Ultramicroscopy 166 (2016) 48–60

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Full length article

Generation and application of bessel beams in electron microscopy

Vincenzo Grillo [a,b,*], Jérémie Harris [c], Gian Carlo Gazzadi [a], Roberto Balboni [d], Erfan Mafakheri [e], Mark R. Dennis [f], Stefano Frabboni [a,e], Robert W. Boyd [c], Ebrahim Karimi [c]

[a] CNR-Istituto Nanoscienze, Centro S3, Via G Campi 213/a, I-41125 Modena, Italy
[b] CNR-IMEM, Parco Area delle Scienze 37/A, I-43124 Parma, Italy
[c] Department of Physics, University of Ottawa, 25 Templeton St., Ottawa, Ontario, Canada K1N 6N5
[d] CNR-IMM Bologna, Via P. Gobetti 101, 40129 Bologna, Italy
[e] Dipartimento di Fisica Informatica e Matematica, Università di Modena e Reggio Emilia, via G Campi 213/a, I-41125 Modena, Italy
[f] H.H. Wills Physics Laboratory, University of Bristol, Bristol BS8 1TL, United Kingdom

ARTICLE INFO

Article history:
Received 13 May 2015
Received in revised form
15 March 2016
Accepted 23 March 2016
Available online 26 March 2016

Keywords:
Bessel beams
Holographic beam shaping
Vortex beams
STEM
Fresnel diffraction

ABSTRACT

We report a systematic treatment of the holographic generation of electron Bessel beams, with a view to applications in electron microscopy. We describe in detail the theory underlying hologram patterning, as well as the actual electron-optical configuration used experimentally. We show that by optimizing our nanofabrication recipe, electron Bessel beams can be generated with relative efficiencies reaching $37 \pm 3\%$. We also demonstrate by tuning various hologram parameters that electron Bessel beams can be produced with many visible rings, making them ideal for interferometric applications, or in more highly localized forms with fewer rings, more suitable for imaging. We describe the settings required to tune beam localization in this way, and explore beam and hologram configurations that allow the convergences and topological charges of electron Bessel beams to be controlled. We also characterize the phase structure of the Bessel beams generated with our technique, using a simulation procedure that accounts for imperfections in the hologram manufacturing process.

1. Introduction

Electron vortex beams have recently drawn significant attention within the electron microscopy community, and have shown great potential for a host of applications [1–3]. The OAM-carrying capacity of free electron beams was highlighted in a seminal theoretical paper by Bliokh [4], which precipitated considerable experimental efforts directed toward the generation of structured electron beams [5]. For example, electron vortex beams have recently been produced with orbital angular momenta as large as $200\hbar$ per electron; such beams show promise for potential applications in magnetic measurement [6]. For this reason, a great deal of effort has been expended in attempts to optimize the efficiency of vortex beam generation. In particular, holographic elements have emerged as promising candidates for high efficiency structured electron beam generation [7–12].

Holographic optical elements can allow electron beams to be shaped by modulating the transverse phase and amplitude profiles of incident electron waves with high precision. Amplitude modulation of incident electron beams can be achieved by alternating thick fringes made from opaque material with regions of high transparency. By contrast, phase modulation is carried out by varying the transverse thickness profile of a nearly transparent material, so as to produce disparities in the electron-optical path lengths experienced by different transverse components of the incident beam [8,9].

Phase-modulating elements have already found a range of applications in electron microscopy [13–15]. Specifically, phase plates can be used in transmission electron microscopy (TEM) to improve the contrast of weak phase objects, or to compensate for spherical aberration effects [16]. Attempts have also been made to produce phase plates for scanning transmission electron microscopy (STEM), in one case resulting in a Fresnel lens analogous to zone plate lenses for X-rays [17]. However, these types of lenses pose a significant nanofabrication challenge.

Beyond the examples mainly focused on vortex beams, relatively little work has been done with a view to shaping electron beams using holographic elements [8,11,12], and still less with reference to specific practical applications. This is not to suggest that this area is entirely unexplored; studies have previously investigated silicon nitride (Si_3N_4) as a candidate holographic material for electron beam shaping, for its low electron-optical density, and its ability to modify the beam phase directly on axis [18].

* Corresponding author at: CNR-Istituto Nanoscienze, Centro S3, Via G Campi 213/a, I-41125 Modena, Italy.
E-mail address: vincenzo.grillo@cnr.it (V. Grillo).

http://dx.doi.org/10.1016/j.ultramic.2016.03.009
0304-3991/

However, no medium, no matter how transparent, can ever act as a perfect phase plate, since atoms in the material always produce inelastic or high-angle scattering that can, in essence, be treated as absorption and/or as loss of coherence, especially in the forward direction. This scattering, along with the limited control that can be exerted over the phase induced in an oncoming beam, can represent a significant hindrance to the use of on-axis phase holograms, producing a “frosted glass” effect, and blurring of the transmitted beam, and a reduction in its quality [19]. The use of Si_3N_4 holograms for on-axis electron beam shaping faces another drawback, in that it requires that thickness modulations be applied with precisions on the nanometer scale, a significant challenge even for state-of-the-art nanofabrication techniques.

In this sense, the introduction of off-axis amplitude holograms can be considered a significant development. These holograms, which consist of a modulated diffraction grating, benefit from the absence of unwanted scattering from their transparent regions by alternating fully absorbing and fully transparent fringes. A second advantage to this approach is that the phase imprinted on the incident wavefront is encoded in the transverse grating profile, and is therefore readily controlled, even when imperfect manufacturing techniques are employed. This technique does suffer from an important drawback, however, in that it typically results in low-efficiency generation of the desired output beam. Recently, we introduced off-axis phase holograms that allow this limitation to be overcome, theoretically reaching efficiencies as large as 100% [8,9]. Here, we report a detailed study of electron Bessel beam generation using this technique.

Bessel beams are widely used in photonics, and have recently been discussed theoretically in the context of a number of electron microscopy applications. In the ideal case, Bessel beams possess a propagation-invariant profile, and are therefore referred to as diffraction-free modes (see the discussion in Section 3). These beams hold great promise for their ability to reduce channeling [20], to control aberrations and their potential applicability to new imaging modes, as well as for the generation of optical tractor beams, and other exotic applications. Apart from their wide range of potential applications, Bessel beams have also drawn considerable interest on theoretical grounds, for their unusual properties [21].

Notably, electron beams of approximately Bessel form have been generated using on-axis techniques such as hollow cone illumination [22,23]. However, electron beams generated in this way suffer from large intensity losses due to the partial blocking of the beam required by the technique. Still more critically, this strategy does not allow for the modification or control of key beam parameters, such as topological charge and convergence.

Here, we report a detailed study of the first off-axis Fresnel phase hologram to generate electron Bessel beams [8], and examine: 1) the conditions under which Bessel beams can be generated and applied to microscopy and imaging; 2) techniques by which key beam and hologram parameters, including topological charge, transverse wavenumber, and hologram aperture radius can be adjusted; and 3) the main practical limitations of electron Bessel beam generation.

2. Holographic generation of structured electron beams

Holographic plates can be used to confer spatial structure upon arbitrary electron beams with high efficiency. These devices are fabricated by inducing spatially varying changes in the optical thickness and transmittance of a material, and therefore amount to optical phase and amplitude masks. When an incident plane wave is transmitted through such a mask, it gains a position-dependent phase $\Delta\varphi(\rho,\phi)$ relative to a reference wave having traveled an identical distance in vacuum, and experiences a spatial amplitude modulation $A(\rho,\phi)$, such that the mask may be described by a transmittance

$$T(\rho,\phi)=A(\rho,\phi)e^{i\Delta\varphi(\rho,\phi)} \quad (1)$$

where ρ,ϕ,z are the standard cylindrical coordinates. The transverse wavefunctions $\psi_{in}(\rho,\phi)$ and $\psi_{tr}(\rho,\phi)$, respectively corresponding to the incident and transmitted beams, are then related by $\psi_{tr}(\rho,\phi)=T(\rho,\phi)\psi_{in}(\rho,\phi)$. Three nontrivial classes of holograms may be distinguished, with reference to Eq. (1). First, *phase holograms* are those for which $\Delta\varphi(\rho,\phi)$ exhibits a spatial dependence, while the hologram's amplitude modulation function is spatially constant, i.e. $A(\rho,\phi)=A_0$. By contrast, *amplitude holograms* induce a spatially varying amplitude modulation, but produce a spatially constant phase in the incident beam, so that $\Delta\varphi(\rho,\phi)=\Delta\varphi_0$. Finally, *mixed holograms* are characterized by spatially varying phase and amplitude modulations, so that neither $A(\rho,\phi)$ nor $\Delta\varphi(\rho,\phi)$ is spatially constant for these masks.

In what follows, we shall restrict our attention to phase holograms, which may in general be associated with a transmittance $T(\rho,\phi)=A_0e^{i\Delta\varphi(\rho,\phi)}$. Physically, the phase modulation $\Delta\varphi(\rho,\phi)$ is induced in the incident beam due to the mean inner potential $V(\rho,\phi,z)$ of the material from which the holographic mask is constructed. This potential results in the addition of an energy term $eV(\rho,\phi,z)$ to the total Hamiltonian governing the time evolution of the electron beam in the material, resulting in a phase shift of the transmitted beam, relative to a reference wave having traveled the same distance in vacuum. From the general solution to the relativistically corrected Schrödinger equation, this phase shift is found to be

$$\Delta\varphi(\rho,\phi) = C_E\int_0^{t(\rho,\phi)} V(\rho,\phi,z)dz, \quad (2)$$

where $t(\rho,\phi)$ is the variation in the thickness of the hologram as a function of position in the transverse plane, and $C_E=\frac{2\pi e}{\lambda}\frac{E+E_0}{E(E+2E_0)}$ is a constant for a particular electron kinetic energy E, rest energy E_0, and λ de Broglie wavelength. In our case, the inner potential of the phase mask may be approximated by its mean value, V_0, such that [24,25]

$$\Delta\varphi(\rho,\phi) \approx C_EV_0\int_0^{t(\rho,\phi)} dz = C_EV_0t(\rho,\phi). \quad (3)$$

Hence, an arbitrary transverse phase profile can be imprinted on the incident beam, provided that variations in the local phase mask thickness $t(\rho,\phi)$ can be controlled with sufficient precision.

3. Generation and propagation of Bessel beams

We shall now focus our attention specifically on the generation of electron Bessel beams, which are described by scalar wavefunctions of the form

$$\Psi(\rho,\phi,z;t) = J_n(k_\rho\rho)e^{in\phi}e^{-i(E/\hbar t-k_zz)}, \quad (4)$$

where J_n represents an nth order Bessel function of the first kind, n is an integer, k_ρ and k_z are respectively the wavefunction's transverse and longitudinal wave vector components; $\hbar$ is the reduced Plank constant. These beams carry an amount of orbital angular momentum (OAM) along their propagation direction given by $L_z=n\hbar$ per electron, as indicated by the presence of a phase term $e^{in\phi}$ in the expression (4).

The generation of a Bessel beam necessarily entails imprinting a phase of the form $\Delta\varphi=\beta=k_\rho\rho+n\phi$ onto the incident wavefunction (see Appendix I). This is equivalent to imposing a conical wavefront on the electron beam [26], and can be achieved by