perp, z) \cdot d\mathbf{r} \\ &= \phi_e + \phi_m\end{aligned}\tag{3.15}$$

due to the Aharonov-Bohm effect, with L the electron's path, $\mathbf{r}_\perp$ the coordinates transverse to the optical axis, C_E the interaction constant and Φ_0 the magnetic flux quantum. ϕ_e and ϕ_m are the contributions from electric and magnetic fields, respectively. Typically the two components are isolated by flipping the sample, which will flip the sign of the magnetic component but not the electric. However, for a relatively uniform thickness sample such as ours, the electric component adds only a constant phase related to the mean inner potential to the probe, and can therefore be ignored.

From the magnetic phase map ϕ_m , the in-plane magnetic induction $\mathbf{B}(\mathbf{r}_\perp)$ can be calculated using the relation

$$\nabla_\perp \phi_m(\mathbf{r}_\perp) = \frac{-e\tau}{\hbar} \mathbf{B}(\mathbf{r}_\perp) \times \hat{\mathbf{z}}\tag{3.16}$$

with $\hat{\mathbf{z}}$ the vector parallel to the electron beam's propagation. Note that from the above relation, the component of magnetic field parallel to the electron beam trajectory does not contribute to signal. This means that only in-plane magnetization is visible – in our case, these are the domain walls rather than the domains themselves. This is a general limitation of magnetic imaging using fast electrons; X-ray magnetic imaging techniques such as XMCD complement electron

microscopy by being sensitive only to the parallel component of magnetization. In both areas, tomography may be used to gain a full picture of magnetization, at the cost of experimental complexity. In addition, the signal is integrated in z , and stray fields can therefore have a non-negligible effect, making a direct link between magnetic induction and magnetization difficult to establish.

Fig. 22a shows the STEMH-acquired phase map, which depicts striped magnetic domains with a bend. See Appendix B for further discussion of the image analysis pipeline. Fig. 22b shows the reconstructed magnetic induction. In these materials, remanence configurations are typically somewhere between straight stripe domains and labyrinthian domains, and depend on the exact field sequence applied prior to imaging. In this case, we set the microscope’s objective lens to 10% strength, immersing the sample in a roughly 330mT applied field, which should saturate it, before decreasing the applied field to remanence. Effects due to the edge of the vacuum region of the sample may also contribute to the domains’ sharp change of direction.

We quantified domain wall spacing for comparison with Lorentz TEM data by measuring the average distance between domain walls. Figs. 22c and d show horizontal and vertical line scans of the in-plane magnetization $|B_{\perp}|^2$, respectively. The distance between neighboring domain walls was measured to be $98 \pm 21.67\text{nm}$ for the horizontal line scan and $100 \pm 27.57\text{nm}$ for the vertical line scan. Fig. 23 shows a Lorentz TEM image of an identical sample, again with a. the acquired phase, b. the reconstructed induction, and c. a vertical line scan of in-plane magnetization shown. Domain wall spacing was measured in the same way, giving an average spacing of $99 \pm 15.67\text{nm}$ between domain walls. Note that the LTEM image shows a mixture of chiral and achiral domains (that is, some

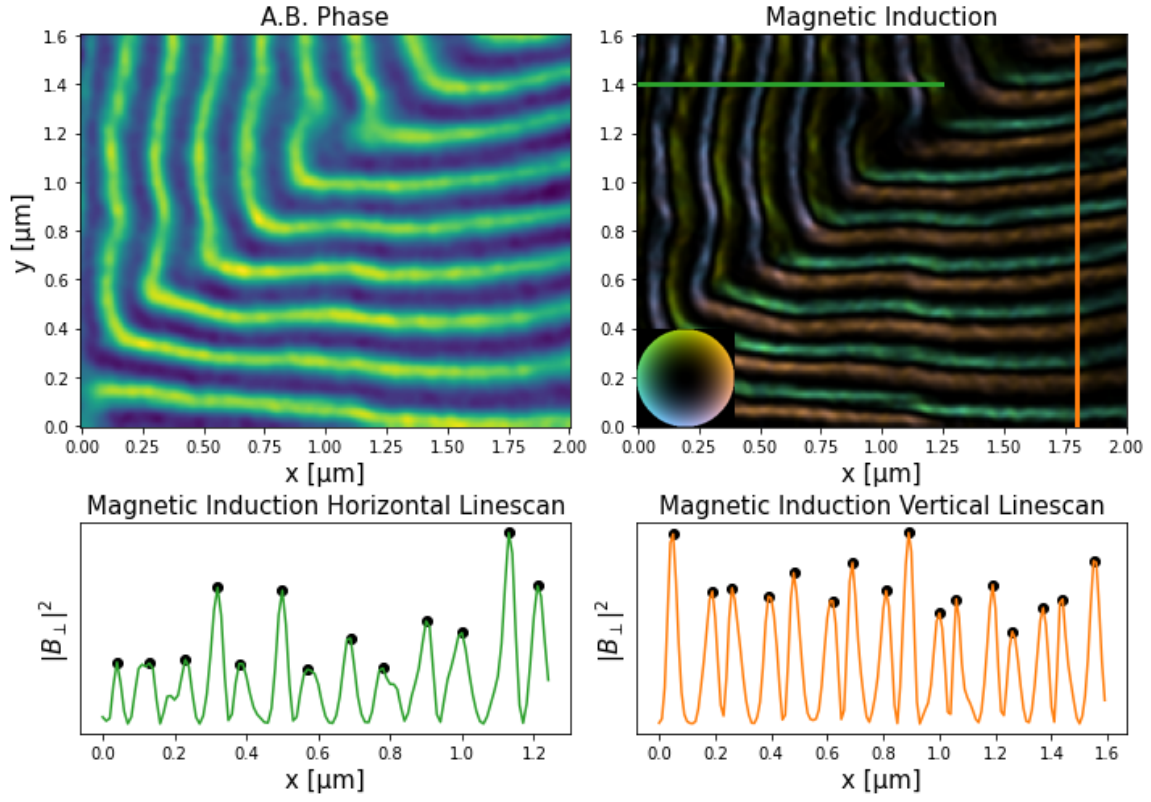


Figure 22. (a) The measured phase, (b) the reconstructed magnetic induction, and (c) and (d) horizontal and vertical line scans of the in-plane magnetic induction, respectively, as measured by STEM holography in a field-free condition.

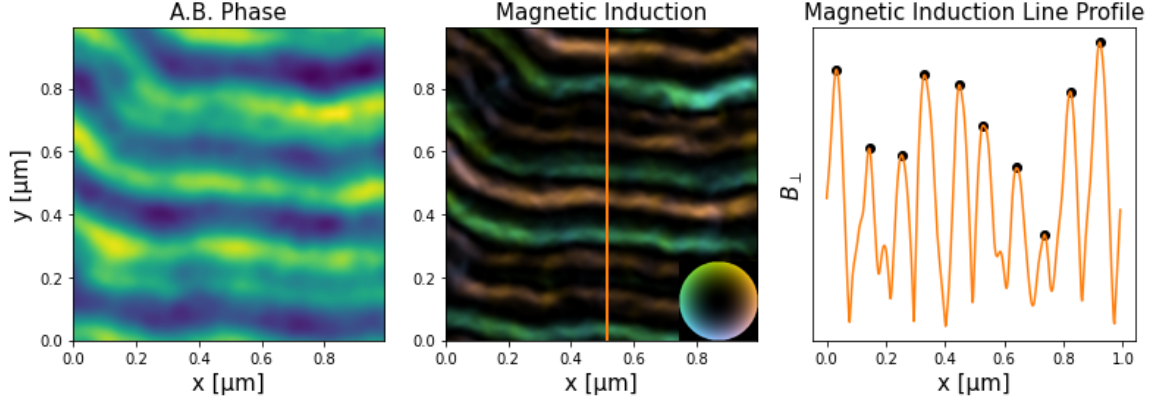


Figure 23. (a) The measured phase, (b) the reconstructed magnetic induction, and (c) a vertical line scan of the in-plane magnetic induction, as measured by Lorentz TEM.

adjacent domain walls are aligned and some are anti-aligned) while the STEMH image shows purely chiral domains. Both of these are common remanence states for Fe/Gd multilayer thin films.

Turning off the objective lens, as is needed for nanomagnetic measurements, pushes ghost images outside the field of view and showcases an interpretable STEMH phase reconstruction. We do not yet trust the quantitative value of the reconstructed phases, but by comparing them to the LTEM results, we can trust the qualitative reconstructed STEMH phase as a reasonable representation of reality.

3.5 High resolution STEMH

Nonetheless, if we want to probe samples at atomic resolution, or in other words, have STEMH be a credible technique, STEMH must be capable of generating interpretable images with the objective lens activated and the beam separation limited to tens of nanometers in length. This is achievable in the case of samples smaller than the field of view. Fig. 24 shows a STEMH reconstruction of a gold nanoparticle roughly 5 nm in diameter using a beam separation of 8.8 nm.

This experiment was performed on the TEAM I at 300 keV using the aberration corrector and K3 direct electron detector. Ghost images are visible in the top left and bottom right of the field of view, but are distinct from the main reconstruction. Of note in the reconstructed nanoparticle are the near-vertical fringes. These lattice fringes are not locations of the atomic lattices [107, 108], but require sub-nanometer resolution to observe.

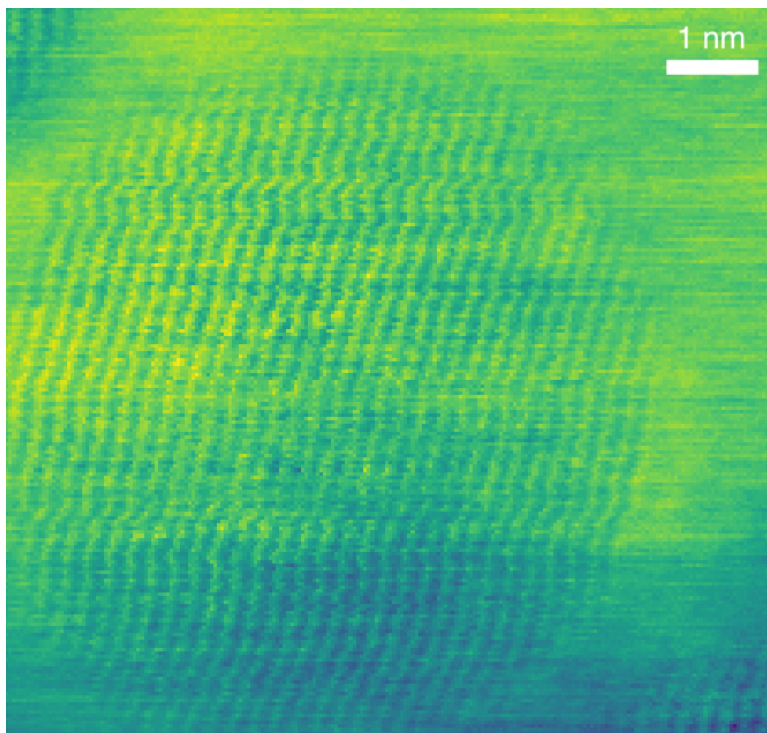


Figure 24. STEMH phase reconstruction of a gold nanoparticle.

The vast majority of samples will not have physical dimensions on the single nanometer scale. A more realistic sample is a half-plane of material from a FIB lamella or atomic-layer deposition. Fig. 25 shows a STEMH reconstruction of a lamella containing several atomic layers of GaN. This experiment was performed on the TEAM I at 300 keV using the aberration corrector, K3 direct electron detector, and probe separation of 9.0 nm. The top half of the measurement is vacuum, but

approximately halfway down the figure, we begin to capture phase contrast images of the sample, including again lattice fringes. Notice the GaN-vacuum interface in the amplitude measurement in Fig. 25a is fairly linear while the interface in the phase reconstruction in Fig. 25b contains a bulge, likely some weakly-scattering carbon contamination. As we scan deeper into the sample, the lattice fringes grow more prominent while more probes pass through the sample. In the phase reconstruction, once the fourth probe enters the sample (at the dashed line in Fig. 25b), the lattice fringes transition from lines oriented at a $+18^\circ$ angle to a two-dimensionally-varying pattern. There is still information to be gained deep within the sample, even if conventional STEMH has irretrievably mixed it together. We will investigate an avenue to extract an interpretable sample phase reconstruction at high resolution in the next Chapter.

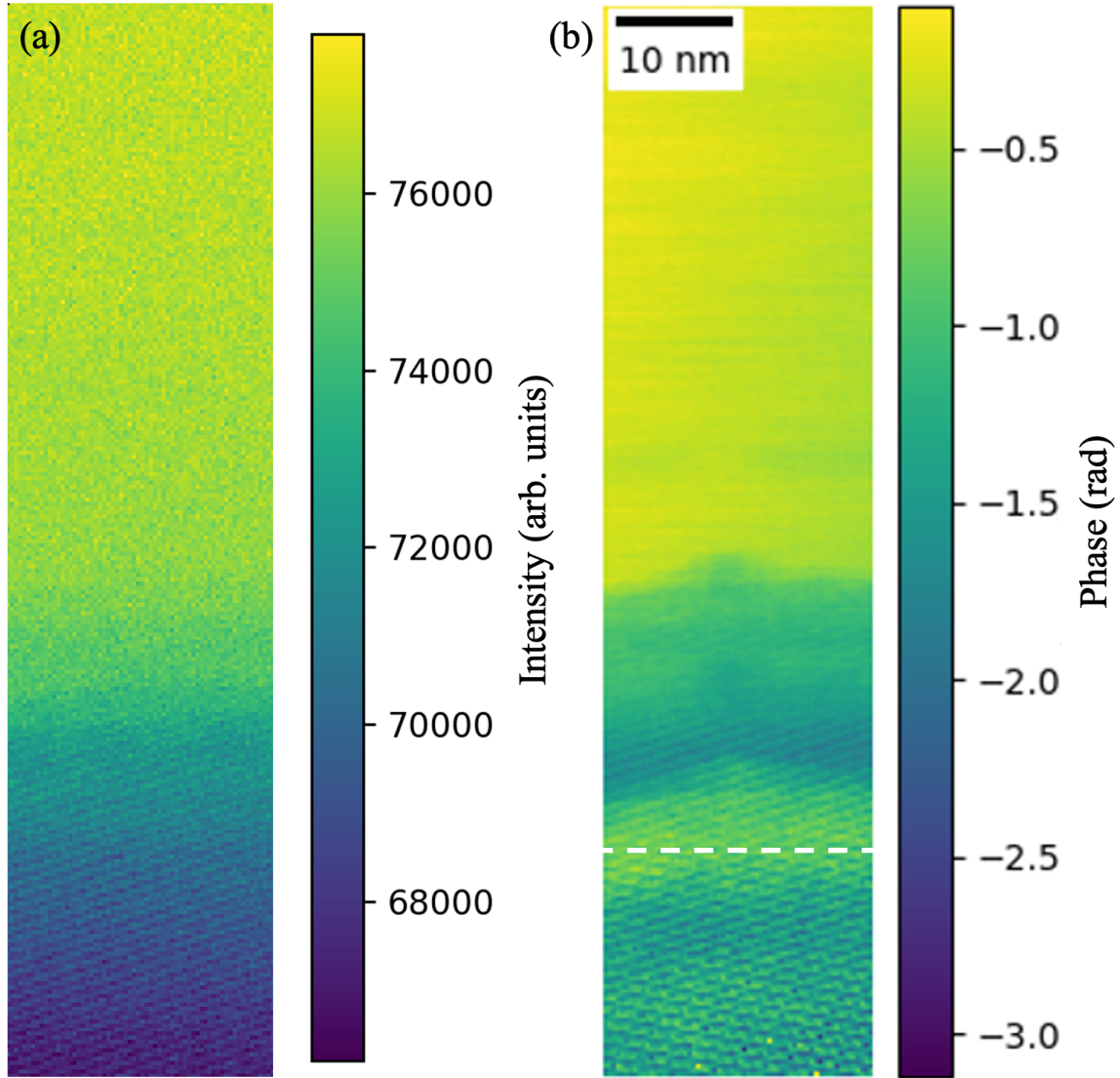


Figure 25. STEMH phase reconstruction of few-atomic-layer GaN. (a) The measured intensity. (b) The reconstructed phase. The scale bar applies identically to both images.

CHAPTER IV

EARLY STEM PTYCHOGRAPHY HOLOGRAPHY RESULTS

This research was a collaboration between myself, Stephanie Ribet of Lawrence Berkeley National Laboratory, Georgios Varnavides of the Delft University of Technology, and Colin Ophus of Stanford University and was the subject of my U.S. Department of Energy Office of Science Graduate Student Research (SCGSR) project. What follows is an original work.

4.1 Introduction

While in the last Chapter we primarily compared STEM holography to TEM holography, the primary STEM phase contrast technique in use today is ptychography. First detailed in the late 1960s [16], ptychography refers to a class of algorithms exploiting shared scattering information in diffraction measurements from neighboring scan positions to reconstruct the sample phase in post-processing [109, 110, 111]. Its key imaging benefit is that its resolution limit is not set by numerical aperture, but by the maximum recorded scattering angle of the diffraction data. Unlike TEM holography, ptychography requires a 4DSTEM detector and no customization of the TEM column. It has been used to set resolution records [18], including reaching the resolution limit set by the inherent thermal vibrations of atoms in a lattice [19], and to perform single particle analysis in cryo-STEM [20], nanomagnetic imaging [112], and electron tomography [113]. Electron ptychography is now a quite mature technique applicable to many samples and experimental conditions [114, 115, 116].

In iterative electron ptychography, we simulate the diffraction from an estimate of our probe interacting with an estimate of our sample, compare the result to the measured diffraction, and update our probe and sample estimates

until they (hopefully) converge on a final image. In the prototypical ptychography experiment, the STEM probe $P(\mathbf{r}_n)$ is rastered across the 2D field of view containing an object described by a transmission function $O(\mathbf{r} - \mathbf{r}_n)$. A 2D diffraction pattern is captured at each beam stop $\mathbf{r}_n$. The result is a four-dimensional dataset containing spatially-localized electron scattering data $I(\mathbf{r}_n, \mathbf{k}) \equiv I_n(\mathbf{k})$. $\mathbf{r}$ is the real space coordinate at the sample plane and $\mathbf{k}$ is the reciprocal space coordinate at the detector plane. The ptychographic reconstruction begins by simulating the interaction of the probe and object wavefunctions. In single-slice ptychography, we assume the electron wavefunction immediately after the sample $\psi_n(\mathbf{r})$ is simply the multiplication of our complex-valued probe and sample wavefunctions

$$\psi_n(\mathbf{r}) = P(\mathbf{r})O(\mathbf{r} - \mathbf{r}_n), \quad (4.1)$$

which is an appropriate model for weakly-scattering samples of several-nanometer thickness. Once a sample is thick enough for multiple scattering to occur, we need multi-slice ptychography. The sample is instead described by N discrete slices where the multiplicative assumption holds, with the scattering probe Fresnel propagated between slices. Multi-slice ptychography is inherently applicable to a wider range of specimens and allows for material characterization along the direction of beam propagation [19, 20, 116].

In either type of iterative ptychography, now armed with the post-sample wavefunction, we Fourier propagate the wavefunction to the detector in the far field and measure the diffraction patterns $J_n(\mathbf{k})$.

$$J_n(\mathbf{k}) = |FT[\psi_n(\mathbf{r})]|^2 \quad (4.2)$$

The measured and simulated diffraction data $I_n(\mathbf{k})$ and $J_n(\mathbf{k})$ are compared and used to independently update the next iteration probe $P'(\mathbf{r})$ and sample wavefunction $O'(\mathbf{r})$ estimates. How exactly $P'(\mathbf{r})$ and $O'(\mathbf{r})$ depend on $I_n(\mathbf{k})$, $J_n(\mathbf{k})$, $P(\mathbf{r})$, and $O(\mathbf{r})$, and the metric used to track convergence, depend on the implementation of the ptychography algorithm in use. Regardless of implementation, the distinct modeling of probe and sample allows the inclusion of physically realistic aberrations in the reconstructed probe, and the effective deconvolution of aberrations from the reconstructed specimen.

While ptychography is successfully being deployed to detect high-spatial-frequency objects like individual atoms, the technique struggles to reconstruct low-spatial-frequency information like constant-thickness substrates and slowly-varying electric potentials in vacuum, especially at low electron dose. At any dose, non-iterative techniques such as single side-band and Wigner-distribution deconvolution ptychography lose all contrast at low spatial frequencies [21, 22]. With infinite electron dose and computation time, iterative ptychography can recover all sample spatial frequency information [23], but with finite dose, i.e., in real operation, iterative ptychography loses its ability to accurately transfer information at low spatial frequencies [24]. STEM and TEM holography, by the dint of their reference beams, perfectly capture contrast at low spatial frequencies [21, 117].

Consequently, STEMH has an experimental niche to fill in electron microscopy if we can solve the multiple-beam issues which plague STEMH. Recall our toy three-beam STEMH example with relative beam amplitudes c_0 and $c_{\pm 1}$, the +1st order acquiring a sample phase ϕ , and the 0th and -1st order passing through vacuum. In the STEMH reconstruction process, this produces a first order interference peak with complex value $c_{-1}c_0 + c_0c_{+1}e^{-i\phi}$. If we knew the values of

the beam amplitudes, we could extract the sample phase ϕ . Iterative ptychography is a technique built to exploit knowledge of the probe wavefunction to solve the sample phase! Therefore, the synthesis of the two techniques, STEM ptychography holography (STEMPH), could capitalize on the reference probes of STEMH to maintain absolute phase contrast at all spatial frequencies while using the iterative ptychography algorithm to combine ghost images into one quantitative result. This Chapter demonstrates simulations of STEMPH which reduce the prevalence of the ghost images even with upwards of seven non-negligible electron beams passing through the sample plane.

4.2 STEMPH Pipeline

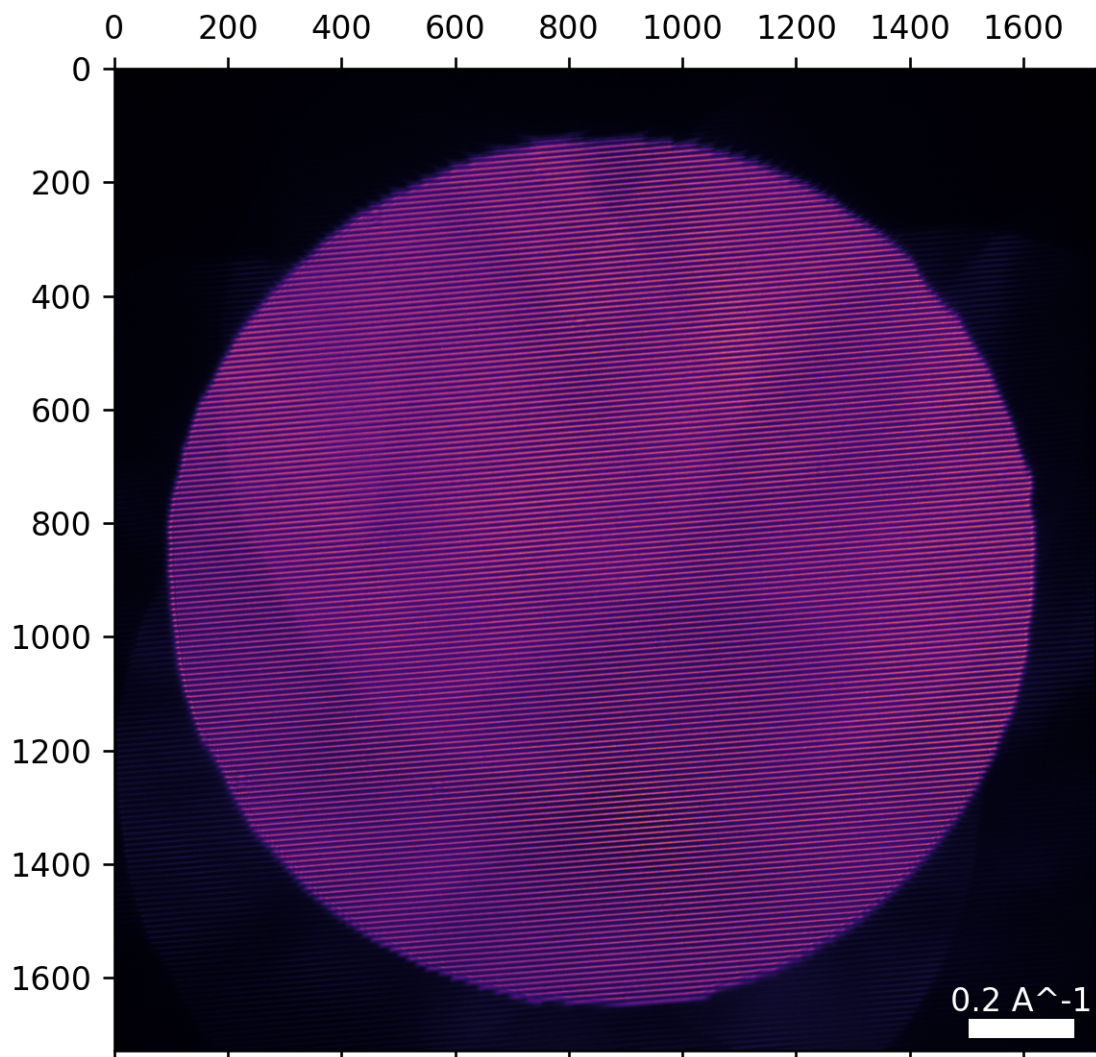
The STEMPH reconstruction pipeline is implemented in the popular 4DSTEM data analysis Python package py4DSTEM [118]. It largely follows the standard single-slice ptychography reconstruction pipeline, with one significant exception: we use non-square arrays to hold our electron probe and scattering data. In standard ptychography, the (single) beam is roughly radially symmetric, so the natural choice, as implemented in py4DSTEM, is to inscribe that circle within a square region of interest. For STEMPH, a square region containing a line of multiple electron beams is much more vacuum than electron probe. Using a full square array to represent the electron probe would waste significant computational resources to confirm many of the component pixels represent vacuum. Additionally, we bin our diffraction measurements along the direction of the interference fringes, as the sample phase shift information is encoded in their movement relative to the fringes. Fig. 26 depicts an example of this binning on real STEMH data. These choices are in response to the large size of STEM holography datasets, typically on the order of tens of gigabytes. The larger the original dataset, the slower and more

computationally expensive the reconstruction. Using non-square arrays allows us to partially ameliorate this drawback to STEMPH.

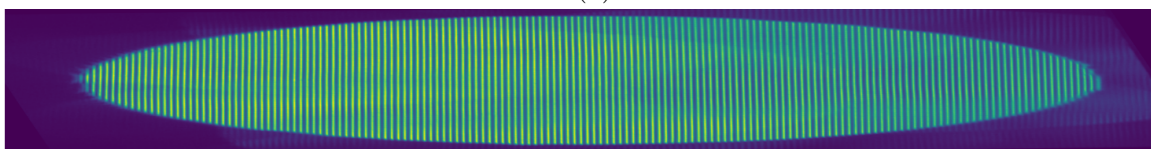
We may also enforce a vacuum region on the probe wavefunction reconstruction after each iteration, an atypical but existing option for single-slice ptychography reconstructions. Fig. 27 depicts a simulated STEMPH reconstruction of an apoferritin protein illuminated by two defocused electron beams. The simulation uses 300 keV electrons with a 5 mrad semiangle and 1000 e^- per pixel. Reconstructions were halted after 64 iterations. Without the probe constraint, the reconstruction generates a third probe (see Fig. 27d) which we know does not truly exist. In general, with N non-negligible beams, the ptychography algorithm outputs $2N + 1$ beams (with the wrong relative intensities). We need some constraint to prevent the reconstruction from placing electrons into non-existent beams. We first tried per-beam regularization, where we replace each updated beam intensity with its measured value when passing through vacuum, but this generated stitching errors which hindered convergence. We find that zeroing out non-existent beams far from the optical axis better directs the convergence of the ptychography algorithm towards the proper relative beam intensities. The probe vacuum constraint also introduces stitching errors, like the discontinuous drop to zero intensity in the right third of Fig. 27f, but these are limited to areas where we know already that the electron dose is quite low.

As for typical ptychographic reconstruction considerations, we use a stochastic gradient descent solver and the error metric of the normalized mean square error (NMSE)

$$NMSE = \frac{\sum_{n,\mathbf{k}} |\sqrt{I_n(\mathbf{k})} - \sqrt{J_n(\mathbf{k})}|^2}{\sum_{n,\mathbf{k}} I_n(\mathbf{k})}. \quad (4.3)$$



(a)



(b)

Figure 26. Example electron diffraction data and its preprocessing for STEM ptychography holography reconstruction. (a) The mean diffraction pattern from a STEMH measurement. (b) The mean diffraction pattern after rotating the interference fringes to be vertical and binning.

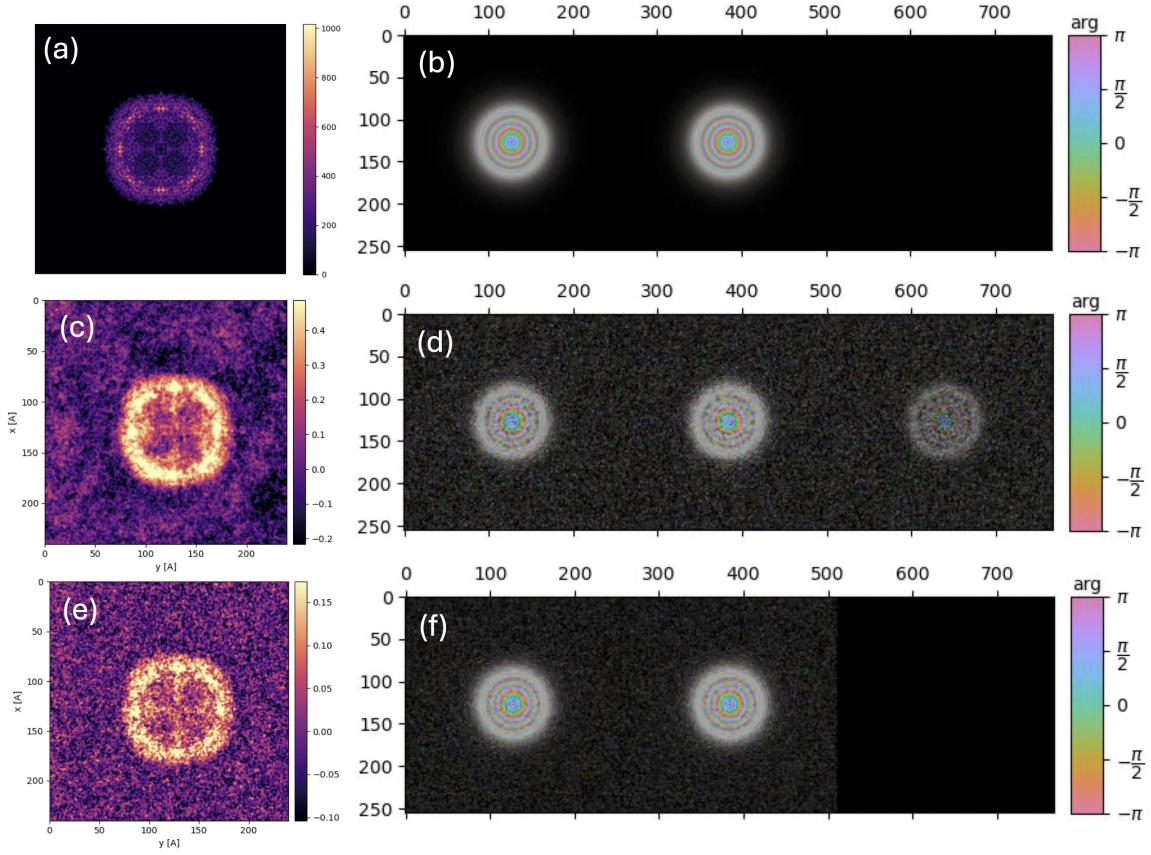


Figure 27. Consequences of probe constraints, or lack thereof. (a) The ground truth projected potential of an apoferritin protein in vacuum. (b) The ground truth probes in real space. (c) STEM ptychography holography reconstruction of the apoferritin protein potential without probe constraint. (d) STEMPH reconstruction of electron probe without probe constraint. (e) STEMPH reconstruction of the apoferritin protein potential with probe constraint. (f) STEMPH reconstruction of electron probe with probe constraint.

We have not attempted hyperparameter tuning. This would be a worthwhile path for future research.

4.3 STEMPH Simulation Results

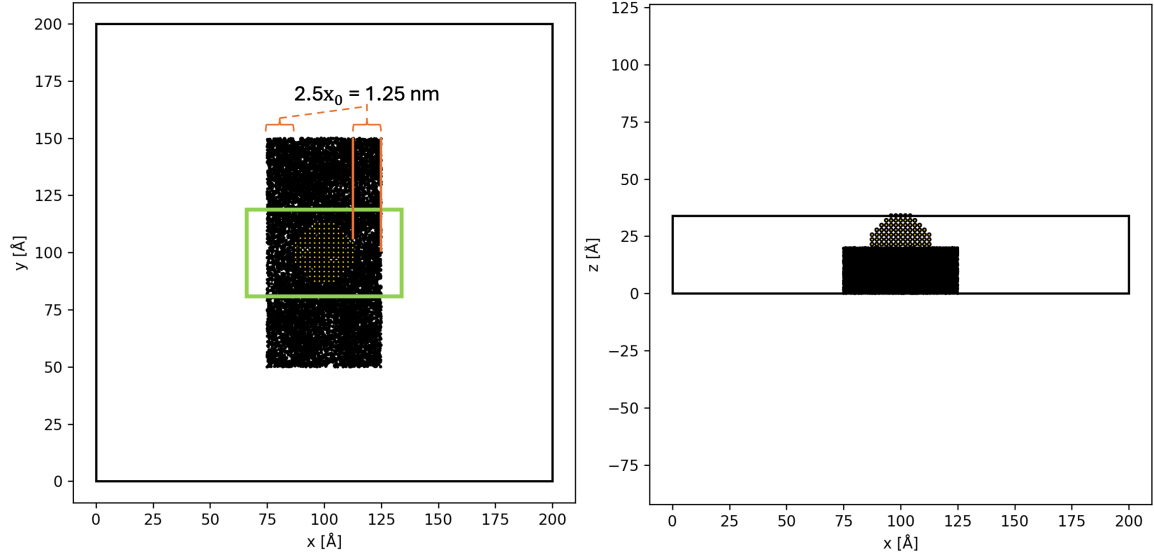
Our main simulated STEMPH results are of a semi-spherical Au nanoparticle of radius 1.3 nm atop 2 nm of amorphous C, depicted from above and from the side in Fig. 28a. The sample is centered in a 2000×2000 gridpoint simulation cell with sampling $0.1 \text{ \AA}^{-1}$. We use a sinusoidal mixed-phase grating with real space beam separation $x_0 = 0.5 \text{ nm}$ to amplitude-divide the electron probe. The carbon substrate is 5 nm wide, placing the Au atomic lattice several beam separations x_0 past the carbon-vacuum interface. This relative size of the sample and beam separation guarantees a simulated STEMH measurement will suffer from ghost images. The simulation cell contains a significant fraction of vacuum to prevent higher order electron beams from looping around the periodic boundaries of the simulation region and interacting with the sample in a non-physical way. The simulated electron probe has an acceleration voltage of 300 keV, convergence semiangle of 25 mrad, and defocus of 50 nm. The real space probe is plotted in the top of Fig. 28b, using the aforementioned sinusoidal thickness profile imparting a phase shift $\frac{7\pi}{16} \approx 1.37 \text{ rad}$ and some weak amplitude modulation incoherently scattering 5% of electrons incident on the grating. Electron diffraction patterns are captured on an 70×40 raster pattern with $1 \text{ \AA}$ step size between scan positions. This scan region is depicted in green in Fig. 28a.

We present two STEM ptychographic holography reconstructions of this nanoparticle on carbon. In both reconstructions, the ptychography algorithm is run for 200 iterations and is seeded with an inaccurate guess for the initial probe plotted in the bottom of Fig. 28b. Vacuum constraints on the probe are not used.

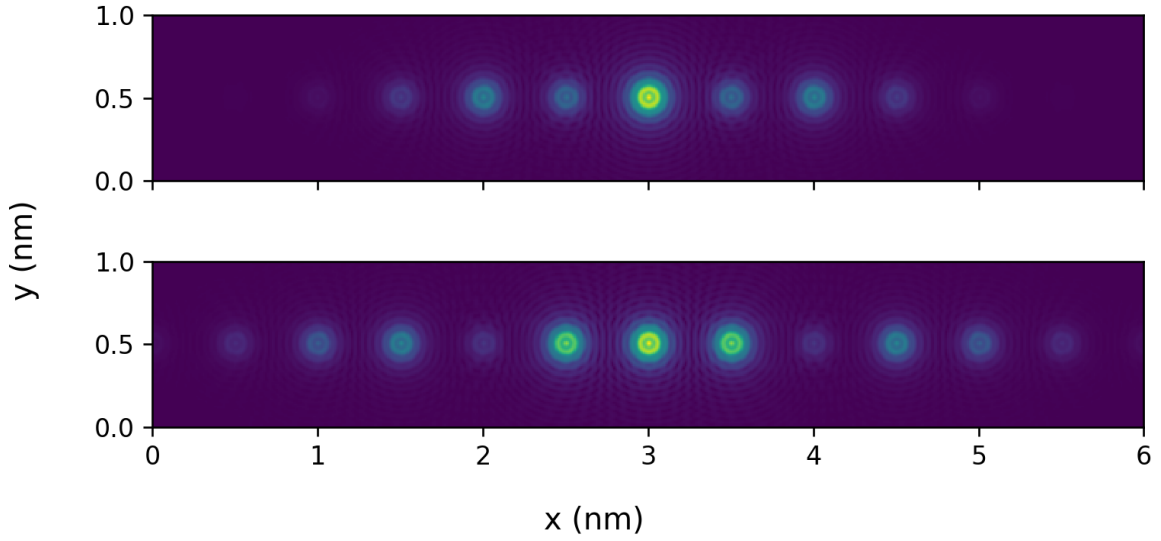
This initial guess was generated by using an identical sinusoidal mixed-phase grating but with an increased phase shift of $\frac{9\pi}{10} \approx 2.83$ rad. This intentionally creates more non-negligible orders, increasing the challenge for the reconstruction algorithm to properly converge to the ground truth.

Fig. 29 shows the first simulated results at several points through the reconstruction. An infinite electron dose is used. Fig. 29a and b show the reconstructed sample potentials and electron probes at varying iteration numbers. Over the first several dozen iterations, ghost images still prevent simple interpretation of the sample potential, but within 64 iterations, the algorithm has significantly addressed the ghost image problem. Over the same period, the probe reconstructions are fairly stable, with the relative beam intensities still much closer to the false initial guess than the true electron probe. While there is improvement in the sample reconstruction between iterations 64 and 96, this is when the algorithm corrects the probe structure. It appears the STEMPH algorithm resolves the ghost images before transitioning to improving its probe reconstruction. This is supported by our error metric, plotted in Fig. 29d, that contains two distinct improvements, one in the first 25 iterations when we notice significant improvement in the sample reconstructions, and another between the 50th and 75th iterations, when we see more accurate relative beam intensities in the probe.

There is an unfortunate artifact where the reconstruction algorithm has shifted the sample and the probe slightly upwards, despite the ground truth being centered vertically along the line $y = 5$ nm. Potentially as a consequence to compensate for this shift, the optical axis of the reconstructed electron probe shifts to the location of the original +1st diffraction order. Still, by iteration 96, the

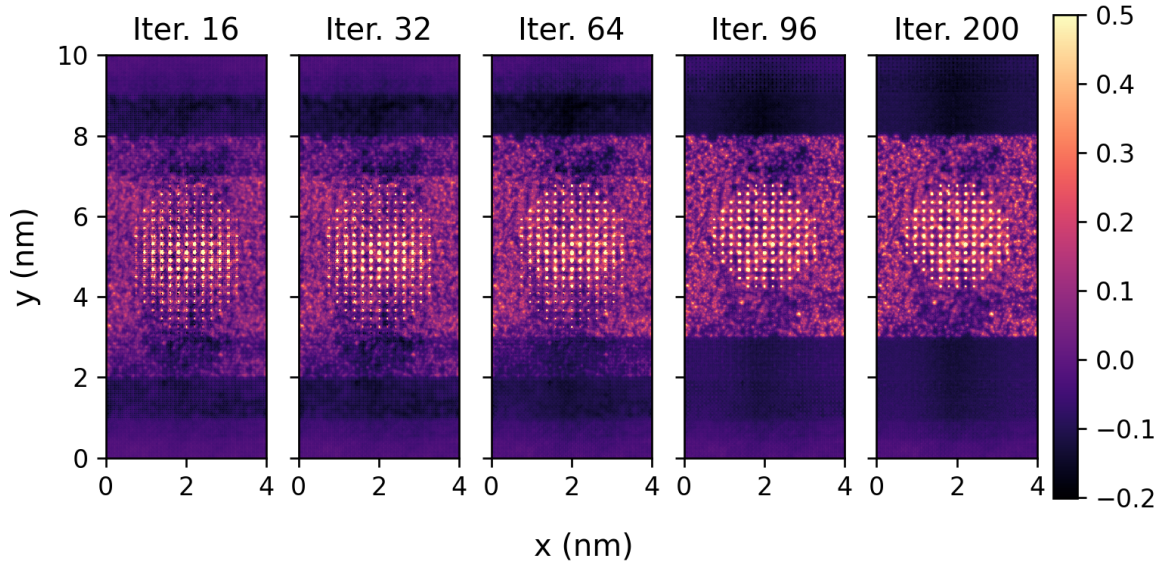


(a)

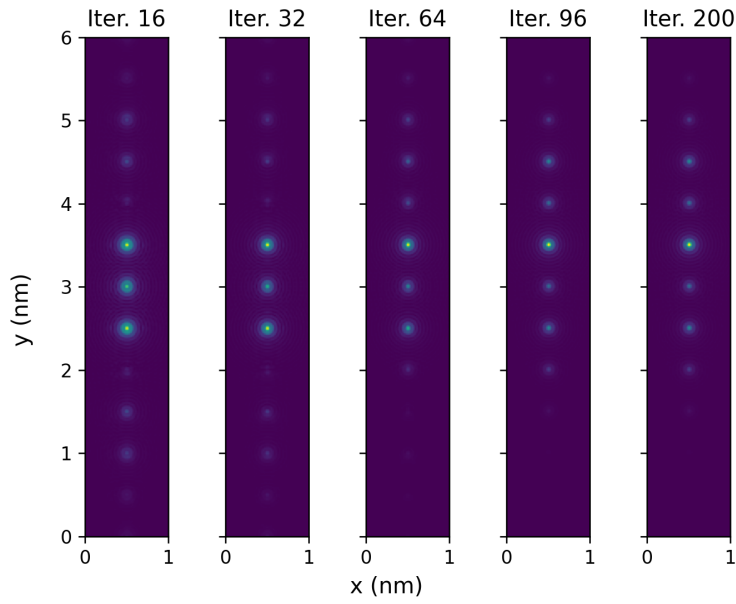


(b)

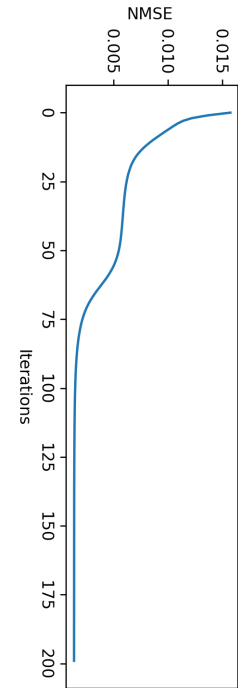
Figure 28. Starting point for STEM ptychography holography simulation. (a) Ground truth atomic model of sample. Left: top-down view of simulated sample with green box depicting scan area and noting the distance between Au nanoparticle and vacuum. Right: side view of simulated sample. (b) Electron probes in simulation. Top: Ground truth real space probe. Bottom: Intentionally incorrect first estimate of real space probe fed to ptychography algorithm.



(a)



(b)



(c)

Figure 29. STEM ptychography holography demonstration with infinite dose. (a) The sample potential reconstruction. (b) electron probe reconstruction. Number of iterations increases from left to right. (c) The normalized mean square error (NMSE) metric plotted versus the number of iterations.

probe wavefunction is again symmetric about the new axis and nearly demonstrates the proper relative beam intensities. To check if this is a scan artifact instead of a STEMH effect, we used an identical raster scan with a single (conventional) electron beam to simulate the bright field and annular dark field STEM images. These appear unshifted relative to their location in the simulation cell, suggesting the STEMPH algorithm is truly to blame for the translations in the reconstruction.

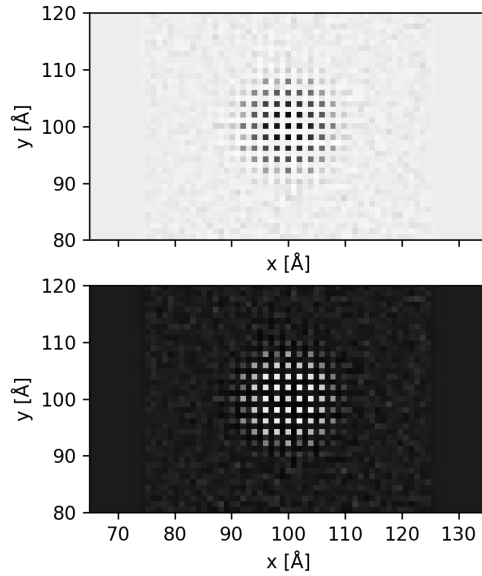
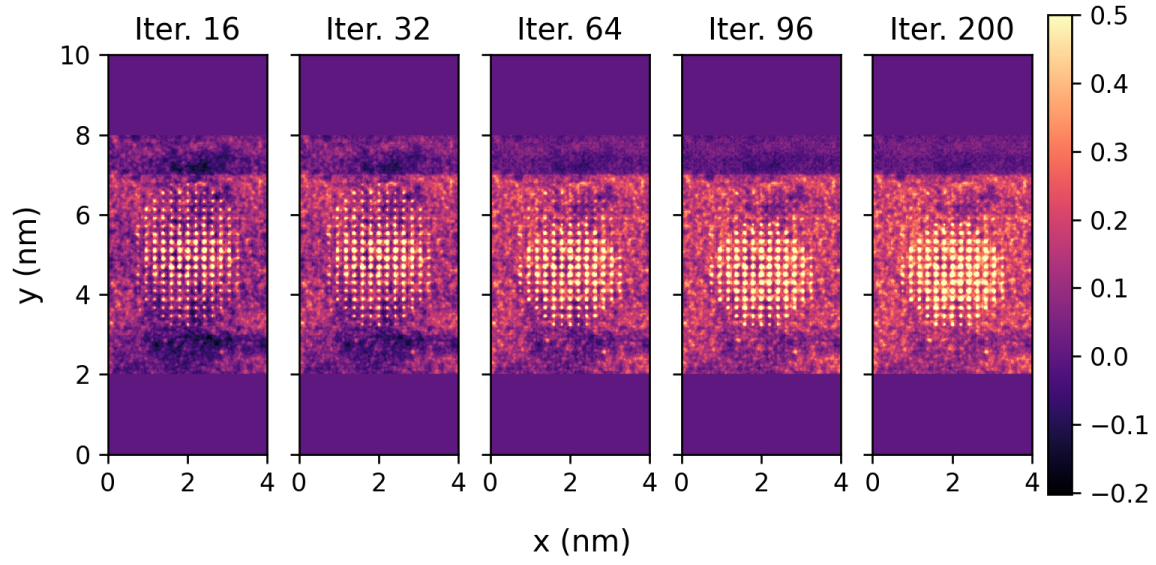


Figure 30. Conventional bright field (top) and annular dark field (bottom) STEM images of the simulated nanoparticle on carbon.

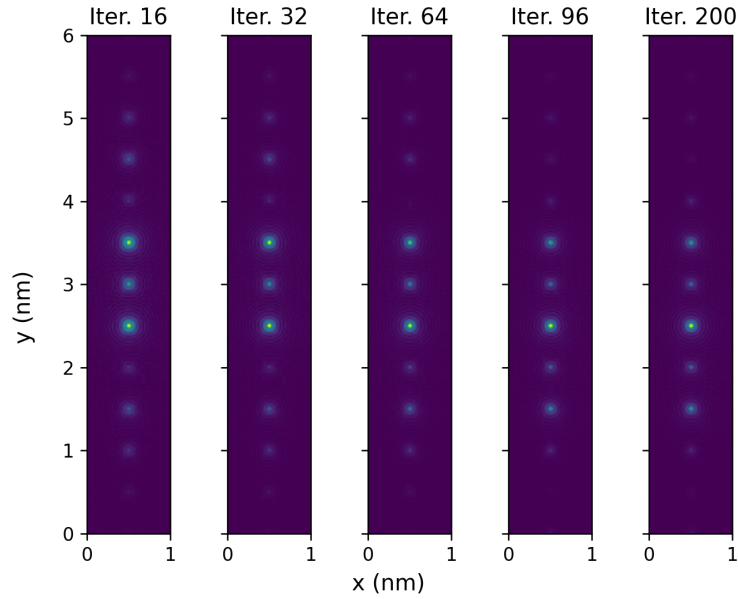
In the second reconstruction, we use finite electron dose ($1000 e^-$ per pixel) and enforce a vacuum region on the sample potential during the reconstruction above and below the lines $y = 8 \text{ nm}$ and $y = 2 \text{ nm}$. The results are plotted in Fig. 31. Our goal with the vacuum constraints on both the probe and sample is to inform the ptychography algorithm that some pixels must be vacuum and some pixels may or may not be vacuum. In the case of the sample potential, we have found the ptychography algorithm struggles to understand that uncertainty. If a

pixel is outside the vacuum-enforced region in the reconstruction, the algorithm wants to keep sample information in that pixel—even if there may not truly be. For example, in Fig. 31a, we still see a ghost image in the region $7 \text{ nm} < y < 8 \text{ nm}$ at iterations 96 and 200, beyond the extent of 5 nm-wide carbon substrate reconstruction with one interface at $y = 2 \text{ nm}$. The relative probe intensities in Fig. 31b are apparently unaffected, and have approached the true probe intensities by iteration 64 (up to another shift of the entire probe down one diffraction order). With nominal change in the NMSE metric after iteration 64, it appears the vacuum constraint has trapped the reconstruction in an incorrect local maximum containing a ghost image. Additionally, we see more evidence that the translation effect is due to the STEMPH reconstruction, as it recurs here. While we do not see a benefit in using vacuum constraints on the sample currently, future work could set a vacuum constraint to limit the translation of the reconstruction relative to the sample for the first several dozen iterations, then turn off the vacuum constraint to allow the algorithm to fully combine ghost images.

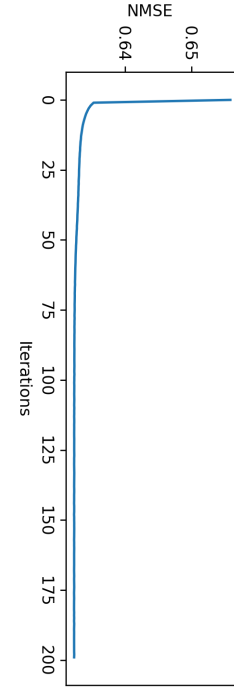
Together, these results show the removal of ghost images from STEMH measurements using a tailored single-slice ptychographic pipeline. This reduction is not perfect, as we can still see some contrast remain outside the sample. Nor has the full potential of the ptychographic postprocessing been optimized, but this represents a large step forward in the development of STEM holography.



(a)



(b)



(c)

Figure 31. STEM ptychography holography demonstration with finite dose and vacuum constraints on the sample potential above and below the lines $y = 8$ nm and $y = 2$ nm. (a) The sample potential reconstruction. (b) electron probe reconstruction. Number of iterations increases from left to right. (c) The normalized mean square error (NMSE) metric plotted versus the number of iterations.

CHAPTER V

CONCLUSION

The potential of Scanning Transmission Electron Microscopy Holography (STEMH) is largely untapped. The mature phase contrast techniques of TEM holography and ptychography grew from several decades of development pushed by several competing research groups worldwide. That level of attention has not yet been paid to STEMH. This dissertation provides an avenue, STEM ptychography holography, to address the main STEMH experimental restrictions, namely being constrained to qualitative measurements and, at high resolution, hard-to-interpret measurements due to ghost images. The obvious next step of STEMH development is successfully performing a single-slice STEM ptychography holography measurement. Near-term follow-up research should also include experimental confirmation that STEMPH retains its low-spatial-frequency information transfer with finite dose. The moderate-term goal should be multi-slice STEMPH, where we account for multiple scattering in the sample during the ptychographic reconstruction, unlocking the measurement of thicker samples and providing a mechanism for three-dimensional imaging. The long-term goal is the adoption of STEM holography as a complementary phase contrast technique ready to answer novel materials science and biological research questions.

APPENDIX A

APPENDIX TO CHAPTER II

A.1 Diffraction symmetry with arbitrary duty cycle phase gratings

We prove how even and odd symmetry in the non-zero region of an arbitrary duty cycle phase grating impose some symmetry in the Fourier coefficients. For context, parity arguments for the generic phase grating proves even symmetry in the thickness profile forces symmetry in the diffraction pattern $c_m = c_{-m}$, while odd symmetry imposes $c_m \in \mathbb{R}$.

Theorem 1. *Let $T(x)$ be a transfer function for a phase grating of arbitrary duty cycle f and even thickness profile $t(x)$. Then if $mf \in \mathbb{Z}$, $\bar{c}_m(f) = \bar{c}_{-m}(f)$.*

Proof. $t(x)$, consequently d_{mf} , and sinc are even functions in m , so the only effect of flipping the sign of m is to map $e^{-imf\pi} \rightarrow e^{imf\pi}$. In general, these functions will not be the same because its imaginary part specifically flips sign: $e^{-imf\pi} = \cos(mf\pi) - i \sin(mf\pi)$ vs. $\cos(mf\pi) + i \sin(mf\pi)$. These equations are equal when the sine term is zero, or when $mf \in \mathbb{Z}$. □

For a rational $f \in \mathbb{Q}$, an infinite subset of diffraction coefficients c_m are equal to c_{-m} . For example, if $f = \frac{1}{3}$, all diffraction orders which are a multiple of three (when $m = 3n, n \in \mathbb{Z}$), will be equal to their reflection on the other side of the optical axis.

Theorem 2. *Let $T(x)$ be a transfer function for a phase grating of arbitrary duty cycle f and odd thickness profile $t(x)$. Then $\text{Im}[\bar{c}_m] = -\text{Im}[\bar{c}_{-m}]$.*

Proof. The odd parity of $t(x)$ forces $d_{mf}, d_{-mf} \in \mathbb{R}$, or $\text{Im}[d_{mf}] = \text{Im}[d_{-mf}] = 0$. Then

$$\text{Im}[\bar{c}_m(f)] = -f \text{Im}[e^{-imf\pi} \text{sinc}(mf\pi)] = f \sin(mf\pi) \text{sinc}(mf\pi), \quad (\text{A.1})$$

$$\text{so } \text{Im}[\bar{c}_{-m}(f)] = f \sin(-mf\pi) \text{sinc}(-mf\pi) = -f \sin(mf\pi) \text{sinc}(mf\pi). \quad \square$$

A.2 Mathematical details for optimal triplicator and duplicator gratings

The final diffraction efficiencies will be written in terms of hypergeometric functions

$${}_pF_q(a_1, a_2, \dots, a_p; b_1, \dots, b_q; z) = \sum_{n=0}^{\infty} \frac{(a_1)_n (a_2)_n \dots (a_p)_n}{(b_1)_n \dots (b_q)_n} \frac{z^n}{n!} \quad (\text{A.2})$$

where $(x)_n = \frac{\Gamma(x+n)}{\Gamma(x)}$ is the Pochhammer symbol. This is a very general structure which can reduce to various special functions and polynomials for specific choices of parameters a_i , b_j , and complex argument z . For example, Bessel functions $J_\alpha(z) = {}_0F_1\left(\alpha + 1; -\frac{z^2}{4}\right)$ and Legendre polynomials $P_\ell(x) = {}_2F_1(-\ell, \ell + 1; 1; -\frac{1}{2}(x - 1))$. Often the specific case ${}_2F_1(a, b; c; z)$ is referred to as the hypergeometric function $F(a, b; c; z)$ [85], but we will use the former notation.

Chebyshev polynomials of the first kind $T_n(\cos x)$ are generalized multi-angle formulas which break $\cos nx$ into a polynomial of cosines. For example, $T_3(\cos x) = 4 \cos^3 x - 3 \cos x$. To solve the optimum triplicator diffraction coefficient integral, we define $T_{n,j}$ to refer to the polynomial coefficients:

$$\cos nx = \sum_{j=0}^n T_{n,j} \cos^j x \quad (\text{A.3})$$

$$T_{n,j} = (-1)^{\frac{n-j}{2}} 2^{j-1} n \frac{\left(\frac{n+j}{2} - 1\right)!}{\left(\frac{n-j}{2}\right)! j!} \quad (\text{A.4})$$

n, j are non-negative integers with $j \leq n$ and $T_{n,j} = 0$ if n, j have differing parities [119]. If n is even, reindexing the sum over powers of $\cos x$ by $j \rightarrow 2j$ retains only terms with nonzero coefficients. The sum now ends at $j = \frac{n}{2}$:

$$\cos nx = \sum_{j=0}^{n/2} T_{n,2j} \cos^{2j} x. \quad (\text{A.5})$$

APPENDIX B

APPENDIX TO CHAPTER III

We detail the image processing performed on the raw data to create the image in Fig. 22a:

- For each FFT of a 4DSTEM frame, grab an $N \times N$ square around the first non-central peak in the FFT. Call this square $I_1(\mathbf{x} - \mathbf{x}_0)$.
- Take the value of the peak $I_1(0)$.
- Reconstruct a phase map $\phi_1(x)$ of the ROI using pixel values of $I_1(0)$
- Select the 3×3 array centered on $I_1(0)$. Find the total of this array
- Reconstruct a phase map $\phi_3(x)$ of the ROI using the sum of the 3×3 array
- Phase unwrap both phase maps
- Subtract the unwrapped phases $\phi_3(x) - \phi_1(x)$
- Apply Gaussian blur

To justify this approach, we applied the same image processing procedure to a second image of the same region in the sample with fewer pixels per fringe and with an energy filter between the sample and detector.

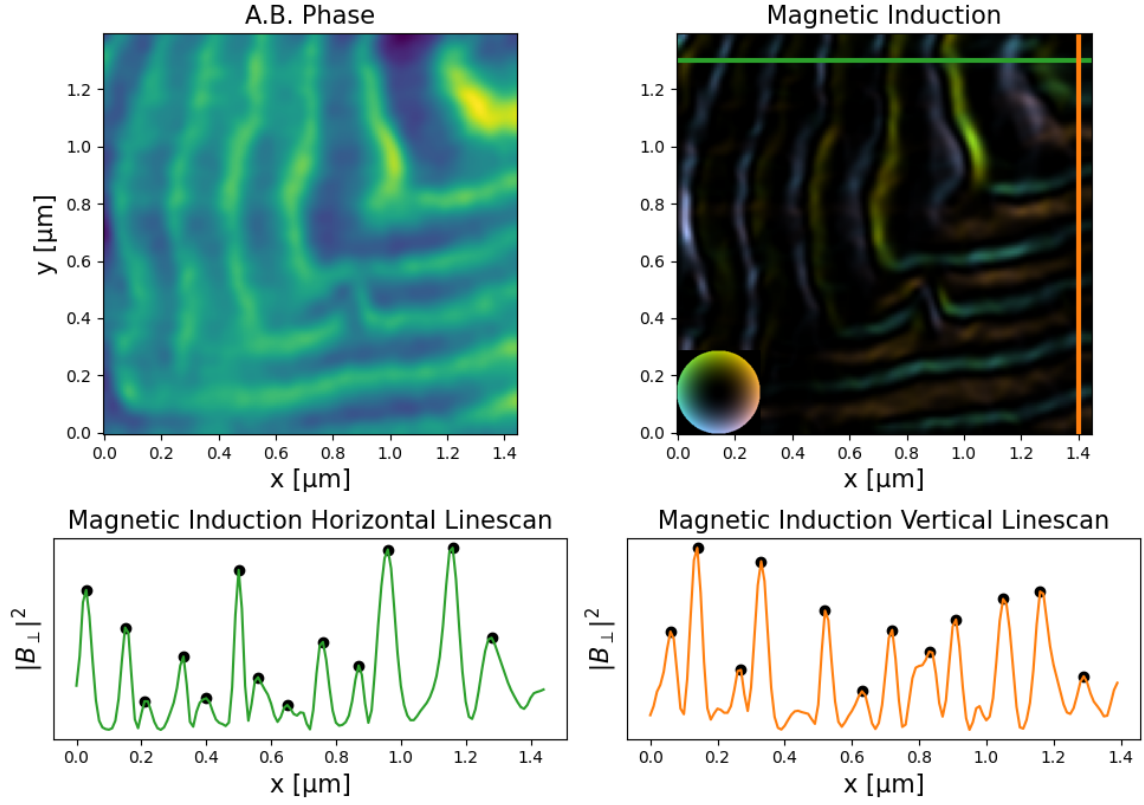


Figure B.1. The reconstructed phase and magnetic induction from the same region, using a more efficient pixel capturing scheme, a day after the original dataset was captured. Horizontal and vertical linescans show a domain wall spacing of 112 nm and 104 nm, respectively.

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