π/2 Mode Converters and Vortex Generators for Electrons

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Abstract

In optics, mode conversion is an elegant way to switch between Hermite Gaussian and Laguerre Gaussian beam profiles and thereby impart orbital angular momentum onto the beam and to create vortices. In optics such vortex beams can be produced in a setup consisting of two identical cylinder lenses. In electron optics, quadrupole lenses can be used for the same purpose. Here we investigate generalized asymmetric designs of a quadrupole mode converter that may be realized within the constraints of existing electron microscopes and can steer the development of dedicated vortex generators for high brilliance electron vortex probes of atomic scale.

Keywords: electron microscopy, vortex beams, mode conversion, orbital angular momentum

1. Introduction

A vortex beam can be characterized by a discontinuity in the phase that dictates a central void in the intensity profile. Further it features a quantized orbital angular momentum (OAM) in units of $\hbar$. The associated magnetic moment and chirality make electron vortex probes sensitive to magnetic excitations and even give them the ability to discriminate chiral crystals [1, 2, 3]. The development in the field was propelled by the close analogy to the established methods for optical vortex generation [4, 5, 6] as well as their application in helical spectroscopy [7, 8, 9]. While light optics has stimulated several methods of electron vortex generation [10, 11, 12], electron vortices can also be formed by multipoles [13] or magnetic fields [14, 15]. Each of these methods has its own merits and challenges, but none of them offers a pure singular OAM state without the need to block out unwanted portions of the intensity.

Yet one particular optical setup holds the promise to work on the entire beam in high purity, so that there would not be any need to filter out other diffraction orders or spurious unwanted angular states: the π/2 mode converter (MC) [16] converts Hermite Gaussian (HG) beams to corresponding Laguerre Gaussians (LG) ones and vice versa. The first order cases are:

$$HG(x,y) \propto 2x \cdot e^{-\frac{x^2+y^2}{w^2}},$$

$$LG(r,\phi) \propto 2r \cdot e^{-\frac{r^2}{w^2}} \cdot e^{i\phi}.$$  

(x,y) and (r,ϕ) are the Cartesian and polar coordinates, respectively. w is the width defining parameter. While two-way LG to HG beam conversion was demonstrated in a proof of principle experiment for electron beams [17], mode matching could not be achieved. Therefore the donut profile was only transient and could not be projected to another plane.

To picture how mode conversion occurs we can replace an incident beam with a straight central phase jump of π (a Hilbert beam) by two sub-waves that possess the same mirror symmetry as vertical and horizontal HG modes. Figure 1 demonstrates quite generally the essence of mode conversion: The two subwaves propagate independently from the entrance to the exit, where they will have accumulated a relative phase (i.e. Guoy shift) of $\pi/2$. Thus their coherent superposition creates an azimuthal ‘stair case’ phase ramp and a corresponding ring current. The beam has now non-zero OAM.

![Figure 1: principle of π/2 mode conversion.](image)

We propose that the quadrupole lenses in existing aberration correctors can be re-purposed to realize a fully functional π/2 MC. When an incoming beam is prepared with a suitable wavefront pattern, it would be completely trans-
formed into a vortex beam without sacrificing intensity. Switching between left and right handed helical operation would be as stable and reproducible as setting electron lenses. The ability to perform mode conversion on electron beams will doubtlessly also open new avenues in mode sorting [18], especially in conjunction with programmable phase masks [19].

We present an analytical treatment of general asymmetric setups and also run simulations for entire electron optical setups of different π/2 MCs.

2. Gaussian Mode Converters

2.1. The optical π/2 mode converter

We reproduce shortly the principle of the symmetric π/2 MC given in Ref. [16]. The schematic of the setup is illustrated in Fig. 2. There is an astigmatic beam waist located at \( z = 0 \) and two cylinder lenses at positions \( z = -a \) and \( z = a \) with a distance of \( d = 2a \). In light optics with static lenses this distance is the only adjustable degree of freedom in the setup. Apart from the incident Gaussian beam, there are 3 conditions to be fulfilled. The conditions are:

- the widths of Gaussian profile evolve as
  \[
  w(z) = \sqrt{1 + \left( \frac{z}{z_r} \right)^2} \cdot \sqrt{\frac{2z_r}{k}}. 
  \]
  (3)
  Here \( z_r \) is the Rayleigh range. The wavenumber \( k \) and wavelength \( \lambda \) follow \( k\lambda = 2\pi \). The widths in the \( xz \) and \( yz \) cuts must be equal at the exit plane \( z = +a \):

  \[
  w_x(a) = w_y(a). 
  \]
  (4)
  - the Gouy phase difference of the \( xz \) and \( yz \) components accumulated at the second cylinder lens must be \( \pi/2 \). Due to center symmetry this is equivalent to:

  \[
  \tan^{-1}\left( \frac{a}{z_{rx}} \right) - \tan^{-1}\left( \frac{a}{z_{ry}} \right) = \frac{\pi}{4}. 
  \]
  (5)

  - the radii of curvature

  \[
  R(z) = z \left( 1 + \left( \frac{z_r}{z} \right)^2 \right) 
  \]
  in the \( xz \) and in the \( yz \) cuts after the second cylinder lens must fulfill

  \[
  R_{x,z}(a) = R_{y,z}(a) = -R(-a). 
  \]
  (7)

  Equations 4 and 5 give immediately

  \[
  z_{rx} = a \left( \sqrt{2} - 1 \right), \quad z_{ry} = a \left( \sqrt{2} + 1 \right). 
  \]
  (8)

  With this result, the widths at the entrance and exit planes follow as:

  \[
  w(-a) = w(a) = \sqrt{\frac{\sqrt{2} \cdot d}{k}}. 
  \]
  (9)

  With Newton’s equations,

  \[
  \frac{1}{R_i} = \frac{1}{R_x(-a)} + \frac{1}{R_y(-a)} + \frac{1}{f} = \frac{1}{R_y(-a)} - \frac{1}{f}. 
  \]
  (10)

  and the previous results for the width Eqn. 9 and the Rayleigh ranges Eqn. 8, this gives

  \[
  d = \sqrt{2} \cdot f 
  \]
  (11)

  for a symmetric optical π/2 MC.

2.2. The quadrupole π/2 mode converter

The key differences between electron and light optics are that the distances and positions are fixed but the focal lengths can be controlled via lens excitations. Also, quadrupoles (QPs) are widely available. For electrons we thus propose to replace the cylinder lenses with QPs. This has no effect on Eqs. 4&5 and hence on the Rayleigh ranges \( z_R \). If the QPs are always focusing the \( xz \) component and defocusing the \( yz \) component, then Newton’s equations read as:

\[
\frac{1}{R_i} = \frac{1}{R_x(-a)} + \frac{1}{f} = \frac{1}{R_y(-a)} - \frac{1}{f}. 
\]
(12)

This modifies the relation from Eqn. 11 to

\[
f = \sqrt{2} \cdot d 
\]
(13)

and the widths at the entrance and exit plane scale accordingly:

\[
w(-a) = w(a) = \sqrt{\frac{2 \cdot \sqrt{2} \cdot d}{k}}. 
\]
(14)

Notably, the curvatures of the incident and outgoing beam simplifies to

\[
R_i = -R_o = -d. 
\]
(15)
Replacing cylinder lenses with QPs simplifies the solutions of the mode matching conditions considerably. The incoming and outgoing curvatures and the quadrupole focusing (or defocusing) are functions of the distance $d$ only, and not of the wavenumber $k$. Only the beam widths scale with $\sqrt{d/k}$. The actions of round lenses and QPs correspond to two orthogonal Zernike polynomials ($Z^0_2$ and $Z^2_3$, respectively), while a cylinder lens is a superposition of the two. QPs are therefore better suited for aligning a $\pi/2$ MC.

2.3. The asymmetric $\pi/2$ mode converter

If the constraint of equal focal lengths of the two QPs is relaxed, then the beam waists will be at different positions for the $xz$ and $yz$ component. In addition, the incoming and outgoing widths will differ, but the radii of curvature in Eqn. 15 are not affected.

If Eqns. 4&7 are met, (i.e. the astigmatism is canceled) then the the relative Guoy shift $\Psi$ is given by the distance $d$ as well as the QPs focal lengths $f_i$ and $f_o$.

$$\tan \Psi = \frac{2u}{1-u^2}, \quad u^2 = \frac{f_i f_o}{d^2} - 1.$$  \hspace{1cm} (16)

In a properly aligned $\pi/2$ MC, $\Psi$ is $\pi/2$ and the dimensionless parameter $u$ becomes 1. The conditions read (see supplementary information):

$$w_i = \sqrt{\frac{2 \cdot f_i}{k}}, \quad w_o = \sqrt{\frac{2 \cdot f_o}{k}}.$$  \hspace{1cm} (17)

The proper choice of quadrupole focal lengths Eqn. 17 for an incoming beam width $w_i$ according to Eqn. 18 allows to achieve $u = 1$ with a magnification of $w_o/w_i$. A schematic example is sketched in Fig. 3.

2.4. Practical considerations

When it comes to electron optical alignment, the very appealing benefit of the asymmetric $\pi/2$ MC design is that there is only one prior requirement on the non-astigmatic incoming beam. Its curvature has to be centered onto the principal plane of QP2. This can be readily achieved by focusing a wide enough beam onto QP2. If the incoming width $w_i$ is several times larger than the width $w$ that would be required in a symmetric $\pi/2$ MC (Eqn. 14), $z_r << d$ will also hold, and the required lens excitations can be found by minimizing the effects of wobbling QP2. Then a smaller condenser aperture can be used with the same lens settings, to provide a smaller $w_i$ with the correct curvature. If the reduced $w_i$ is comparable to the $w$ of the symmetric $\pi/2$ MC, the required $f_i$ and $f_o$ will also be comparable. Since the outgoing radius of curvature does not depend on the Guoy shift, pairs of $f_i$ and $f_o$ can be realized by choosing any one and adjusting the other one, until $u$ from Eqn. 16 becomes 1. There is no need to match the width of the symmetric $\pi/2$ MC exactly.

3. Spherical Mode Converters

3.1. Guoy phase

Sculpting a Gaussian intensity profile is impractical if not impossible in electron microscopy. Instead we consider standard spherical waves as an input. The Guoy phase of an astigmatic higher order HG$_{nm}$ beam follows a $\tan^{-1}$ function.

$$\Psi = (n+\frac{1}{2})\tan^{-1}\left(\frac{z-z_{rx}}{z_{rx}}\right) + (m+\frac{1}{2})\tan^{-1}\left(\frac{z-z_{ry}}{z_{ry}}\right)$$  \hspace{1cm} (19)

where $z_{rx}$ and $z_{ry}$ are the positions of the line foci and $z_{rx}$ and $z_{ry}$ are the respective Rayleigh lengths. A spherical wave has a different Guoy phase. A typical example of an incoming electron beam in the geometric optic regime is shown in Fig. 4.

![Figure 4: Guoy phase of a spherical wave (full lines) and an HG$_{00}$ beam (dashed) at different radii. A lens with a focal length of 120 mm is positioned at $z = -119.4$ mm. The aperture radius and Gaussian width at the lens are $w = r/\sqrt{2} = 1250$ nm. Acceleration voltage $U_a = 200$ kV. The inset shows the Gaussian waist and Airy disk formed at $z = 0$, the dots correspond to the different radii. Horizontal gridlines are at $\pi/4$, $\pi/2$ and $3\pi/4$, vertical gridlines count Rayleigh ranges $z_r = 7.3$ mm of the Gaussian beam.](image-url)
Guoy phase at roughly 1/3 and 2/3 of the radius of the Airy disk are also very linear up to \( z = z_R \). This comparison illustrates that a Gaussian input for the \( \pi/2 \) MC can be replaced by a spherical wave with a scaled diameter.

### 3.2. Hilbert beams

A feasible approach to produce an electron beam similar to a HG is a Hilbert plate that induces a phase shift of \( \pi \) between the two halves of a round aperture. One may also use a magnetic bar to this aim [21, 22]. In the following, we shall refer to such a phase shifter as a Hilbert device, independent of the principle used. Beams produced with such a device are henceforth called spherical Hilbert beams.

The Guoy shift of HG beams in the mode converter can be calculated analytically with Eqn. 19. For spherical Hilbert beams we have to resort to wave optical simulations. To this aim we performed two independent simulations of an asymmetric QP \( \pi/2 \) setup for the horizontal and vertical components as suggested in Fig. 1. The magnification is the same as in Figs. 6&9 with \( r_i = 357 \) nm, \( d = 120 \) mm and quadrupole focal lengths of \( f_i = 80 \) mm and \( f_o = 360 \) mm. The acceleration voltage is \( U_a = 200 \) kV.

\[
\langle \hat{L}_z \rangle = \frac{\langle \psi | \hat{L}_z | \psi \rangle}{\langle \psi | \psi \rangle} = \hbar \sum_m |m| \int |c_m(r)|^2 r dr. \tag{21}
\]

Figure 6 shows the phase structure before and after the second quadrupole for a \( HG_{0,1} \) and a Hilbert beam. The parameters for the \( \pi/2 \) MC are identical to those in Figs. 5&9. Note that the phase structure has been compensated for the diverging curvature Eqn. 15. The remaining purely azimuthal phase structure at the entrance to QP2 is visibly astigmatic for both beam profiles. Indeed, the decomposition according to Eqn. 20 reveals a broadened distribution. After QP2 the astigmatism is corrected, and the \( m = 1 \) contribution increases. The \( HG_{0,1} \) beam is transformed into a clean \( m = 1 \) LG state. QP1 did already exert the full torque of \( \langle m \rangle = 1 \), while QP2 establishes mode purity. The Hilbert beam picks up angular momentum on QP1 and QP2 and does also acquire a total of \( \langle m \rangle = 1 \), albeit with a slightly lower \( m = 1 \) mode purity. The mode
purity may be further increased by another aperture, as the central region shows an ideal linear azimuthal phase spiral.

4. Numerical Simulations

So far the setups for $\pi/2$ MCs were very much simplified. They were modeled by a composite input of an aperture, a Hilbert device, a lens and a QP followed by one single propagation step and a composite output of a lens and a QP. The analytic treatment of Gaussian beams passing through such stylized $\pi/2$ MCs as well as the very similar behavior of HG and Hilbert beams in test scenarios suggest that vortex generation is possible in an actual aberration corrected TEM. Numerical simulations on more realistic and complete setups are indispensable to confirm and possibly retune the parameters for real world electron optical designs. The obvious challenge of simulating an entire electron optical setup, is keeping track of multiple optical devices and propagation steps in between them.

4.1. Rescaled propagation

In an extended optical system, like an entire TEM column, different sections of the beam have very different lateral extend or magnification. One very efficient way to adapt the lateral scale to a propagated beam can be to replace the combined action of a lens with focal length $f$ and further propagation over a distance $d$ with the combined action of a propagation over a distance $d'$, a lateral rescaling and a lens with focal length $f'$. So instead of propagating forward to the imaging plane, the incident wavefront is propagated backwards to the object plane. Then the magnification of the imaging and a new lens with focal length $f'$ are applied. The transformed distance $d$ does not need to be the full distance to the next lens or aperture. In fact it can be chosen freely, and the signs of the rescaled and remaining distance are arbitrary. With the introduction of $s$ and $s'$ for the original and the re-scaled grid resolution, the transformations can be written as:

$$\frac{1}{d'} = \frac{1}{d} - \frac{1}{f}$$

$$s' = 1 - \frac{d}{f'}$$

$$f' = f - d.$$  (24)

Sign changes in $d$ are equivalent to propagating backwards. Sign changes in $f$ and $s$ trigger a mirror inversion and a phase shift of $\pi$.

4.2. Virtual microscope

In this section, we present a detailed numerical study of the propagation behavior of HG and Hilbert beams through an ensemble of lenses and QPs. To this aim, we have developed a JAVA plugin for ImageJ. The graphical user interface represents a fully editable virtual microscope. Different setups can be stored in human readable and editable xml files which define among other parameters a unique order in which lenses, apertures and propagation distances are applied to an initial plane wave. Wavefronts of the propagated beam can be viewed as stacks of images.

Numerically, lenses $L$, apertures, quadrupoles $QP$ and other devices are represented as a complex map for the real and the imaginary part of their action, respectively.

$$L(f, x, y) = \exp \left[ i \cdot \left( x^2 + y^2 \right) \cdot \frac{k}{2 \cdot f} \right]$$  (25)

$$QP(f, x, y) = \exp \left[ i \cdot \left( x^2 - y^2 \right) \cdot \frac{k}{2} \right]$$  (26)

The simulation starts with a plane wave with phase 0 at $z = 0$. At every plane, the current cross section $\psi_z$ is multiplied with the complex sheets at this plane. This step can account for arbitrary apertures, gratings, Hilbert devices, wavefront deformations by lenses and multipoles. It can also define for instance a Hermite Gaussian. Then the wavefront is propagated through free space to the next plane. The propagation over a distance $d$ from $\psi_z$ to $\psi_{z+d}$ can be individually configured to be carried out in customizable steps. We always employ the par-axial approximation, since lateral dimensions are $\mu$m and relevant distances are at least mm. Each step can be propagated in frequency or spatial domain.

$$\psi_{z+d} = \mathcal{F}^{-1}(\mathcal{P}(d) \cdot \mathcal{F}(\psi_z))$$

$$\psi_{z+d}(x, y) = -i \int_{x', y'} \psi_z(x', y') \cdot \exp \left[ \left( x-x' \right)^2 + \left( y-y' \right)^2 \right] \cdot \frac{d}{2} \cdot dx' dy'$$  (28)

$\mathcal{F}$ denotes Fourier transformation and the propagator $\mathcal{P}$ in Eqn. 27 is defined in frequency range $\hat{x}, \hat{y}$

$$\mathcal{P}(d, \hat{x}, \hat{y}) = \exp \left[ i \cdot \left( \hat{x}^2 + \hat{y}^2 \right) \cdot \frac{d}{2 \cdot k} \right]$$  (29)

Propagation steps in spatial domain (Eqn. 28) may also contain a custom zoom between the planes at $z$ and $z + d$.

The custom splitting and scaling and per step choice between spatial and frequency domain are found to be versatile in circumventing the need for excessive oversizing or oversampling of the complex sheets and wavefronts. All simulations could be carried out on a grid of 512x512 pixels with dynamic resolution. The phase information is consistent with the Guoy shift, but there is an arbitrary global phase factor for different planes.

The first test case for the virtual microscope is the symmetric cylinder lens setup. Figure 7 illustrates the propagation of the phase colored wavefronts through a basic symmetric $\pi/2$ MC setup. The incoming rotated HG$_{1,0}$ ($U_a = 200$ kV) has a width $w_i = 367$ nm. The round lenses with $f_L = 409.7$ mm are on the inside but share the same plane with the cylinder lenses with $f_c = 84.9$ mm which are 120 mm apart. In this ordering the effects of the first and
Figure 7: Propagation of an incident rotated $HG_{0,1}$ with width $w = 367\,\text{nm}$ and $U_a = 200\,\text{kV}$ through cylinder lenses with $f = 84.9\,\text{mm}$ and round lenses with $f = 409.7\,\text{mm}$. The last frame is a Laguerre Gaussian (LG) with the same $w$. The scalebar is 500 nm. The wavefront is shown for every 10 mm.

second cylinder lens are not obscured by the isotropic curvature. The first cylinder lens introduces horizontal bands and a vertical phase curvature. The following intermediate wave fronts visualize the continuous mode conversion. And finally the second cylinder transforms the asymmetric phase pattern after the second lens into the exact LG pattern for $m = 1$.

4.3. Spherical waves and multi-scale simulation

Moving towards a more realistic virtual setup necessitates to include round apertures and spherical waves as well as the condenser and the objective lens systems.

The full asymmetric $\pi/2$ MC setup is sketched in Fig. 8. The Hilbert device is assumed to be mounted in the condenser system and has a diameter of 10 $\mu$m. The black dots mark actual images. The lens labeled "demag" would form another image (gray dot) at the principal plane of the second QP (QP2). The first QP (QP1) introduces astigmatism and forms one real line focus (red dot). The corresponding perpendicular line focus is virtual (blue dot). After the second QP (QP2) the beam is mode matched and appears as if emanating from an image (gray dot) in the principal plane of QP1. It is refocused in another real image in front of the condenser/objective system. The last black dot is in the focus of the objective.

The full wave optical simulation for the extended asymmetric $\pi/2$ MC setup in Fig. 8 is shown in Fig. 9. The Hilbert device acts as a beam splitter and introduces a phase shift of $\pi/2$. The Hilbert beam is incident on a Hilbert device in an aperture with a diameter of 10 $\mu$m. A 1 $\mu$m wide magnetic bar induces a phaseshift of $\pi$ between the two sides. The Hilbert device is excited asymmetrically with $f_i = 80\,\text{mm}$ and $f_o = 360\,\text{mm}$ to match the incoming width (Eqn. 18) and to provide the correct Guoy shift (Eqn. 16). The output of the $\pi/2$ MC is clearly a vortex beam and the spiraling phase pattern has a diverging curvature centered at QP1. The magnification inside the $\pi/2$ MC is $w_o/w_i = 2.12$. The next frames are before the "mag" lens and after the condenser. The magnified beam is focused by the objective lens to form a donut shaped STEM probe. Changing the helicity of the STEM probe is as straightforward and reproducible as rotating the QPs by 90°, which is a crucial aspect for measuring dichroism [23]. Notably the central bar in the Hilbert device and its diffractive blurring upon propagation to the first QP contribute to the resemblance of a HG$_{1,0}$ beam.

The second cylinder lens in Fig. 9. Lenses (purple) and quadrupoles (red/blue) focus an incoming Hilbert beam. Black and gray dots mark real images, the red/blue dot mark the real/virtual astigmatic line focus of the first quadrupole. The given diameters $\varnothing$ are according to geometric optics.

Figure 8: Complete optical setup for vortex generation. This setup is used in Fig. 9. Lenses (purple) and quadrupoles (red/blue) focus an incoming Hilbert beam. Black and gray dots mark real images, the red/blue dot mark the real/virtual astigmatic line focus of the first quadrupole. The given diameters $\varnothing$ are according to geometric optics.

Figure 9: Propagation from a Hilbert device in the condenser to a STEM probe with orbital angular momentum $\pm \hbar$. Captions are explained in the text. The hue coloring is identical to Fig. 7 is at QP2 (Eqn. 15). The distance $d$ between the QPs is 120 mm. The quadrupoles are excited asymmetrically with $f_i = 80\,\text{mm}$ and $f_o = 360\,\text{mm}$ to match the incoming width (Eqn. 18) and to provide the correct Guoy shift (Eqn. 16). The output of the $\pi/2$ MC is clearly a vortex beam and the spiraling phase pattern has a diverging curvature centered at QP1. The magnification inside the $\pi/2$ MC is $w_o/w_i = 2.12$. The next frames are before the "mag" lens and after the condenser. The magnified beam is focused by the objective lens to form a donut shaped STEM probe. Changing the helicity of the STEM probe is as straightforward and reproducible as rotating the QPs by 90°, which is a crucial aspect for measuring dichroism [23]. Notably the central bar in the Hilbert device and its diffractive blurring upon propagation to the first QP contribute to the resemblance of a HG$_{1,0}$ beam. Except for the magnification the cross sections from the interior of the $\pi/2$ MC closely resemble the internal cross sections shown in Fig. 7. The differences in the spiraling phase pattern before (120 mm) and after QP2 might seem subtle in direct phase coloring, but they are the same as in the isophasal representation shown in Fig. 6. The second QP

$\varnothing 10\,\mu m$
$\varnothing 0.7\,\mu m$
$\varnothing 1.5\,\mu m$
$\varnothing 2.4\,\mu m$
$\varnothing 2.6\,\mu m$

$1000\,nm$
$2000\,nm$
$4000\,nm$
$10,000\,nm$
$10,000\,nm$
$2\,nm$
5. Conclusion

We have explored the realm of possible designs for $\pi/2$ MCs in electron optics based on reconfiguring well established and relatively wide spread probe correctors. Using already existing quadrupoles is an appealing aspect, because there is no need for mechanical modifications of the TEM column and helicity switching would be straightforward. The most relevant parameters are the distance between the two quadrupoles $d$, the possible excitations of the quadrupoles or minimal $f_l$ and $f_o$, as well as the incident virtual aperture size. Allowing for asymmetry in the quadrupole excitations introduces a magnification or de-magnification and leads to an effective decoupling of the constraints on achieving isotropic width and curvature as well as $\pi/2$ mode conversion at the exit plane. We propose that a $\pi/2$ MC can be used to generate a very pure, and switchable $m = \pm 1$ vortex beam. The design of the Hilbert device we have considered here numerically is minimalistic, and there are conceivable aperture designs that could mimic a HG$_{0,1}$ input beam even more closely.

Significantly smaller probe diameters could be envisaged in dedicated setups with intermediate magnification stages and additional apertures.

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