

INTERACTION-FREE MEASUREMENTS WITH ELECTRONS

by

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DISSERTATION ABSTRACT

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Quantum-enhanced electron imaging promises highly efficient, low-damage transmission electron microscopy, transforming high-resolution imaging of samples affected by beam irradiation. One proposed quantum protocol for damage-free imaging is interaction-free measurement. However, this quantum phenomenon had yet to be experimentally realized with electrons. In this dissertation, we demonstrate the first interaction-free measurement with electrons. To realize this quantum protocol with electrons required the development of a novel, free-electron interferometer constructed in a conventional transmission electron microscope. Here we design and create a flexible two-grating electron Mach-Zehnder interferometer with high contrast in discrete outputs. We test this balanced interferometer with equal-intensity paths to map the electrostatic potential around a nanorod and perform quantitative phase imaging of a nanoparticle. We then achieve a quantum-enhanced, counterfactual measurement

with electrons - the interaction-free measurement. Comparing this novel electron interferometer to conventional electron holography, which is a well-established technique in electron microscopy, we find that the interferometer is more suitable for electron interaction-free measurements due to the high contrast in discrete outputs. The two-grating interferometer is then optimized for higher efficiency interaction-free measurements by forming a weak interaction probe that decreases the probability of interacting with the sample. The modified configuration more than doubles the interaction-free measurement efficiency of the original balanced interferometer. This body of work not only includes the development of a versatile electron interferometer, capable of a broad scope of measurements, but realizes a fundamental quantum experiment. Achieving the interaction-free measurement with electrons takes us one step closer to developing a functional quantum-enhanced electron microscope.

This dissertation contains previously published and unpublished material.

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CHAPTER I

INTRODUCTION

1.1. History and Motivation

The electron played an essential role in developing and establishing quantum mechanics. Before the discovery of the electron, in the early 1700s, Charles François du Fay first identified positive and negative electrical charge [1]. He rubbed a glass rod with silk and a piece of amber with wool and found that the glass would repel a charged gold-leaf while the amber would attract it. However, when the amber or glass contacted the gold, the “electrical fluids” would combine and neutralize. This experiment prompted two centuries of theories and research, searching for the fundamental unit of electricity. Finally, in 1897, J.J. Thomson discovered the free electron as a unique particle [2]. Using a cathode ray, Thomson measured the electron’s charge-to-mass ratio, establishing the electron as a subatomic particle with a fraction of the mass of hydrogen. Furthermore, Thomson claimed that the electron exists in all atoms of any element. Soon thereafter (1909), Robert Millikan and Harvey Fletcher developed the oil droplet experiment, where a charged oil droplet was suspended in midair, balanced between the forces of gravity and an external electric field [3]. The experiment was sensitive to the individual number of charged ions in each droplet, uncovering the discrete mass and quantized charge of the electron. These experiments inspired a new era of atomic theory, establishing the electron as an elementary fermionic particle with quantized negative charge and mass.

In 1923, Louis de Broglie hypothesized that all matter, including the electron, can be described as a wave [4], revolutionizing the understanding of the electron. This

wave-like interpretation of electrons initiated a surge of theoretical and experimental work. In 1926, Erwin Schrödinger proposed the non-relativistic wave equation, which described the propagation, diffraction, and interference of low-energy electron waves [5]. Meanwhile, George Paget Thomson passed electrons through thin metal foils [6] while Clinton Davisson and Lester Germer reflected electrons off a nickel crystal [7] to experimentally demonstrate electron diffraction. Only one year after Schrödinger's wave equation, Paul Dirac unified quantum mechanics and special relativity with the Dirac equation, realizing the relativistic wave equation of an electron.

The unification of particle and wave theory generated both confusion and interest in quantum mechanics. Wave-particle duality is counterintuitive; a quantum entity can at times diffract and interfere like a wave, or transfer momentum and collide like a particle. Furthermore, the position of the electron is unknowable, while the probability of finding an electron in any given position is knowable. Scientists were uncomfortable that nature was based on a highly probabilistic theory. Even Einstein stated, "God does not play dice with the universe" [8]. However, experiments continued to support that electrons exhibit wave-particle duality and are quantum mechanical by nature. The quantum mechanical nature of the electron led to many technological applications such as lasers, transistors, electronics, and electron microscopes.

Harnessing the wave-like nature of the electron, the first electron microscope was constructed by Max Knoll and Ernst Ruska in 1931 [9]. The motivation to develop the electron microscope was to reach new magnitudes of high-resolution imaging. Due to the significantly smaller wavelength of high-speed electrons (picometers) in comparison to light (hundreds of nanometers for visible light), electron microscopes were theoretically capable of 1000 times the resolution of conventional optical

microscopes. The original electron microscopes incorporated multiple solenoids to focus and manipulate the beam, acting as magnetic lenses that were first conceived by Hans Busch in 1926 [10]. The microscope passed electrons through the sample and used the transmitted electrons to form an image, coining the name transmission electron microscope (TEM). Although modern TEMs feature significant advancements since the 1930s, the framework and principles of the original design remain.

In the subsequent fifteen years, the scanning electron microscope (SEM) was designed and constructed [11, 12]. Unlike the TEM, which creates an image using transmitted electrons, the SEM uses reflected or secondary electrons to form an image to look at the surface of materials. The advantages of the SEM are minimal sample preparation, a tolerance for larger samples, lower total cost to purchase and operate, quicker imaging, and a larger field of view. In contrast, the TEM requires meticulous sample preparation (a sample thickness of less than 150 nm) and is expensive, but offers higher resolution and imaging of internal structures. Furthermore, the design of the TEM creates a manipulable electron optics workbench, capable of diverse experiments exposing nanoscale features such as magnetic structures [13, 14], electrostatic fields [15, 16], nanoplasmonic modes [17, 18, 19], and atomic arrangements [20, 21]. In numerous cases, the high-resolution and broad scope of retrievable information in the TEM outweigh the speed and lower cost of the SEM.

The modern advancements of the TEM make it a diverse tool capable of many different modes and techniques. TEMs can now be equipped with direct electron detectors, electron spectrometers, aberration correction, probe correction, monochromators, and more. With these advancements, the TEM is capable of

subatomic orbital imaging [22], atomic reconstructions of materials in their hydrated state [23, 24, 25, 26], mapping nanoplasmonic modes [17, 18, 19], nanoscale magnetic imaging [14, 27], atomic resolution elemental mapping [20, 21], precision quantum experiments [28, 29], as well as myriad other applications. Some of these feats have been achieved through electron interferometry, relying on the wave-like nature of the electron. In 1953, Marton created the first electron interferometer using three crystals as beamsplitters, resulting in a “very skew Mach-Zehnder” interferometer [30]. Since this first electron interferometer, interference- and diffraction-based imaging in the TEM has improved greatly. Beamsplitting mechanisms expanded from crystals to nanofabricated material beamsplitters [31], biprisms [32, 33], standing light waves [34], and microwave chips [35]. With these improved beamsplitters, interferometric techniques progressed, including holography [36, 37], ptychography [38, 39], and scanning TEM holography [40, 41].

When the TEM achieved atomic resolution imaging, one sample type of particular interest was low-Z materials (materials made up of atoms with low atomic numbers). Low-Z materials include biomaterials, proteins, salts, viruses, and metal organic frameworks. However, these fragile molecular structures pose a substantial hurdle for atomic resolution electron imaging. Due to the low number of protons in the nuclei, these materials create minimal contrast in TEM images. Low-Z materials are also easily damaged under the high-energy electron beam in conventional TEM imaging; the energy required to displace an atom is related to the material’s bond strength, lattice structure and the atomic weight of the constituent atoms [42]. The main sources of damage are radiolysis (decomposition by ionizing radiation) and elastic knock-on damage (displacing an atom due to an electron-atom collision) [43]. Currently, one effective technique for imaging low-Z materials is single particle

analysis [44, 45]. Single particle analysis involves taking thousands of noisy images of identical samples in different orientations, using an electron fluence ($e^-/\text{\AA}^2$) below the damage threshold of the sample. The noisy images are then aligned, filtered, analyzed, and combined to create a high-resolution, three-dimensional reconstruction of the sample. Single particle analysis has several disadvantages, including requiring thousands of identical samples, irreversible damage inflicted by the beam, and costly and time consuming sample preparation, data acquisition, and data analysis. Imaging an individual, low-Z sample at atomic resolution without damage is an unresolved goal in the field of electron microscopy.

Applying quantum protocols to electron imaging could provide new avenues for repetitive, damage-free imaging of fragile samples. One proposed quantum protocol for low dose ($e^-/\text{\AA}^2/s$) electron imaging [46, 47] is the interaction-free measurement [48], first proposed in 1993 by Elitzur and Vaidman. The thought experiment captures the non-intuitive weirdness of quantum physics by occasionally measuring the presence of an object without any sample interaction. Experimentally realizing the interaction-free measurement with electrons is not only a novel, fundamental quantum experiment, but could have immediate applications in low-dose electron imaging.

In this dissertation, I (1) develop a versatile free-electron Mach-Zehnder interferometer in the transmission electron microscope (TEM) with scanning capabilities for imaging, (2) use the interferometer to realize the first interaction-free measurement with electrons, and (3) optimize the interferometer to increase the interaction-free measurement efficiency, creating a viable pathway toward quantum-enabled, damage-free electron imaging.

1.2. Fourier Optics, the Electron Microscope and Diffraction Gratings

Before diving in to the bulk of this dissertation, we explore the background information required, including a brief review of linear Fourier optics, an overview of the TEM, and a theoretical description of electrons interacting with thin films and diffraction grating holograms.

1.2.1. Linear Fourier Optics for Electrons

Electrons exhibit both wave and particle behaviors; in other words they embody wave-particle duality. The electron can be diffracted just like light; however, the electron also delivers energy to a single point like a particle. De Broglie showed that an electron can be described as a quantum of an electron wave. The photon has an energy $E = hc/\lambda$ and momentum $\mathbf{p} = \hbar\mathbf{k}$, where c is the speed of light, h is Planck's constant, and $\mathbf{k}$ is the wave vector. The wave vector is related to the wavelength by $|\mathbf{k}| = 2\pi/\lambda$ and $|\mathbf{p}| \equiv p$, giving $p = h/\lambda$. De Broglie showed that analogously the momentum of an electron $\mathbf{p} = m_e\mathbf{v}$, where m_e is the mass of the electron and $\mathbf{v}$ is the velocity vector, can similarly be related to a wave vector by $\mathbf{p} = \hbar\mathbf{k}$. Thus the de Broglie wavelength of an electron is defined as $\lambda = h/p$. When thermal energy is the source of the kinetic energy of a moving particle, the de Broglie wavelength depends on temperature (the average velocity of the particle or ideal gas). However, when an electron is emitted, this wavelength has no temperature dependence and instead is dependent on the momentum.

To create a mathematical model of a free electron propagating down the barrel of a TEM, we turn to the Schrödinger equation. Because the free electrons in a TEM can travel at velocities which are a considerable fraction of the speed of light, a full description of the electron requires the Dirac equation [49], which incorporates

both special relativity and quantum mechanics for massive, spin-1/2 particles [50]. However, the Dirac equation is mathematically complex and the energies used in this dissertation only reach electron velocities of $v \approx 0.7c$. Thus, we employ the Dirac equation's non-relativistic counterpart, the Schrödinger equation and use corrections to the wavelength to account for any subtle relativistic effects.

In the experiments in this thesis, beyond the magnetic lenses used to focus the beam, the electrons only interact with electrostatic potentials allowing us to set the magnetic vector potential to zero, $\mathbf{A} = 0$. Thus we start with the time dependent Schrödinger equation,

$$\left[-\frac{\hbar^2}{2m} \nabla^2 + V(\mathbf{x}, t) \right] \Psi(\mathbf{x}, t) = i\hbar \frac{\partial}{\partial t} \Psi(\mathbf{x}, t) \quad (1.1)$$

with $\hbar$ being the reduced Planck constant, m the mass of the particle, $V(\mathbf{x}, t)$ the spatially and time dependent potential and $\Psi(\mathbf{x}, t)$ defining the particle wave function dependent on both time and position. Because the potential $V(\mathbf{x}, t)$ does not vary with time and the system's energy is constant, we simplify the time dependent Schrödinger equation to the time independent Schrödinger equation,

$$\left[-\frac{\hbar^2}{2m} \nabla^2 + V(\mathbf{x}) \right] \Psi(\mathbf{x}, t) = E \Psi(\mathbf{x}, t), \quad (1.2)$$

where E is the energy of the electron. The time independent Schrödinger equation is a good model when the energy E is well below the rest energy of the electron $E_0 = m_0 c^2 = 511 \text{keV}$, such that $E \ll E_0$.

In the TEM, the electron is accelerated by a static potential, $V_{\text{acc}} = -eU_{\text{acc}}$, with e the charge of the electron. Given this potential, the kinetic energy of the electron is $E_K = E - V_{\text{acc}}$, which simplifies the time-independent Schrödinger equation to the

form of the Helmholtz equation,

$$\nabla^2 \Psi(\mathbf{x}, t) + k^2 \Psi(\mathbf{x}, t) = 0, \quad (1.3)$$

where k is the wavenumber of the electron, $k = \frac{2\pi}{\lambda} = \frac{\sqrt{2mE_K}}{\hbar}$. Note that if $V \ll E$ then the wavefunction is separable in $\mathbf{x}$ and t , giving $\Psi(\mathbf{x}, t) = \psi(\mathbf{x})e^{-iEt/\hbar}$.

To account for the electron travelling at a substantial fraction of the speed of light in a TEM and avoid the complexity of the Dirac equation, we use relativistic corrections to the Helmholtz equation to better approximate the system. Substituting the relativistic correction for the electron's wavelength [51],

$$\lambda_{rel} = \frac{hc}{\sqrt{2E_K E_0 + E_K^2}}, \quad (1.4)$$

the wavenumber k corrects to $k_{rel} = \frac{2\pi}{\lambda_{rel}}$.

In a conventional TEM, the electrons are accelerated in one direction, down the column of the microscope; the beam of electrons travels close to the optical axis throughout the entire system with only a small beam divergence. Therefore we can model the electron beam as a plane wave with a wavefunction $\psi(\mathbf{x}) = A(\mathbf{x})e^{ikz}$, where the amplitude function $A(\mathbf{x})$ is independent of or slowly varying in z . Inserting this wavefunction into the Helmholtz equation, we use the paraxial approximation to simplify the Helmholtz equation further. The paraxial approximation holds so long as

$$\left| \frac{\partial^2 A(\mathbf{x})}{\partial z^2} \right| \ll \left| k \frac{\partial A(\mathbf{x})}{\partial z} \right|. \quad (1.5)$$

In other words, the angle between the $\mathbf{k}$ vector and the optical axis is small and can be approximated as zero. Substituting $\psi(\mathbf{x})$ into the Helmholtz equation and

applying the paraxial approximation results in the paraxial Helmholtz equation,

$$\nabla_{\perp}^2 \psi + 2ik \frac{\partial \psi}{\partial z} = 0, \quad (1.6)$$

where $\nabla_{\perp}^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$ is the transverse Laplacian. The Fresnel diffraction integral is an exact solution of this equation,

$$\psi_1(x_1, y_1) = -\frac{ik}{2\pi} \frac{e^{ikz}}{z} \iint_{-\infty}^{\infty} \psi_0(x_0, y_0) e^{\frac{ik}{2z} [(x_1-x_0)^2 + (y_1-y_0)^2]} dx_0 dy_0, \quad (1.7)$$

where $\psi_0(x_0, y_0)$ describes the initial field or wavefunction at $z = 0$, $\psi_1(x_1, y_1)$ describes the final field or wavefunction at z after propagation, and $k = k_{rel} = 2\pi/\lambda_{rel}$ is the wavenumber for the particle in free space. The Fresnel paraxial approximation holds true in and beyond the Fresnel near-field (where the incident and diffracting wave fronts are spherical).

The Fresnel diffraction integral can be simplified further in the far-field (where the incident and diffracting wave fronts are plane waves) by expanding the exponential term in the integral and assuming $|z| \gg \frac{ka^2}{2}$, where $a = \sqrt{x_0^2 + y_0^2}$ is the maximum radial extent of the input signal. This far-field simplification gives

$$\begin{aligned} \psi_1(x_1, y_1) &= -\frac{ik}{2\pi} \frac{e^{ikz}}{z} \iint_{-\infty}^{\infty} \psi_0(x_0, y_0) e^{ik_x x_0} e^{ik_y y_0} e^{\frac{ik}{2z} (x_1^2 + y_1^2)} dx_0 dy_0 \\ &= -\frac{ik}{2\pi} \frac{e^{ikz}}{z} e^{\frac{ik}{2z} (x_1^2 + y_1^2)} \mathcal{F}\{\psi_0(x_0, y_0)\}, \end{aligned} \quad (1.8)$$

where $\mathcal{F}\{\psi_0\}$ is the Fourier transform of ψ_0 , and $k_x = \frac{kx_1}{z}$ and $k_y = \frac{ky_1}{z}$ can be thought of as the spatial frequencies. The integral above is also known as the Fraunhofer diffraction integral. The paraxial Helmholtz equation and its solutions

are well established linear Fourier optical theory and allow us to analytically describe our system using Fourier transforms [52, 53].

1.2.2. The Transmission Electron Microscope

The TEM is a highly advanced piece of equipment, incorporating high-brightness electron sources, magnetic lenses, a high vacuum system, cooling mechanisms, aberration correctors, stigmators, monochromators and a system of complex electron detectors. To provide a quick overview of the instrument, an electron source is housed at the top of the instrument where electrons are accelerated to a fraction of the speed of light using electrostatic potentials. Magnetic lenses made of coils focus and direct the electron beam down the evacuated column. The beam interacts with the sample and then is projected on a detector where an image is formed. A labelled illustration of the TEM can be found in Fig. 1.1 The experimental work reported in this dissertation was completed on two TEMs: the Titan at the Center for Advanced Materials Characterization in Oregon (CAMCOR) and TEAM I at the National Center for Electron Microscopy (NCEM).

The Titan housed in CAMCOR is an FEI Titan 80-300 keV scanning TEM with an image aberration corrector. It is equipped with at least six detectors, eleven tunable magnetic lenses, and four aperture holders. TEAM I at NCEM is the same base instrument but has incorporated significant modifications and upgrades. The instrument is double-aberration-corrected (probe and image) with a high-brightness Schottky-field emission “X-FEG” electron source with a monochromator. TEAM I has all the same detectors as the Titan with the addition of three extra detectors for high-resolution, single-electron, high-speed imaging.

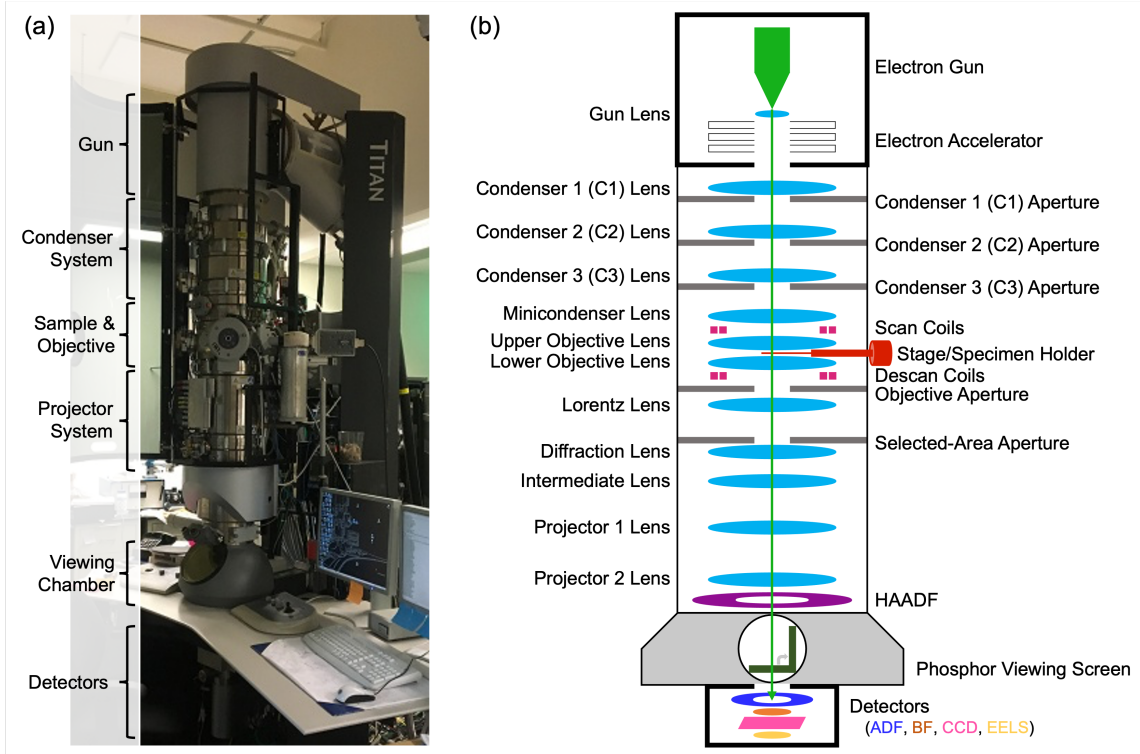


FIGURE 1.1. (a) Photograph and (b) illustration of the transmission electron microscope. The picture (of the Titan in CAMCOR) labels sections of the microscope, while the illustration labels individual apertures, lenses, gun components and detectors. (Illustration is not to scale).

The TEM has two predominant modes of imaging. The primary TEM imaging mode uses a parallel electron beam to illuminate the sample. The plane wave of electrons interacts with the sample, which ripples and modifies the plane wave. The image formed relies on contrast in the probability current at the detector. The contrast of an image arises from mass or thickness differences (inelastic and elastic scattering increases with the atomic number and sample thickness), diffraction (electrons are diffracted in accordance with Bragg's law), and phase variations (samples impart different phase shifts on the passing electron wavefront due to the mean inner potential of the material), described in Section 1.2.3 and 1.2.4.

The other TEM imaging mode is scanning TEM (STEM). Instead of passing a plane wave through the sample, the pre-specimen optics are adjusted to form a focused, narrow probe at the sample. Scan (pre-specimen) and descan (post-specimen) coils are then used to raster the probe across the sample. These scanning coils are deflection coils; they create a magnetic field that forces passing electrons to take a different route down the column of the microscope. The descan coils undo the geometric shift in the transverse direction that the scan coils impart, such that the beam appears stationary at the camera during a scan. STEM images can be formed from distinct electron-sample interactions which results in secondary, backscattered, transmitted, and diffracted electrons (as shown in Fig. 1.2(a)).

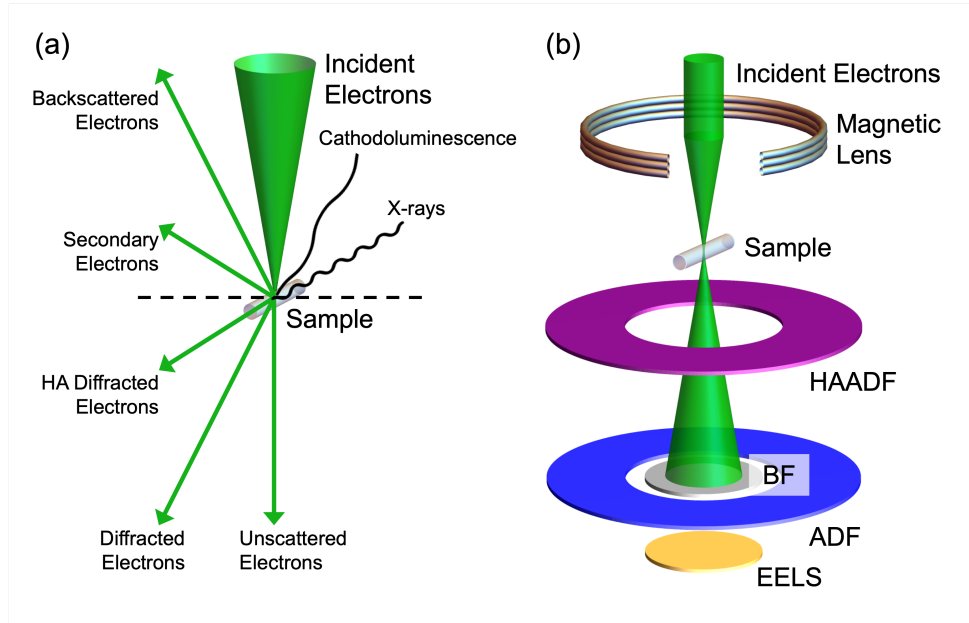


FIGURE 1.2. Schematics of (a) the outcomes of distinct electron-sample interactions and (b) the different detectors for scanning TEM imaging. The detectors shown include the high-angle (HA) annular dark-field (HAADF), bright-field (BF), and annular dark-field detector (ADF) detectors, with the electron energy-loss spectrometer below. All detectors can be retracted and inserted into the microscope's column.

For TEM imaging, there are nine main types of detectors to filter for different electron-sample interactions. The charge-coupled device (CCD) and direct electron detector are most common when using the instrument in TEM mode. Both detectors are mounted at the bottom of the column, made of a 2D pixelated array to form an imaging sensor, and capable of imaging in TEM and STEM mode. In STEM mode, a wider variety of detectors are used: bright-field, annular dark-field, high-angle annular dark-field (HAADF), secondary-electron, backscattered, and energy-dispersive X-ray (EDX) detectors are all common, in addition to the electron energy-loss spectrometer (EELS). Here we focus on the CCD, direct electron detector, bright-field and HAADF detectors as those were the detectors used in this dissertation. In the CCD, electrons hit a scintillator, convert into photons, which are then transferred to a pixelated sensor, via fiberoptic bundle, where the output can be read. The major pitfalls of the CCD are the smearing and blooming effects when overexposed, slow read-out speeds, no direct pixel access and a low detective quantum efficiency (DQE). The CCD measures the intensity or the modulus square of the wavefunction at the detector. Direct electron detectors do not have a scintillator or fiber optic coupling, instead the sensor is directly illuminated with the electron beam. The electrons directly interact with a CCD or complementary metal oxide semiconductor (CMOS), leaving an ionization trail that is directly collected (integrated or counted) in pixels. These detectors count single electrons. A recent upgrade, the Gatan K3, can read out many pixels at high speeds with a high DQE. The largest drawback to direct electron detectors is the large image size; for 4D image stacks (where a 2D diffraction pattern is captured for every pixel position at the sample), the datasets become arduous to analyze and store.

The bright-field detector is made of a scintillator and a photomultiplier. The detector is located at the bottom of the column, near the optical axis of the microscope. As the STEM probe is rastered across the sample, it measures the transmitted, unscattered electrons at each scan location to form the image. The HAADF detector works similarly but instead detects single electrons which have scattered incoherently at high angles. The detector is a disk with a hole in the middle to allow unscattered and low angle scattered electrons through. Because the amount of scattering is related to the atomic number Z , the contrast between different materials is enhanced in comparison to other detectors. The HAADF is located just below the sample and objective aperture, and is made of a scintillator and photomultiplier.

1.2.3. Electron Transmission through Matter

The following theory builds from work detailed by Reimer et. al. [51]. In optics, light travels at a specific speed in vacuum $c = f\lambda_{\text{vac}}$, where f and λ_{vac} are the frequency and wavelength of light in vacuum, respectively. When light travels through a material, the wavelength changes to λ_{mat} and thus the velocity changes to $v = f\lambda_{\text{mat}}$. The ratio of these velocities gives a dimensionless number called the index of refraction, $n = c/v = \lambda_{\text{vac}}/\lambda_{\text{mat}}$. The same concept can be applied to electron optics. When an electron travelling through vacuum passes through a thin material, the electron's velocity is modified by the matter.

Because electrons are charged, the change in velocity is predominantly due to the attractive Coulomb potential of the matter acting on the passing electron. Each atom of the material creates a radially dependent potential that modifies the velocity of each passing electron based on the electron's relative position to the nucleus. Looking

at an electron wave passing through a thin material, we can average the spatially dependent potential across the material and use the mean inner potential (MIP), V_{mip} , of the material to approximate the total velocity change of the electron wave; the modified kinetic energy of the electron being $\tilde{E}_K = E_K - V_{\text{mip}}$. Note that the mean inner potential is independent of the thickness of the material. With this and Eq. 1.4, the refractive index of an electron wave with no magnetic field becomes

$$\begin{aligned} n &= \frac{\lambda_{\text{vac}}}{\lambda_{\text{mat}}} \\ &= \left[\frac{2E_0(E_K - V_{\text{mip}}) + (E_K - V_{\text{mip}})^2}{2E_0E_K + E_K^2} \right]^{1/2}. \end{aligned} \quad (1.9)$$

Assuming a thin material, we find the Taylor series for the index of refraction n around $V_{\text{mip}} = 0$, assuming $V_{\text{mip}} \ll E_0, E_K$. This expansion simplifies the index of refraction to being linear with respect to the V_{mip} ,

$$n = 1 - \frac{V_{\text{mip}}}{E_K} \frac{(E_K + E_0)}{(E_K + 2E_0)} + \mathcal{O}[V_{\text{mip}}^2]. \quad (1.10)$$

Note that for electrons, $n \geq 1$ because the mean inner potential in matter is negative. Using this definition of the index of refraction allows us to calculate the optical path difference $\Delta s = (n - 1)t$ of an electron passing through a material with the mean inner potential V_{mip} and thickness t . From the path difference Δs , we calculate the phase shift ϕ imparted on the electron wave,

$$\begin{aligned} \phi &= \frac{2\pi}{\lambda} \Delta s \\ &= \frac{2\pi}{\lambda} \frac{eV_{\text{mip}}}{E_K} \frac{(E_K + E_0)}{(E_K + 2E_0)} t \\ &= \sigma V_{\text{mip}} t, \end{aligned} \quad (1.11)$$

where $\sigma = \frac{2\pi}{\lambda} \frac{e}{E_K} \frac{(E_K + E_0)}{(E_K + 2E_0)}$ is the object-independent interaction parameter for a free electron with de Broglie wavelength λ , which only depends on the energy of the beam. Using this equation that directly relates the thickness of a material to the imparted phase shift on a passing electron, we can both model phase contrast induced by transparent specimens and design material diffraction gratings for electrons.

However, an electron wavefunction passing near or transmitting through an object can accumulate both phase shifts and amplitude losses. In the weak phase approximation [54], these effects are proportional to the longitudinal extent of the interaction, e.g., the thickness of the material. This can be described by a complex index of refraction $\kappa = \sigma V_{\text{mip}} + i\gamma$, where γ is a material-dependent decay coefficient that models coherent amplitude loss due to high-angle scattering. We use this complex index of refraction model to describe both the diffraction holograms we employ as beamsplitters and the semi-transparent specimens we image.

1.2.4. Electron Diffraction Grating Holograms

The following theory builds from work done by Johnson et. al. [55] and Harvey et. al. [56]. As a specific and practical example of the concept above, consider the spatial phase induced on an electron plane wave passing through a thin grating. Such gratings arbitrarily shape the incoming electron beam and have applications in vortex beams [57], holographic experiments [40], symmetry matching of nanoplasmonic modes [58], as well as myriad others. Assuming a diffraction grating is infinite and periodic in x , the thickness profile $g_a(x, y)$ can be written as

$$g_a(x, y) = d_a - h_a \sum_n c_n^{\{a\}} e^{ink_a x} \quad (1.12)$$

where a indicates the grating of interest, h_a is the membrane thickness, d_a is the groove depth, $k_a = 2\pi/p_a$ where p_a is the grating pitch and $c_n^{\{a\}}$ are the Fourier coefficients for a unit height periodic grating profile. Assume the input wavefunction $\psi_i(x_i, y_i)$ is a time-independent electron plane wave at normal incidence

$$\psi_i(x_i, y_i) = \frac{1}{\sqrt{V}} e^{ikz}, \quad (1.13)$$

with $k = 2\pi/\lambda$ the electron wavevector in vacuum and V the volume of the box in which the electron plane wave is normalized. When the electron plane wave interacts with the diffraction grating, the wavefunction just after the grating positioned at $z = 0$ is

$$\psi_{t1}(x_1, y_1) = \frac{1}{\sqrt{V}} t_1(x_1, y_1) = \frac{1}{\sqrt{V}} e^{i\kappa g_1(x_1, y_1)} \quad (1.14)$$

where $t_1(x_1, y_1)$ is the transmission function of the grating that is defined by the grating's depth profile $g_1(x_1, y_1)$ and κ [59]. κ is the complex wavenumber of the electrons in the material, which depends on the material's mean inner potential, the amplitude decay coefficient, and the energy of the passing electrons (as described in Section 1.2.3).

After the electron plane wave interacts with the grating and picks up a spatially dependent phase, we can use a magnetic lens to focus the wavefunction at an imaging plane further down the microscope. This process allows us to determine the diffraction efficiency of the grating, or how much current is split into each individual path created by the diffraction from the grating. To propagate $\psi_{t1}(x_1, y_1)$ to the far-field, we take a Fourier transform of the wavefunction according to Eq. 1.8 [54]. To simplify the integral, instead of expanding $g_1(x_1, y_1)$ as a Fourier series as shown

in Eq. 1.12, we can expand the wavefunction $\psi_{t1}(x_1, y_1)$,

$$\psi_{t1}(x_1, y_1) \propto e^{i\kappa g_1(x_1, y_1)} = \sum_n \tilde{c}_n^{\{1\}} e^{in k_1 x_1} \quad (1.15)$$

where $\tilde{c}_n^{\{1\}}$ are the Fourier coefficients describing the field immediately after a specific grating denoted by the superscript $\{1\}$. Thus, the wavefunction in the far field where probes form, $\psi_{p1}(x_2, y_2)$, is simply

$$\psi_{p1}(x_2, y_2) \propto \sum_n \tilde{c}_n^{\{1\}} \delta(q_{x_2} - n k_1). \quad (1.16)$$

where $q_{x_2} = k x_2 / z_2$.

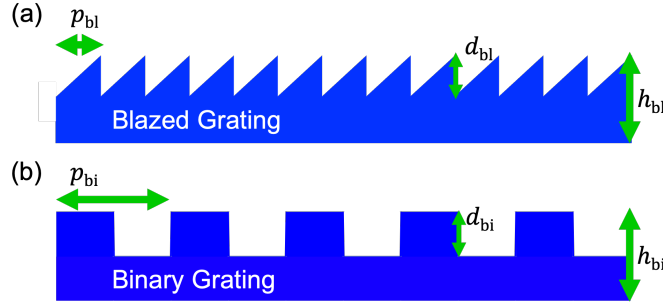


FIGURE 1.3. Illustrations of the cross-sectional profiles of two gratings with different groove shapes: (a) a blazed grating and (b) a binary grating.

In this dissertation, we use two periodic grating profiles, a blazed and binary profile (as shown in Fig. 1.3). Here we ignore the finite extent of the gratings. To find the Fourier coefficients of the fields immediately after a binary grating, we note that binary gratings consist of two mill depths, translating to two constant phase values in the transmission function. Thus the binary transmission function, $t_{bi}(x, y)$,

can be written as a sum,

$$\begin{aligned}
t_{\text{bi}}(x, y) &= e^{i\kappa g_{\text{bi}}(x)} \\
&= e^{i\kappa(h-d)} \sum_n c_n^{\{\text{bi}\}} e^{ink_{\text{bi}}x} + e^{i\kappa h} \left(1 - \sum_n c_n^{\{\text{bi}\}} e^{ink_{\text{bi}}x} \right) \\
&= e^{i\kappa h} + (e^{i\kappa(h-d)} - e^{i\kappa h}) \sum_n c_n^{\{\text{bi}\}} e^{ink_{\text{bi}}x} \\
&= \sum_n \tilde{c}_n^{\{\text{bi}\}} e^{ink_{\text{bi}}x}.
\end{aligned} \tag{1.17}$$

Solving for the Fourier coefficients of the field, we find

$$\tilde{c}_n^{\{\text{bi}\}} = \begin{cases} (e^{i\kappa(h-d)} - e^{i\kappa h}) c_n^{\{\text{bi}\}} + e^{i\kappa h} & \text{if } n = 0 \\ (e^{i\kappa(h-d)} - e^{i\kappa h}) c_n^{\{\text{bi}\}} & \text{if } n \neq 0 \end{cases} \tag{1.18}$$

where $c_n^{\{\text{bi}\}}$ describe the Fourier coefficients for a unit height binary profile, which are defined as

$$c_n^{\{\text{bi}\}} = \begin{cases} b & \text{if } n = 0 \\ \frac{1}{\pi n} \sin(\pi b n) & \text{if } n \neq 0 \end{cases} \tag{1.19}$$

These coefficients depend on the duty cycle b , and can easily be inserted into Eq. 1.16 to determine the diffraction efficiency of the grating.

To find the Fourier coefficients of the fields immediately after a blazed grating, we expand the transmission function as a Fourier series

$$t_{\text{bl}}(x) = e^{i\kappa g_{\text{bl}}(x)} = \sum_n \tilde{c}_n^{\{\text{bl}\}} e^{ink_{\text{bl}}x} \tag{1.20}$$

where $c_n^{\{\text{bl}\}}$ describe the Fourier coefficients for a unit height blazed profile. We rearrange the equation to find

$$\tilde{c}_n^{\text{bl}} = \frac{1}{p_{\text{bl}}} \int_0^{p_{\text{bl}}} e^{i(\kappa g_{\text{bl}}(x) - nk_{\text{bl}}x)} dx. \quad (1.21)$$

With a groove depth d_{bl} , membrane thickness h_{bl} , and a grating period p_{bl} , we can rewrite the thickness profile assuming $x \in (0, p_{\text{bl}}]$ as $g_{\text{bl}}(x) = d_{\text{bl}} - h_{\text{bl}}x/p_{\text{bl}}$. Inserting $g_{\text{bl}}(x)$ into Eq. 1.21, we find

$$\tilde{c}_n^{\text{bl}} = e^{i(\kappa(d_{\text{bl}} + h_{\text{bl}}/2) + n\pi)} \text{sinc}\left(\frac{\kappa h_{\text{bl}} + 2n\pi}{2}\right). \quad (1.22)$$

Using these Fourier coefficients and Eq. 1.16, we can determine the diffraction efficiency of each profile given the groove depth, membrane thickness, membrane material and accelerating voltage of the microscope. If the groove depth is tuned to impart a π phase difference between the troughs and crests of the thickness profile, the binary profile creates two dominant beams in the ± 1 diffraction orders. The blazed profile on the other hand, skews the diffraction pattern such that the 0 and -1 orders are dominant, as shown in Fig. 1.4.

To nanofabricate blazed and binary diffraction grating holograms, we use gas-assisted etching in the dual-beam focused ion beam (FIB) [31]. We mill into 30 or 50 nm thick silicon nitride membranes coated with an approximately 5 nm thick charge alleviation layer. Using an 80 keV electron beam, $\kappa = i0.008 - \pi/21\text{nm}^{-1}$ is the complex wavenumber for our diffraction gratings [31]. Gas assisted etching quickens the milling process and mitigates material redepositing. Nanofabricated diffraction grating holograms have proven to controllably shape and manipulate the electron beam in a TEM [31].

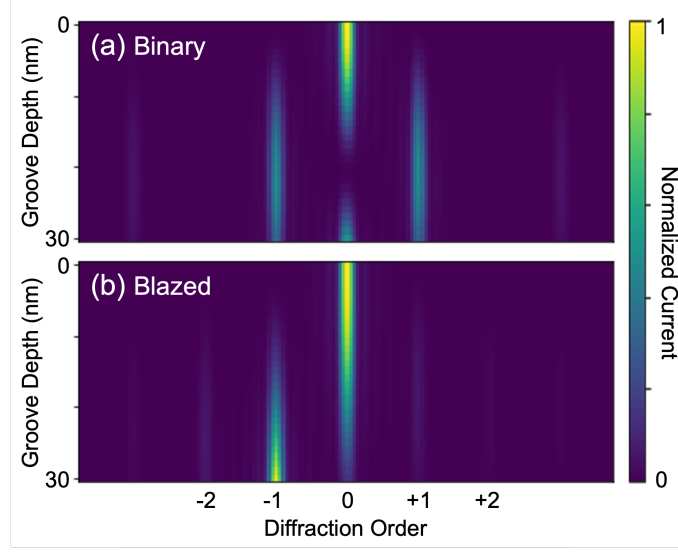


FIGURE 1.4. Simulations of the diffraction efficiency of an ideal (a) binary and (b) blazed grating with no apertures. This simulation is for an 80 keV electron beam and 30 nm thick silicon nitride membrane. A mill depth of 21 nm is associated with a π phase shift.

1.3. Dissertation Outline

In this dissertation, we will cover the construction of a novel electron interferometer, the first interaction-free measurement with electrons, and an optimized interferometer for interaction-free measurements. Chapter 2 discusses the construction of a path-separated, two-grating free electron Mach-Zehnder interferometer in a conventional TEM. As proof of the interferometer's capabilities, we then use the interferometer for quantitative phase imaging of a nanoparticle and mapping the electrostatic potential of a nanowire. Chapter 3 applies this interferometer to make the first interaction-free measurement with the electron. We explore the quantum protocol, discuss the limitations of the current apparatus, and consider possibilities of applying this feature to quantum-enhanced, low-damage imaging in the future. In chapter 4, we compare the two-grating interaction-free measurement technique to an alternative holography scheme, proving that the two-

grating interferometer is truly advantageous. Subsequently, the interferometer is optimized in Chapter 5 for high efficiency interaction-free measurements with single electrons. The experiment and simulations in this chapter create a viable platform for low-damage imaging in the future. Finally, Chapter 6 highlights the theoretical and experimental achievements in this dissertation, summarizing the dissertation and describing impactful, future directions tangential to this work.

This dissertation contains previously published and unpublished material.

CHAPTER II

SCANNING TWO-GRATING FREE ELECTRON MACH-ZEHNDER INTERFEROMETER

In pursuit of realizing the interaction-free measurement with electrons, we must construct a suitable electron interferometer. In this chapter, we design and develop an electron Mach-Zehnder interferometer in a conventional transmission electron microscope. To exhibit its capabilities, we employ the interferometer to map the electrostatic potential of a grounded versus insulated silver nanowire, and quantitatively measure the phase of a latex nanoparticle.

Notes for Ch. III:

This chapter is adapted from the published research article:

[55] **A. E. Turner***, C. W. Johnson*, and B. J. McMorran. Scanning two-grating free electron Mach-Zehnder interferometer. *Physical Review Research*, 3(4):043009, 2021.

*CWJ and I were co-authors with equal contributions. CWJ and I had equal parts in writing the paper, developing the theory, designing and executing the experiments, and analyzing the data. BJM provided experimental guidance and significant edits to the paper.

We demonstrate a two-grating free-electron Mach-Zehnder interferometer constructed in a transmission electron microscope. A symmetric binary phase grating and condenser lens system forms two spatially separated, focused probes at the sample which can be scanned while maintaining alignment. The two paths

interfere at a second grating, creating constructive or destructive interference in the output beams. This interferometer has many notable features: positionable probe beams, large path separations relative to beam width, continuously tunable relative phase between paths, and real-time phase information. Here we use the electron interferometer to measure the relative phase shifts imparted to the electron probes by electrostatic potentials as well as a demonstration of quantitative nanoscale phase imaging of a polystyrene latex nanoparticle.

2.1. Introduction

Electron holography and interferometry can enable nanoscale phase imaging [37, 60], the exploration of the Aharonov-Bohm effect [29, 61], interaction-free measurements and quantum electron microscopy [46, 47, 62], the measurement of coherence properties [63, 64, 65], quantum state tomography [66, 67], and the coherent control of the free-electron wavefunction [68]. While interferometry is widely used in optics and photonics fields such as astronomy [69], optical metrology [70], and quantum optics [71], electron interferometry has advanced at a slower pace, partially due to a lack of basic optical elements such as beamsplitters and mirrors that can be used to build a versatile system. Here we use two diffraction gratings as beamsplitters in a conventional transmission electron microscope (TEM) to create a flexible, path-separated Mach-Zehnder interferometer for free electrons, emulating the canonical example for phase-sensitive interferometry.

Electron interferometers are currently limited and defined by the electron optical elements used to construct the interferometer. Electrostatic mirrors for free electrons show great promise, but are in their early development [72, 73, 74]. Beamsplitters have existed for decades, the most prevalent being electrostatic biprisms [75] that

divide wavefronts. Biprisms generate interferograms from which the phase of the sample can be extracted after image processing. However, they require high spatial coherence [76]; thus, they are inherently limited by modern electron emission sources that are only partially coherent. Using recently improved diffraction gratings as amplitude dividing beamsplitters [34, 77, 78] to create spatially separated paths evades the high spatial coherence requirement, if the separated paths are perfectly overlapped after the sample [79]. We note that amplitude division with microwave chip beamsplitters has also been demonstrated, but is not yet practical for electrons with kinetic energies above 200 eV [35].

Various demonstrations of matterwave interferometry have proceeded for decades [80]. Path-separated Mach-Zehnder interferometers specifically have been demonstrated for different kinds of matterwaves, including neutrons [81], atoms [82], BECs [83], conduction and quantum Hall valley electrons in 2D devices [84, 85], and SQUIDS [86]. Free electron Mach-Zehnder interferometers with discrete outputs have predominantly been constructed using nanofabricated diffraction gratings or crystals as beamsplitters. Two-plane electron interferometers fabricated from monolithic uniform crystals [87, 88] and three-plane Mach-Zehnder interferometers with discrete crystal planes [80, 89] have been demonstrated. A three-grating Mach-Zehnder interferometer for free electrons was demonstrated with nanofabricated transmission amplitude gratings [90]. These free electron interferometer variants have advanced electron interferometry and could potentially be used for imaging applications, but the compact longitudinal designs significantly restrict the types of experiments that can be performed.

Here we demonstrate a two-grating electron Mach-Zehnder interferometer (2GeMZI) constructed inside a conventional TEM that provides clearly defined

isolated probes, arbitrary probe phase shifts, and scanning/imaging capabilities. Furthermore, since this is a scanning probe technique, the magnification can be changed without adjusting the setup, unlike electron holography. This is accomplished by placing diffraction gratings in apertures above and below the specimen plane of a TEM operated in scanning TEM (STEM) mode. The small deviations of the lens and aperture positions from the nominal STEM settings allow us to maintain the high resolution imaging capabilities afforded by the TEM while retaining the precise interferometer alignments. To demonstrate the phase sensitivity of the 2GeMZI we map electrostatic potential differences in the vicinity of both grounded and charged silver nanorods and demonstrate quantitative nanoscale phase imaging of a spherical latex nanoparticle.

2.2. Theoretical Description of Electron Interferometer

In this electron interferometer, nanoscale diffraction gratings are used as amplitude-dividing beamsplitters and the standard TEM imaging optics are used to separate, scan, and recombine the beams. An illustration of this system can be seen in Figure 2.1 where successive transverse planes are defined in relation to the previous plane by Fourier transform. The first beamsplitter in the interferometer is a grating (G1) in the x plane. The electron beam then passes through a lens (L1) that focuses and projects path path separated probes at the specimen, the k plane. The second lens (L2) projects an image of G1 onto the second grating (G2) in the $\mathbf{x}'$ plane. Finally, the third lens (L3) projects the discrete outputs of the interferometer into the $\mathbf{k}'$ plane at the detectors. Here we describe how the evolution of the electron wavefunction can be modeled throughout the interferometer.

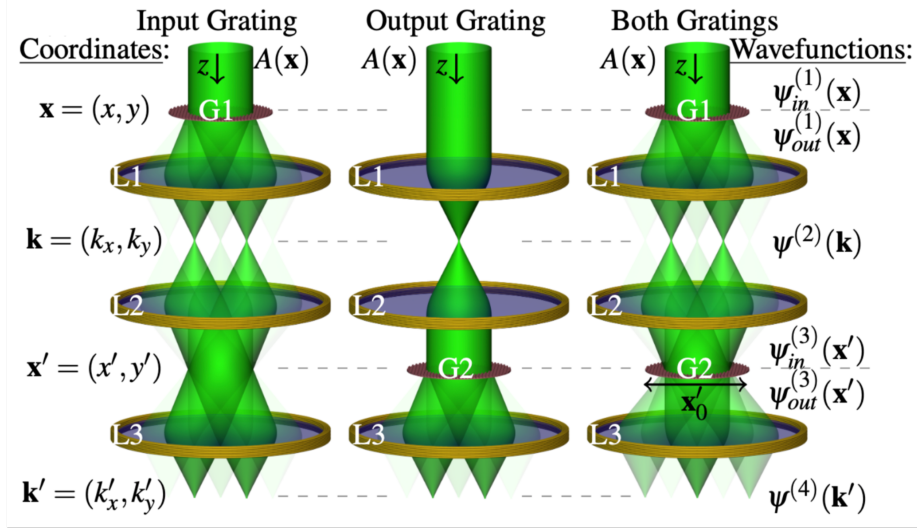


FIGURE 2.1. Diagram for 2GeMZI showing definitions of the different transverse planes as well as labels for the transverse wavefunctions in each plane, the magnetic lenses (L1, L2, L3), gratings (G1, G2), and beam-defining aperture $A(\mathbf{x})$ are also shown. The three different images depict the 3 cases of when the different gratings are inserted or removed. The position of the second grating relative to the beam is denoted as $\mathbf{x}'_0$.

The evolution of an electron wavefunction propagating through free space, neglecting spin, can be described by the Schrödinger equation with relativistic corrections. In a typical TEM with a field emission electron source, electrons are accelerated to beam energies of 40 to 300 keV with a 500 meV energy spread and the electron beam has very small beam divergence (on the order of one milliradian) [91]. The evolution of electron wavefunctions can therefore be modeled using Fresnel and Fourier optical theory [54], consistent with the Schrödinger equation while assuming the electrons are largely quasimonochromatic, non-interacting, and collimated. We use the complex index of refraction model (as described in Section 1.2.3) to describe the accumulated phase shifts and amplitude losses of an electron wavefunction passing near or transmitting through an object. The complex index of refraction

is $\kappa = \sigma V_{\text{mip}} + i\gamma$. This theory can describe an electron beam interacting with both a diffraction grating hologram and a specimen we image.

The transverse electron wavefunction incident on the input grating of the interferometer is assumed to be a plane wave with an outer edge defined by an aperture $\psi_{in}^{\{1\}}(\mathbf{x}) = A(\mathbf{x})$. When transmitted through a grating the wavefunction is modified by its transmission function

$$\psi_{out}^{(1)}(\mathbf{x}) = A(\mathbf{x})e^{i\kappa t_1(\mathbf{x})}, \quad (2.1)$$

where $t_1(\mathbf{x})$ is the periodic thickness profile of the grating. For a straight grating with pitch p_1 and diffraction wavevector $\mathbf{k}_1 = 2\pi/p_1$, we can expand Eq. (2.1) by the Fourier series representation of the exponential term

$$\psi_{out}^{(1)}(\mathbf{x}) = A(\mathbf{x}) \sum_n c_n^{\{1\}} e^{in\mathbf{k}_1 \cdot \mathbf{x}}, \quad (2.2)$$

where the Fourier coefficients are given by

$$c_n^{\{1\}} = \frac{1}{p_1} \int_0^{p_1} d\tilde{x} e^{i\kappa t_1(\tilde{x}) - in|\mathbf{k}_1|\tilde{x}} \quad (2.3)$$

where $\tilde{x}$ is the coordinate in the direction of the grating pitch. The unnormalized probes in the far-field after G1, focused in the back focal plane of the L1, can be expressed as

$$\psi^{(2)}(\mathbf{k}) \propto \sum_n c_n^{\{1\}} \tilde{A}(\mathbf{k} - n\mathbf{k}_1), \quad (2.4)$$

where $\tilde{A}$ is the Fourier transformation of A . Upon propagation to the $\mathbf{x}'$ plane the wavefunction $\psi_{in}^{(3)}$ is an image of $\psi_{out}^{(1)}$. If G2 is allowed to translate by an amount $\mathbf{x}'_0$, then the output of the second grating is

$$\psi_{out}^{(3)}(\mathbf{x}') \propto A(\mathbf{x}') \sum_{n,n'} c_n^{\{1\}*} c_{n'}^{\{2\}} e^{in'\mathbf{k}_2 \cdot (\mathbf{x}' - \mathbf{x}'_0) - in\mathbf{k}_1 \cdot \mathbf{x}'}, \quad (2.5)$$

where $c_n^{\{2\}}$ and $\mathbf{k}_2$ are similarly defined for the second grating. When the image of the input grating is projected onto the output grating with the same pitch and orientation, i.e., $\mathbf{k}_0 = \mathbf{k}_1 = \mathbf{k}_2$, the wavefunction in the final $\mathbf{k}'$ plane can be written as

$$\psi^{(4)}(\mathbf{k}') \propto \sum_{n,n'} c_n^{\{1\}*} c_{n'}^{\{2\}} e^{-in'\mathbf{k}_0 \cdot \mathbf{x}'_0} \tilde{A}(\mathbf{k}' - (n - n')\mathbf{k}_0). \quad (2.6)$$

The output of the interferometer is divided into distinct $m = n - n'$ diffraction orders. Using this to re-index the double sum, we can write Eq. (2.6) as a sum of output diffraction orders

$$\begin{aligned} \psi^{(4)}(\mathbf{k}') &\propto \sum_m \left(\sum_n c_n^{\{1\}*} c_{n-m}^{\{2\}} e^{-i(n-m)\mathbf{k}_0 \cdot \mathbf{x}'_0} \tilde{A}(\mathbf{k}' - m\mathbf{k}_0) \right) \\ &= \sum_m \psi_m^{(4)}(\mathbf{k}'). \end{aligned} \quad (2.7)$$

When the gratings are symmetric and put a majority of the the transmitted intensity into the ± 1 diffraction orders, i.e., $|c_{\pm 1}| \gg |c_{|n| \neq 1}|$ and $|c_{+1}| = |c_{-1}|$, then the 0th output diffraction order where $n = n'$, up to a global phase, is

$$\psi_0^{(4)}(\mathbf{k}') \propto |c_1^{\{1\}*} c_1^{\{2\}}| \left(1 + e^{-2i\mathbf{k}_0 \cdot \mathbf{x}'_0} \right) \tilde{A}(\mathbf{k}') + \dots \quad (2.8)$$

If the probes are allowed to scan in the specimen plane, $\mathbf{k} \rightarrow \mathbf{k} + \mathbf{k}_s$, while passing through some electrostatic potential $V(\mathbf{k}, z)$, we can use the weak phase approximation and write the phase accumulated by each probe as being proportional to the projected potential along the z direction $\Phi(\mathbf{k}) = \sigma \int dz V(\mathbf{k}, z)$.

Incorporating the phase due to a potential, the 0th order output of the interferometer is approximately

$$\psi_0^{(4)}(\mathbf{k}') \propto \left(1 + e^{i\varphi(\mathbf{k}_s, \mathbf{x}'_0)}\right) \tilde{A}(\mathbf{k}'), \quad (2.9)$$

where the total phase difference between the two highest intensity probes is

$$\varphi(\mathbf{k}_s, \mathbf{x}'_0) = -2\mathbf{k}_0 \cdot \mathbf{x}'_0 + \Phi(\mathbf{k}_s - \mathbf{k}_0) - \Phi(\mathbf{k}_s + \mathbf{k}_0). \quad (2.10)$$

We should note that the static potential V can have contributions that extend into the vacuum region due to the build up of surface charge on a material as well as inside materials from the mean inner potential as was defined in κ (as described in Section 1.2.3).

2.3. Construction of Interferometer

To realize such an electron interferometer, we fabricated two arrays of 350 nm pitch, 30 μm diameter binary phase gratings that were each nanofabricated onto a 250 $\times$ 250 μm^2 , 30 nm thick, free-standing Si_3N_4 membrane using focused ion beam (FIB) gas-assisted etching [31]. To aid with alignment, the gratings were patterned at multiple orientations in a 6 $\times$ 6 array. In an image-corrected 80-300 keV FEI Titan TEM, one grating array was installed in the second condenser aperture and used as the first grating G1. The second grating array was installed in the post-

specimen selected area aperture as the beam-combining grating G2. Both of these grating apertures only take one of many aperture slots, so their installation does not impede the conventional functionalities of the TEM. The TEM was operated at 80 keV in STEM mode with approximately a 1 mrad convergence angle. An independently positionable circular aperture at the third condenser lens was used to select the output of a single grating out of the widely illuminated array of gratings. The diffraction orders created by the selected input grating were focused to narrow probes at the sample, the $\mathbf{k}$ plane. The lenses were used in free lens control with assistance of the “Lorentz” lens in the image corrector to form a correctly magnified, oriented, and in-focus image of G1 onto G2. Finally, the post-G2 projection lenses were used to image the far-field diffraction pattern from both beamsplitters onto the detectors at the bottom of the TEM column. The relative grating shift parameter $\mathbf{x}'_0$ was controlled by the diffraction alignment coils in the image corrector that shifted the image of G1 relative to G2 allowing for arbitrary relative phase shifts between the two specimen plane probes in the interferometer output. The $\mathbf{k}$ plane probes at the specimen section could be set to have up to 1 μm separation between the ± 1 probes, however the spot size increases proportionally with the probe separation. Using these beamsplitter gratings, the ratio between separation between the paths and the width of the probes is approximately 20.

The magnitude of the $\{c_n^{\{1\}}\}$ and $\{c_n^{\{2\}}\}$ Fourier coefficients were measured by inserting only one of G1 or G2 at a time while collecting an image of the probe intensities with a scintillator fiber-coupled CCD. A single diffraction order was integrated and divided by the total integrated intensity to determine normalized diffraction efficiencies $|c_n^{\{1\}}|^2$ and $|c_n^{\{2\}}|^2$; the measured grating outputs are shown in Fig. 2.2(a,b). Ideally, the gratings would be perfect binary gratings with 50% groove

duty cycle with up to 40.5% of the transmitted intensity going into the ± 1 probes. However, edge rounding and non-ideal duty cycles from nanofabrication with a finite width ion beam as well as over and under milling from the ideal groove depth caused deviations from the optimal diffraction efficiency. Even so, we were able to achieve dominant ± 1 coefficients allowing for efficient two-beam scanning in the 2GeMZI.

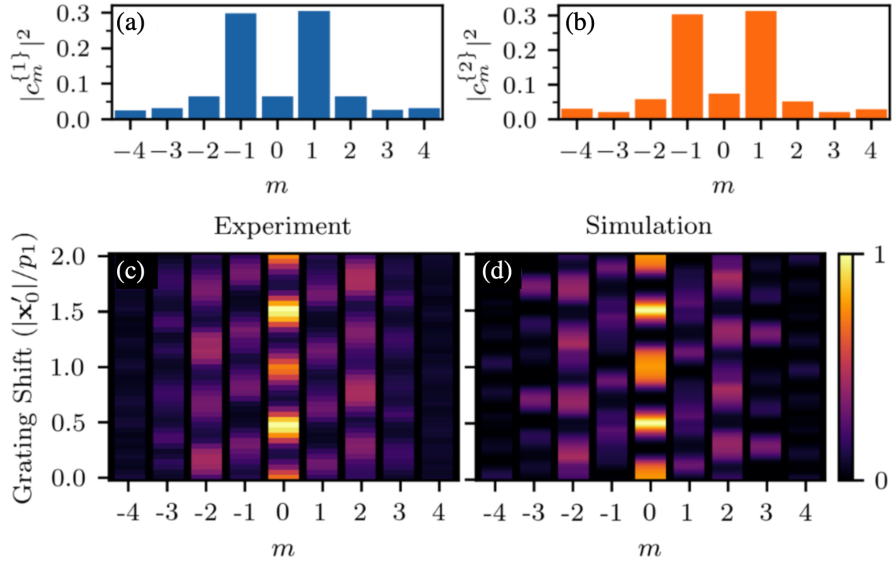


FIGURE 2.2. (a) Diffraction efficiencies of G1, measured when G2 is removed. (b) Diffraction efficiencies of G2, measured when G1 is removed. (c) Measured output intensities of the 2GeMZI for different relative grating shifts normalized to maximum output intensity. (d) Simulated output intensities of the 2GeMZI for different relative grating shifts.

With G1 and G2 both inserted, we collected CCD images of the output beams for different values of $\mathbf{x}'_0$ by tuning the previously mentioned diffraction alignment coils. The measured output intensities are in good agreement with the expected result, Fig. 2.2(c,d). Without the presence of an electrostatic potential, the intensity of the 0th order interferometer output is expected to be proportional to the modulus square of Equation (2.9), i.e., sinusoidal in the argument $\mathbf{k}_0 \cdot \mathbf{x}'_0$. We see this dependence

in the experimental data, but it is also accompanied by a beating at half the spatial frequency $\mathbf{k}_0/2$. This amplitude beating is caused by a combination of grating duty cycle mismatch and contributions of the higher order terms from the sum of probe coefficients and is confirmed in the simulations that use simulated grating groove profiles that produce the experimentally measured far-field diffraction patterns in Fig. 2.2(a,b). Due to these higher order effects, the fringe visibility $\mathcal{V} = (I_{max} - I_{min})/(I_{max} + I_{min})$ is $\mathcal{V} = 0.76$ when the output is aligned for maximally destructive interference and $\mathcal{V} = 0.82$ when aligned for maximally constructive interference. The maximum theoretical fringe visibility $\mathcal{V} = 1$ can be approached through the continued improvement of gratings.

Scan and descan coils can be used to raster both beams across a scan region up to 3 times the probe separation while keeping the electron interference pattern (the image of G1) stationary on G2, ensuring the interferometer output was constant while beams were scanned across a flat phase region. The scan/descan system is independent of the diffraction alignment used to control $\mathbf{x}'_0$, so the relative phases between the interferometer probes remains constant throughout the scan. While scanning, a bright field (BF) monolithic detector can be inserted such that it is illuminated by only the 0th interferometer output order. With this configuration we can perform direct phase imaging with the 2GeMZI. The relative phase between the probes at any scan position in the specimen plane $\mathbf{k}_s$ can be reconstructed from the intensity of the interferometer output

$$I_0^{(4)}(\mathbf{k}_s, \mathbf{x}'_0) \approx \langle I_0^{(4)} \rangle [1 + \mathcal{V} \cos(\varphi(\mathbf{k}_s, \mathbf{x}'_0))]. \quad (2.11)$$

It should be noted that phase shifts due to path length differences during the scan certainly exist, but are much smaller than the longitudinal coherence length, which

for 80 keV electrons with a 500 meV energy spread is on the order of a micron. Throughout a scan, the scan/descan coils maintain the path length difference within a fraction of the electron’s wavelength, as can be seen in flat intensity of the large vacuum scan region of Fig. 2.4(a), and so we can safely neglect this contribution to the relative probe phase.

2.4. Electrostatic Potentials in the Interferometer

One application of the 2GeMZI is mapping electrostatic potentials in real-time. In the last 30 years, quantitative potential maps measured with electron holography have been used to accurately determine charge distributions of nanoscale devices [92, 93]. However streamlined this method has become, it still requires image post-processing or proprietary live analysis software [94, 95]. While the high spatial and phase resolution of electron holography cannot yet be matched by the 2GeMZI in this initial demonstration, the interferometer provides a live interpretation of the electrostatic potential, whereas electron holography requires post-scan image processing. Each pixel’s intensity indicates the electrostatic potential difference at the two probe positions in the specimen plane. Here we use the 2GeMZI to show the fringes in raw interferometric BF images of a grounded and insulated vertical nanorod.

We fabricated two 550 nm tall vertical silver nanorods surrounded by a vacuum window; one with a conductive lead to ground and the other electrically insulated such that it would support a surface charge throughout an imaging scan. Using a Ga^+ FIB operated at 30 keV with a 7.7 pA beam current, we fabricated vertical silver nanorods on a nitride cantilever from a 100 nm thick silver film thermally deposited on a 50 nm thick Si_3N_4 membrane in the following steps:

- (i) Mill completely through silver and nitride forming cylindrical silver bead along nitride tether.
- (ii) Mill only through silver defining bottom edge of nanorod, optionally leaving a small lead of silver between the rod and the film.
- (iii) Mill completely through silver and nitride defining top edge of rod and nitride cantilever.
- (iv) Flip membrane over and raster FIB over the bare nitride section of cantilever to induce bending until the nanorod is vertical, normal to the silver film and nitride membrane [96].

These nanorods were fabricated with a clear vacuum region around the rods for easy access for imaging in the 2GeMZI. One of the two nanorods was given a small lead to ground to the rest of the silver film, while the other was electrically insulated by removing all of the silver between the nanorod and the film. An illustration of the fabrication process can be seen in Figure 2.3(a) accompanied by scanning electron microscopy (SEM) micrographs displaying the sample geometry and orientation, Fig. 2.3(b-d).

This device was inserted in the specimen plane of the 2GeMZI which was adjusted to have a larger path separation of 500 nm with a probe size of about 25 nm. We recorded interferometric BF images over a scan region of $1.5 \times 1.5 \mu\text{m}^2$ of the grounded and insulated nanorods shown in Fig. 2.4(a,b). The electric potential from the semiconductor nitride substrate cantilever and the grounded nanorod was small; as shown in Fig. 2.4(a), the interferometer output was only modulated close to the surface of the nitride and the relative phase between two probes in the vacuum region far away from the object is constant. However, the electrically insulated

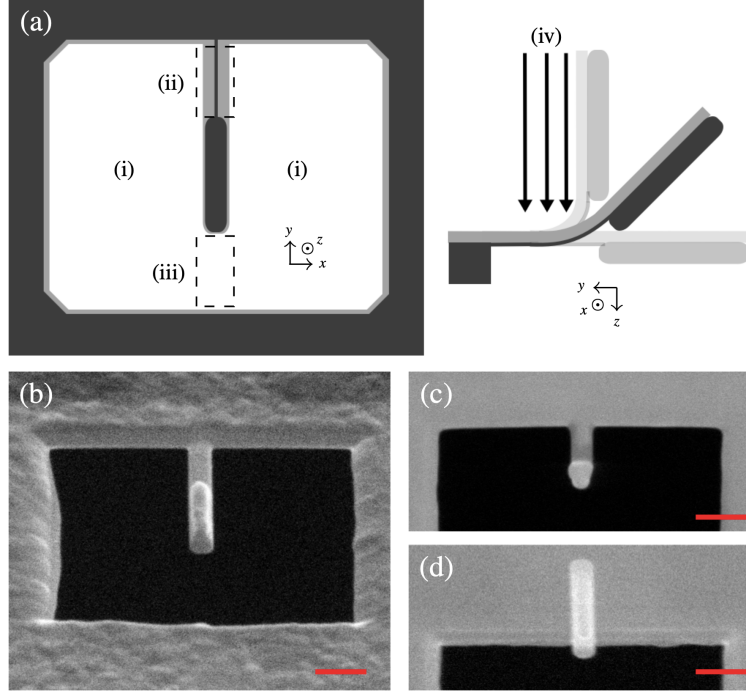


FIGURE 2.3. (a) Vertical silver nanorod FIB nanofabrication steps (i-iv). (b) SEM micrograph at 52° tilt after fabrication steps (i-iii). SEM micrographs after fabrication steps (i-iv) imaging from bottom of membrane at (c) 0° tilt and (d) 52° tilt. All scale bars are 200 nm.

nanorod charged when exposed to the beam until reaching a static surface charge, creating a larger static potential. The resulting interference fringes for the probe potential differences can clearly be seen far into the surrounding vacuum region, as shown in Fig. 2.4(b). Close to the nanorod, the larger gradient in electrostatic potential induces a phase that varies within the width of the probes, resulting in a loss of fringe visibility.

We use a $1/r$ potential to approximate the nonlinear monotonically decreasing behavior that is expected surrounding a charged vertical nanorod (Fig. 2.4(c,d)). In the experimental interferometric BF images there is elongation in the probe

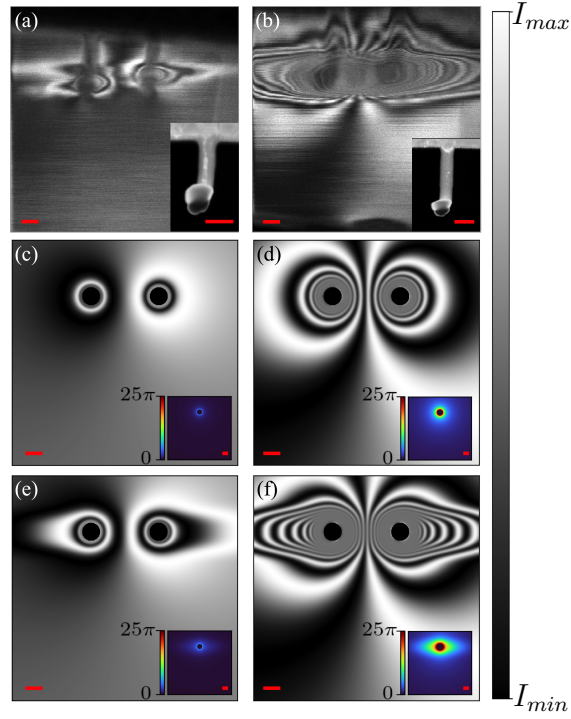


FIGURE 2.4. Bright field (BF) STEM images showing the interferometer output spatially oscillating between the intensity minimum and maximum for (a) grounded vertical Ag nanorod, (b) insulated vertical Ag nanorod. Insets are STEM HAADF images of each nanorod. (c,d) Simulated 2GeMZI output for 2 probes passing through a $1/r$ electrostatic potential where (d) has 10 times the charge of (c). (e,f) Same as (c,d) but with a horizontally elongated Gaussian included with the potential to simulate the increased induced charging by the incidence of the higher diffraction orders. Insets in (c-f) are the projected potentials used to create the simulated images. All scale bars are 100 nm.

separation direction that is not shown in this simple model, but this can be explained by an increased surface charge on the nanorod when the higher order probes ($m \neq \pm 1$) are incident on the specimen. Including a Gaussian background to the $1/r$ potential elongated in the probe separation direction to account for this increase in surface charge creates an interference pattern that is qualitatively consistent with the experimental images (Fig. 2.4(e,f)).

This initial demonstration shows that the 2GeMZI is sensitive to differences in electrostatic potentials at the locations of the two probes: spatially varying

electrostatic potentials impart a phase to the specimen probes and the phase difference modulates the intensity at the BF detector. With moderate improvements, the real-time interferometric BF images can provide nanoscale maps of the static projected potential with higher phase resolution. Some challenges still need to be overcome to achieve quantitative real-time potential mapping, especially for determining static charge distributions. First, is to ensure the static charge is independent of the scanning probes which can be accomplished with adequately grounded, conductive materials. Second, is to limit the samples of interest such that the spatial extent of the potential does not extend over multiple probes. This ensures there is a reference probe and a measurement probe with a phase difference that is directly proportional to the projected potential. This second point can be relaxed with a careful analysis and full understanding of the accumulated relative phases between all the probes as they are scanned into the potential. Third, smaller probe sizes must be used to probe potentials with large spatial gradients.

2.5. Quantitative Phase Imaging of a Latex Nanoparticle

To demonstrate an example of quantitative phase imaging with the 2GeMZI, we imaged polystyrene latex spherical nanoparticles on a suspended single layer of graphene. Polystyrene latex has a well-characterized mean inner potential, $V_{\text{mip}}^{\text{latex}} = 8.5 \pm 0.7$ volts [97], and small amplitude decay coefficient, $\gamma^{\text{latex}} \approx 0$. We use a nanosphere with a diameter of 60 nm. The ratio of probe size and separation is fixed, but can be simultaneously tuned by changing the lens magnification settings. Here the 2GeMZI was tuned for a probe separation of 92 ± 2 nm and a focused beam width of approximately 5 nm such that one of the two 2GeMZI probes could be scanned through the nanosphere while the other passed through uniform graphene

in the specimen plane (Fig. 2.5). The phase imparted by the graphene is expected to be about 45 mrad [98]. Individual atoms are not resolvable at the resolution in this initial demonstration, so we treat the sample as a homogeneous latex sphere with a small uniform phase background. Another benefit of the graphene substrate is that it efficiently alleviates charge, allowing us to disregard extraneous static fields due to sample charging and only consider the mean inner potential from the latex as the source of the probe phase shift. Due to the size of the nanoparticle in comparison to the large probe separation, the elimination of electrostatic fields, and the negligible decay coefficient of latex, we can assume the phase difference between the two probes is

$$\varphi(\mathbf{k}_s, \mathbf{x}'_0) = -2\mathbf{k}_0 \cdot \mathbf{x}'_0 + \sigma V_{\text{mip}}^{\text{latex}} t_{\text{sph}}(\mathbf{k}_s). \quad (2.12)$$

The first term, $2\mathbf{k}_0 \cdot \mathbf{x}'_0$, is due to the interferometer alignment and the last, $\sigma V_{\text{mip}}^{\text{latex}} t_{\text{sph}}(\mathbf{k}_s)$, is the phase accumulated by the probe passing through the sphere of projected thickness $t_{\text{sph}}(\mathbf{k}_s)$ at the scan location $\mathbf{k}_s$. σ is a beam dependent parameter as defined in Section 1.2.3. The latex spheres were interferometrically imaged by scanning the probes over a $100 \times 100 \text{ nm}^2$ scan region while the 0th order interferometer output was recorded by the BF detector. Each BF image is smoothed using a Gaussian filter and then normalized. The normalization limits are determined by the constructive and destructive interferometer outputs when both probes are in vacuum.

We recovered the phase image of the 60 nm diameter latex nanoparticle from two interferometric BF images, one with the interferometer initially aligned for maximally constructive 0th order output, $2\mathbf{k}_0 \cdot \mathbf{x}'_0 = \pi$ (Fig. 2.5(ii)) and one aligned

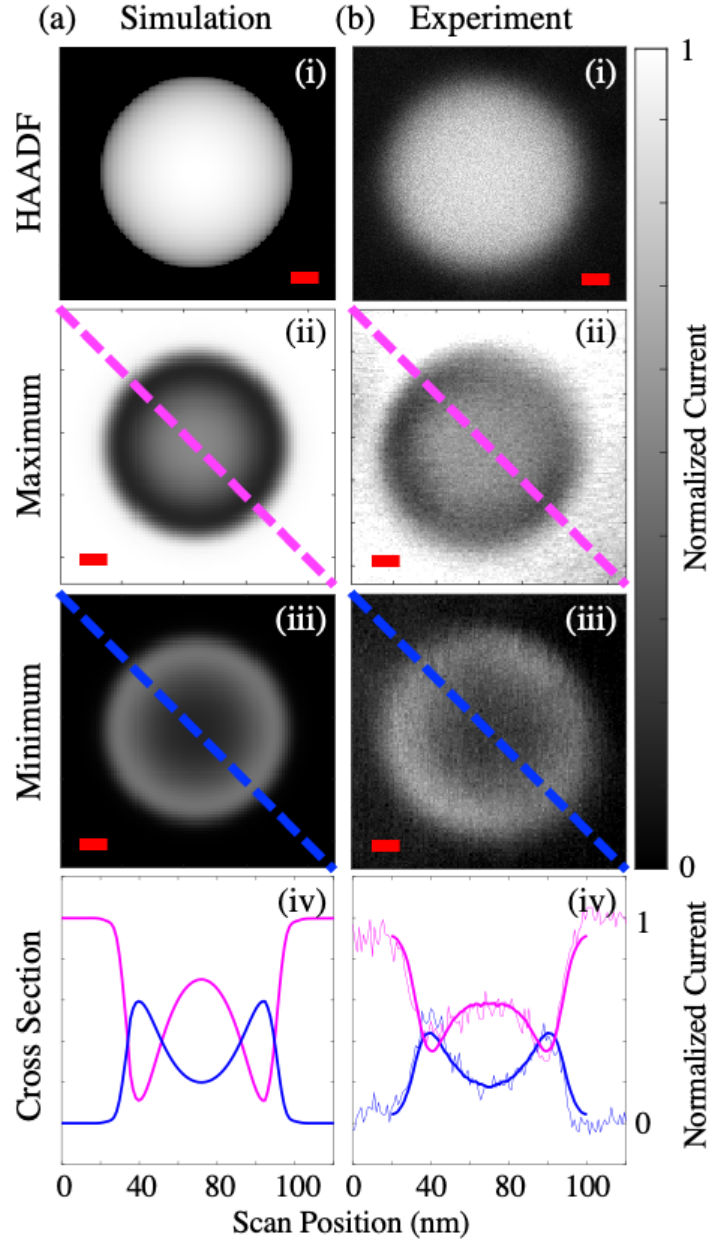


FIGURE 2.5. HAADF and 2GeMZI BF image scans of a latex nanoparticle both from (a) simulation and (b) experiment. The rows display (i) the HAADF image, the 2GeMZI image aligned at the maximally (ii) constructive (magenta) and (iii) destructive (blue) interferometer output, and (iv) the respective line profiles. The experimental cross section (b(iv)) shows a raw cross section (thin line) and the radially averaged signal (thick line). All scale bars (red) are 10 nm.

for destructive output $2\mathbf{k}_0 \cdot \mathbf{x}'_0 = 0$ (Fig. 2.5(iii)). To map each pixel's intensity onto a phase, we first find the center of the nanoparticle and take an azimuthal average of the intensity to exploit the particle's symmetry. This provides an intensity line profile across the nanoparticle and graphene from the constructive and destructive interferometric images (Fig. 2.5(iv)). Exploiting the phase continuity of the spherical nanoparticle, we note that the phase should be monotonically increasing from the graphene substrate to the center of the nanoparticle. We set the phase of the graphene substrate to zero, as a reference. Using the co-sinusoidal relation between phase and intensity found in Equation (2.11), we map the intensity profile to a phase profile. From these radial phase profiles, we reconstruct a phase image of the particle, as shown in Fig. 2.6.

Using the experimental Fourier coefficients which define each grating, a 5 nm spot size, 100 nm probe separation and assuming the nanoparticle is a perfectly spherical phase object, we simulate the expected intensity output in the BF detector when the positive first order probe interacts with the sample for both alignment schemes (Fig. 2.5(a)). The simulated intensity profile is then mapped to phase using the same mechanism as described above. The experimental phase profiles undershoot the simulated results (Fig. 2.6(d)) because the nanoparticle's amplitude decay coefficient is not exactly zero. The discrepancy between the constructively and destructively interfering experimental results can also be attributed to the amplitude decay. Decoupling the amplitude loss and imparted phase is a subject of ongoing work. The experimental phase profile of the nanosphere is slightly wider than the simulated results, due to the large probe size and the induced charging effects.

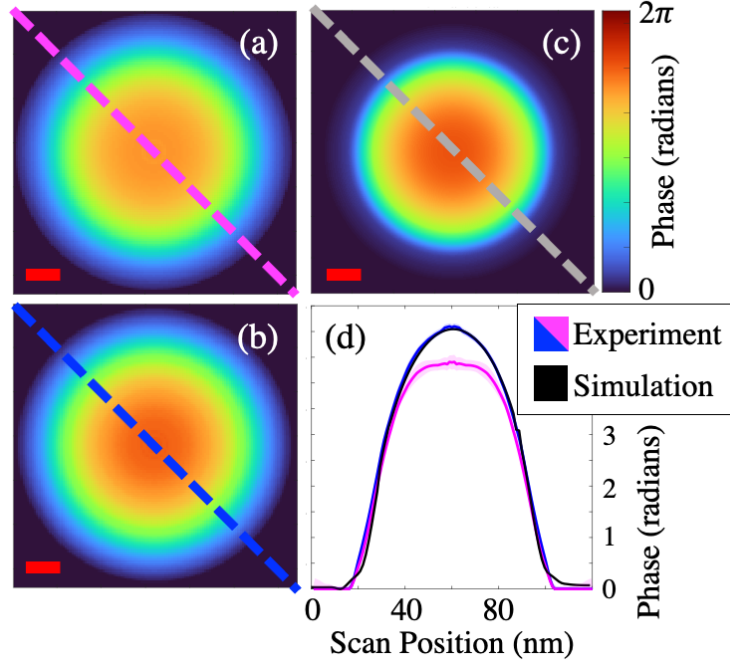


FIGURE 2.6. Reconstructed azimuthally averaged phase images of a latex nanoparticle from the raw 2GeMZI BF images with (a) constructive alignment (magenta) and (b) destructive alignment (blue). The simulated outcome (c) is also shown. (d) The experimental and simulated cross-sections of the reconstructed particle phase with shaded regions to show the error. All scale bars (red) are 10 nm.

2.6. Discussion

The 2GeMZI achieves qualitative real-time phase imaging and quantitative phase recovery providing a platform for low-dose imaging, interaction-free measurements, sub-nanometer live phase imaging, and experiments probing the Aharonov-Bohm effect.

A phase resolution of $\sigma_{ph} = 240 \text{ mrad}$ has been achieved; the phase precision could be improved with enhanced gratings, a smaller probe size, longer exposure times with efficient charge alleviation, or a lower noise BF detector. Another benefit of an improved detector is that the 512×512 pixel image, 1 Hz frame rate of this

initial demonstration can be significantly increased. At the single electron detection level, with improved grating efficiency [31], and a high frame rate, unambiguous interaction-free imaging could be realized; these improvements have already been shown individually, so incorporation into the 2GeMZI is not an insurmountable task and would significantly improve our previous demonstration of interaction-free measurements with electrons [62].

Considering grating-based STEM holography achieved 30 mrad phase and sub-nanometer spatial resolutions [40], there is promise for atomic resolution phase imaging with this technique, but in real-time without image post processing. The spatial resolution of the 2GeMZI is limited by the focused probe width, which is tunable by selecting different convergence angles using the lens system. Sub-nanometer resolution can be achieved in our device with a smaller grating pitch and larger aperture diameter. The limiting factor in this case is the stability of the scan/descan coils in the microscope keeping the image of G1 on G2 stationary to within a small fraction of the grating pitch throughout an imaging scan, making it difficult to reach a sufficient probe separation with sub-nanometer probes. The $1\text{ }\mu\text{m}$ that was demonstrated here could be safely improved by up to a factor of 2 without a loss of efficiency by decreasing the grating pitch.

It has been shown that the path separation between the probes from a grating interferometer can be increased by more than order of magnitude by the inclusion of biprisms in intermediate planes, between the $\mathbf{x}$ and $\mathbf{k}$ planes [41]. Although this would require a highly specialized microscope, such large path separations would allow for a more isolated reference beam for phase imaging and could more readily enable fundamental experiments such as testing the limits of the Aharonov-Bohm effect [99] and decoherence theory [65].

2.7. Conclusion

We constructed a scanning, path-separated, two-grating Mach-Zehnder interferometer for free electrons by employing two nanofabricated diffraction grating holograms in a conventional TEM. Although each figure of merit can be tuned or improved, the initial implementation shown here has an interference contrast of 82%, a path separation of up to 1 μm , a demonstrated phase resolution of approximately $\sigma_{ph} = 240 \text{ mrad}$, and an output current on the order of tens of picoamps. We qualitatively showed that the 2GeMZI is sensitive to phase shifts due to electrostatic potentials in vacuum by imaging the potential differences around both grounded and insulated silver vertical nanorods. We then quantitatively recovered the phase of a polystyrene latex nanoparticle on graphene. The 2GeMZI is particularly impactful in free electron interferometry due to its tunable probe separation, the accessibility of individual paths, the ability to arbitrarily apply phase shifts between separate paths, its scanning capabilities, and the real-time phase information at the nanoscale.

With incremental improvements in grating beamsplitters and detectors, the 2GeMZI could be used for interaction-free electron imaging [46, 47], low-dose STEM imaging [100, 101], nanoscale magnetic imaging [27], fundamental quantum physics experiments such as the Aharonov-Bohm effect [61], and furthering decoherence theory [102, 103]. Subjects of ongoing work are decoupling the imparted phase and amplitude loss, enhancing the contrast at the detector, and improving the spatial and phase resolution. Due to the flexible design and broad applications, the 2GeMZI is uniquely positioned in electron microscopy to open doors to sub-nanometer electron interferometry and low-dose, high-resolution microscopy.

2.8. Chapter Summary

In this chapter, we constructed a versatile electron interferometer with many features: scanning capabilities for imaging, spatially separated paths at the sample and detector, independently manipulable electron optical components, and the ability to arbitrarily tune the relative phase in separated paths. We exhibit the broad capabilities of the interferometer by performing live, electrostatic field mapping and direct phase imaging.

CHAPTER III

INTERACTION-FREE MEASUREMENT WITH ELECTRONS

In this chapter, we employ the two-grating electron Mach-Zehnder interferometer developed in the previous chapter, and use it to make the first interaction-free measurement with electrons.

Notes for Ch. II:

This chapter is adapted from the published research article:

[104] **A. E. Turner**, C. W. Johnson, P. Kruit and B. J. McMorran. Interaction-Free Measurement with Electrons. *Physical Review Letters*, 127(11):110401, 2021.

BJM and I developed the study for the paper. CWJ assisted in grating nanofabrication and provided advice on experimental design. PK contributed essential advice on experimental limitations. BJM, CWJ and PK all provided paper edits. I performed the grating nanofabrication, developed the experimental setup, took measurements and wrote the paper.

Here we experimentally demonstrate interaction-free measurements with electrons using a novel electron Mach-Zehnder interferometer. The flexible two-grating electron interferometer is constructed in a conventional transmission electron microscope and achieves high contrast in discrete output detectors, tunable alignment with independently movable beamsplitters, and scanning capabilities for imaging. With this path-separated electron interferometer, which closely matches theoretical expectations, we demonstrate electron interaction-free measurements with an

efficiency of $14 \pm 1\%$. Implementing this quantum protocol in electron imaging opens a path toward interaction-free electron microscopy.

3.1. Introduction

The quantum bomb tester proposed by Elitzur and Vaidman was named one of the seven wonders of the quantum world because it exploits the nonlocality and self-interference of a particle to non-interactively detect an object [48, 105]. In this thought experiment, Elitzur and Vaidman considered the use of an interferometer to detect the presence of a bomb so sensitive that even a single probing particle could trigger it. The interferometer is configured such that the wavefunction of a probing particle traverses a superposition of two paths in an interferometer that destructively interfere at one of the outputs, the dark port, and constructively interfere at the other, the bright port (Fig. 3.1(a)). The interference is disrupted if an opaque object obstructs one of the interferometer paths, increasing the probability of a detection event at the dark port - the outcome corresponding to an interaction-free measurement (IFM) of the object (Fig. 3.1(b)). Here we use a flexible, path-separated electron interferometer with good contrast between discrete output detectors to demonstrate the first experimental IFM with electrons.

IFMs have been demonstrated experimentally with photons [106], neutrons [107], and neutral atoms [108] but never before with charged particles such as electrons, which interact uniquely with matter and electromagnetic potentials. Electrons can be focused to much finer length scales, enabling widely used nanoscale and atomic resolution electron microscopies. Electron IFM configurations have recently been proposed for high-resolution, damage-free electron microscopy [46, 47], which would improve imaging of radiation-sensitive samples such as biomolecules [109].

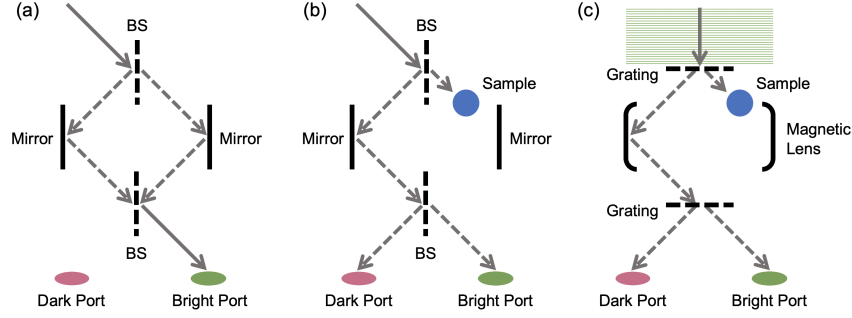


FIGURE 3.1. Schematic of an optical Mach-Zehnder interferometer (a,b) juxtaposed with the electron two-grating interferometer (c). The first two schematics (a,b) illustrate the effect of an opaque sample on the output of a Mach-Zehnder interferometer.

Interaction-free imaging with photons [110] has been shown to reduce the dose required to image semitransparent phase and amplitude samples [111, 112, 113, 114, 115, 116], even in lossy experiments [117, 118]; these protocols could revolutionize electron microscopy. Proposals to reduce the electron dose to theoretical limits feature advanced reillumination and multi-pass designs [101, 119] that require substantial advancements in electron optics. To progress toward fully interaction-free designs, we developed a suitable electron matter wave interferometer to complete a single-pass proof-of-principle experiment.

Electron interferometers constructed using electrostatic biprisms as wavefront-dividing beamsplitters have provided high-resolution phase images for decades [120, 121, 122] in a technique called electron holography. Multiple biprisms have been employed to provide separated path geometries [123, 124]. However, these biprism beamsplitters are not conducive to efficient IFMs because they do not provide discrete interferometer outputs - in electron holography the interference pattern is imaged directly. In principle, an IFM could be demonstrated in such a setup by defining the dark detector to be the narrow strips along each dark trough of the sinusoidal

interferogram, yet there would be an inherent tradeoff between the IFM efficiency and errant detections. Efficient, real-time IFMs require an interferometer with discrete outputs.

The earliest Mach-Zehnder electron interferometer with discrete outputs was constructed of single crystals [89]. Decades later, two crystals were imaged onto each other in a transmission electron microscope (TEM) to create an interferometer [125, 126], with more recent implementations built from a monolithic crystal [87, 88], but these compact interferometers are not adjustable and are difficult to use for imaging applications. Independently adjustable nanoscale gratings were used in a custom low-energy electron Mach-Zehnder interferometer [90]. Electron interferometers using a single phase grating as a beamsplitter in a TEM achieved high-resolution imaging by directly recording interference patterns [40, 79]. These experiments were made possible due to improvements in nanofabricating diffraction grating holograms, which are now more efficient and are precisely milled to shape the electron beam [31, 56].

In this chapter, we construct and discuss a novel two-grating electron Mach-Zehnder interferometer in a conventional field emission TEM with discrete, complimentary outputs like traditional optical Mach-Zehnder interferometers (Fig. 3.1). We adjust the relative phase in our interferometer and compare the output to both theory and simulation. This electron interferometer has high contrast in discrete output detectors, independently movable beamsplitters, and scanning capabilities for imaging, which opens doors for future electron interferometric research and quantum measurement protocols. Here we employ the flexible, path-separated electron interferometer to demonstrate the first experimental IFM with electrons.

3.2. Flexible Two-Grating Interferometer

Modern TEMs provide a versatile, highly configurable platform for coherent electron optics experiments [127]. Whereas an optical Mach-Zehnder interferometer uses partially reflecting mirrors as beamsplitters and fully reflecting mirrors to redirect the separate paths, here we use nanofabricated phase gratings as electron beamsplitters and magnetic lenses to redirect the electron paths, as shown in Fig. 3.2(a). Both nanogratings consist of shaped grooves milled to precise depths into free-standing, thin silicon nitride membranes (Fig. 3.3(a,c)) [31]. Each grating is installed in an available TEM aperture that allows the gratings to be independently manipulated in the transverse plane.

Incident electron wavefunctions transmitted through the input grating coherently divide into multiple diffraction orders. The input grating has a blazed profile that forms two primary interferometer paths, the -1^{st} and 0^{th} diffraction orders, of nearly equal current (Fig. 3.3(a,b)). Positive diffraction orders with smaller amplitudes are also present but are blocked with an aperture. Analyzing the interaction (0^{th}), reference (-1^{st}) and negative higher ordered probes, the input current to the interferometer splits such that $52 \pm 3\%$, $46 \pm 3\%$ and $2 \pm 1\%$ is diverted into each path, respectively. Thus, if an object were present, the probability of an interaction is $P_{\text{int}} = 0.52 \pm 0.03$. A magnetic lens after the input grating focuses these diffraction orders down to sharply peaked probes at the interaction plane. The probe spacing for this configuration is on the order of hundreds of nanometers.

A second magnetic lens system defocuses and overlaps the multiple paths, projecting the resulting interference pattern onto the output grating. In this case, we chose to use a symmetric binary phase grating for the output beamsplitter, as shown in Fig. 3.3(c,d), designed to yield two primary diffraction orders (-1 and $+1$) with

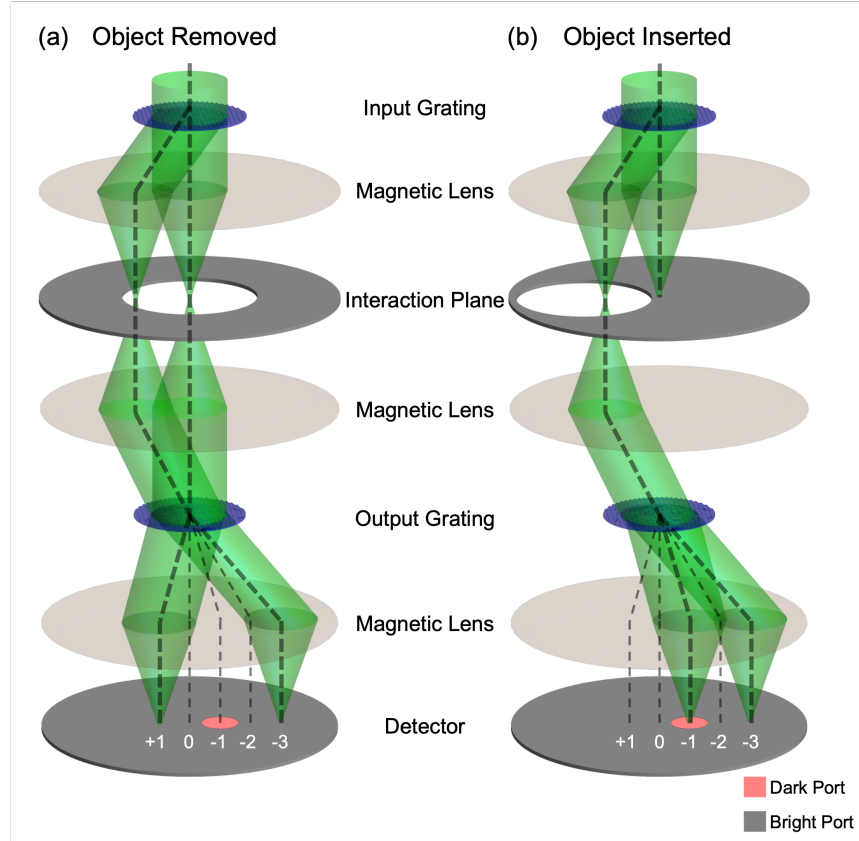


FIGURE 3.2. Schematic of an electron two-grating interferometer in a TEM (not to scale) with the object removed (a) and inserted (b), overlaid with ray diagrams. The detector coloring illustrates the selectable regions on the camera that define the bright and dark output ports. The interferometer relies on nanofabricated diffraction gratings as beamsplitters, both blazed and binary.

the same angular separation as those from the input grating. To reach the necessary alignment conditions, the image of the input grating formed by the interference of all diffraction orders is projected onto the output grating such that $2p_1 = p_2$, where p_1 and p_2 are the pitches of the input and output gratings, respectively. The interferometer output is directly imaged using the final lenses to project the far field diffraction pattern onto a scintillator screen optically coupled to a CCD at the bottom of the TEM column.

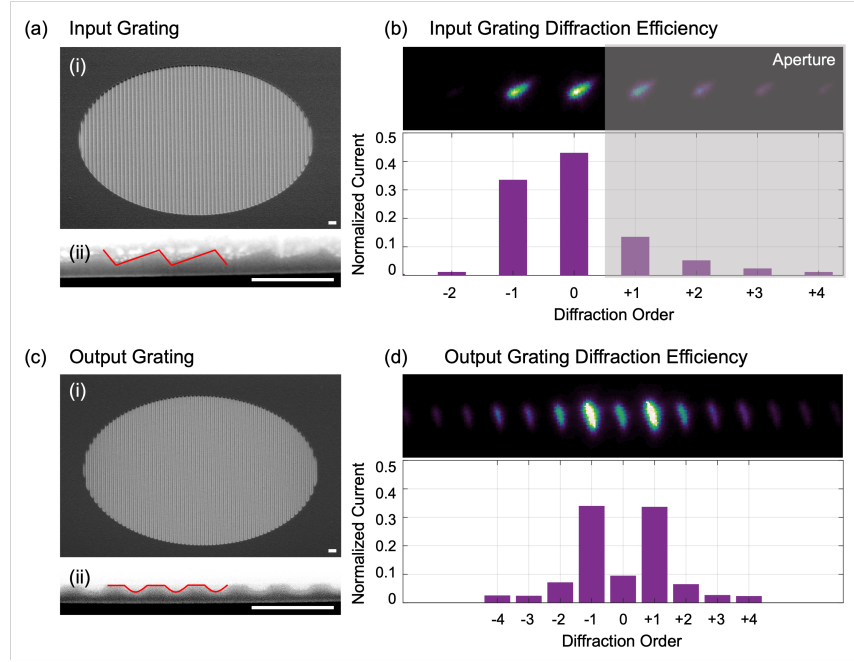


FIGURE 3.3. The nanofabricated diffraction grating beamsplitters, both blazed (a,b) and binary (c,d). The full gratings (i) and capped cross-sections (ii) are shown; the red lines highlight the profiles muddled by the platinum cap. All scale bars represent 400 nm. (b,d) Raw images of the diffracted outputs of the two gratings used in the interferometer and their diffraction efficiencies below. The two-path interferometer is constructed by inserting an aperture, as shown in (b).

This experiment was conducted in an FEI Titan 80-300 TEM with a field emission source, operated at 80 keV. An FEI Helios Dual-beam FIB was used to mill the input and output diffraction grating holograms into 50 nm thick silicon nitride membranes coated with a thin charge alleviation layer. The input blazed grating had a diameter of 50 μm and a pitch of 200 nm. The input grating was inserted into the second condenser aperture. The TEM condenser lens settings were adjusted to form probes at the third condenser aperture, which was used as the beam-blocking interaction object in this Letter. The binary output grating had a diameter of 20 μm and a pitch of 800 nm. The output grating was held by a single tilt sample holder and inserted like a traditional TEM sample, thus the second grating could be transversely shifted

similar to a sample. The objective lens was turned off such that the objective aperture could be used to limit the field of view. A 20 μm objective aperture was inserted to select for the most coherent and well-aligned portion of the overlapping gratings. The projection and diffraction lenses were used to project the output diffraction pattern of the overlapped gratings onto the camera, a CCD at the bottom of the TEM column. The dark and bright port detectors were isolated regions of pixels on the camera defined during post-processing.

The far-field diffraction pattern from the aligned two-grating configuration results in isolated beams that constructively and destructively interfere at the detector. As the output grating is laterally shifted by a distance δx_2 , the probability P_n of an event in each diffraction order n is periodic over the interval, $\delta x_2/p_1 \in [0, 1]$:

$$P_n(\delta x_2) \propto \frac{TI_n}{2} \left[1 + V_n \cos \left(\frac{2\pi\delta x_2}{p_1} + \phi_n \right) \right] \quad (3.1)$$

where T is the coherent transmission coefficient of the output grating, and I_n , $V_n \in (0, 1]$, and ϕ_n are the current, visibility, and phase, respectively. The n^{th} output diffraction order is determined by the Fourier coefficients of the two phase gratings chosen during nanofabrication [31, 56] and then measured experimentally (Fig. 3.3(b,d)). We find that the second grating coherently transmits $T = 60 \pm 3\%$ of incident electrons. This is the main source of inefficiency in our IFM experiment, but it can be improved with refined gratings [31] or lossless diffraction elements [34, 73].

A dark port and bright port, similar to an optical Mach-Zehnder interferometer, can now be established. Because the blazed grating has an asymmetric output and the binary grating has a symmetric output, it is the -1 diffraction order that has highest contrast and complete destructive interference. Thus the pixels of the

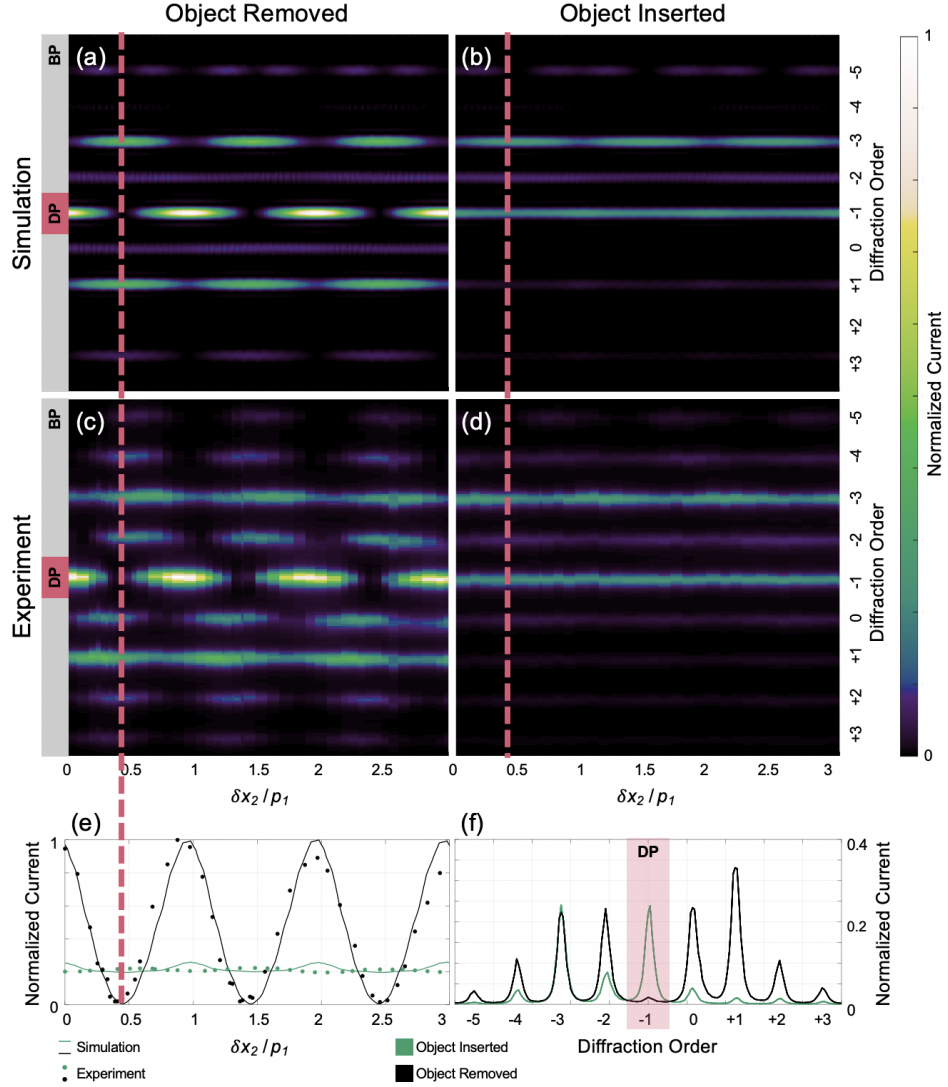


FIGURE 3.4. Simulated (a,b) and experimental (c,d) output diffraction profiles of a two-grating interferometer as the second grating is moved relative to the projection of the top grating. The relative grating shift is reported as a fraction of the blazed grating pitch, $\delta x_2 / p_1$. The left column (a,c) shows the output currents for the object removed; the two-path case. The right column (b,d) shows the output currents for the object inserted; the one-path case. The colored regions on the left illustrate the defined dark port (pink) and bright port (gray) regions of the detector. The dashed, pink line illustrates an interaction-free measurement alignment. All values are normalized to the maximum pixel current in the two-path case. (e) The normalized dark port current as a function of the relative grating shift. (f) The experimental diffraction profile along the dashed, pink line with the dark port highlighted.

detector that enclose the -1 diffraction order up to halfway to the adjacent diffraction orders are defined as the dark port (DP) detector, such that $P_{\text{DP}} = P_{-1}$. All other pixels on the detector are labelled as the bright port (BP) such that $P_{\text{BP}} = \sum_{n \neq -1} P_n$ (as denoted by the colored regions on the left of Fig. 3.4). The position $\delta x_2/p_1$ of the output grating is tuned such that the -1 diffraction order at the detector is minimized, denoted by the vertical dashed lines in Fig. 3.4.

3.3. Interaction-Free Measurement

To demonstrate IFMs, we use the edge of a platinum aperture as the opaque target object that selectively blocks one of the two interferometer paths, as shown in Fig. 3.2(b). When the object (knife-edge) is inserted, the rate of total events at the output decreases. Yet, the number of events at the dark port *increases* due to there no longer being destructive interference at the output; the hallmark sign of quantum IFMs. Ideally, the output diffraction pattern would be unvarying when shifting the output grating with an object inserted. However, the position of the second grating subtly modifies the output diffraction pattern due to the interference between the faint higher orders and the dominant reference path, shown in Fig. 3.4(b,d). Regardless, the dark port events increase significantly.

Here we compare the dark and bright port events with the object inserted and removed. All values are normalized to the total detector intensity for the two-path case and a one second exposure. All error bars are the standard deviation from multiple measurements at different IFM alignments. As seen in Fig. 3.5, when the object is removed, the dark port holds $1.2 \pm 0.1\%$ of the events detected during an exposure. When an object is inserted to block one interferometer path, the dark port event rate increases to $13.6 \pm 0.2\%$ relative to the two-path total event rate.

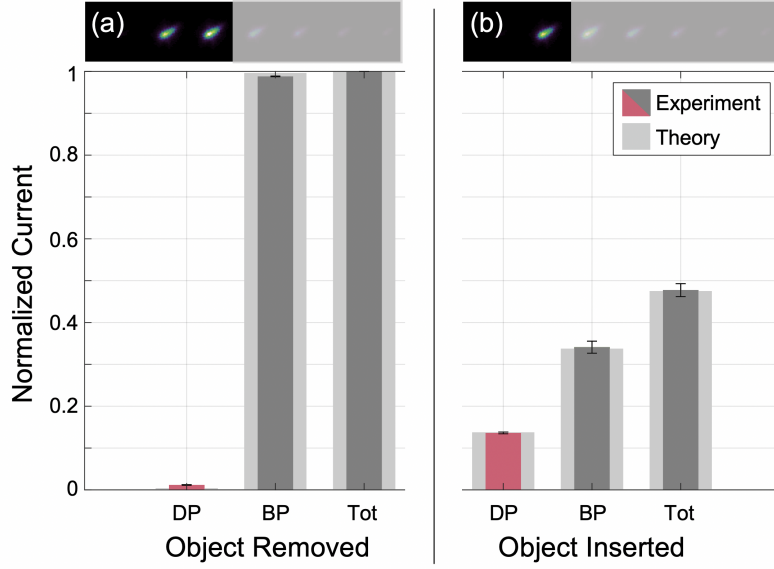


FIGURE 3.5. Normalized output currents of a properly aligned two-grating interferometer with the object (a) removed and (b) inserted. The top row contains raw images of the interaction plane with a mock aperture to illustrate the object’s position. The bar chart displays the dark port (DP), bright port (BP) and total (Tot) current in each configuration, normalized to the maximum output of the two-path configuration (Tot in (a)). The light gray bars show the expected outputs from simulation. The error bars are the standard deviation of multiple measurements in distinct alignments.

For each electron entering our interferometer when an object is present, the outcome probability of a dark port event (an IFM) is $P_{\text{DP}} = 0.082 \pm 0.004$, an interaction event $P_{\text{int}} = 0.52 \pm 0.03$, a bright port event $P_{\text{BP}} = 0.20 \pm 0.01$, and an electron scattered out of the experiment by the second grating is $P_{\text{loss}} = 0.19 \pm 0.02$. The probability P_{NI} of no interaction and a dark port detection count using multiple electrons or sample reillumination [107] is $P_{\text{NI}} = P_{\text{DP}} \sum_{n=0}^{\infty} [P_{\text{BP}}]^n = 10.3 \pm 0.5\%$. In this initial demonstration, the incoherent scattering significantly reduces the IFM signal, though this could be minimized with improved beamsplitters or eliminated with lossless diffractive elements [34, 128].

The probabilities reported above disregard the inefficiency of the electron detector, which also results in IFM signal loss. The detective quantum efficiency (DQE) of our imaging detector at this beam energy can be greater than 0.5 [129, 130, 131, 132], but collecting multiple events over longer exposure times provides better statistics. Future experiments will use improved detectors with single-electron sensitivity and a DQE approaching unity [133, 134, 135]. For a given detector, our experiment demonstrates that the fraction of *detected* events that can be interaction-free [106] is

$$\eta = \frac{P_{\text{DP}}}{P_{\text{DP}} + P_{\text{int}}} = 14 \pm 1\%. \quad (3.2)$$

Due to the incomplete suppression of the dark port during alignment, there is a small probability error P_{E} that a dark port detection will misidentify the sample

$$P_{\text{E}} = \frac{P_{\text{DP}}^{\text{SR}}}{P_{\text{DP}}^{\text{SI}} + P_{\text{DP}}^{\text{SR}}} = 8.1 \pm 0.7\%, \quad (3.3)$$

where SI and SR denote sample inserted and removed, respectively.

Even with single-particle sensitivity and optimized beamsplitters, an interaction event is more likely than a dark port event in a single-pass IFM experiment; thus, there is ongoing conversation about what constitutes an “interaction-free” measurement [136]. We do not utilize a single-electron source, but we emphasize that with a beam current on the order of 50 pA and a longitudinal coherence length on the order of microns, there was about one electron in the microscope at a time. Recording single-electron IFM events has diminished value given a thermal source, though it can still enable reduced-dose imaging [101]. The detector used in this experiment is not capable of single electron detection, but the reported signal is directly proportional to the rate of single electrons arriving at the detector, analogous

to previous IFM demonstrations using light [110]. The dark port signal is a record of electrons that did not interact with the object yet indicate its presence, and thus reasonably demonstrates interaction-free measurements. Chapter 5 discusses an experiment performed on a different instrument that uses a sensor capable of single electron detection.

3.4. Discussion

In this chapter, we report the first interaction-free measurement with electrons. This was enabled by the inclusion of two independently movable nanofabricated phase gratings in a conventional TEM to create an electron Mach-Zehnder interferometer. This versatile electron interferometer is path-separated at the sample and detector, with high contrast in the discrete outputs. Furthermore, the interferometer can be configured to incorporate scanning, which enables interferometric imaging. The demonstrated IFM efficiency $\eta = 14 \pm 1\%$ (with an overall IFM detection probability of $P_{\text{DP}} = 8.2\%$), is less than the optimal efficiency ($\eta_o = 33\%$) of a single-pass Mach-Zehnder interferometer using 50/50 beamsplitters. However, it is a significant signal even with an error probability of $P_E = 8.1 \pm 0.7\%$. The undesired higher diffraction orders produced by the holograms, unequal splitting into the two interferometer paths, and incoherent scattering at the second beamsplitter grating decrease the efficiency of our system. Fortunately, the behavior of the interferometer is well-modeled using standard Fourier analysis in linear optics, which provides a path to hone grating nanofabrication, increase the interferometer efficiency and decrease the number of errant detections.

The flexible two-grating interferometer can be utilized for inelastic interferometry, direct phase imaging [40], magnetic imaging [27, 137], Aharonov-Bohm experiments

[29, 99], and low-dose imaging [100, 101]. Furthermore, the electron interferometer enables interaction-free microscopy at the sub-nanometer length scale. The interferometer's dose limits for imaging beam sensitive materials could be characterized [138] by looking at the fading electron diffraction spots of a degrading material [139, 140, 141] or by observing chemical changes using electron energy-loss spectroscopy (EELS) [142] in materials such as poly(methyl methacrylate) (PMMA). More advanced multi-pass electron IFM implementations will reduce the interactions to zero [46, 47, 119]. By applying quantum protocols to TEM imaging, the tension between high-resolution and damage-free imaging is reduced, which opens a path toward imaging individual biological structures at atomic resolution with negligible damage [46, 47].

3.5. Chapter Summary

Here, we employed the novel two-grating electron Mach-Zehnder interferometer, previously employed for electrostatic mapping and direct phase imaging, to realize a quantum protocol with electrons. This chapter not only discussed the realization of a modern quantum experiment, but exemplified the flexibility and versatility of the interferometer.

CHAPTER IV

HOLOGRAPHY VERSUS TWO-GRATING ELECTRON MACH-ZEHNDER INTERFEROMETRY

In the previous chapter we described the first interaction-free measurement (IFM) with electrons using the two-grating electron Mach-Zehnder interferometer (2GeMZI). The 2GeMZI is a novel technique with presumed advantages over conventional electron holography such as the discrete high-contrast outputs, live phase imaging, and individually alignable components for tuning the interference at the output. Although, holography is capable of atomic level imaging with a simpler alignment scheme. Here we analyze the suitability of holography versus the 2GeMZI for IFMs, comparing the IFM efficiency and probability error for each technique.

Notes for Ch. IV:

This chapter contains unpublished material.

4.1. Introduction

Electron holography is a broadly applicable and recognized technique in electron microscopy. In 1948, Dennis Gabor invented electron holography [36]. He described interfering two wavefronts (a reference and a diffracted wave from the object) and reconstructing an image representation of the object from the recorded hologram (the image of the superimposed wavefronts). Gabor proposed an inline experimental scheme, developed to overcome the resolving power of the electron microscope by removing the limits set by spherical aberrations of the electron lenses. For inline

holography, the two interfering wavefronts propagate collinearly. In 1952, Haine and Mulvey recorded the first holograms of zinc oxide crystals and reconstructed an image of the crystals with 1 nm resolution [143]. Möllenstedt and Düker invented the biprism in 1955 [32], which enabled the realization of off-axis holography, where the object and reference waves are spatially separated and then combined by the electrostatic field of the biprism (Fig. 4.1). Although inline holography is more phase sensitive and provides advantages at lower energies, off-axis holography provides more accurate quantitative spatial measurements and a higher signal-to-noise ratio in the TEM [144]. Currently, over 20 electron holography setups exist, each designed to overcome experimental limitations, image particular samples, and to accentuate specific material characteristics.

Because electron holography recovers both the phase and amplitude of an object, it has become a well-established, diverse imaging technique for electron microscopists. Holography is advantageous over conventional TEM imaging because it is quantitative both in phase and amplitude, linear (whereas an intensity image may contain nonlinearities), exploits *a posteriori* aberration correction, and selects the zero-loss electrons [145]. Holography is especially useful for imaging weak phase objects which are virtually invisible in conventional TEM imaging, and semitransparent objects where the phase and amplitude of the sample are otherwise blended and indistinguishable in intensity imaging. Furthermore, electron holography can image static electric and magnetic fields in thin films with high-resolution. The diverse applications [146, 147] include materials analysis on the atomic scale [40, 98], locating magnetic domains [148], mapping electric potential distributions [149, 150], quantifying sample thickness [151], and measuring the Aharonov-Bohm effect [29], among others.

Even though electron holography is a powerful tool, the technique has weaknesses. Holography requires high spatial and temporal coherence, and thus is typically limited by the source. Extraneous electromagnetic fields in the microscope impart phase backgrounds to the interference pattern, demanding precise grounding and shielding of microscope components and samples for stable imaging. However, if the fields are static, they can be removed using a reference image. Electron holography is also subject to geometric distortions (e.g. charging, background electromagnetic inhomogeneities, and projector lens aberrations) and time-dependent changes (e.g. sample drift, atomic vibrations, and structure changes due to sample irradiation) [152]. Additionally, because the hologram at the detector must have resolvable fringes, the magnification must keep the fringe resolution well above the Nyquist frequency, placing additional constraints on the image resolution and field of view.

In this chapter, we compare idealized electron holography to the two-grating electron Mach-Zehnder interferometer (2GeMZI). We then discuss whether a holographic technique could be used for interaction-free measurements (IFM). Finally, we compare the IFM efficiency and error probability for the two schemes, showing the 2GeMZI is better suited for IFMs than electron holography.

4.2. Comparison of Two-Grating Interferometry and Holography

The distinguishing factors when comparing electron holography and the two-grating interferometer are the resolution, the required number of modified apertures, the number of interfering waveforms at the detector, and the recorded output. Electron holography is currently capable of high phase and spatial resolution using a single hologram (no scanning required) [145]. It can achieve high resolution without a modified aperture (inline holography) or with one modified aperture (off-

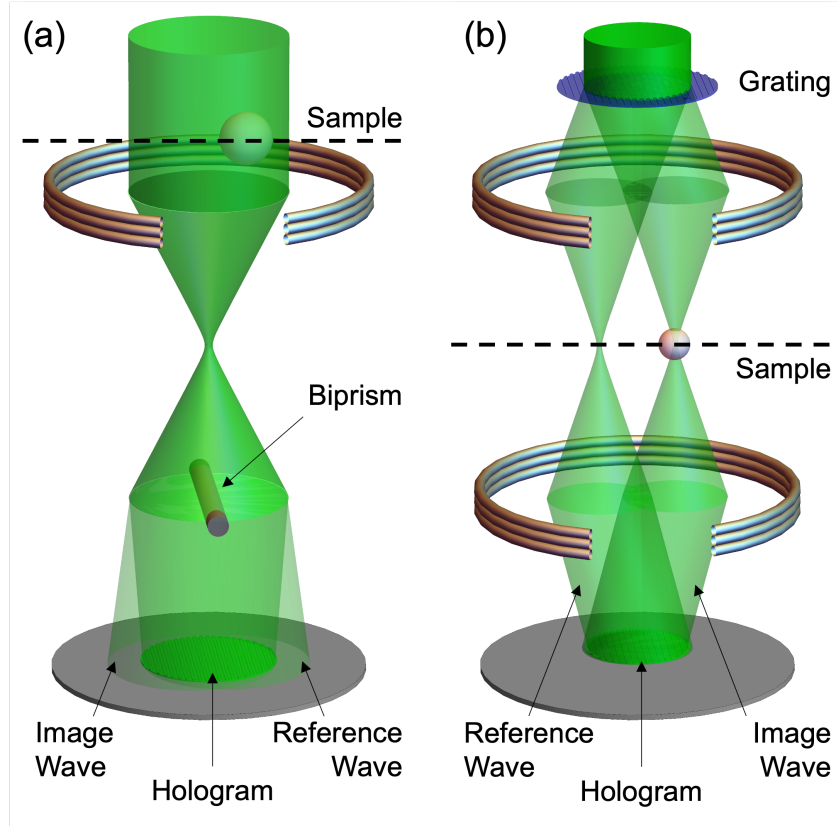


FIGURE 4.1. Two illustrations of different holography setups. (a) A typical off-axis holography setup, using a charged biprism to overlap the separated paths to form a hologram at the detector. (b) A scanning TEM holography setup where a diffraction grating is used to split the beam, magnetic lenses focus the separated paths to probes at the sample, and then the paths are focused and overlapped at the detector. Not to scale.

axis holography with a biprism or diffraction grating). Because holography can be achieved with fewer beamsplitting mechanisms than the 2GeMZI, the alignment is simpler and quicker. Furthermore, holography involves only two paths, a reference and object wavefront. One of the main drawbacks of holography in comparison to the 2GeMZI is the output, holograms. Each hologram must be computationally processed to reconstruct the phase and amplitude of the sample, precluding live imaging without supplementary processing software. Overall, holography poses as a high-resolution imaging technique with a straightforward alignment procedure.

Two-grating interferometry presents different benefits than holography. The 2GeMZI is capable of direct interferometric imaging, no post-processing required. The interferometer scans across the sample and creates an image of the sample in discrete outputs, avoiding post-processing interferograms to recover the phase and amplitude of the sample. Although the 2GeMZI requires two modified apertures, the individually modifiable and movable interferometer components create a highly manipulable interferometer. The 2GeMZI also comes with several drawbacks. The interferometer has both low spatial and phase resolution (currently limited by the detector noise and by the alignment scheme which generates a large probe diameter on the order of 1 nm). These are easily modifiable to match the resolutions of electron holography with additional lenses in the column and/or a probe corrector, which are available in many top-tier TEMs such as TEAM I, which is also equipped with a low-noise direct electron detector. The alignment of the multiple diffraction grating beamsplitters in the 2GeMZI is time consuming, requiring meticulous adjustments. Furthermore, the material diffraction gratings are lossy and do not produce only two paths; instead, they create an infinite number of diffraction orders that must be taken into account while interpreting the results (though rapidly decreasing to negligible amplitudes). Essentially, the main benefits of the flexible 2GeMZI are the discrete, high-contrast outputs and that it is capable of direct imaging.

4.3. Interaction-Free Measurements in Holography

To determine if IFMs can be implemented using conventional off-axis electron holography, we discuss a theoretical and idealized holographic formulation. For a simplistic analysis, we consider the hologram produced by an idealized two-path holographic setup. We assume that, when no sample is present, all emitted electrons

from the source interfere at the detector to form the hologram; there is no loss and the two paths completely and perfectly overlap at the detector. When the two-paths interfere, they produce sinusoidal fringes at the detector. The fringes are spatially dependent only in the direction of the change in transverse momentum (here the x direction). The sinusoidal output fringe profile can be described as

$$I_{\text{SR}}(x, \mathcal{V}) = I_{\text{ave}} (1 + \mathcal{V} \sin(kx)) \quad (4.1)$$

where x is the position along the detector, I_{SR} is the intensity profile at the detector with the sample removed (SR), I_{ave} is the average intensity across the fringes, $\mathcal{V}$ is the visibility of the fringes, and $k = 2\pi/p$ where p is the period of the fringes. When a fully opaque sample blocks the object wavefront (resembling the IFM experiment with the 2GeMZI), the interference across the detector disappears. The intensity I_{SI} at the detector with the sample inserted (SI) is

$$I_{\text{SI}}(x, \mathcal{V}) = \frac{I_{\text{ave}}}{2}. \quad (4.2)$$

The IFM experiment requires two outputs: a dark port and a bright port. In the absence of a sample, the interferometer is tuned such that the dark port does not register any counts due destructive interference. To maximize visibility $\mathcal{V}$ at the detector, we set the current in the reference and object wave to be equal. The two paths of equal current causes the probability of interaction to be $P_{\text{int}} = 50\%$. In a holographic setup, where the interference is imaged directly (two wavefronts interfering), we designate the dark port as the trough of each sinusoidal cycle, masking a pixelated detector with stripes to denote the dark port and bright port, as shown in Fig. 4.2. The dark port contains all pixels around each minimum with

width δx and period p , determined by the fringes with the sample removed. The probability $P_{\text{DP}}^{\text{SR}}$ of a dark port (DP) detection with the sample removed (SR) is

$$\begin{aligned} P_{\text{DP}}^{\text{SR}}(\delta x, \mathcal{V}) &= \frac{n \int_{p/2-\delta x/2}^{p/2+\delta x/2} I_{\text{SR}}(x, \mathcal{V}) dx}{\int_0^d I_{\text{SR}}(x, \mathcal{V}) dx} \\ &= \frac{2d \left(\pi \delta x - p \mathcal{V} \sin \left(\frac{\pi \delta x}{p} \right) \right)}{p \left(2\pi d + p \mathcal{V} \sin \left(\frac{2\pi d}{p} \right) \right)} \end{aligned} \quad (4.3)$$

with δx the width of the dark port, p the period of the fringes, $\mathcal{V}$ the visibility of the fringes, d the width of the hologram on the detector, and $n = d/p$ the number of periods visible in the field of view. When the sample is inserted, blocking the object wavefront, the fringes at the detector disappear. Thus, the probability $P_{\text{DP}}^{\text{SI}}$ of a dark port detection with the sample inserted (SI) is

$$\begin{aligned} P_{\text{DP}}^{\text{SI}}(\delta x, \mathcal{V}) &= \frac{n \int_{p/2-\delta x/2}^{p/2+\delta x/2} I_{\text{SI}}(x, \mathcal{V}) dx}{\int_0^d I_{\text{SI}}(x, \mathcal{V}) dx} \\ &= \frac{\delta x}{p}. \end{aligned} \quad (4.4)$$

From these probabilities, we can determine the IFM efficiency and probability of erroneously assigning the state of the sample (inserted or removed) given a dark port detection.

In this context of IFMs from interference fringes, the IFM efficiency $\eta(\delta x, \mathcal{V})$ (Eq. 3.2) now becomes

$$\begin{aligned} \eta(\delta x, \mathcal{V}) &= \frac{P_{\text{DP}}^{\text{SI}}(\delta x, \mathcal{V})}{P_{\text{DP}}^{\text{SI}}(\delta x, \mathcal{V}) + P_{\text{int}}} \\ &= \frac{\delta x}{\delta x + p}. \end{aligned} \quad (4.5)$$

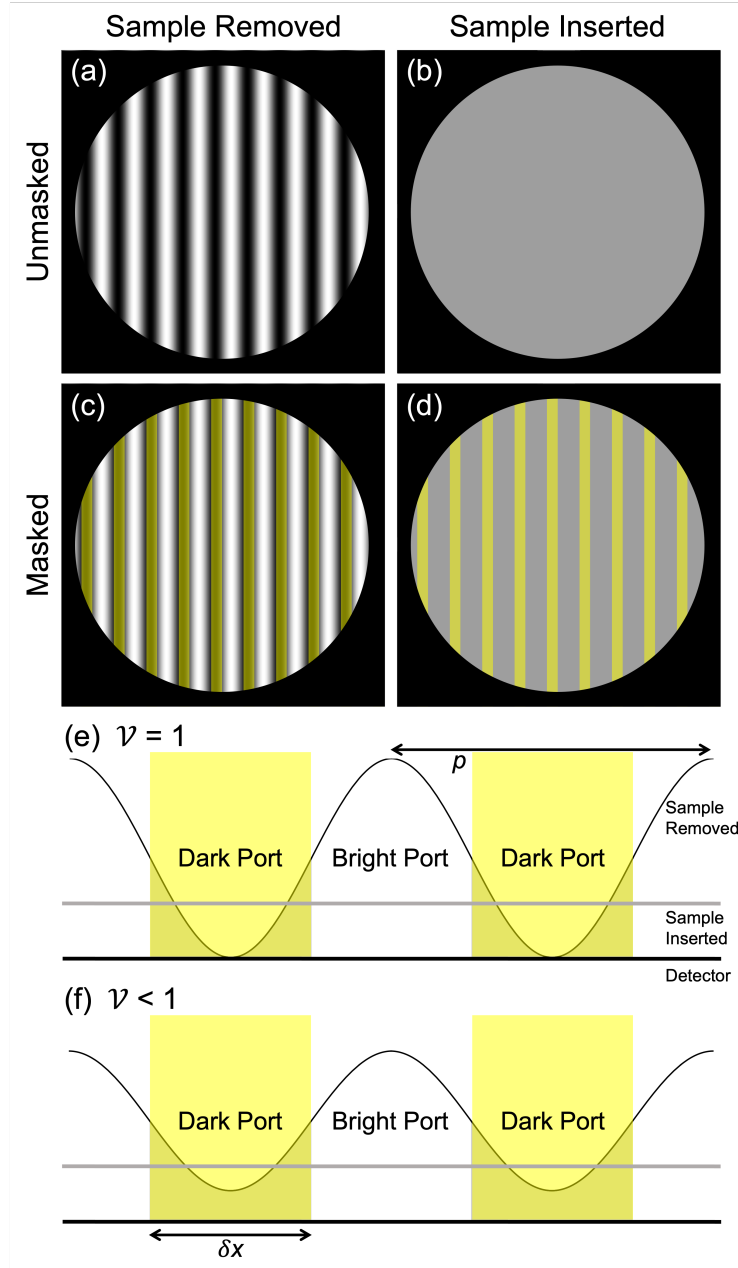


FIGURE 4.2. Illustrations of the masking process for holographic interaction-free measurements. The top row shows the output with the sample (a) removed and (b) inserted. The second row depicts the dark port mask overlayed on the holography outputs with the sample (c) removed and (d) inserted. (e) A cross-section of the sinusoidal fringes with the dark port mask in yellow. (f) The increased counts in the dark port with imperfect visibility, $\mathcal{V} < 1$.

We can rewrite δx as a fraction of the pitch p , such that $f = \delta x/p$, giving

$$\eta(\delta x, \mathcal{V}) = \frac{f}{1+f}. \quad (4.6)$$

The efficiency is shown in Fig. 4.3(a) as a function of the dark port width fraction f and the visibility $\mathcal{V}$. However, the efficiency is independent of visibility.

The probability of getting a dark port detection and incorrectly assigning the sample state (Eq. 3.3) now becomes

$$P_E(\delta x, \mathcal{V}) = \frac{P_{\text{DP}}^{\text{SR}}(\delta x, \mathcal{V})}{P_{\text{DP}}^{\text{SR}}(\delta x, \mathcal{V}) + P_{\text{DP}}^{\text{SI}}(\delta x, \mathcal{V})}. \quad (4.7)$$

The probability error P_E is dependent on both the dark port width δx and the visibility $\mathcal{V}$. As the visibility diverges from unity, the probability error quickly increases.

We must choose the width of our dark port δx such that the number of IFM detections is maximized while minimizing the probability error. If we limit the probability error to $P_E \leq 20\%$, the dark port width can maximally be $\delta x \approx 0.28p$ assuming perfect visibility $\mathcal{V} = 1$, producing an IFM efficiency of $\eta = 22\%$. However, a visibility of unity is not experimentally feasible. Lowering the visibility requirement to $\mathcal{V} = 0.9$ (relatively high but achievable for electron experiments), the dark port width narrows to $\delta x \approx 0.13p$, giving $\eta = 12\%$. This idealized two-path holographic technique is already providing lower IFM efficiencies for greater error tolerances than the 2GeMZI experimental results with lossy gratings, detector inefficiencies and imperfect visibility.

To mimic the most recent two-path IFM experiment with the 2GeMZI, we set $P_E \leq 10\%$. In the perfect visibility case ($\mathcal{V} = 1$), the dark port width becomes

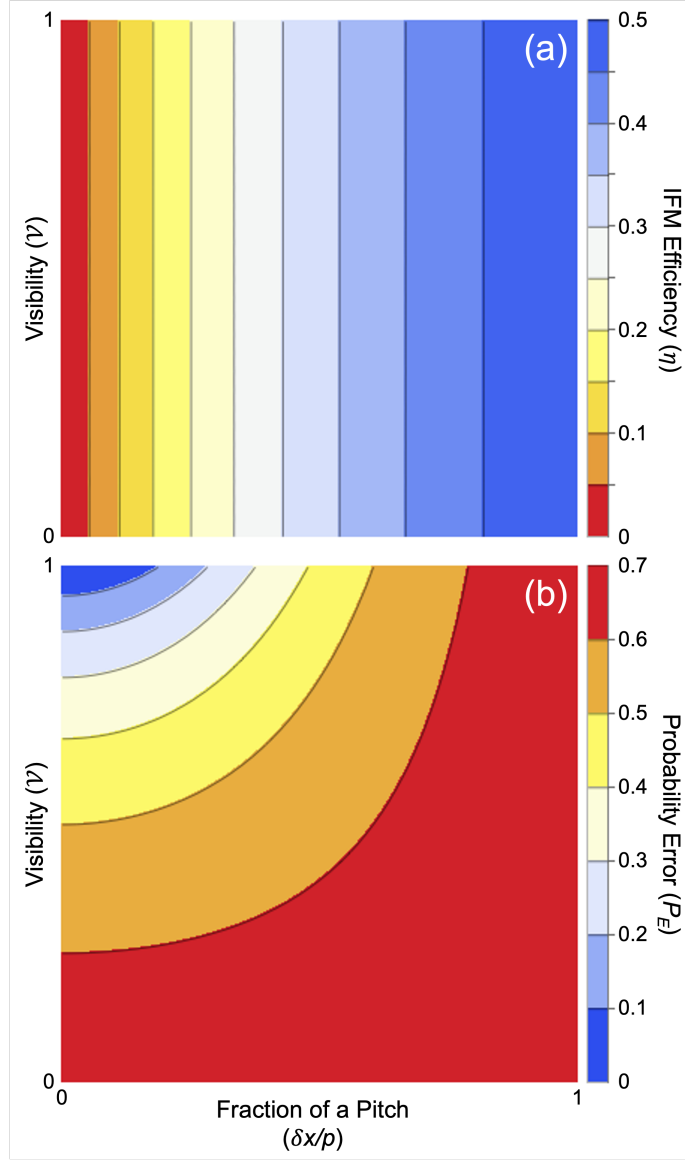


FIGURE 4.3. The (a) interaction-free measurement efficiency and (b) probability error as a function of the visibility $\mathcal{V}$ and the dark port width fraction $f = \delta x/p$.

$\delta x \approx 0.19p$ resulting in an IFM efficiency of $\eta = 16\%$. This idealized holographic setup is similar to the experimental efficiency measured with the two-path 2GeMZI, which produced $\eta = 14 \pm 1\%$. With a realistic visibility of $\mathcal{V} = 0.9$, no δx generates a $P_E \leq 10\%$.

4.4. Discussion

Holography is a powerful tool in many cases. However, the lack of discrete outputs renders holography impractical for interaction-free measurements (IFMs). With discrete outputs, the two-grating electron Mach-Zehnder interferometer (2GeMZI) thus outperforms any holographic technique. For an idealized two-path holography system, assuming a visibility of unity and allowing for a probability error of $P_E = 10\%$, we see a maximum efficiency of $\eta = 16\%$. Using the two-path 2GeMZI [104], we achieved an IFM efficiency of $\eta = 14 \pm 1\%$ with a probability error of $P_E = 8.8 \pm 0.7\%$ with a lossy experimental setup and imperfect visibility.

For the 2GeMZI, it is theoretically possible to achieve $\eta = 25\%$ (the theoretical maximum IFM efficiency for an equal current, two-path interferometer) with a probability error of zero. For any holographic technique, because the dark port is defined by its finite width, the probability error will always be nonzero and the efficiency is dependent on the width. Thus, the 2GeMZI proves a more useful and practical technique for IFMs than any holographic technique.

4.5. Chapter Summary

Compared to well-established holographic techniques in TEMs, the 2GeMZI theoretically outperforms holography for IFMs. The lack of discrete outputs in holography implicates an intrinsic error in every measurement, even for idealistic cases where the visibility of the hologram is unity.

CHAPTER V

HIGH-EFFICIENCY INTERACTION-FREE MEASUREMENTS WITH SINGLE ELECTRONS

For a single-pass interferometer with two paths of equal current, the theoretical maximum interaction-free measurement (IFM) efficiency is $\eta = 33\%$. Here we increase the IFM efficiency further by modifying the beamsplitters to decrease the probability of interacting with the sample. By weighting the current in each interferometer path, we more than double the experimental IFM efficiency in comparison to the previous two-path configuration, while maintaining a low probability error.

Notes for Ch. V:

This chapter is adapted from a manuscript that is in preparation:

A. E. Turner and B. J. McMorran. High Efficiency Interaction-free Measurements with Single Electrons. *{Manuscript in Preparation}*, 2022.

BJM and AET developed the study for the paper. AET nanofabricated gratings, designed the experiment, performed the experiment, analyzed the data, produced the theory and simulations, and wrote the paper. BJM provided essential advice, ideas and edits. BJM and AET collaborated to develop the statistical analysis.

We demonstrate high-efficiency, single-pass interaction-free measurements with single electrons by developing a low-interaction electron interferometer featuring optimized diffraction grating holograms. The interferometer is constructed in a conventional transmission electron microscope with two modified apertures to

create a flexible, path-separated, electron Mach-Zehnder interferometer. While a traditional Mach-Zehnder interferometer with two equal paths maximizes the visibility at the output, here we optimize two gratings to produce a weak interaction probe while maintaining complete interference in the output of interest. A low-current probe beam reduces the probability of sample interaction and maximizes the interaction-free measurement efficiency. We experimentally demonstrate interaction-free measurements with single electrons, achieving an efficiency of $\eta = 30.7 \pm 1.6\%$. Applying this high-efficiency quantum protocol to electron imaging could improve imaging beam-sensitive materials and is one step closer to interaction-free quantum electron microscopy.

5.1. Introduction

Quantum-enhanced imaging in a transmission electron microscope (TEM) could enable atomic-resolution, damage-free imaging. While a conventional TEM can image matter at the atomic scale [39], the high energy electron beam damages fragile samples while forming an image, limiting or eliminating the ability to structurally map single fragile samples such as biomolecules [153, 154], carbon structures [141, 155], metal-organic frameworks (MOFs) [156, 157], and salts [158]. Fortunately, the TEM has proven to be a highly manipulable electron optics workbench suitable for quantum experiments [159, 160, 161], creating an avenue to achieve damage-free, high-resolution imaging. A notable quantum-enabled imaging proposal, the quantum electron microscope [46, 47], exploits the interaction-free measurement (IFM) [48] to detect and image a sample without interaction. In pursuit of damage-free electron imaging, IFMs have recently been realized with electrons using a two-grating electron Mach-Zehnder interferometer (2GeMZI) in a TEM, as described

in Chapter 3 [104]. Here we optimize the 2GeMZI and implement single-electron detection to demonstrate higher efficiency, single-pass IFMs with electrons.

Elitzur and Vaidman theorized that one could use an interferometer operated in the single photon regime to detect an object without interacting with it [48]. Consider a perfectly aligned Mach-Zehnder interferometer (Fig. 5.1(a)) such that the input constructively interferes at one output (the bright port) and destructively interferes at the other (the dark port). A single input quantum (a photon, in the original thought experiment) traverses both paths, interferes with itself, and is always detected at the bright port. When a sample is inserted into the interaction path (Fig. 5.1(b)), the interference at the output is disrupted and the dark port becomes viable: this dark port detection reveals that a sample is present without sample interaction, an interaction-free measurement (IFM). This IFM scheme has been realized with photons [106], neutrons [107], neutral atoms [108], and electrons [104]. The IFM efficiency can be enhanced using the quantum Zeno effect, either by passing a single probing quantum through an interferometer multiple times [162] or by constructing a series of identical interferometers [163, 164]. While these multi-pass configurations detect objects with minimal exposure to probing particles, such configurations are difficult to implement in an electron microscope; although custom designs capable of multi-pass imaging are being developed [47]. Novel electron optics [73, 74] are required to implement the quantum Zeno effect for electrons; if successful, these innovations could enable an electron interferometer with an IFM efficiency approaching unity.

In Chapter 3, we demonstrated IFM with electrons [104], enabled by the construction of an electron interferometer constructed in a conventional electron microscope [55]. This interferometer used two diffraction gratings as beamsplitters

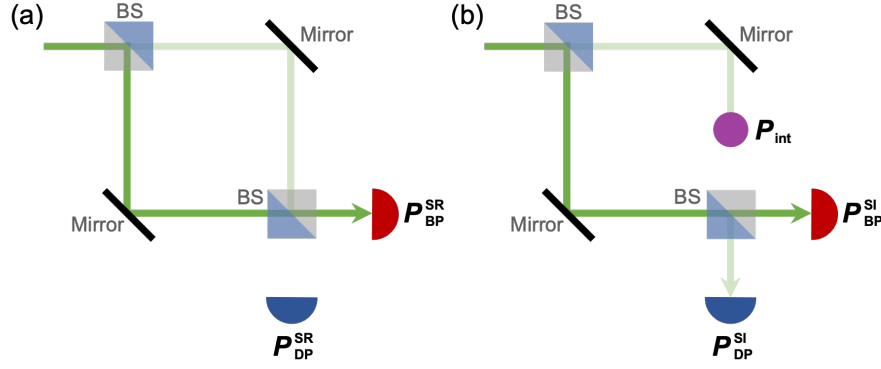


FIGURE 5.1. Illustration of a Mach-Zehnder interferometer constructed to have a low probability of interacting with the sample, P_{int} . The transparency of the paths illustrates the unequal splitting of the beam at each beamsplitter (BS). As depicted, when the sample is (a) removed, there is no probability of detection in the dark port, $P_{\text{DP}}^{\text{SR}}$, and maximal probability of a detection at the bright port, $P_{\text{BP}}^{\text{SR}}$. But when the sample is (b) inserted, there is a low probability of a detection at the dark port, $P_{\text{DP}}^{\text{SI}}$, due to the lack of destructive interference at the output.

in a Mach-Zehnder arrangement with two paths of equal current to maximize the visibility at the output. Other electron interferometric techniques have existed prior to this two-grating electron Mach-Zehnder interferometer (2GeMZI), including electron holography [37, 146], scanning TEM holography [40], crystal interferometers [87, 88], and three-grating interferometers [90]. However, these configurations either do not have discrete outputs, are not capable of scanning/imaging, or have low contrast at the output, making them inopportune for IFMs. The 2GeMZI is unrivaled for IFM experiments because it has discrete paths at the sample and detector, imaging capabilities, arbitrary phase tuning, and a manipulable design [55]. While our previous work demonstrated IFMs with electrons, the experiment was not optimized.

Here, we improve the electron IFM efficiency by more than a factor of 2 through the implementation of a weak probe beam and a single-electron detector. The two holographic diffraction grating beamsplitters are nanofabricated to form a weak

probing beam while maintaining complete constructive and destructive interference in the dark port. We use a pixelated direct electron detector to count the electrons in discrete outputs, emphasizing the quantum nature of IFMs even with massive, charged particles. Using the optimized interferometer and direct detector, we demonstrate high-efficiency, single-pass interaction-free measurements with electrons. To do so, we first establish the theoretical limits of a single-pass IFM, then develop a theoretical description for the 2GeMZI, optimize the 2GeMZI for single-pass IFMs, measure the experimental efficiency and error of the optimized interferometer, and finally simulate “interaction-free” imaging with our interferometer.

5.2. Theoretical Limit of Single-Pass Interaction-free Measurements

The figure of merit in various optical Elitzur and Vaidman experiments [106, 110] is the IFM efficiency η (Eq. 3.2), defined as

$$\eta = \frac{P_{\text{DP}}^{\text{SI}}}{P_{\text{DP}}^{\text{SI}} + P_{\text{int}}} \quad (5.1)$$

where $P_{\text{DP}}^{\text{SI}}$ is the probability of a dark port (DP) detection with the sample inserted (SI) and P_{int} is the probability of interacting with the sample. Physically, η is the fraction of measurements that are interaction-free, assuming every bright port detection (the null outcome) triggers a re-measurement (an additional input quantum); η is the probability of detecting the object through IFM before interacting with it, assuming no detection loss. The efficiency is predominantly dependent on the beamsplitting mechanisms, which determine the probability of traversing each trajectory through the interferometer. A conventional optical Mach-Zehnder interferometer using 50/50 beamsplitters produces a maximum IFM efficiency of

$\eta = 33\%$. However, this maximum can be increased by creating an asymmetric interferometer, modifying the beamsplitters to create paths of different probability currents. By creating a low-current interaction path relative to the reference path, the probability of interaction for a single measurement is small (as shown in Fig. 5.1), increasing the IFM efficiency. To maintain complete interference at the output, the beamsplitters in an asymmetric optical interferometer must be identical but opposite, specifically the reflectivity of the first beamsplitter matches the transmissivity of the second. In this case, $P_{\text{DP}}^{\text{SI}} = P_{\text{int}}(1 - P_{\text{int}})$, assuming no loss in the system. If the interaction probability approaches zero, the IFM efficiency approaches the theoretical maximum, $\eta = 50\%$ [48, 106].

5.3. Theoretical Description of the Weak-interaction Interferometer

To construct a weak-interaction Mach-Zehnder interferometer in a TEM, we use two weakly diffracting transmission gratings as beamsplitters and magnetic lenses as mirrors, as shown in Fig. 5.2. The two nanofabricated diffraction grating holograms serving as amplitude-dividing beamsplitters are installed in accessible aperture locations in the column of the microscope, one pre-specimen and one post-specimen. The electron wavefunction is collimated using magnetic lenses to create a plane wave at the input grating. The input grating then splits the electron wavefunction into distinct paths and magnetic lenses focus the paths to form narrow, spatially separated probes at the interaction plane. The paths are then refocused and overlapped to form an image of the input grating at the output grating, which recombines and interferes the separated paths. The diffracted beams are then focused again to narrow probes at the detector where the output of the interferometer is recorded. The interferometer output (a diffraction pattern consisting of electrons

doubly diffracted by the two gratings) is imaged with a single-electron sensitive detector. This interferometer is well modelled using linear Fourier optical theory; every grating or probe plane accessible in the TEM is related to the previous plane by a Fourier transform. The minute features of these components determine the overall IFM efficiency. Here we develop a theoretical model to understand these effects in order to improve the 2GeMZI and optimize the IFM efficiency.

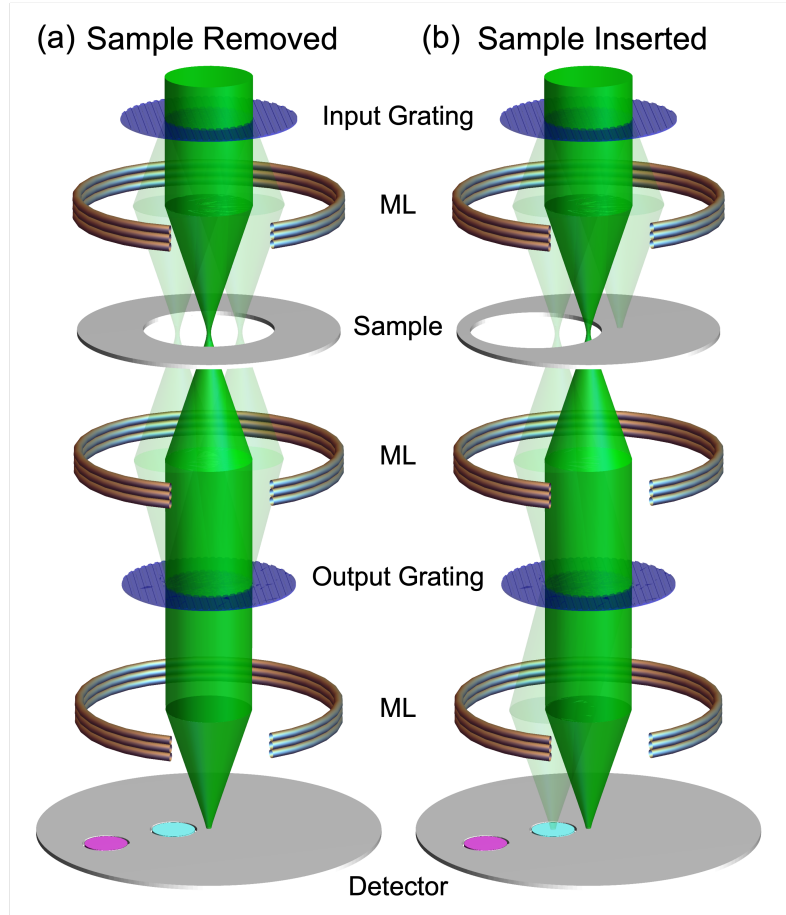


FIGURE 5.2. Schematic of the two-grating Mach-Zehnder interferometer with optimized input and output gratings constructed in a transmission electron microscope. When the object is removed (a) deconstructive interference results in a low probability for events detected at the dark port (cyan region on the detector). When the object is inserted (b), the interference is interrupted and there is an increased probability for events at the dark port.

The output wavefunction incident on the single-electron sensitive detector can be described by a comb of delta functions weighted by the Fourier coefficients describing the gratings (see [55] and Chapter 1),

$$\psi(\mathbf{k}) \propto \sum_{n,m} \tilde{c}_n^{\{1\}} \tilde{c}_m^{\{2\}} e^{i\mathbf{k}_2 m \Delta} \delta(\mathbf{k} - (n\mathbf{k}_1 - m\mathbf{k}_2)). \quad (5.2)$$

where Δ is the shift of the output grating and $\mathbf{k}_1 = 2\pi/p_1$ and $\mathbf{k}_2 = 2\pi/p_2$ with p_1 and p_2 the spatial periods of the input and output gratings, respectively. The Fourier coefficients, $\tilde{c}_n^{\{1\}}$ and $\tilde{c}_m^{\{2\}}$, describe the electron wavefunction after the input and output gratings, denoted by $\{1\}$ and $\{2\}$ respectively, assuming an incident plane wave. These coefficients are determined by the diffraction efficiencies of the gratings and depend on the grating material, membrane thickness, the groove shape (depth, duty cycle, and periodic profile), and the accelerating voltage of the microscope. We designate each diffracted output probe with the subscript μ , where $\mu = n - m$ for all combinations of $n, m \in \mathbb{Z}$.

To improve the 2GeMZI for efficient IFMs, we nanofabricate diffraction gratings with shallow grooves, such that the interaction probe is weak relative to the reference path (as shown in Fig. 1.4(a)), while retaining complete interference at the output. The two grating beamsplitters should be identical to maximize interference contrast at the output, such that $\mathbf{k}_1 = \mathbf{k}_2$ and $\tilde{c}_n^{\{1\}} = \tilde{c}_m^{\{2\}}$ for $n = m$. The first diffraction orders also must be relatively weak, $\tilde{c}_1 \ll \tilde{c}_0$, to maximize the IFM efficiency. For a binary transmission grating with shallow grooves, the output current in the zeroth order is dominant while the first order probes are weak and the higher order probes are negligible, as shown in Fig. 5.5(a,b). Here we use the positive first order beam, $n = +1$, as the interaction probe. When an electron enters the interferometer, the

probability that it interacts with an opaque sample blocking the interaction probe P_{int} is

$$P_{\text{int}} = |\psi_{n=+1}|^2 = |\tilde{c}_{+1}^{\{1\}}|^2, \quad (5.3)$$

where $\psi_{n=+1}$ is the wavefunction of the positive first order probe.

With two weakly-diffracting gratings and an aligned the interferometer, we establish the dark port and predict the output with a beam-blocking sample removed and inserted. We defined the positive first-order beam at the sample, $n = +1$, as the interaction probe, causing the dark port to be the $\mu = -1$ probe at the detector. With the sample removed (SR), the probability of a dark port (DP) detection, $P_{\text{DP}}^{\text{SR}}$, is

$$P_{\text{DP}}^{\text{SR}} = |\psi_{\mu=-1}^{\text{SR}}(\mathbf{k})|^2 \propto (1 - P_{\text{loss}}) |\tilde{c}_{+1}^* \tilde{c}_0|^2 4 \cos^2(\mathbf{k}_2 \Delta / 2) \quad (5.4)$$

where P_{loss} is the probability that an electron is scattered away by the second grating and $*$ denotes the complex conjugate. With the image of the input grating properly oriented and magnified relative to the output grating, we can align the interferometer by shifting the output grating in the transverse direction to minimize the counts in the dark port. $P_{\text{DP}}^{\text{SR}}$ is minimized when $\Delta_{\text{DP}} = p_2(\alpha + 1/2)$ for $\alpha \in \mathbb{Z}$. Thus to properly align the interferometer for IFMs the two gratings must be physically shifted in the transverse direction such that the groove offset at the output grating is Δ_{DP} . When a sample is inserted and blocks the weak interaction probe, the probability of a dark port detection changes to

$$P_{\text{DP}}^{\text{SI}} = |\psi_{\mu=-1}^{\text{SI}}(\mathbf{k})|^2 \propto (1 - P_{\text{loss}}) |\tilde{c}_{+1}^* \tilde{c}_0|^2 \quad (5.5)$$

where the output in the dark port no longer depends on the shift of the second grating, Δ .

During the construction of the interferometer, a precise balance must be struck such that P_{int} is inconsequential while $P_{\text{DP}}^{\text{SI}}$ remains a measurable signal above the noise of the detector. Physically, the grooves of the gratings must be shallow to minimize sample interaction, but concurrently must be deep enough that the probability of a dark port detection with the sample inserted is significant.

5.4. Grating Optimization for Interaction-free Measurements

To determine the optimal mill depth that maximizes the IFM efficiency using a 2GeMZI, we first need to determine how the output of the interferometer depends on the groove depths of the gratings. Using the groove depth dependent Fourier coefficients for a binary grating (Eq. 1.18), we insert them into Eq. 5.3 to model the relative current in the interaction probe at the sample and Eq. 5.2 for the relative current in each probe formed at the detector, allowing us to relate the output of the interferometer to the groove depths of both gratings.

We also simulate the output of a two-grating interferometer constructed with two identical binary gratings milled in 30 nm thick silicon nitride membranes and illuminated by an 80 keV electron beam (the experimental parameters). At this beam energy for silicon nitride membranes, we equate the complex wavenumber to $\kappa = i0.008 - \pi/21\text{nm}^{-1}$ (described in Section 1.2.3) [31]. Using these experimental parameters, we simulate the output of the interferometer with the sample removed and inserted (Fig. 5.3). From the interferometer outputs, we calculate the IFM efficiency and probability error for each groove depth and output grating displacement, defining the probability error P_{E} as

$$P_{\text{E}} = \frac{P_{\text{DP}}^{\text{SR}}}{P_{\text{DP}}^{\text{SI}} + P_{\text{DP}}^{\text{SR}}}. \quad (5.6)$$

where P_E describes the probability of misidentifying the state of the sample. In Fig. 5.3(e,f), we mask the IFM efficiency map with the regions of low probability error (less than 20%). The gratings must be precisely aligned to minimize error and milled to maximize the IFM efficiency.

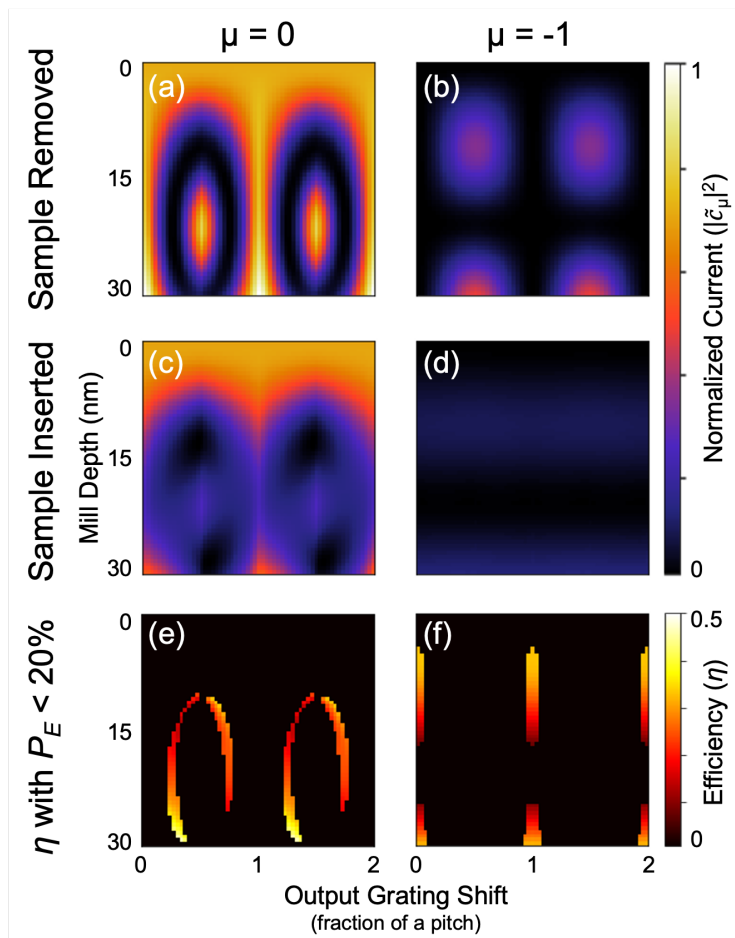


FIGURE 5.3. Simulated output of the two-grating interferometer as a function of the position of the output grating and the groove depth of the binary gratings. The output of the interferometer is found with the sample (a,b) removed and (c,d) inserted, specifically looking at the (a,c,e) zeroth and (b,d,f) negative first-order outputs. Using the top two rows and the probability of interacting with the sample (which is dependent on the mill depth), we then simulate the interaction-free measurement efficiency and the probability error. We then mask the efficiency to find where we have a high efficiency while maintaining a low probability error, $P_E < 20\%$. All simulations are for two identical binary gratings using linear Fourier optics and using assuming an 80 keV electron beam.

The simulation shows that there are multiple mill depths and alignments that produce high IFM efficiencies in different output probes while maintaining a low probability error. Using the zeroth order ($\mu = 0$) as the dark port, the optimal mill depth is nearly through each membrane. If instead we use the first order probe ($\mu = \pm 1$) as the dark port, two mill depths provide a maximized IFM efficiency; a mill depth of approximately 7 nm or through the entire membrane. All these mill depths are theoretically achievable, however, milling nearly or all the way through the material creates uneven tension in the grating membrane and can cause pocketing in the grating troughs. Thus, we aim for a 7 nm mill depth. Another notable characteristic illustrated by the simulation is the repercussions of loss due to the gratings; the deeper the grooves, the less scattering loss in the gratings, and thus the higher IFM efficiency.

Experimentally, the two gratings used to construct the 2GeMZI are unique. Due to the magnification between grating planes in the interferometer, the output grating must have a different physical diameter and pitch than the input grating. Thus, the milling parameters in the Focused Ion Beam (FIB) are distinct for each grating. The difference in grating dimensions and milling parameters results in subtle mismatches in groove shape, mill depth and diffraction efficiency, which in turn decreases the IFM efficiency and/or increases the probability error. Furthermore, each successive mill is changed by the time dependent vacuum pressure. The vacuum in the FIB improves over long periods of time (order hours), which causes the ion beam to increase in intensity during a mill, producing deeper grooves over time. As seen in Fig. 5.4, if the groove depths of the two gratings are not identical, the probability of an erroneous detection increases rapidly. To keep the probability error limited to less than 20%, the gratings must be milled to within 3 nm of one another.

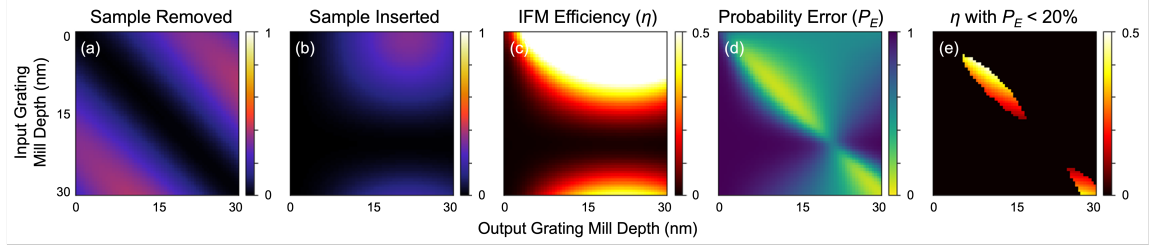


FIGURE 5.4. Simulated dark port output of the two-grating interferometer perfectly aligned to maximize the IFM efficiency as a function of the groove depths of the input and output gratings. The output of the interferometer with the sample (a) removed and (b) inserted, (c) the IFM efficiency, (d) the probability error and (e) the IFM efficiency masked for a low probability error, $P_E < 20\%$.

5.5. Experimental Interaction-free Measurement Efficiency

This experiment was conducted on a double-aberration-corrected (scanning) transmission electron microscope, a modified FEI Titan, operated at 80 keV. The instrument is equipped with a high-brightness Schottky-field emission electron source and a source monochromator which was used to control the current at the detector. The probe and image correctors were not used. Two 30 nm thick silicon nitride membranes were coated with a thin < 5 nm charge alleviation layer and then milled with a dual-beam focused ion beam (FIB) using gas assisted etching [31]. Each membrane was milled with an array of binary diffraction grating holograms of differing groove depths and pitches. The input grating used in this experiment had a $50 \mu\text{m}$ diameter and 300 nm pitch while the output grating had a $35 \mu\text{m}$ diameter and 550 nm pitch. The input grating was installed in the condenser 2 (C2) aperture and the output grating was inserted into the sample plane using a double tilt holder. The diffraction efficiencies of each of the input and output gratings used in this experiment are shown in Fig. 5.5(a,b).

To align the interferometer, we used a conventional Compustage goniometer to manipulate the position of the output grating. The output of the interferometer

while aligning the two-gratings is shown in Fig. 5.5(c,d) (using gratings similar to those in Fig. 5.5(a,b) but with slightly stronger ± 1 diffraction orders to emphasize the complete interference in the dark port). The condenser lenses were tuned such that the image of the input grating was focused on the output grating and a crossover existed at the condenser 3 (C3) aperture. The C3 aperture was used to both select a grating from the grating array in C2 and as the beam-blocking sample. The minicondenser and objective lenses oriented and magnified the image of the input grating to match the physical output grating at the sample plane. The projection and diffraction lenses were used to focus probes at the detector. The detector was a Gatan K3 direct detection camera which can collect 1500 frames per second with a detective quantum efficiency (DQE) greater than 0.9 at 80 keV. For this experiment, 100 frames were saved from 1 second exposures using the camera's full field of view without binning at multiple alignments. The data was analyzed using Python (described in detail below). A complete theoretical description of the 2GeMZI can be found in Chapter 2.

To experimentally demonstrate high efficiency two-grating IFMs, we use a platinum beam-blocking aperture as our sample and record the output of the interferometer using a direct electron detector. We use a Gatan K3 detector because the 24 megapixel detector provides single-electron sensitivity with a high DQE while operating at a fast camera speed. The combination of high-resolution and high-speed decreases the coincidence loss, providing single electron counting in real time. To further mitigate electron counting errors, we use the diffraction stigmators to smear the narrow probes out perpendicular to the direction of diffraction, increasing the DQE [165]. Because the probes have been smeared and are no longer circular, we use a rectangular dark port. To determine the boundaries of the dark port, we record

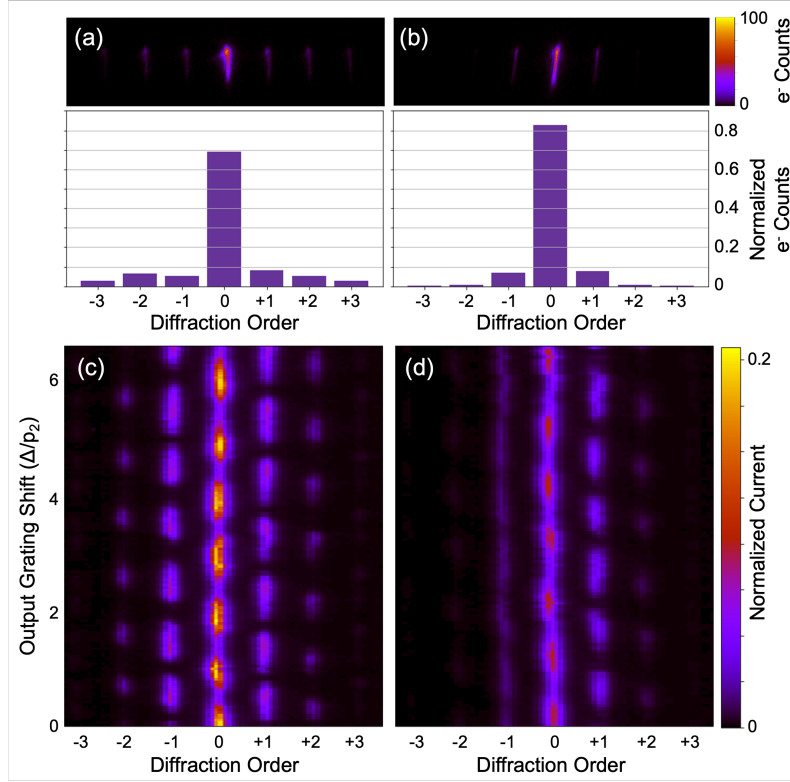


FIGURE 5.5. Experimental data showing the diffraction efficiencies of the gratings used in this chapter and a representative alignment process. The recorded images of the diffraction patterns of the (a) input and (b) output gratings using the direct electron detector. The second row shows the output of the two-grating interferometer as a function of the shift of the output grating, Δ , normalized by the pitch the output grating, p_2 , with the sample (c) removed and (d) inserted.

a full second exposure with the sample removed. The image of the output probes is then segmented perpendicular to the diffraction direction, each section having a width equal to the path separation and centered on each diffraction order. The dark port segment is then summed along the two dimensions of the section and the dark port boundaries are set using a threshold of $10e^-/\text{pixel}$, as shown by the blue box in Fig. 5.6(a,d).

One hundred 0.7 ms exposures were recorded within a 1 second window with the sample removed and then inserted. The probability of a dark port detection

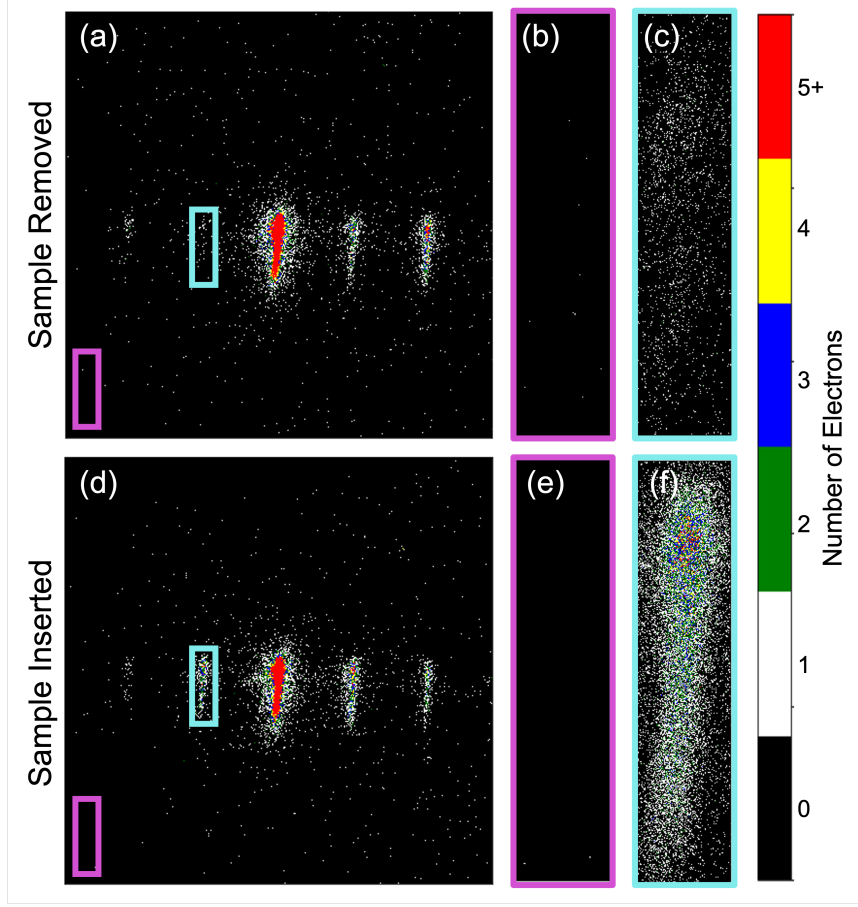


FIGURE 5.6. Images of the raw diffracted outputs from the two-grating interferometer with (a) the sample removed and (d) inserted, counting individual electron detection events. These raw images are the sum of one hundred 0.7 ms frames in a 1 second exposure. The dark port (cyan box) and vacuum (purple box) areas highlighted. The vacuum regions (b,e) show that the background noise is negligible and complete destructive interference was not achieved (comparing to (c)). The dark port close ups show the increase in counts from when the sample is removed (c) and then inserted (f).

with the sample removed is $P_{\text{DP}}^{\text{SR}} = 0.19 \pm 0.05\%$ (Fig. 5.6(c)). When the sample is inserted to block the $n = +1$ diffraction order, the probability of an interaction is $P_{\text{int}} = 4.7 \pm 1.4\%$. The beam-blocking aperture disrupts the destructive interference in the $\mu = -1$ diffraction order such that the probability of a dark port detection increases to $P_{\text{DP}}^{\text{SI}} = 2.1 \pm 0.2\%$ (Fig. 5.6(d)). This increase in dark port counts is the

signature of IFMs. As depicted in Fig. 5.7(a), the counts in the dark port during a single 0.7 ms exposure increase from 22 ± 6 electrons with the sample removed to 249 ± 19 electrons when the sample is inserted. Here we use a beam current on the order of 50 pA with longitudinal coherence length on the order of microns, providing us with approximately one electron in the microscope at any given moment. Thus, each electron count detected in individual pixels in Fig. 5.6 can be interpreted as single-electron experiments run in parallel, producing single-electron IFM events.

To characterize the efficacy of this single-pass weak-interaction experiment, we find the interaction-free efficiency, $\eta = 30.7 \pm 1.6\%$ (Fig. 5.7(b)). This is more than a 200% increase from previous single-pass, electron IFM experiments [104]. However, the dark port is not truly dark when the sample is removed, as shown in Fig. 5.6(b,e). The incomplete destructive interference in the dark port is primarily due to slight mismatches in the two gratings' mill depths, orientations, and pitches; that is, slightly mismatched wavevectors $\mathbf{k}_1$ and $\mathbf{k}_2$. There is also a small undesired background due to the higher diffraction orders produced by the gratings. The incomplete suppression of the dark port results in a small probability of misidentifying the state of the sample, $P_E = 8.3 \pm 0.3\%$ (Eq. 5.6).

Even though this experiment increases the IFM efficiency significantly in comparison to previous electron IFM experiments [104], the experimental setup does not yet reach the theoretical limit of ideal single-pass IFMs, $\eta = 50\%$. To highlight the cause of the discrepancy, we explicitly insert Eq. 5.5 into our efficiency calculation (Eq. 5.1) to see how the loss at the output grating affects the IFM efficiency

$$\eta = \frac{|\tilde{c}_{+1}^* \tilde{c}_0|^2 (1 - P_{\text{loss}})}{|\tilde{c}_{+1}^* \tilde{c}_0|^2 (1 - P_{\text{loss}}) + P_{\text{int}}} \quad (5.7)$$

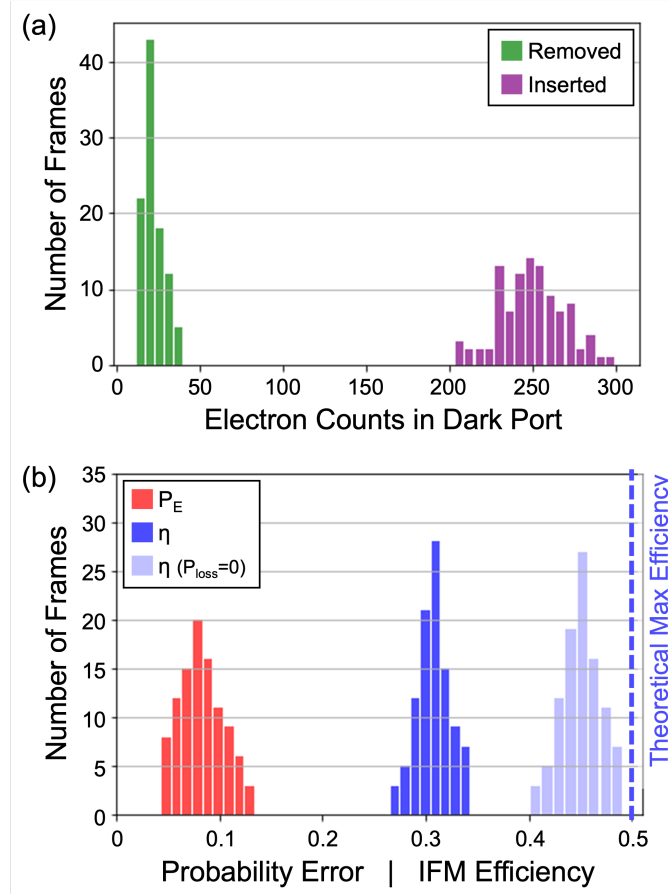


FIGURE 5.7. Experimental data showing the distribution of results when looking at 100 individual 0.7 ms frames. The top histogram (a) shows the number of counts in a set exposure with the sample removed (green) and inserted (purple). The clear increase in counts per frame when the sample is inserted is the signature of interaction-free measurements (IFMs). Below (b) is a histogram of the IFM efficiency with the experimental loss (blue) and zero loss (light blue), and the probability error (red) calculated from one hundred individual exposures. The blue dashed line on the right shows the theoretical maximum efficiency, $\eta = 50\%$, for single-pass IFMs.

where $P_{\text{DP}}^{\text{SI}} = |\tilde{c}_{+1}^* \tilde{c}_0|^2 (1 - P_{\text{loss}})$ with P_{loss} the probability that an electron is scattered away by the second grating. We also note that $|\tilde{c}_{+1}^* \tilde{c}_0|^2 = P_{\text{int}} (1 - P_{\text{int}})$. In the limit

that $P_{\text{int}} \ll 1$,

$$\begin{aligned}
\eta &= \frac{P_{\text{int}}(1 - P_{\text{int}})(1 - P_{\text{loss}})}{P_{\text{int}}(1 - P_{\text{int}})(1 - P_{\text{loss}}) + P_{\text{int}}} \\
&= \frac{1 - P_{\text{int}} - P_{\text{loss}} + P_{\text{int}}P_{\text{loss}}}{2 - P_{\text{int}} - P_{\text{loss}} + P_{\text{int}}P_{\text{loss}}} \\
&\approx \frac{1 - P_{\text{loss}}}{2 - P_{\text{loss}}}.
\end{aligned} \tag{5.8}$$

Thus, for a small interaction probability, the IFM efficiency is limited by loss. Here, using an 80 keV electron beam and a 30 nm thick silicon nitride grating membrane, $P_{\text{loss}} = 45.9 \pm 0.7\%$. The lossy nature of the second grating is the limiting factor in high efficiency IFMs with this experimental design. However, the loss could be mitigated by nanofabricating gratings on thinner membranes, increasing the energy of the beam, or implementing an optical standing wave as a lossless output diffractive element [34]. Low loss diffractive elements, such as microwave chips [35] and standing light waves [34], have yet to be incorporated into electron interferometers but show promise for low loss interferometry. With a lossless diffractive element ($P_{\text{loss}} = 0$) employed in the experimental setup described here, the IFM efficiency would increase to $\eta = 45.0 \pm 1.9\%$ (Fig. 5.7(b)), which approaches the theoretical limit for a single-pass IFM. The remaining discrepancy between our lossless experimental IFM efficiency and the theoretical efficiency maximum, $\eta = 50\%$, is due to the two gratings not being perfectly identical.

5.6. Poissonian Source and Statistical Analysis of Imaging

In parallel to the recent experimental advancements in IFMs with electrons, theoretical descriptions have progressed the field by investigating semitransparency [113, 116], non-uniform transparent samples [115, 166], conditional reillumination

[101], and experimental flaws such as detector efficiency and loss [117, 118]. Here we augment these statistical and theoretical works by applying binomial and Poissonian statistics to the 2GeMZI. The analysis directly relates the exposure time (and thus the dose) to the probability of damaging the sample and a dark port detection.

Before diving into the specifics of statistics, we briefly denote the main difference between conventional scanning TEM (STEM) and IFM imaging. In STEM imaging, all emitted electrons from the source are incident on the same location at the sample. If no sample is inserted, all electrons are detected at the camera. If a beam-blocking sample is inserted, all the electrons interact with the sample and thus no electrons are detected at the camera. The insertion of a sample decreases the number of electrons incident on the detector. The opposite is true for IFMs. If a sample is inserted, the number of detections increases at the dark port in comparison to when the sample is removed. Thus, for STEM imaging, a null detection indicates a sample is inserted, whereas for IFM imaging, a positive detection indicates the sample's presence.

Conventional TEM electron sources, and specifically the high-brightness Schottky-field emission “X-FEG” electron source used in this paper, are Poissonian by nature. The Poissonian random variable k denotes the number of electrons emitted in a given exposure with mean $\lambda = rt$, where r is the beam current and t is the illumination time per pixel. The probability that k electrons are emitted in a given exposure is described by the Poissonian distribution

$$P(k) = \frac{\lambda^k e^{-\lambda}}{k!}. \quad (5.9)$$

Each of the k electrons is then split into a superposition of each of the separated paths by the diffraction grating. The probability of the electron being detected in either

the interaction path or any of the other paths can be described using a binomial distribution.

The probability of damaging the sample P_{dam} as a function of λ , is

$$P_{\text{dam}}(\lambda) = \sum_{k=0}^{\infty} \frac{\lambda^k e^{-\lambda}}{k!} \left[1 - \sum_{x=0}^{n_{\text{dam}}-1} \frac{k!}{(k-x)!x!} (P_{\text{int}})^x (1 - P_{\text{int}})^{(k-x)} \right], \quad (5.10)$$

where P_{int} is the probability that the electron interacts with the sample (is diffracted into the interaction probe) and n_{dam} is the average damage threshold. The summation over x determines the probability that the sample will be damaged given the number of electrons in the interferometer $k > n_{\text{dam}}$. We then sum over the all possible numbers of electrons in the exposure, $k \in \mathbb{Z}_{\geq 0}$, to find the total probability of damaging a sample. To image biological samples in the TEM at high-resolution, no more than $5 \text{ e}^-/\text{\AA}^2$ are used to mitigate damaging the fragile samples [167, 168]. We set the damage threshold to 5 electrons ($n_{\text{dam}} = 5$) to determine when a pixel (of arbitrary units) is damaged and to emulate imaging a biomaterial [153, 169].

We use a similar formalism to find the probability of a dark port detection, an IFM. The erroneous dark port detections are due to the background noise of the camera. To mitigate incorrectly assigning the opaque sample as beam-blocking, we require that two electrons must be detected in the dark port to claim there is an opaque sample present in the interaction path, setting the dark port threshold for IFM to $n_{\text{IFM}} = 2$ events. To find the beam current at the detector $\tilde{\lambda}$, we modify the mean electrons in an exposure λ to account for the electrons that interacted with the sample or were scattered and lost at the second grating, such that $\tilde{\lambda} = \lambda(1 - P_{\text{int}})(1 - P_{\text{loss}})$. The probability P_{IFM} of getting n_{IFM} events or more in the

dark port for a given λ is

$$P_{\text{IFM}}(\tilde{\lambda}) = \sum_{k=0}^{\infty} \frac{\tilde{\lambda}^k e^{-\tilde{\lambda}}}{k!} \left[1 - \sum_{x=0}^{n_{\text{IFM}}-1} \frac{k!}{(k-x)!x!} (P_{\text{DP}})^x (1 - P_{\text{DP}})^{k-x} \right], \quad (5.11)$$

where P_{DP} is the probability that a detected electron is observed in the dark port, not the bright port.

With these representations of P_{IFM} and P_{dam} as a function of the mean number of electrons per measurement location (as shown in Fig. 5.8), we further discuss the dose implications for IFM imaging. To determine half of the pixels using IFMs, we use 80 electrons per pixel. However, while correctly determining 50% of the pixels, approximately 30% of the sample will be damaged. With a lossless system, the mean number of electrons per pixel decreases to 42 electrons, decreasing the probability of damage to $P_{\text{dam}} \approx 5\%$.

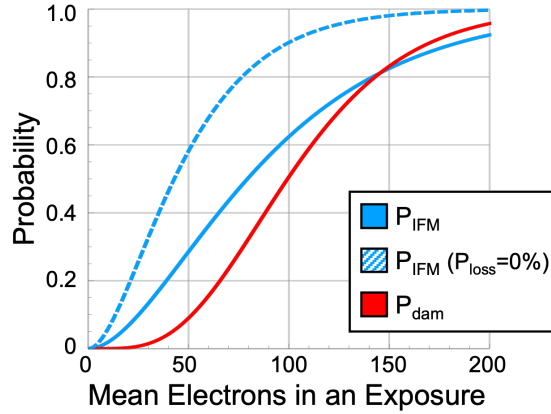


FIGURE 5.8. Using a Poissonian probability distribution to describe the source and the experimental beamsplitting efficiencies, this graph shows the interaction-free measurement and damage probabilities as a function of the mean number of electrons in an exposure. P_{IFM} is shown incorporating the experimental loss probability and $P_{\text{loss}} = 0\%$. The damage threshold was set to 5 electrons and the interaction-free measurement threshold was 2 electrons.

To illustrate the outcome of imaging with the existing interferometer (for experimental and negligible loss), we simulate imaging an opaque nano-moose (Fig. 5.9(a)). The opaque sample's largest dimension must be less than the probe spacing of the interferometer, such that no two probes are ever simultaneously incident on the sample while imaging. The interaction probe is rastered across the sample while the interferometer alignment is preserved. At each raster location, the detector counts the number of electrons detected in the dark port. In our simulation, we can follow the trajectory of every electron that enters the interferometer and count the number of electrons that interact with the sample. Counting the interactions allows us to understand the inflicted damage to the sample. In practice, unless the electron is detected at the camera, we do not know whether the electron interacted with the sample or was lost due to scattering. As described above, we set the damage threshold to 5 electrons; if 5 or more electrons interact with the sample, that pixel is designated as damaged (red pixels in Fig. 5.9). Due to our high signal to noise ratio using the Gatan K3 detector, we set the dark port threshold to 2 electrons; if we detect 2 or more electrons in the dark port, we mark that pixel as having an opaque sample (blue pixels in Fig. 5.9). Because the dark port is not completely dark with the sample removed, there is a possibility that two dark port detections occur while the sample is removed. This erroneous designation is highlighted in yellow in Fig. 5.9.

In the simulation, we input a random number of electrons per pixel into the interferometer, chosen from a Poissonian distribution with mean number of electrons λ (each column in Fig. 5.9). Using the experimental beamsplitting probabilities and associated errors, we randomly choose the number of electrons to traverse each interferometer path from a binomial distribution. We then sum all electron counts

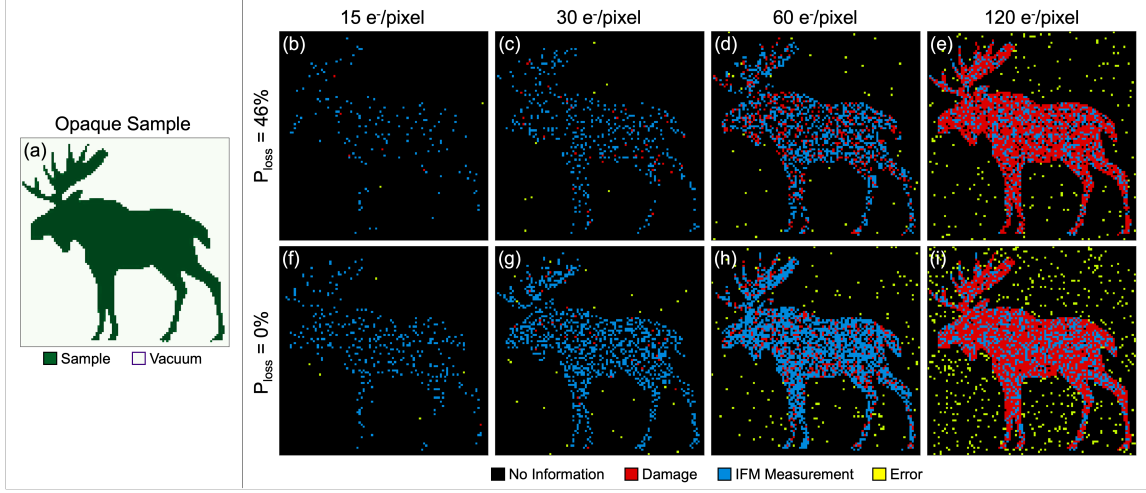


FIGURE 5.9. Simulation depicting the high efficiency interaction-free measurement (IFM) scheme applied to imaging an opaque sample. (a) Shows the shape of the opaque sample. (b-i) The rest of the panes show simulated IFM imaging results using different exposures: (b,f) 15, (c,g) 30, (d,h) 60, and (e,i) 120 electrons per pixel. The top row (b-e) shows the outcomes with the experimental loss probabilities and the bottom row (f-i) shows the outcomes with zero loss. In all of the IFM images, blue depicts the pixels correctly identified without sample damage (requiring at least 2 dark port detections), the red shows the pixels which were damaged by the beam (interacted with 5 or more electrons), the yellow pixels were assigned erroneously, and black represents the pixels that provided no information.

at the sample and the dark port to create the visualizations shown in Fig. 5.9. The simulated interferometric images are shown for the experimental loss (Fig. 5.9(b-e)) and the idealized zero loss configuration (Fig. 5.9(f-i)). While the damage rate is independent of loss, the probability of exceeding the dark port threshold increases with the mean electrons in an exposure. However, at higher exposures, $\lambda \geq 60e^-$, the probability of sample damage overtakes the probability of dark port detections, as shown in Fig. 5.9 (e,i).

From this analysis, we see that using an exposure time that generates a mean number of electrons less than $30 e^-$ mitigates the probability of damaging the opaque sample ($P_{\text{dam}} < 1\%$). With the current experimental system, $30 e^-$ gives the probability of IFM of $P_{\text{IFM}} \approx 14\%$; with a lossless system this increases to

$P_{\text{IFM}} \approx 33\%$. Thus for negligible damage to the sample ($P_{\text{dam}} < 1\%$), one could correctly identify a tenth of the pixels for an opaque sample, and a third of the pixels in a lossless system.

5.7. Discussion

Here we demonstrated high efficiency interaction-free measurements (IFMs) using an optimized two-grating electron interferometer in a transmission electron microscope (TEM). The two-grating interferometer was modified from the original two-path configuration [55] to a weak probe beam interferometer by employing two diffraction grating holograms with shallow grooves. These optimized gratings produce a weakened probe beam, decreasing the probability of interacting with the sample which in turn increases the IFM efficiency. We present an improved IFM efficiency of $\eta = 30.7 \pm 1.6\%$, which includes the experimental loss due to the second grating. However, we discussed future experimental advancements to mitigate or even remove the loss due to the output grating, which would increase the IFM efficiency to $\eta = 45.0 \pm 1.9\%$, approaching the theoretical maximum for a single-pass IFM scheme of $\eta = 50\%$. Moreover, we were able to achieve these substantial increases in efficiency while maintaining a low probability error, $P_E = 8.3 \pm 0.3\%$.

The two-grating electron Mach-Zehnder interferometer (2GeMZI) is a powerful, versatile tool due to the tunable path-separation, positionable probes, individually manipulable components and the ability to arbitrarily apply phase shifts to individual paths. It has already been employed in inelastic interferometry [170], phase imaging [55], and traditional two-path IFMs [104], and could be used in Aharonov-Bohm experiments [29, 61], interferometric magnetic imaging [27], and low-dose scanning TEM imaging [100, 167]. Here, the 2GeMZI exhibits its flexibility, substituting

in modified apertures to achieve the desired diffraction efficiency with a weak interaction probe. Because the other advantages of the 2GeMZI are maintained, two-grating, single-pass interferometry poses as a viable low-dose imaging platform [110]. Incorporating conditional reillumination [101] and a controllable electron source [171], multi-pass [119], or the quantum Zeno effect [46, 47], could increase the IFM efficiency to the theoretical limit of unity, which would eliminate sample damage while imaging. Applying quantum protocols, such as weak probe beam IFMs, to electron imaging creates new prospects for imaging delicate samples damage-free at the atomic scale.

5.8. Chapter Summary

In this chapter, we further examined the modifiable parameters of the two-grating electron Mach-Zehnder interferometer (2GeMZI), specifically the diffraction grating holograms, in attempt to increase the interaction-free measurement (IFM) efficiency. We analyzed the impact of the gratings' mill depths and relative alignment on both the IFM efficiency and the probability error, both through Fourier linear optical theory and simulations. After optimizing the mill depth, we experimentally achieved a higher efficiency, single-pass IFM configuration, limited by the grating loss. The enhanced 2GeMZI also incorporated a single electron sensitive detector. Using the experimental beamsplitting probability currents, we simulated "interaction-free" imaging using a Poissonian source for a given dose rate.

CHAPTER VI

FUTURE DIRECTIONS AND CONCLUSION

Notes for Ch. VI:

This chapter contains unpublished material.

6.1. Summary of the Dissertation

In this dissertation, I designed and constructed a versatile electron interferometer with broad applications and employed it to demonstrate a fundamental quantum experiment, the interaction-free measurement with electrons. Specifically, I developed a two-grating electron Mach-Zehnder interferometer (2GeMZI), demonstrated the first interaction-free measurement (IFM) using electrons, proved the suitability of the 2GeMZI over holography for IFMs, and optimized the 2GeMZI to achieve high-efficiency, single-pass IFMs.

The 2GeMZI is a versatile, unique tool with significant advantages over existing electron interferometric and holographic techniques. The interferometer has high contrast in discrete outputs, independently manipulable and modifiable components for a flexible alignment, scanning capabilities for imaging, the ability to arbitrarily tune the phase in separate interferometer paths, spatially separated paths at the sample and detector, and can be implemented in any conventional three condenser lens transmission electron microscope (TEM). In the 2GeMZI's brief existence, the newfangled interferometer has already achieved quantitative phase imaging [55], electrostatic potential mapping [55], inelastic interferometry [172], and a fundamental

quantum experiment [104]. Beyond these applications, the 2GeMZI could be applied to magnetic imaging [27], high-resolution interferometric imaging of phase and semitransparent samples [40, 55], low-dose imaging [100, 167], beam irradiation damage measurements [43, 173], spatial spectrum imaging of plasmonic modes [19, 172], Aharonov-Bohm experiments [28, 29, 61], coherence and decoherence measurements [102, 103, 159, 174], and quantum eraser experiments [175, 176].

The development of the 2GeMZI led to the realization of the first IFM with electrons. The interferometer employed a blazed input grating and binary output grating to create two equal current paths at the sample and a high-contrast, off-axis diffraction order dark port at the detector. This experimental design achieved an IFM efficiency of $\eta = 14.1 \pm 1\%$ and a probability error of $P_E = 8.1 \pm 0.7\%$. To optimize the 2GeMZI for high efficiency IFMs, we explored unequal beamsplitting as a method to decrease sample interaction. The ideal configuration involved a weak probe beam and strong reference beam, which decreased the probability of interacting with the sample. To experimentally realize this skewed electron Mach-Zehnder interferometer, we optimized two binary diffraction gratings to weakly diffract while maintaining complete interference at the detector. Using this modified 2GeMZI, we achieved an IFM efficiency of $\eta = 30.7 \pm 1.6\%$ with a probability error of $P_E = 8.3 \pm 0.3\%$.

6.2. Future Directions

While the 2GeMZI has proven a powerful technique for IFMs, simple experimental advances could improve the method by overcoming current efficiency and resolution limits. To overcome the current spatial resolution limits set by the probe size, the alignment scheme could be adjusted to minimize the beam waist. As shown in previous grating-based interferometry techniques, subatomic beam diameters are

achievable, generating atomic resolution images [40]. In addition to a minimized beam waist, other experimental upgrades could be beneficial and effective. Ideally, the 2GeMZI would be implemented in a monochromated TEM with a single electron detector and a probe corrector. The improved electron detector would increase the signal to noise ratio, improving the phase resolution. The probe corrector reduces the probe diameter and adjusts the electron wavefunction to a pure mode through the beam waist, which enhances the spatial resolution. The most impactful experimental upgrade for IFMs with the 2GeMZI would be incorporating a lossless diffractive element as the output beamsplitter, such as a standing wave of light known as the Kapitza-Dirac beamsplitting mechanism [34]. The standing wave of light used to diffract the electron beam has a wavelength on the order of hundreds of nanometers, like the period of the nanofabricated diffraction gratings exhibited in the 2GeMZI, and is weakly diffracting, which is ideal for high efficiency IFMs. Eliminating the electron loss due to the second grating would reduce the gap between the experimental and theoretical IFM efficiencies.

The diffraction pattern output of the 2GeMZI contains currently unexplored information. For IFMs, the diffraction order denoted as the dark port is the only required output, and thus has been the focus in this dissertation. However, observing and analyzing the full diffraction pattern could provide quantitative phase and amplitude information at higher resolutions. Incorporating post-processing could allow the microscopist to quantitatively separate the amplitude and phase of a sample, like holography, resolving the challenge of semitransparent samples. Furthermore, the total counts at the detector results in a low visibility, bright field image of the sample. Applying post-processing to 2GeMZI data, in tandem with the benefit of direct imaging, could reveal new layers of sample characterization.

The 2GeMZI appears to be supremely set up for “interaction-free” imaging [110] due to its proven IFM [104] and scanning capabilities [55]. However, this seemingly simple next step involves numerous challenges. Notably, most samples imaged in a TEM are not completely opaque. TEM samples are primarily less than 200 nm thick and thus are either semitransparent or transparent to the beam. Theoretical work has started to apply the IFM to damage-free imaging of phase and semitransparent samples [111, 112, 113, 114, 115, 116], but the problem is unresolved. Furthermore, the 2GeMZI is sensitive to electromagnetic fields. As the fields in the microscope change over time, the phase of the interferometer is modified. These fields can be the result of settling lenses, sample charging, or plasmonic excitations. Further investigation into separating the phase and amplitude information when imaging semitransparent objects, and filtering out or controlling background time-dependent electromagnetic potentials is required before realizing a practical “interaction-free” imaging technique.

To truly achieve damage-free, quantum-enhanced electron imaging, more elaborate innovations are required. Conditional reillumination [101] theoretically decreases the dose applied to a semitransparent sample while imaging; however, it requires real-time feedback and a shuttered electron source. To achieve an IFM efficiency approaching unity, the quantum Zeno effect [106, 162] needs to be applied by increasing the number of times the electron interacts with the specimen by passing through it multiple times. However, implementing a multipass electron microscope effect requires a complex tabletop electron optics setup or a combination of nonexistent electron optics (a single electron source, electron resonator, two-state coupler, and high-precision electron shutters [46, 47]). Nevertheless, an international

collaboration is currently developing such an ambitious instrument, the quantum electron microscope.

A viable alternative of low-damage IFM imaging with modern electron optics is low-dose inelastic interferometry. The interference signal from inelastically and elastically scattered electrons is π out of phase [172]. A monochromated 2GeMZI could be tuned to only pass the elastically or inelastically scattered electrons into the dark port of the interferometer. With each successive interacting electron, the probability of inflicting permanent damage to a fragile sample increases. Statistically, the first five electrons to interact with a biological sample can inelastically lose energy without inflicting permanent damage, though they do modify electronic processes and increase atomic vibrations [167, 168]. With a perfectly aligned interferometer and detector with a high detective quantum efficiency, electrons could interact with the sample, damage-free, while reporting the presence of a fragile, transparent sample.

6.3. Big Picture

A prominent goal in electron microscopy is achieving three-dimensional atomic-resolution imaging of biological specimens and macromolecules with negligible damage. Current atomic resolution imaging has already impacted drug development [23, 24, 177] and disease treatment [25, 26]. However, the fragile samples are damaged while imaging, limiting the achievable resolution. With the ability to image biomolecules at even slightly reduced doses, new sample targets could include gene expression and regulation complexes, membrane proteins (the target in 60% of drugs), viruses, and the proteins involved in neurodegenerative diseases [178]. Furthermore, damage-free imaging could create opportunities to understand biological processes in real-time at the atomic level. Currently, electron optics are

swiftly evolving in pursuit of the modern revolution in electron imaging, hopefully exposing unobserved subatomic structures, unknown electromagnetic realities, and strange quantum phenomena. Applying quantum measurement protocols to electron microscopy could alleviate the tension between high-resolution and damage-free imaging, providing a path toward imaging individual biological samples and macromolecules with negligible damage [47].

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