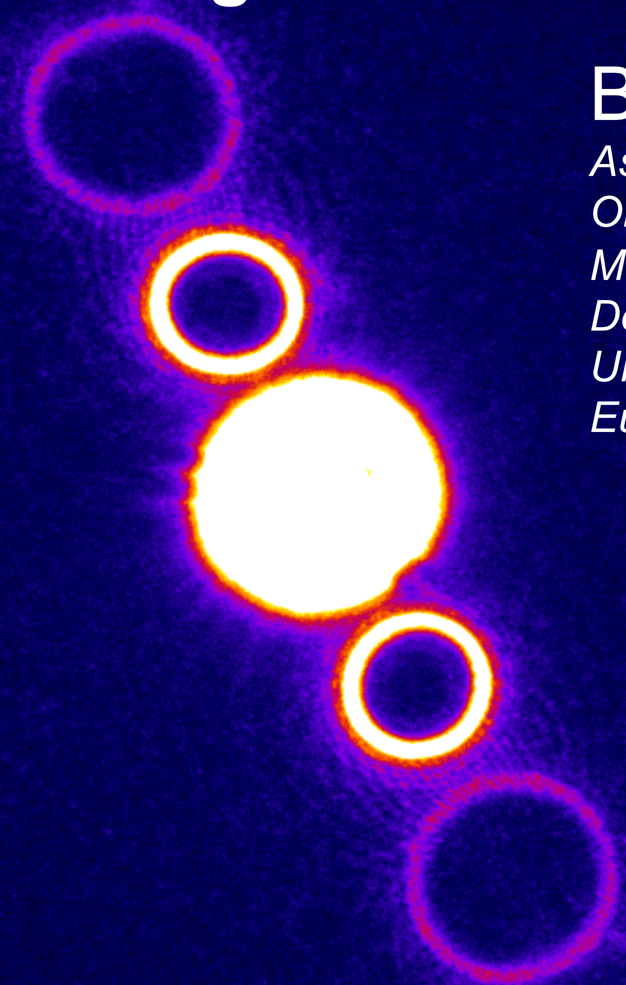
Electron Microscopy with Vortex Beams Carrying Orbital Angular Momentum



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Thanks to

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Oregon's High-Tech Extension Service

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LDRD Carbon Cycle 2.0
"Electron Microscopy with Vortex Beams"

U.S. DEPARTMENT OF ENERGY | Office of Science

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This data thanks to

UNIVERSITY OF OREGON

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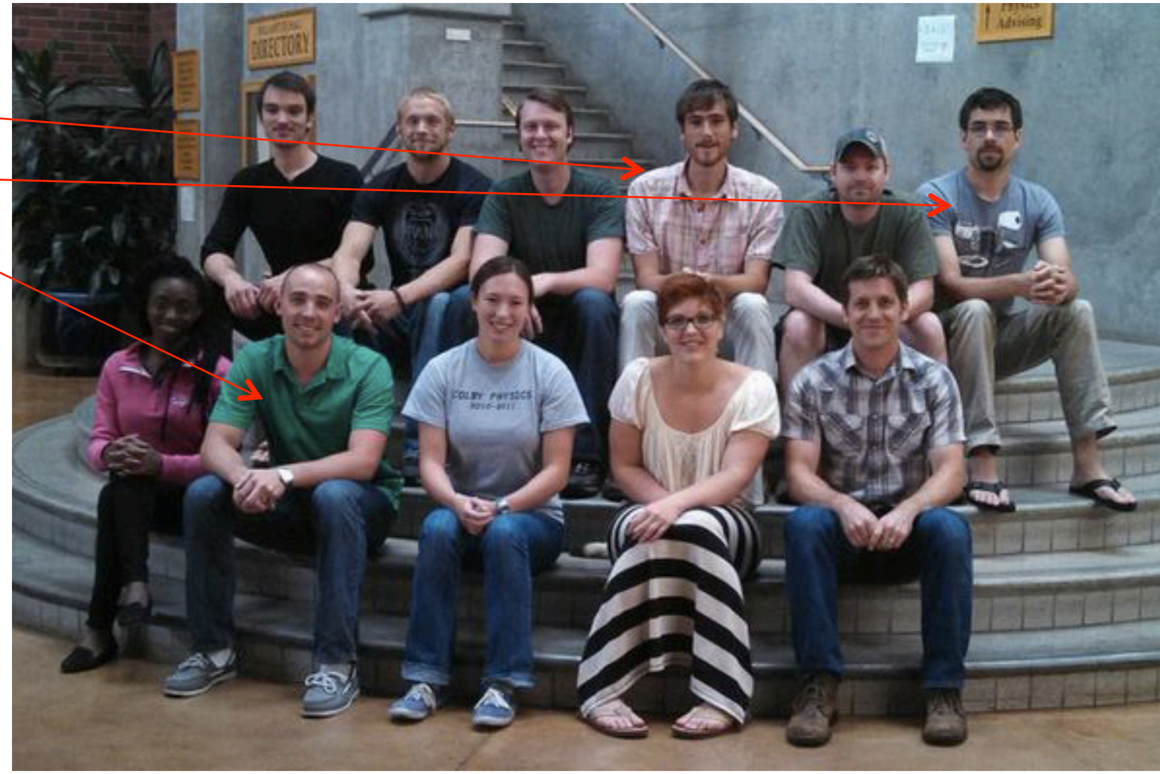

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A National Nanoscience Research Facility

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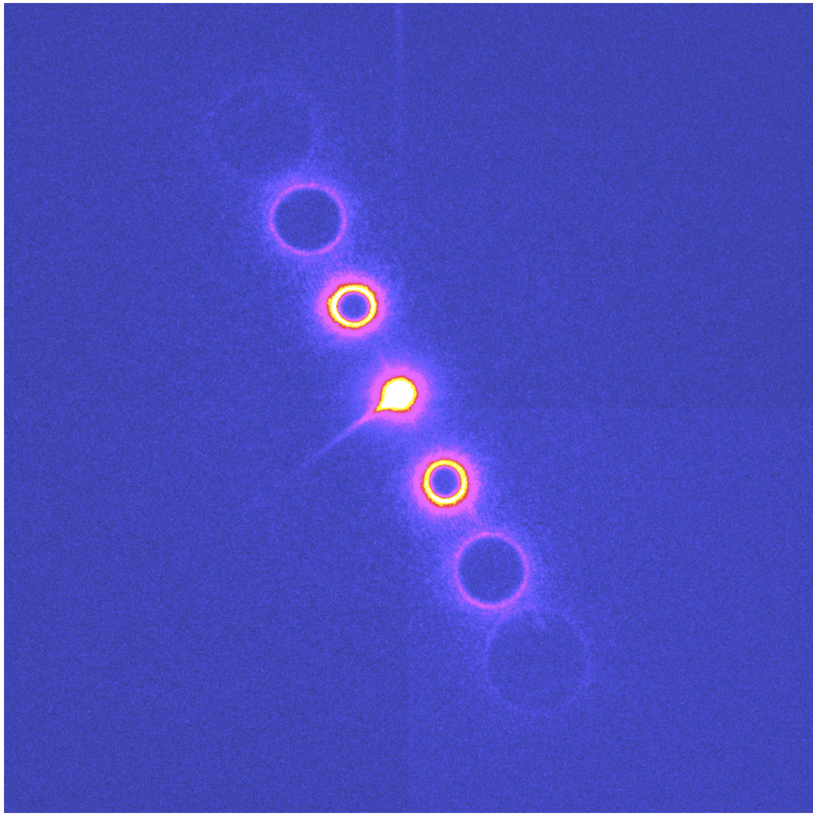
Overview

What is a vortex beam?:

- helical phase
- quantized orbital angular momentum



How is a vortex beam produced?:

- spiral phase plate
- holographically (diffractive optics)
- detected?

Is a vortex beam useful?:

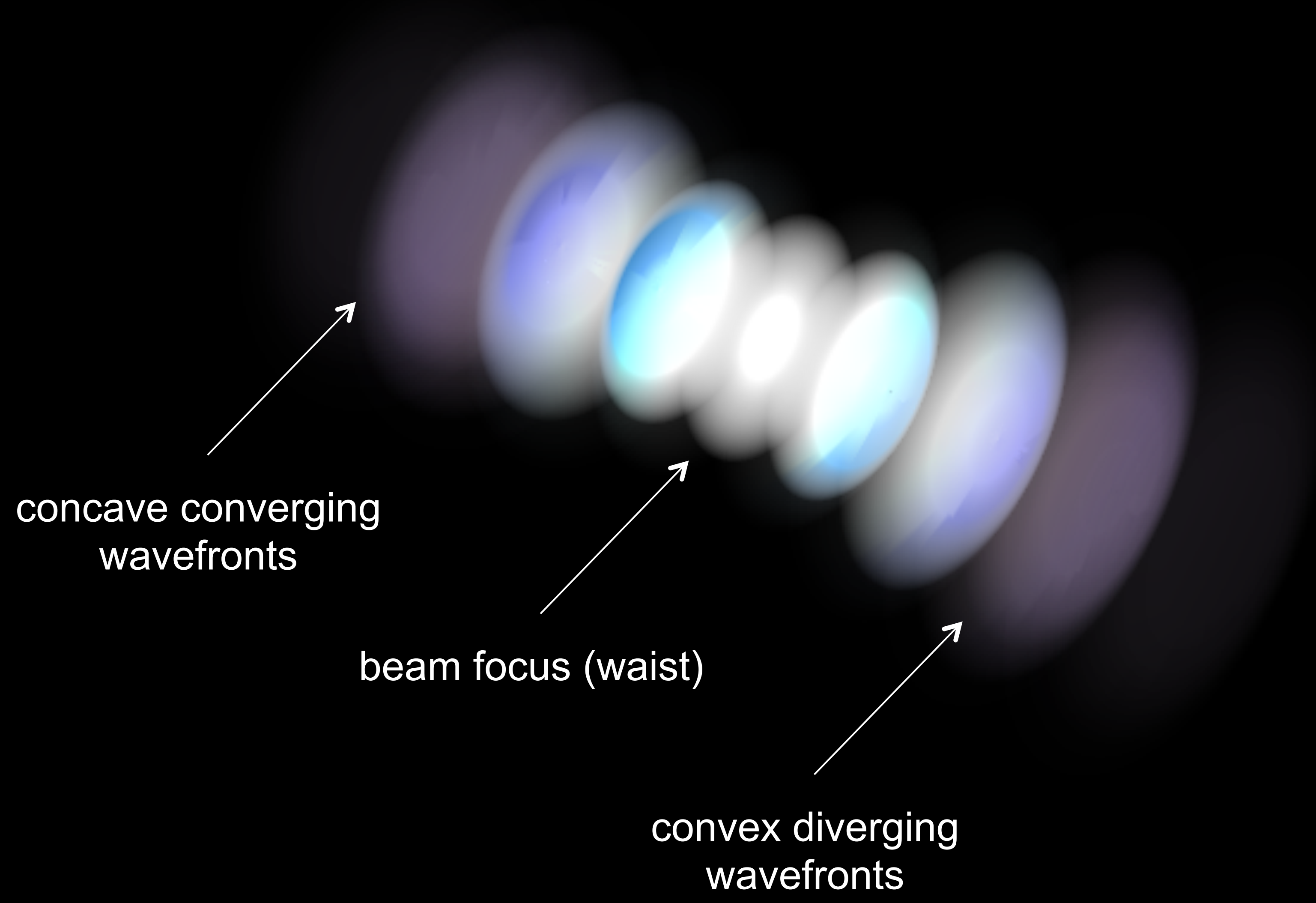
- optical trapping/manipulation
- phase imaging/coherence filter
- magnetic sensitivity

What is a vortex beam?

Example:

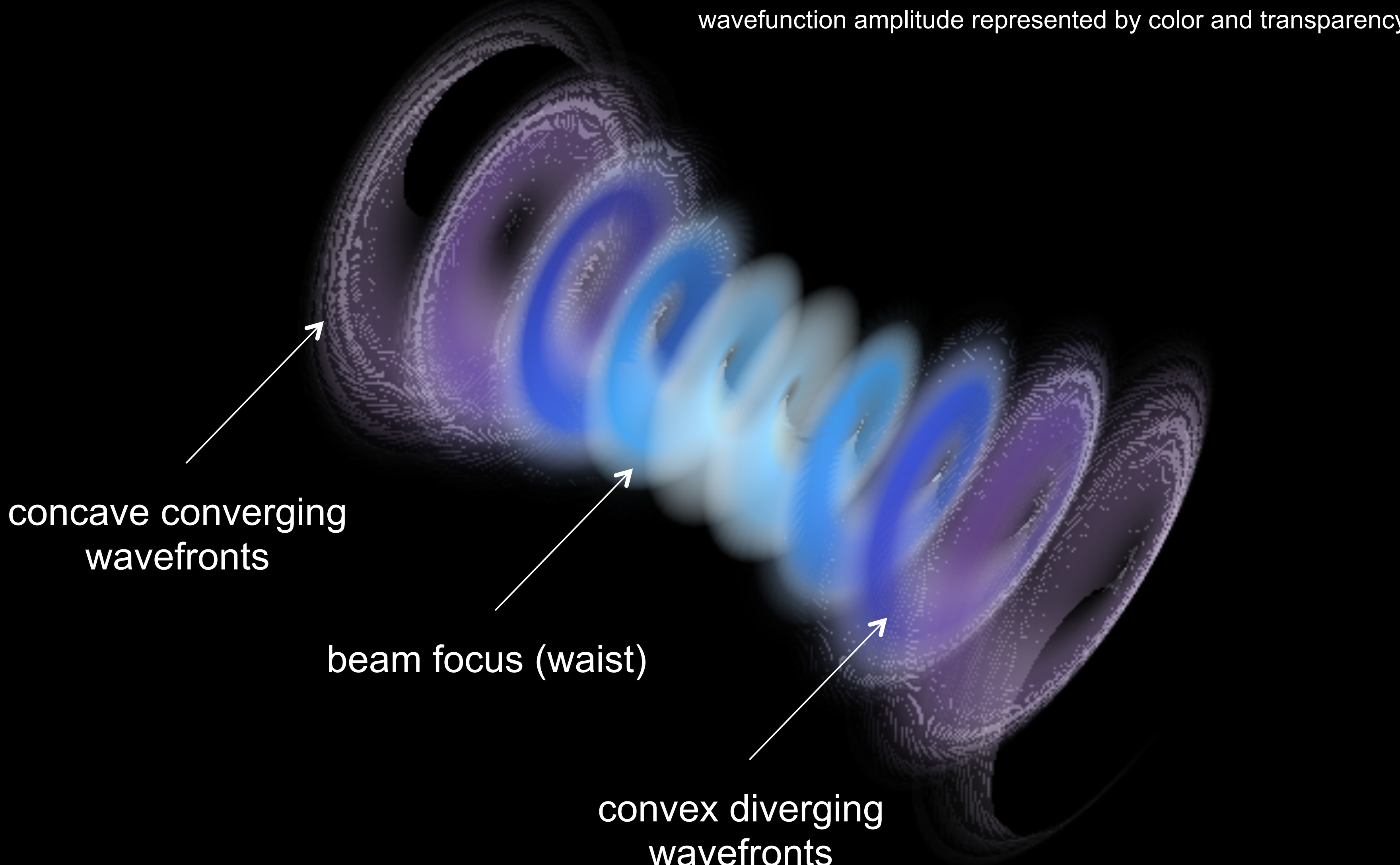
Suppose we want a regular beam with flat wavefronts, a plane wave:

wavefunction amplitude represented by color and transparency



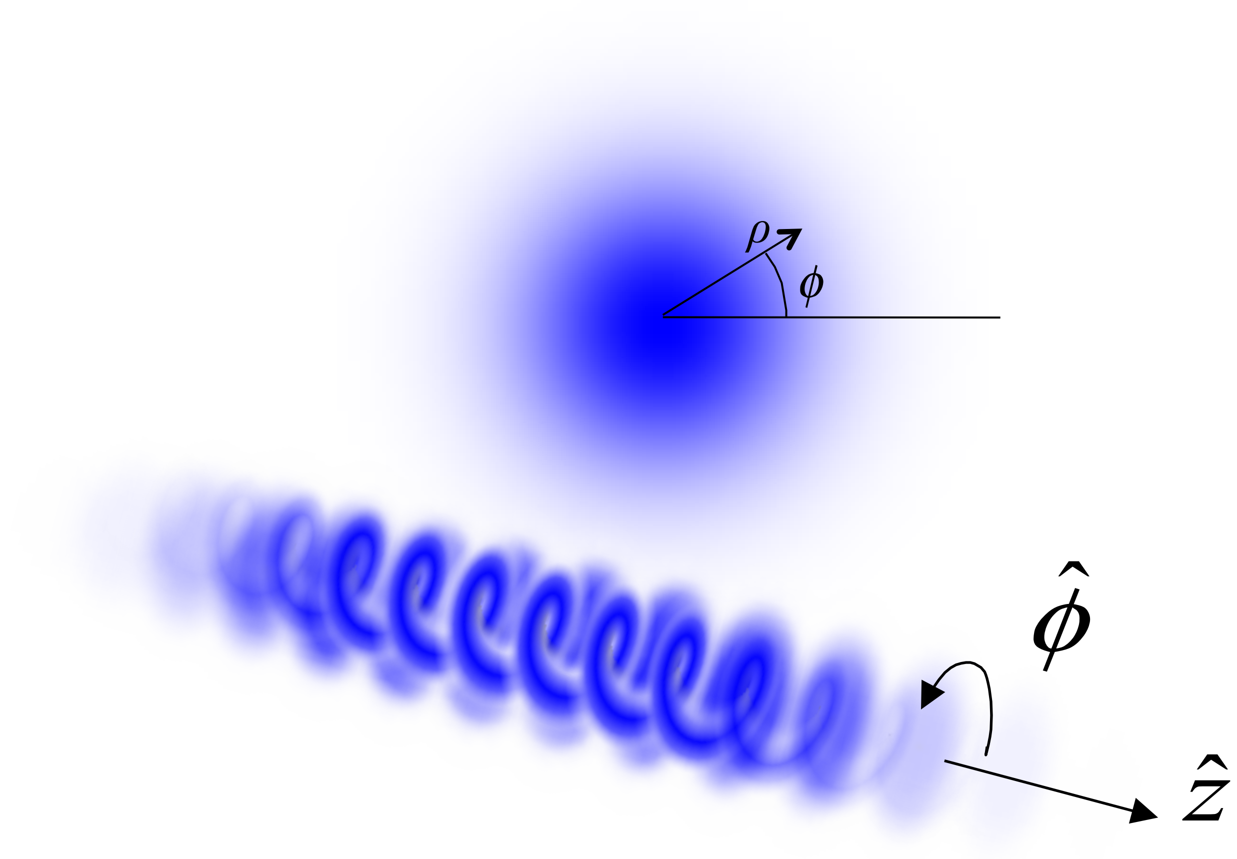
Another Example:

Suppose we want a "vortex beam" with azimuthal phase topological screw dislocation in wavefronts



consider a plane wave:
imprint with azimuthal phase

$$u(\rho,\phi,z) \propto e^{-\left(\rho^2/w^2\right)} e^{ikz} e^{-im_\ell \phi}$$



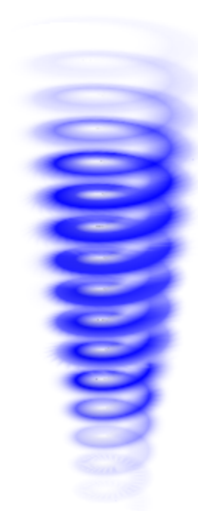
simplest optical vortex - Laguerre-Gaussian beams:

$$LG_{0m_\ell}(\rho,\phi,z) \propto \rho^{m_\ell} e^{-\left(\rho^2/w^2\right)} e^{ikz} e^{-im_\ell\phi}$$

$\underline{\ell = 0}$

$\underline{\ell = 1}$

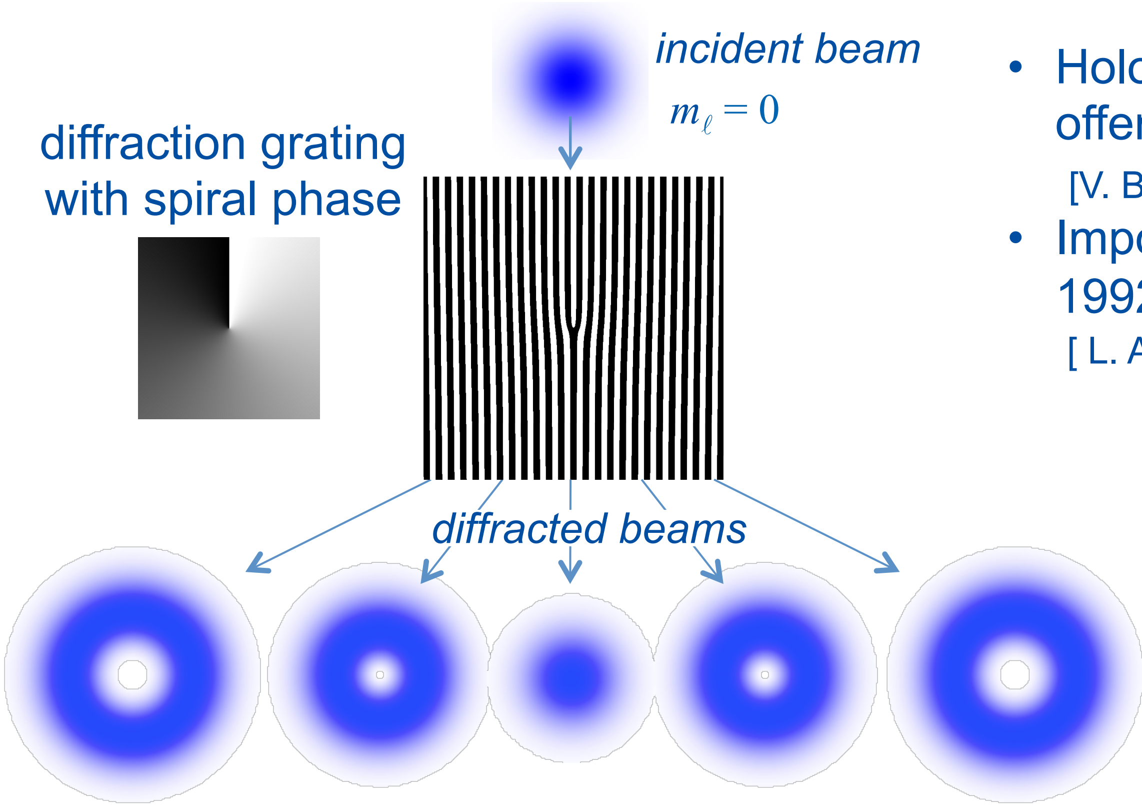
$\underline{\ell = 2}$



"screw"-shaped wavefronts

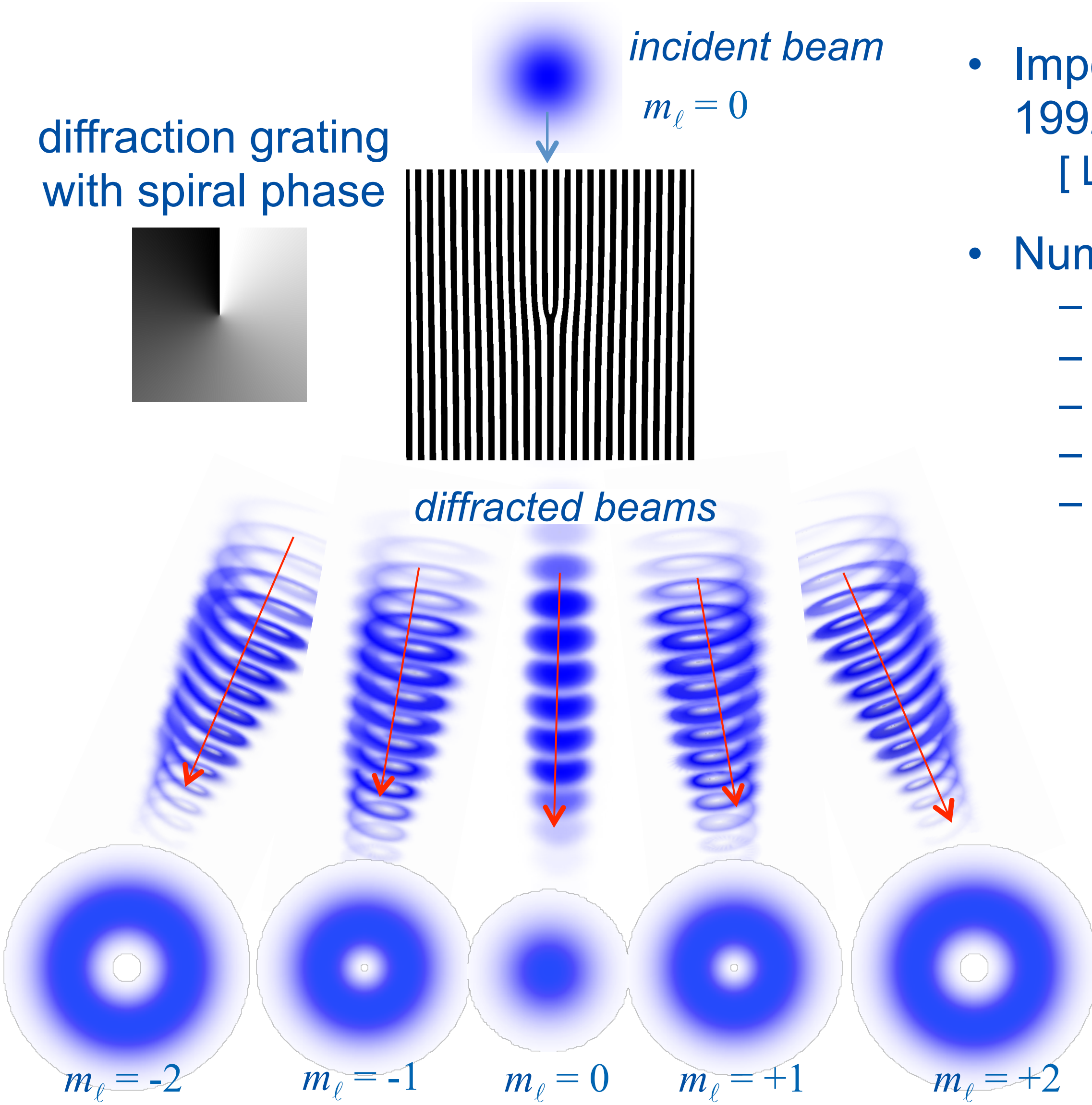
- "thread pitch" λ
- "number of starts" m_ℓ
- "lead" $m_\ell \lambda$

Light Optics: beams with helical phase made by physical holograms



- Holograms (spatial light modulator) offer *precise* way to produce
  [V. Bazhenov *et al.*,JETP Letters. 52, 429 (1990)]
- Important topic in light optics since 1992*
  [L. Allen *et al.*, PRA **45**, 8185 (1992)]

Light Optics: beams with helical phase made by physical holograms



- Important topic in light optics since 1992*
  [L. Allen *et al.*, PRA **45**, 8185 (1992)]
- Numerous applications:
  - microscopy
  - astronomy
  - micromanipulation
  - communications
  - quantum state manipulation

L. Allen *et al.*, PRA 45, 8185 (1992):
"Vortex beams" carry orbital angular momentum (OAM)

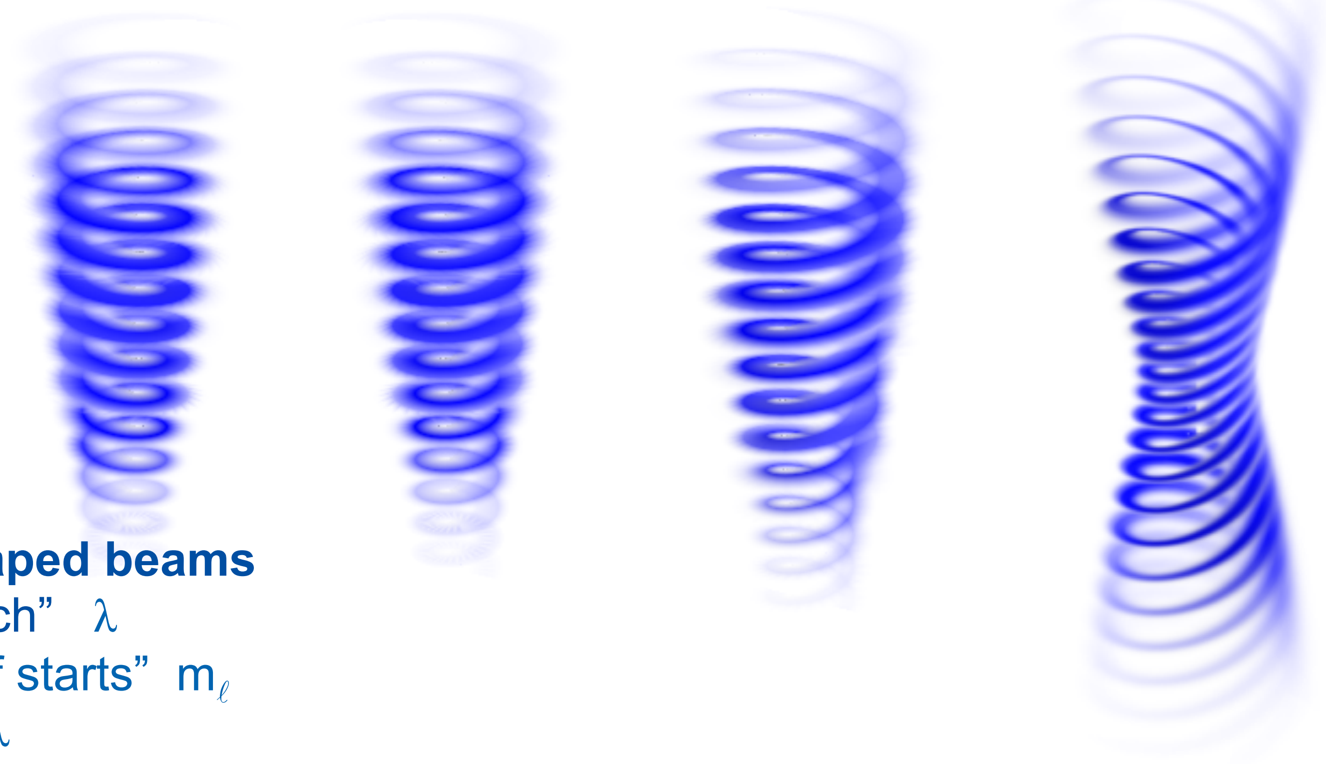
$$\psi(\rho,\phi,z) = f(\rho,z)\,e^{-im_\ell\phi}$$

OAM operator: $\hat{L}_z = i\hbar\dfrac{\partial}{\partial\phi}$

$$\hat{L}_z\left[\psi_{m_\ell}\right] = \hbar m_\ell\left[\psi_{m_\ell}\right]$$

$m_\ell = -1$ | $m_\ell = 1$ | $m_\ell = 2$ | $m_\ell = 5$

$L_z = -\hbar$ | $L_z = +\hbar$ | $L_z = +2\hbar$ | $L_z = +5\hbar$

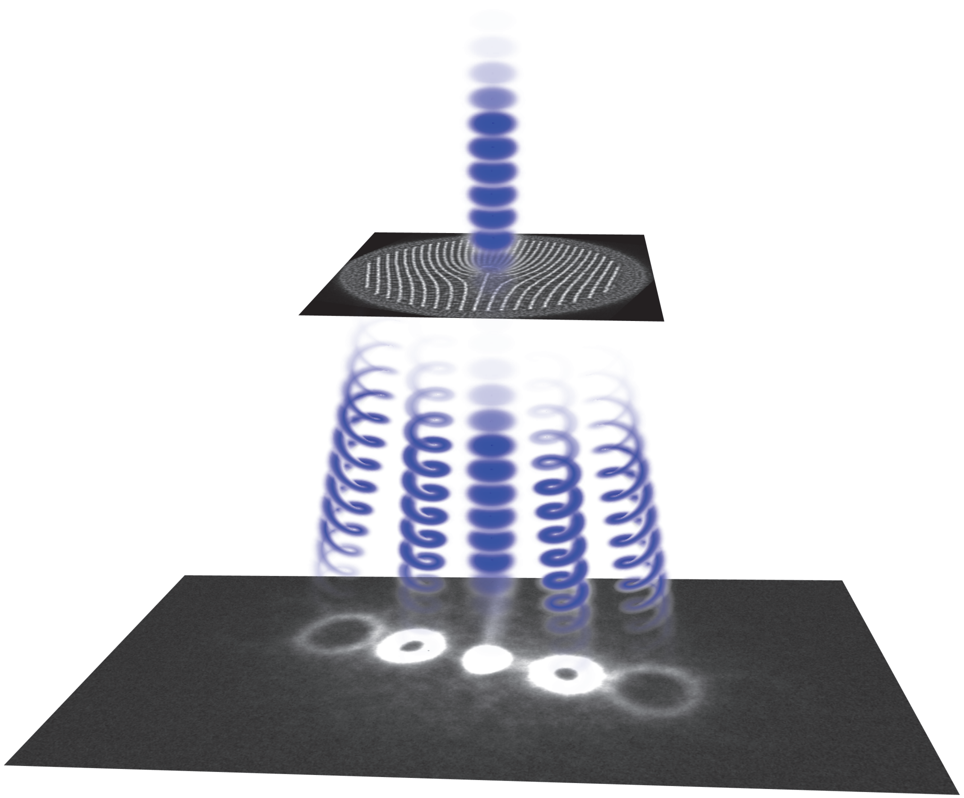


"screw"-shaped beams

- "thread pitch" λ
- "number of starts" m_ℓ
- "lead" $m_\ell\,\lambda$

Can also make *electron* vortex beams

- 300 keV electrons (λ = 2 pm)
- nanofabricated diffractive holograms
- 39 μrad diffraction angle
- TEM




back scatter

Electron vortex beams

Just as electron wavefunctions in atomic orbitals have orbital angular momentum (OAM), so too can light waves in optical beams (see "Light's orbital angular momentum," by Miles Padgett, Johannes Courtial, and Les Allen, PHYSICS TODAY, May 2004, page 35). Over the past year, several different research groups have demonstrated that OAM can also be conferred on electron beams.

This CCD image comes from recent work by Benjamin McMorran and colleagues at NIST. The bright center spot has zero OAM; each spot further to the left of center has an additional OAM of $-25\hbar$ and to the right, $+25\hbar$, up to $\pm 100\hbar$. Destructive interference along the axis of the diffracted beams yields an intensity node at each vortex core and generates the characteristic ring shapes (here in false color). Whereas the electron wavefunction has the characteristic $\exp(im\varphi)$ phase dependence on azimuthal angle, the electrons themselves don't spiral but travel straight.

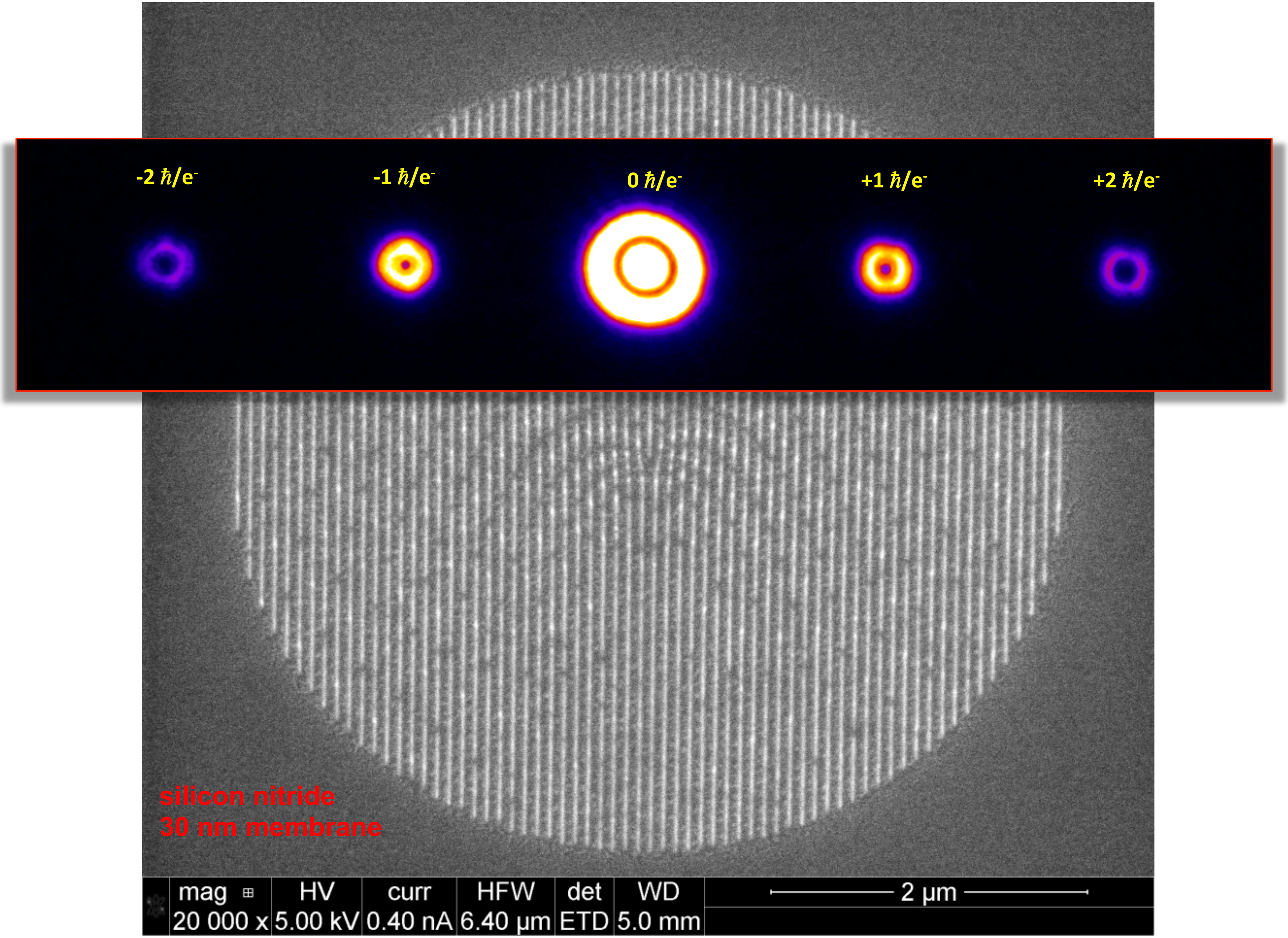
To make electron vortex beams, the researchers passed the Gaussian beam of a transmission electron microscope (TEM) through a nanoscale hologram consisting of a diffraction grating with so-called fork dislocations—in this case 25 extra lines inserted on one half of the grating. Electron vortex beams could provide new magnetic, crystallographic, chemical, and phase probes for TEM samples. (B. J. McMorran et al., *Science* **331**, 192, 2011; image © B. McMorran, A. Herzig/NIST.)

To submit candidate images for Back Scatter, visit http://www.physicstoday.org/backscatter.html.

84 March 2011 Physics Today www.physicstoday.org

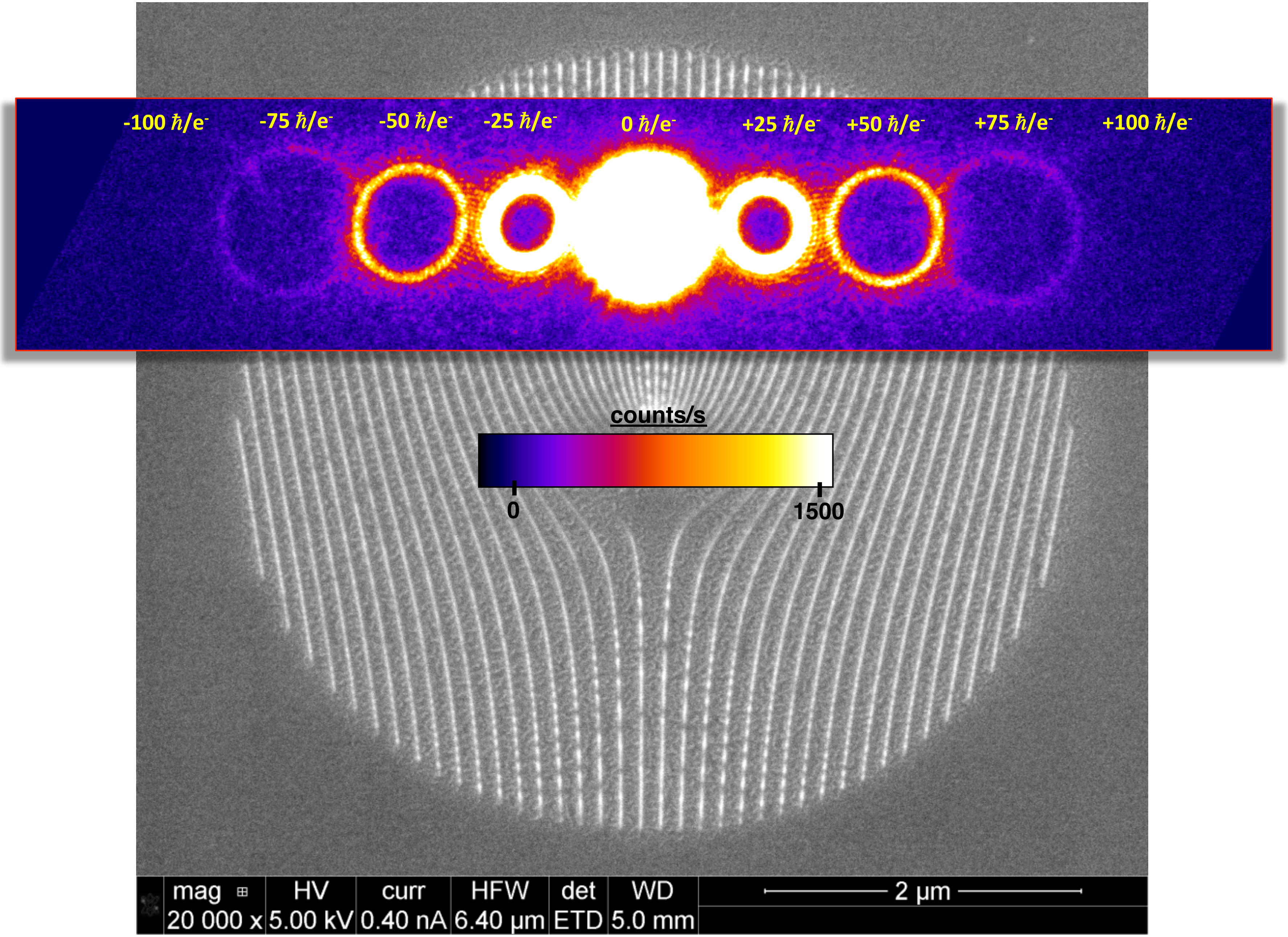
B.J. McMorran *et al.*, *Science* **331**, 192 (2011)

Nanofabricated diffraction hologram for electron vortex beams:

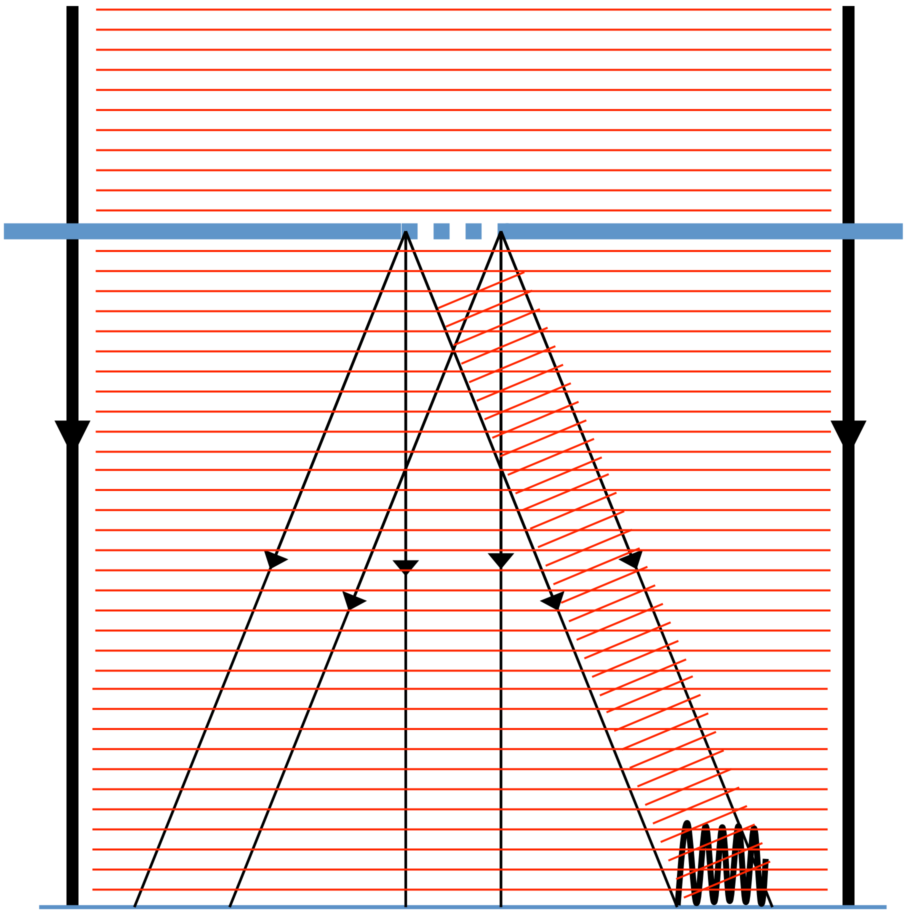


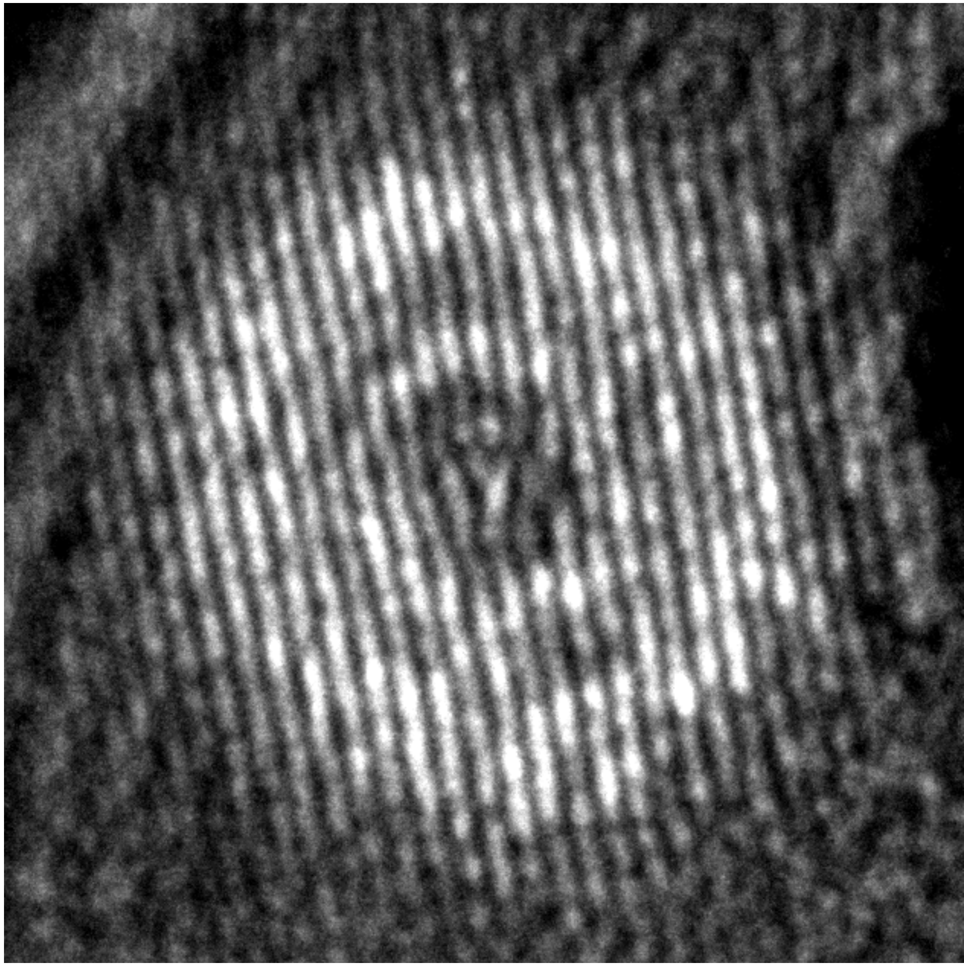
B. McMorran

Nanofabricated diffraction holograms for electrons

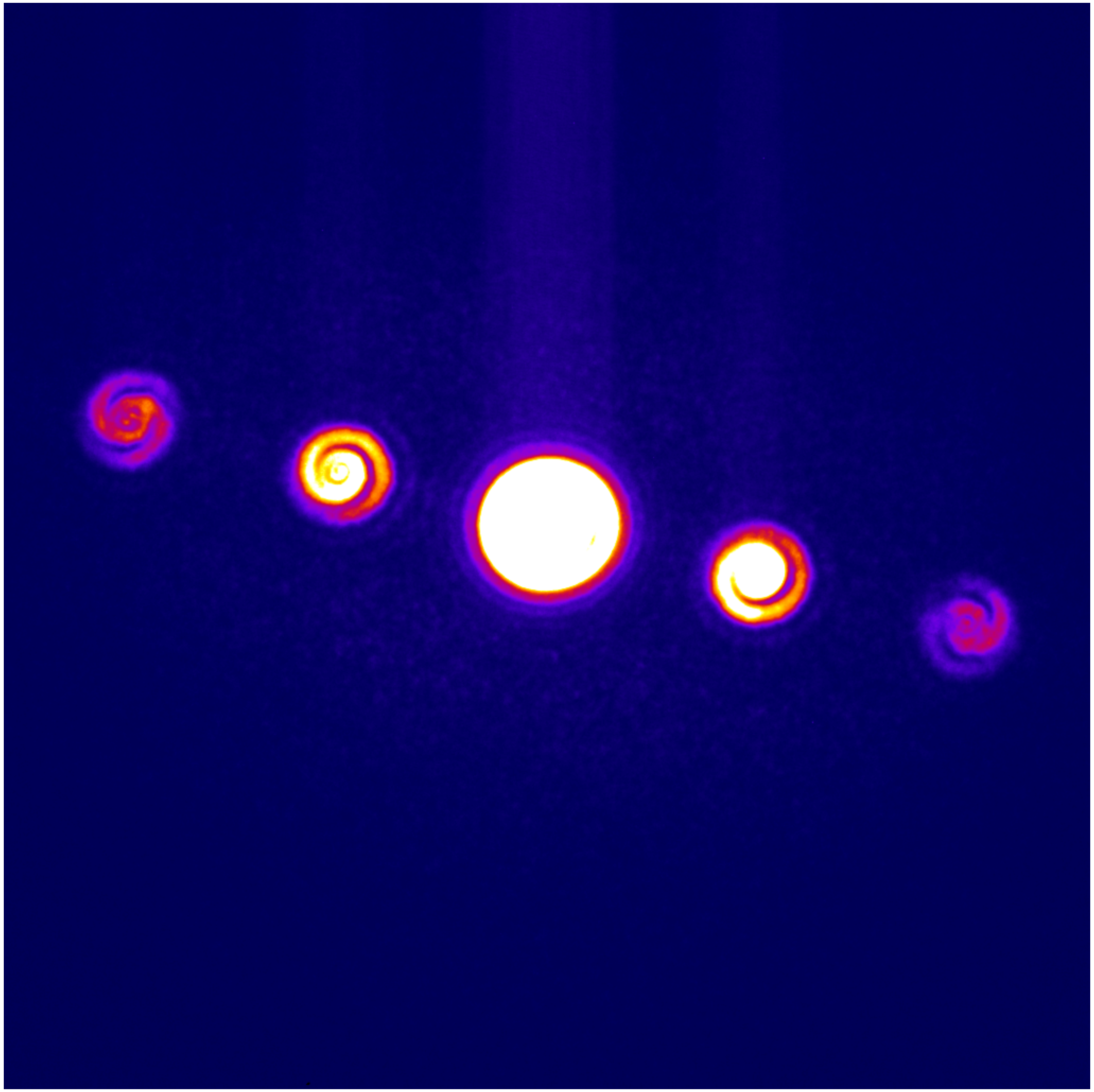


Confirming the helical phase of an electron vortex beam using interferometry





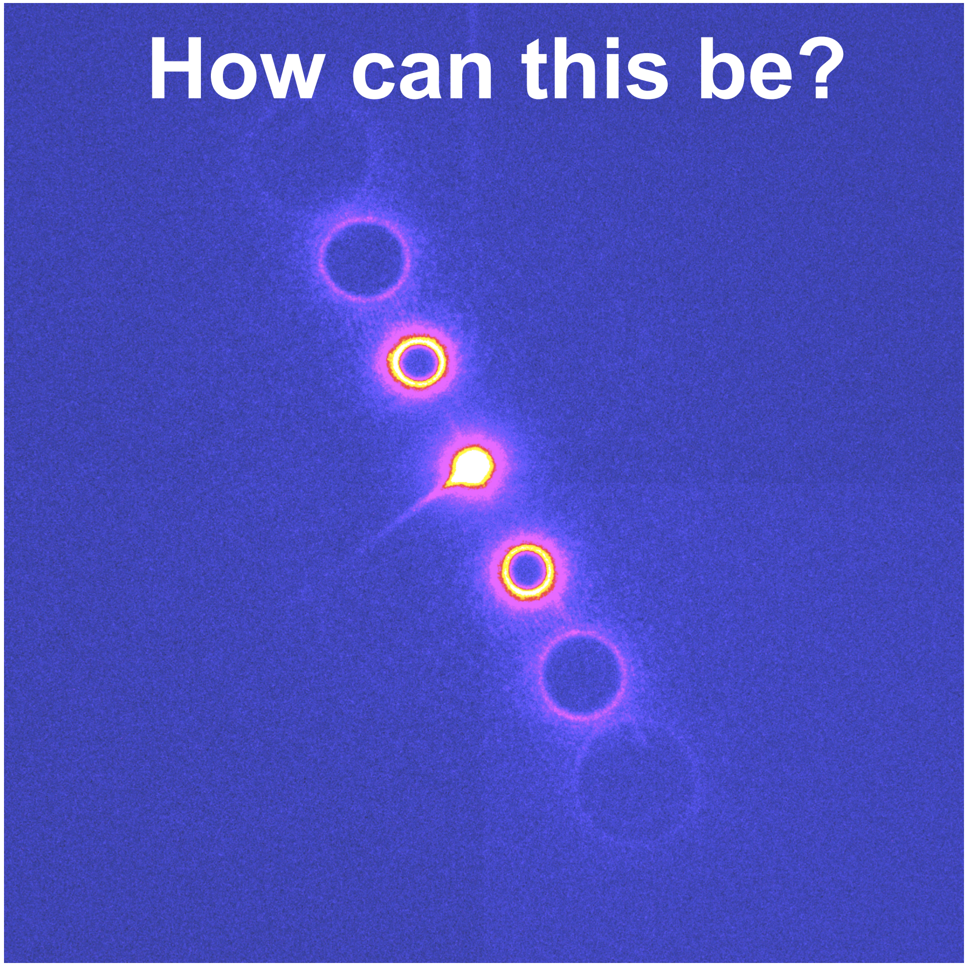
Measuring helical phase using electron interferometry



Stable orbital motion of an unbound *massive* particle

Stable orbital motion of an unbound *charged* particle

Orbital eigenstates in free space?

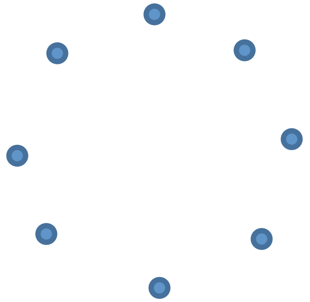


The electron is <u>not</u> orbiting in circles (not a helical trajectory)

The wavefunction represents the *shape* of the electron

The electron’s propagation looks like this:

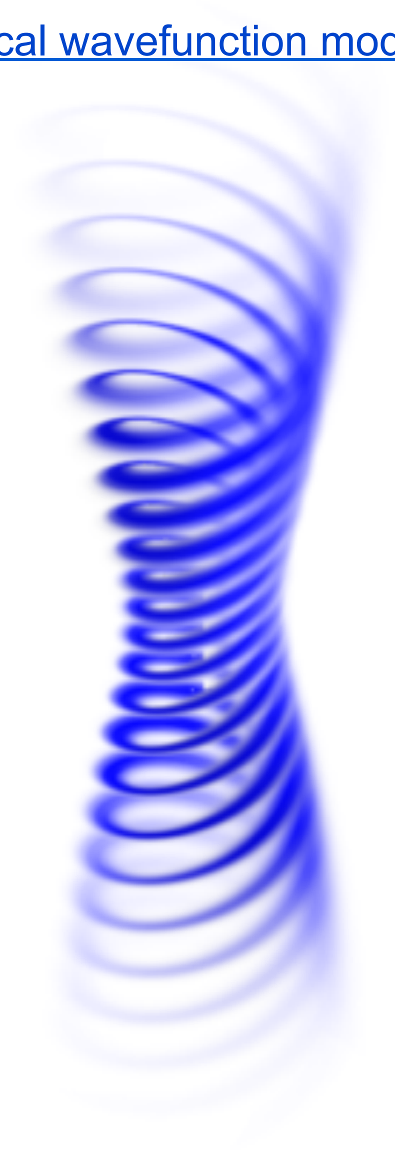
in the electron's reference frame
orbital motion does not require closed trajectories



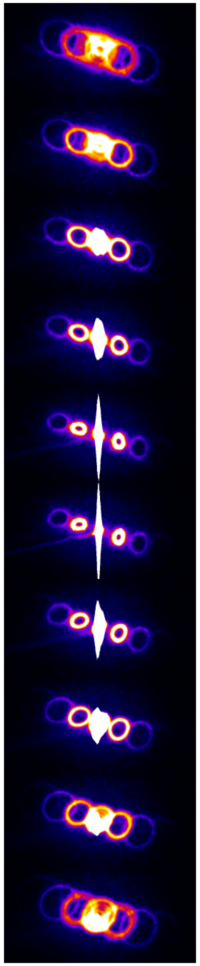
The electron vortex beam: a superposition of straight line trajectories

How we model free electrons with orbital angular momentum

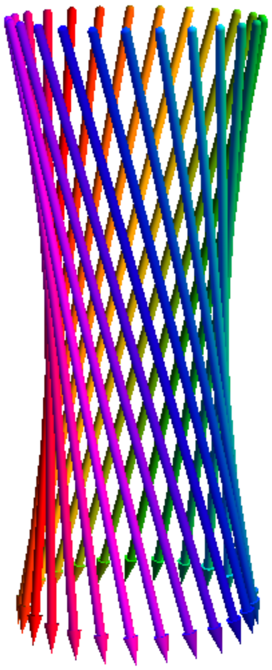
helical wavefunction model



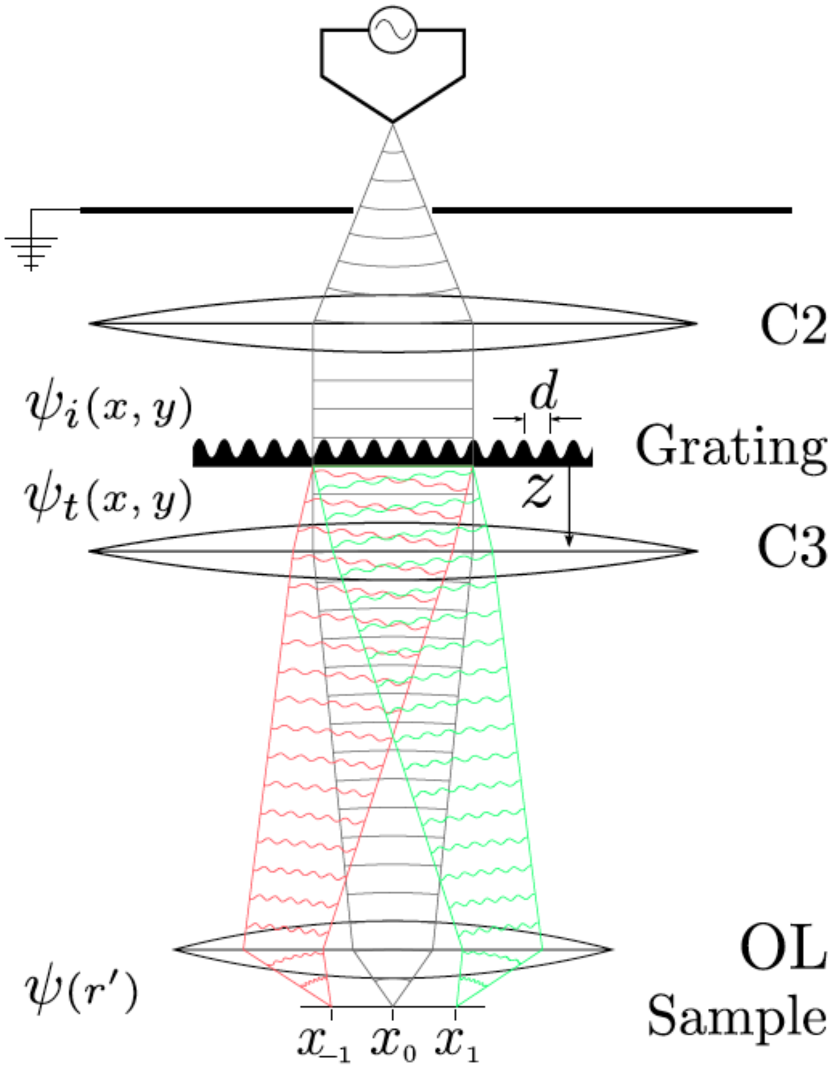
current loop model

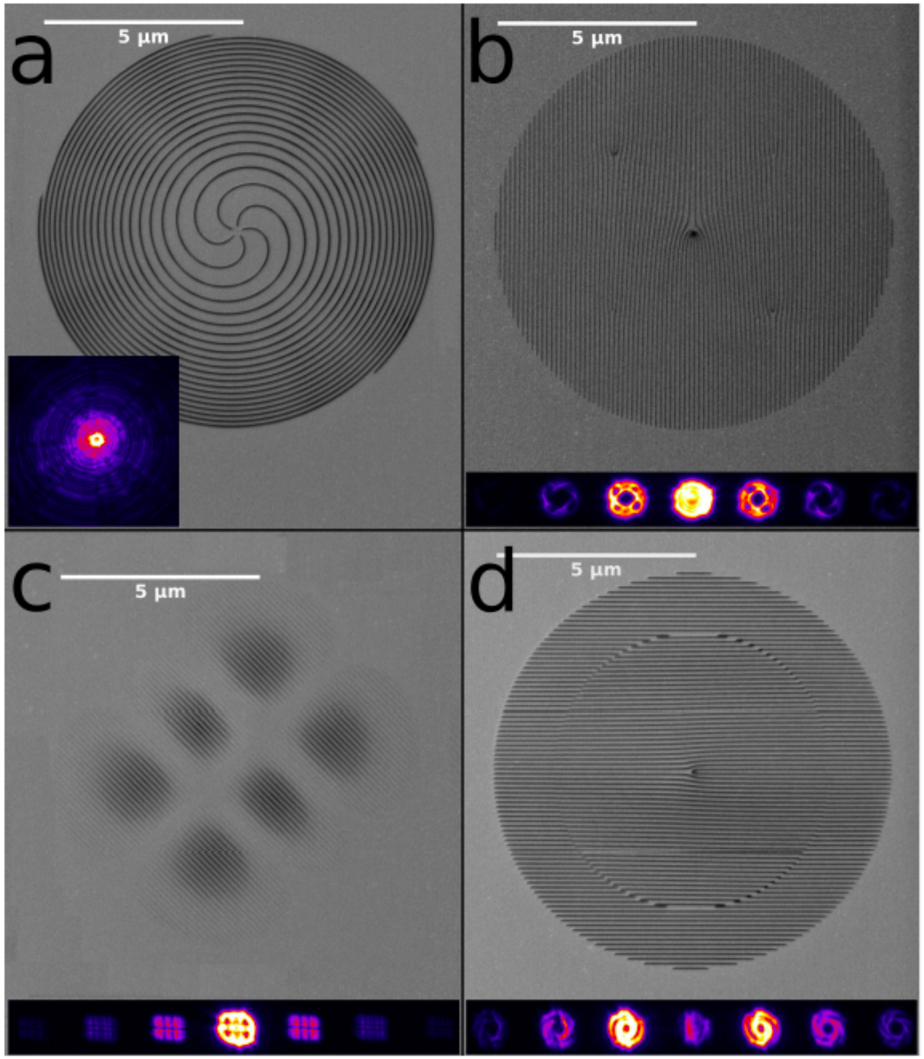


semiclassical trajectory model

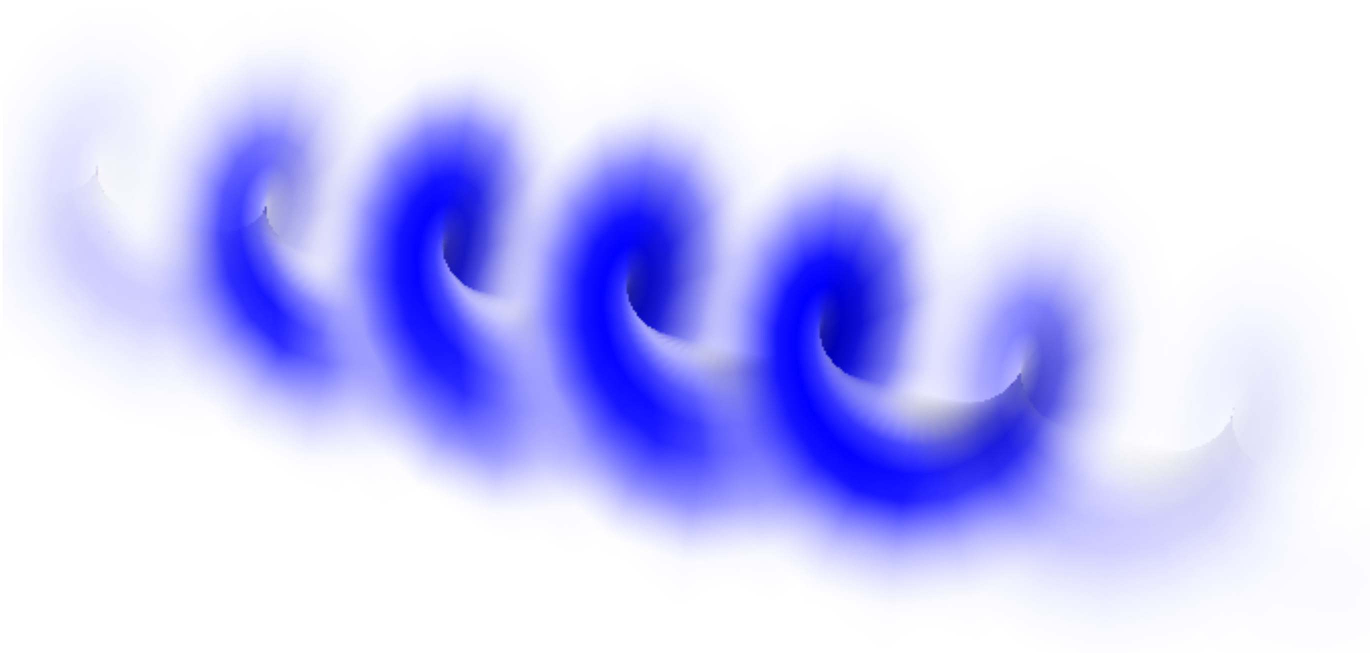


This is one example of a more general ability to engineer electron wavefronts (or other short wavelength beams)



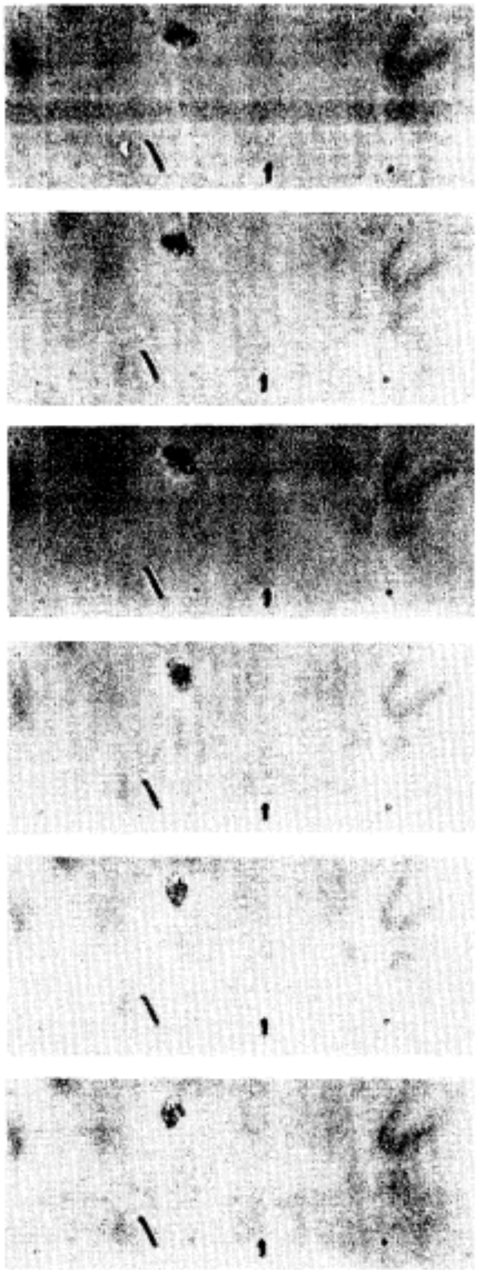


What do we do with an electron vortex beam?



exchange angular momentum with specimens

Optical ("light") vortex beams carry real angular momentum



H He *et al.*, PRL **75**, 826 (1995)

- An optical vortex beam can torque objects
- This torque is quantized; i.e., $\ell\hbar$ per photon
- Optical beams (and particle beams) can carry <u>two</u> types of angular momentum:
  -- spin angular momentum (SAM), AKA polarization
  -- orbital angular momentum (OAM)

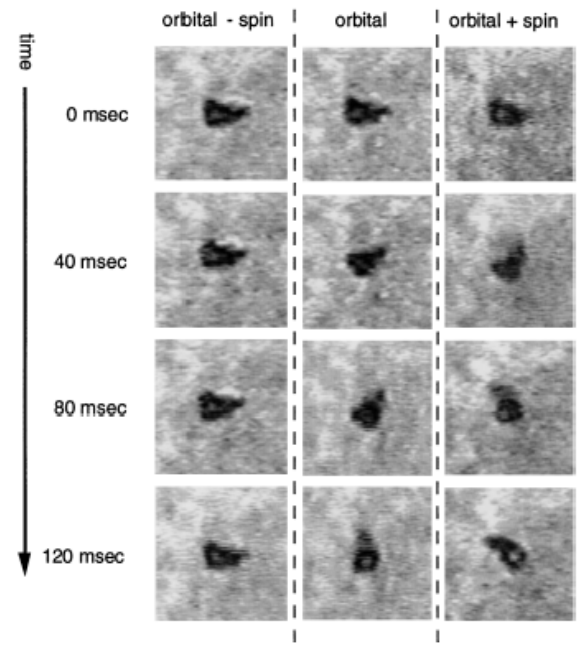
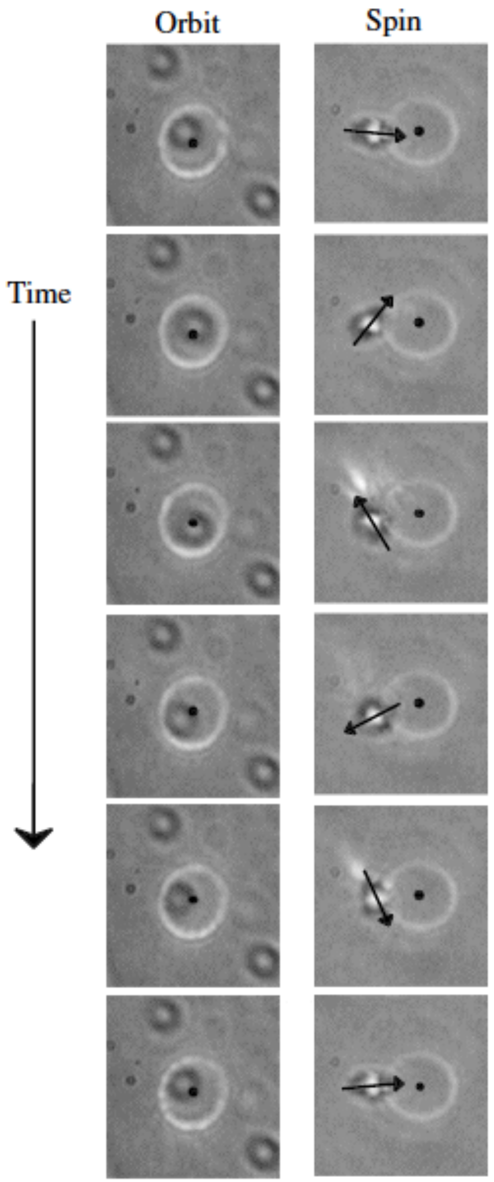


Fig. 2. Successive frames of the video image showing the stop–start behavior of a 2-μm-diameter Teflon particle held with the optical spanner.

N. B. Simpson *et al.*, Opt. Lett., **22**, 52 (1997)

Optical ("light") vortex beams carry real angular momentum



A. T. O'Neil *et al.*, PRL **88**, 053601 (2002)

- Spin angular momentum (SAM) is "intrinsic"
  - circular polarization
  - transferred wherever photon is absorbed

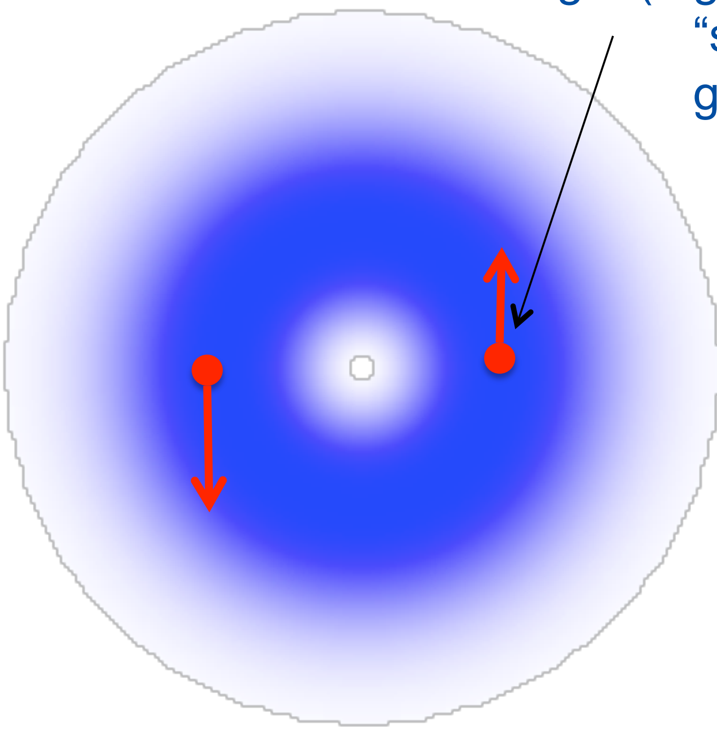
- Orbital angular momentum (OAM) is "extrinsic"
  - property of wavefunction shape
  - transfer depends on relative beam/specimen size

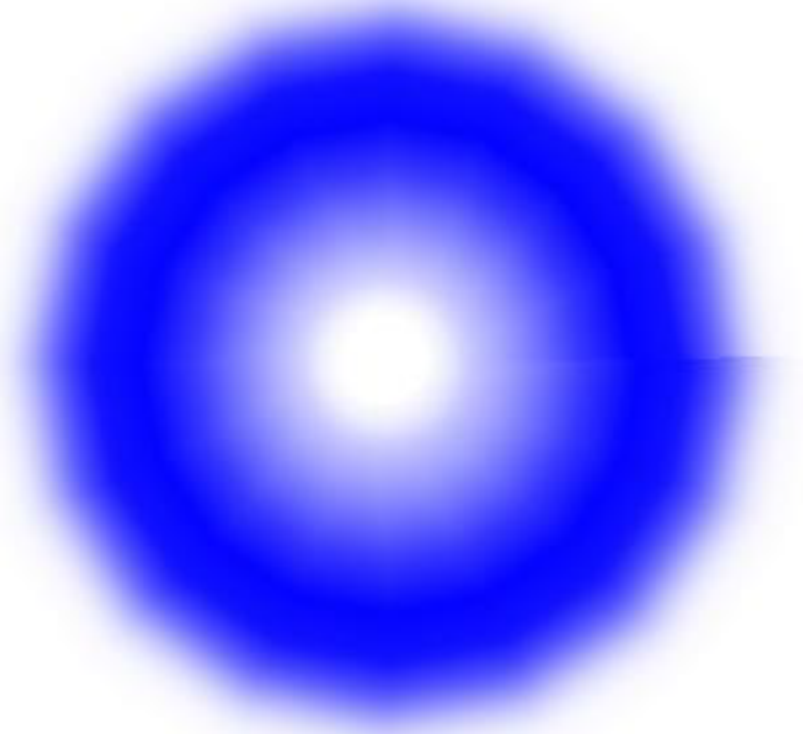
Miles Padgett, "Light's twist", Proc. Royal Soc. Lon. A **470**, 20140633 (2014):

> "For beams that are large compared with the particle, the behaviour of SAM and OAM is different. Whereas the transfer of SAM causes particles to spin around their own axis, the transfer of OAM causes them to orbit around the beam axis"

“Extrinsic” OAM – carried by the entire wave

target (e.g,.atom, nucleus)
“sees” a slightly canted plane wave,
gets a slight kick in the canting direction



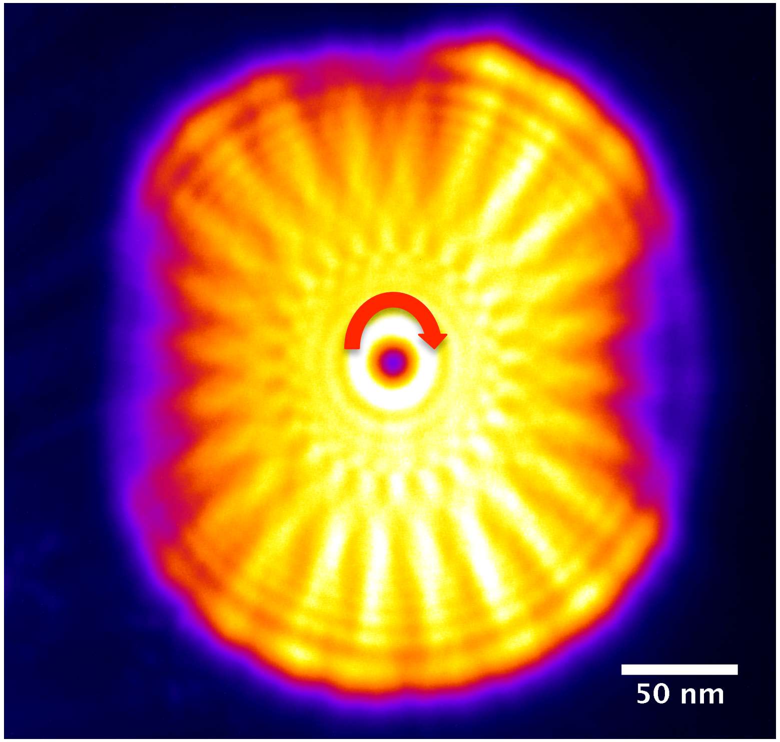


Can focused electron vortex beams induce angular momentum-dependent transitions?

How could we test this?

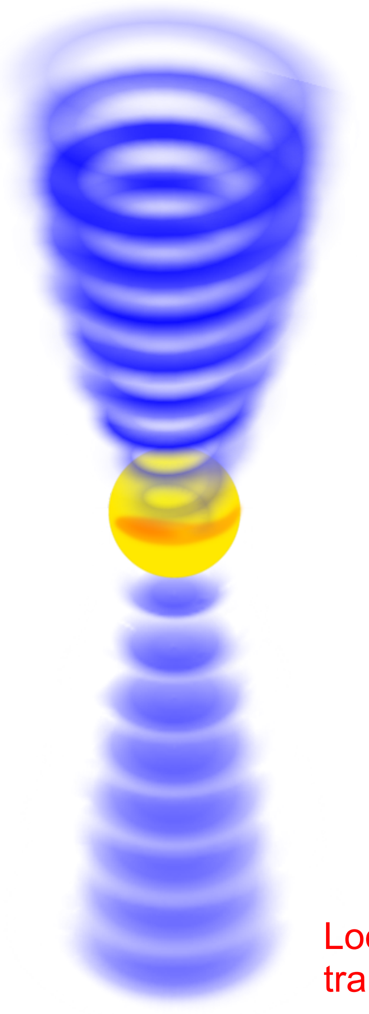
Central node (dark spot) indicates OAM

- Carries quantized orbital angular momentum ↔ azimuthal phase dependence
- Intensity drops to a minimum as $r \rightarrow 0$
- present throughout the entire path of the beam (remains through focus)
- The only way to make central dark spot disappear is by losing OAM



Basic experiment: Look at the donut hole!

Electron vortex beam incident on plasmonic structure transfers ΔE_p *and* $\pm 1\ \hbar$ (OAM)



E_{acc} (= 300 keV), $+1\ \hbar$ (OAM)

$E_{acc} - \Delta E_p$ *and* 0 OAM

Look for transfer of energy

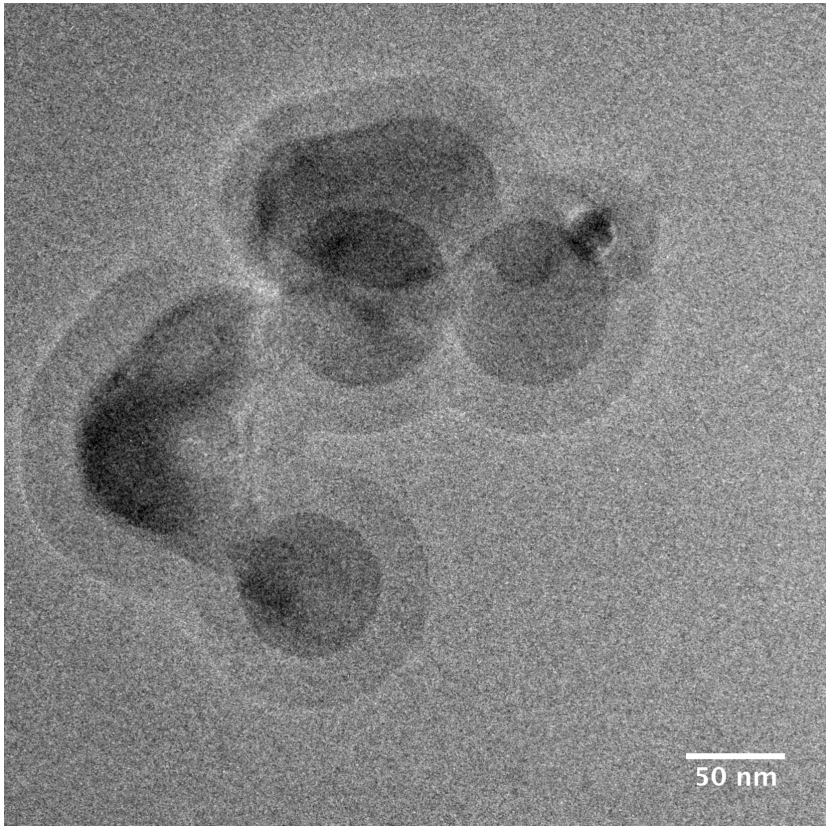
Look for transfer of OAM

Electron vortices on chiral nanocluster

Al/Al_2O_3 nanoparticles
3.2 – 3.8 eV surface plasmon resonance [Scholl *et al.*, Nature **483**, 421 (2012)]

→ high-enough E_p for us to measure in EELS!

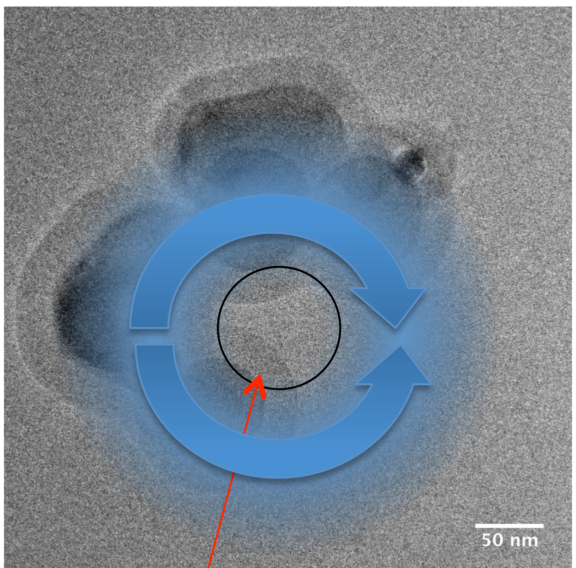
chiral nanocluster:



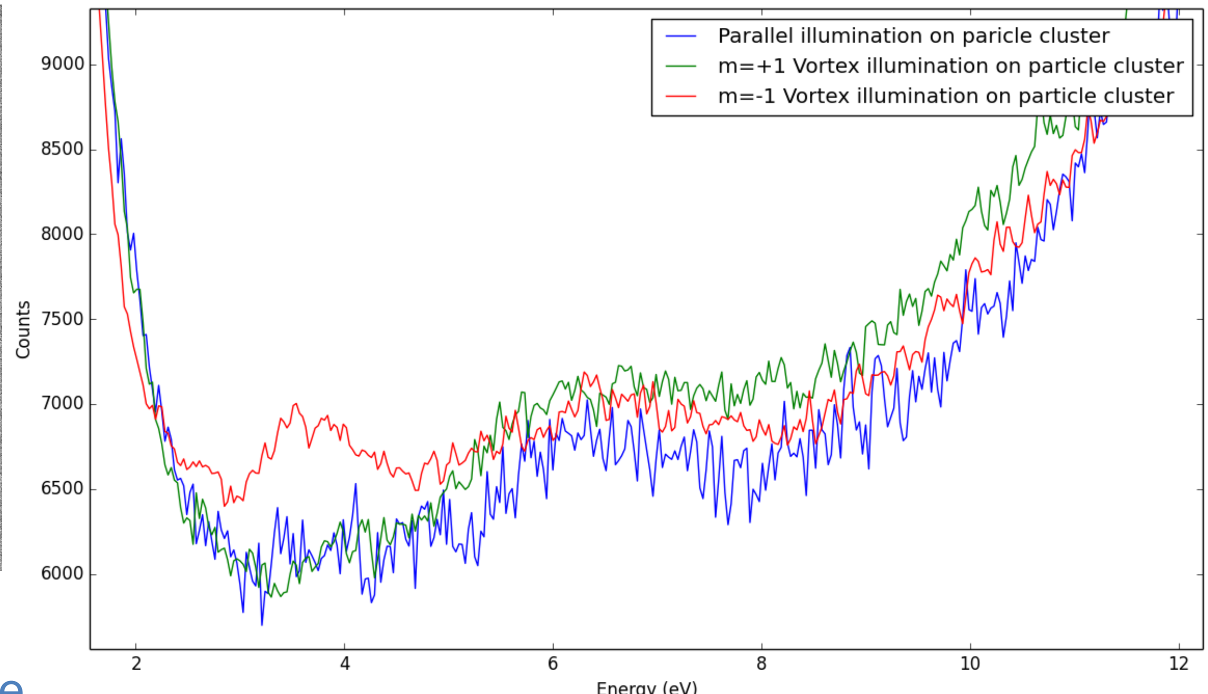
Electron vortices on chiral nanocluster

- Illuminate with three electron beams: -1, 0, and +1 OAM
  NOTE: -1 and +1 beams have identical intensity distributions

- Record EELS spectra only for electrons scattered into the dark spot



EELS entrance aperture



T. R. Harvey *et al., M&M* (2014)

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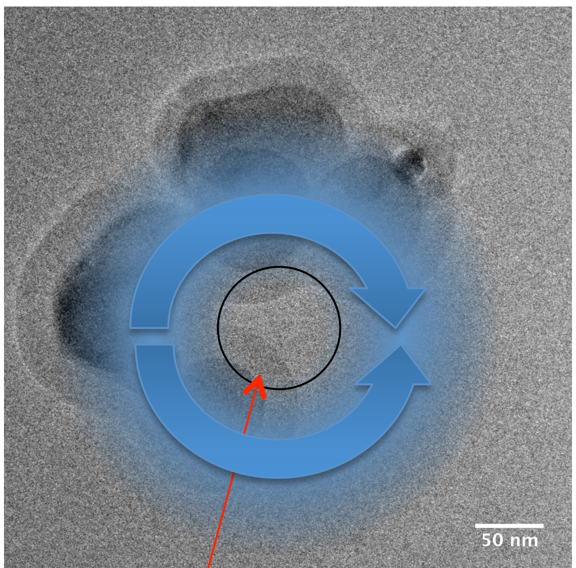
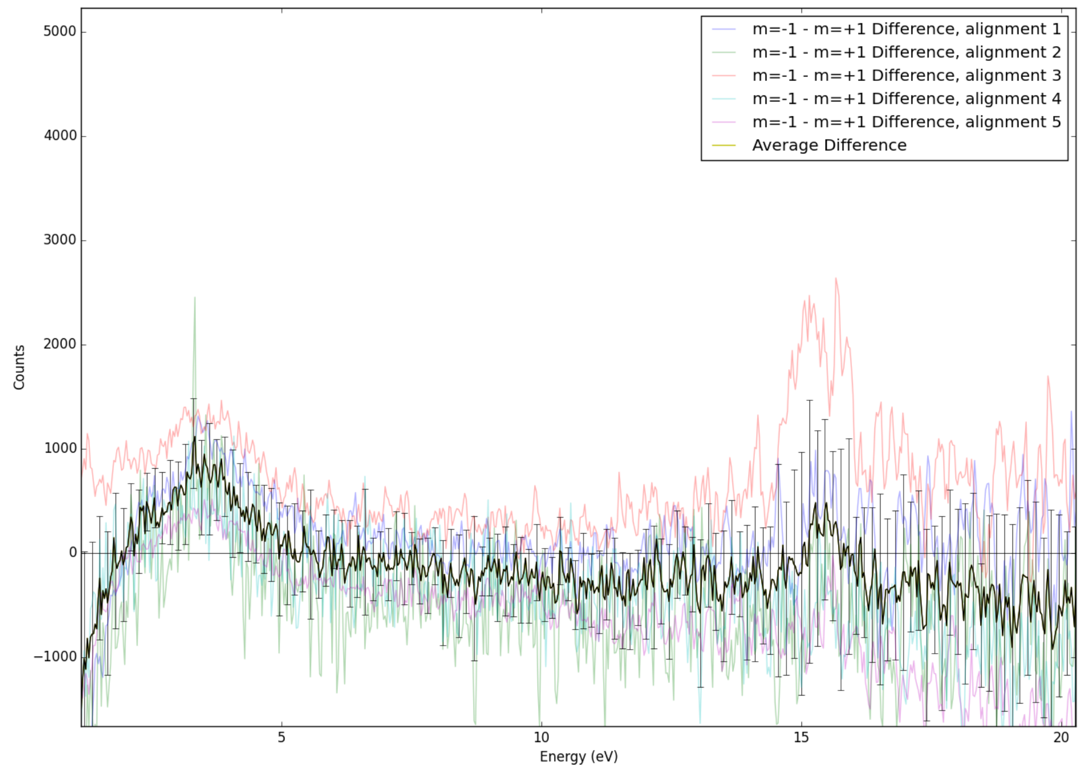


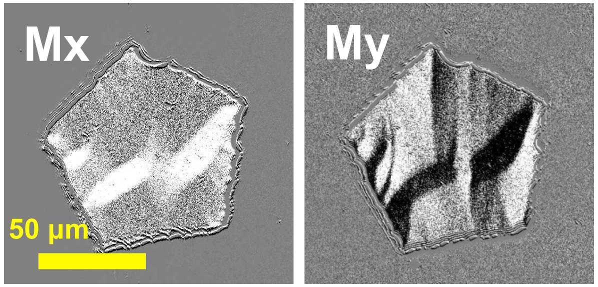
EELS entrance aperture



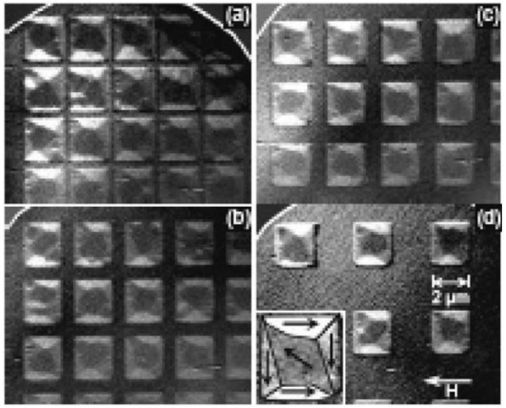
T. R. Harvey *et al.*, *M&M* (2014)

magnetization is due to angular momentum of charges
➔ see it using beams that carry angular momentum

polarized light

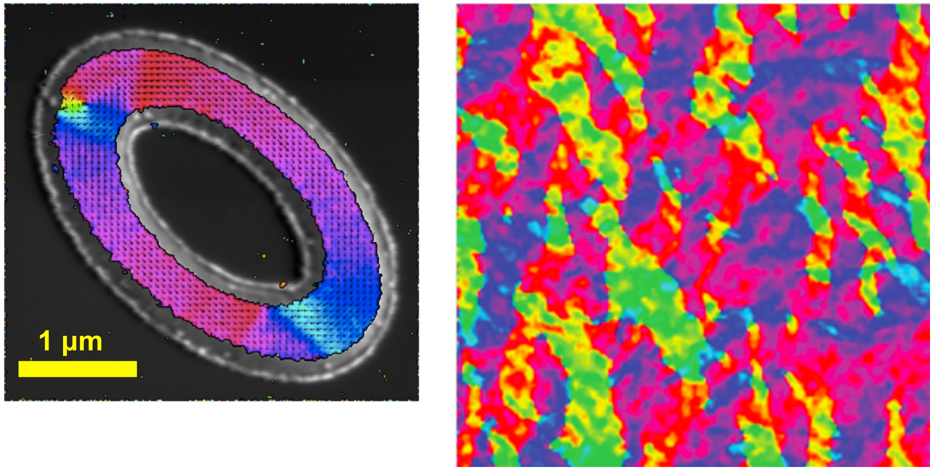


polarized X-rays

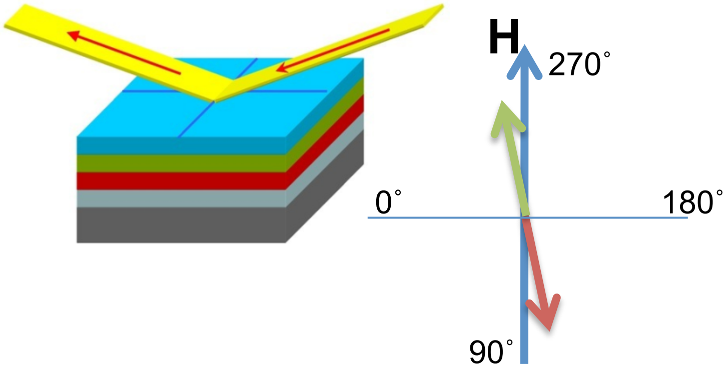


P. Fischer *et al.*, *Materials Today* **9**, 26 (2006)

spin polarized electrons



spin polarized neutrons



Magnetic dichroism at higher resolutions?
OAM-dependent *atomic* scattering

Calculate scattering amplitude:

$$P_{i\to f} = \frac{2\pi}{\hbar}\left|\left\langle n'\ell'm'\right|H_{\text{int}}\left|n\ell m\right\rangle\right|^2$$

(Fermi's Golden rule)

interaction Hamiltonian:

$$H_{\text{int}} = \left\langle \psi_2\right|V(r)\left|\psi_1\right\rangle$$

$$\psi_2(\mathbf{r}) = A_2 e^{i\mathbf{k}_2\bullet\mathbf{r}}$$

scattered electron

$$\psi_1(\mathbf{r}) = A_1(\rho)e^{i\mathbf{k}_1\bullet\mathbf{r}}e^{-im_\ell\phi}$$

incoming electron vortex

Coulomb scattering:

$$V(r) = \frac{e^2}{4\pi\varepsilon_0}\left(\frac{-Z}{|\mathbf{r}|} + \sum_j^Z \frac{1}{|\mathbf{r}-\mathbf{r_j}|}\right)$$

electron vortex STEM probe ≈ circularly polarized X-rays
→ circular dichroism using electron orbital angular momentum

new selection rules!

Transmitted electron energy loss (EELS):

$$P_{i\to f} \propto \delta\left(E_f - E_i - \Delta E\right)\begin{pmatrix} \ell' & 1 & \ell \\ m' & \Delta m_\ell & m \end{pmatrix}^2$$

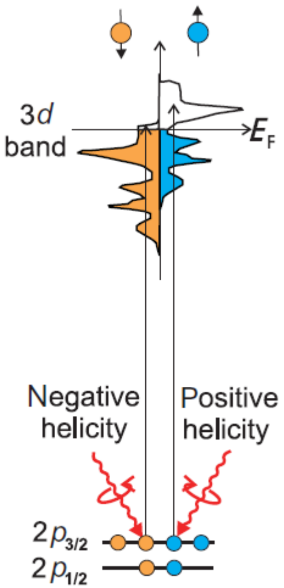
X-ray absorption (XMCD):

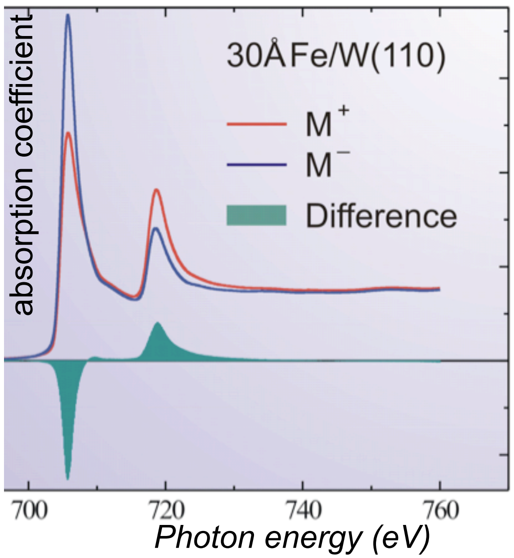
$$P_{i\to f} \propto \delta\left(E_f - E_i - E_\gamma\right)\begin{pmatrix} 1' & 1 & 1 \\ m' & \sigma_\pm & m \end{pmatrix}^2$$

XMCD-like contrast?

photons transfer energy and angular momentum to atom
➔ X-ray magnetic circular dichroism (XMCD)

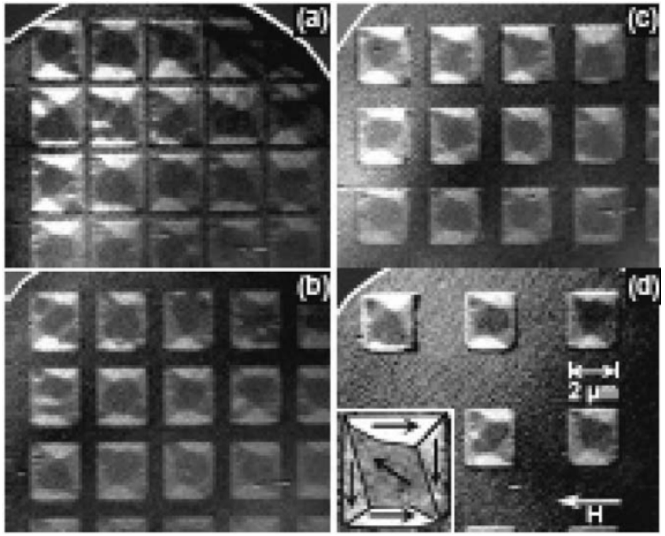
Helicity-Dependent X-ray Absorption





imaging with
circular polarized
X-rays

Magnetic Imaging
(resolution ~10 nm)



P. Fischer *et al.*, *Materials Today* **9**, 26 (2006)

electrons with *orbital* angular momentum (1 $\hbar$) may also provide this sensitivity?

Electron vortex: inelastic collisions with atoms

Scattering amplitude:

$$P_{i\to f} \propto \delta\left(E_f - E_i - \Delta E\right)\left|\sum_{j}^{Z}\sum_{\lambda=0}^{\infty}\boldsymbol{R}_{n'\ell',\lambda|m_\ell|,n\ell}(q)\,\Omega_{\ell'm',\lambda m_\ell,\ell m}\right|^2$$

<u>**E**</u>lectron
<u>**E**</u>nergy
<u>**L**</u>oss
<u>**S**</u>pectroscopy

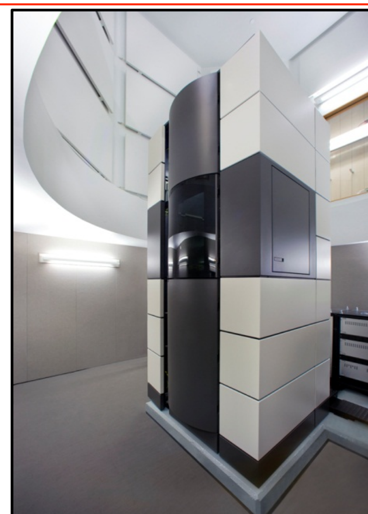
Need atomic-sized beam
(integral over radial coordinates)

Optics-like selection rules
(integral over angular coordinates)

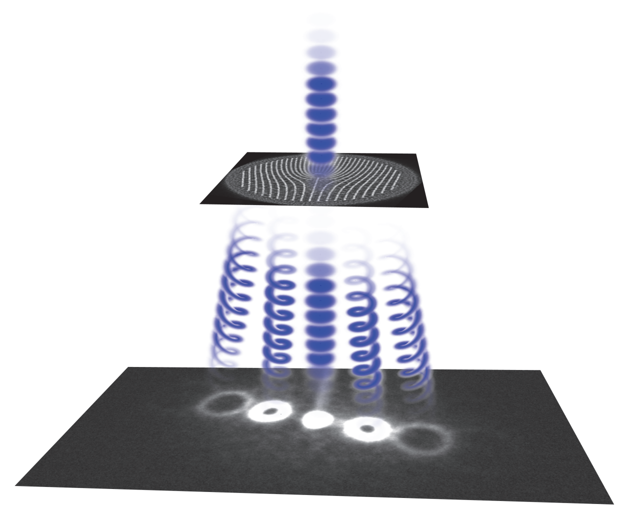
quantized energy transfer
➔ **map elements**

wavefunctions must overlap
➔ **<u>need atomic scale beam</u>**

quantized ang. momentum transfer
<u>➔ magnetic dichroism</u>



TEAM 1

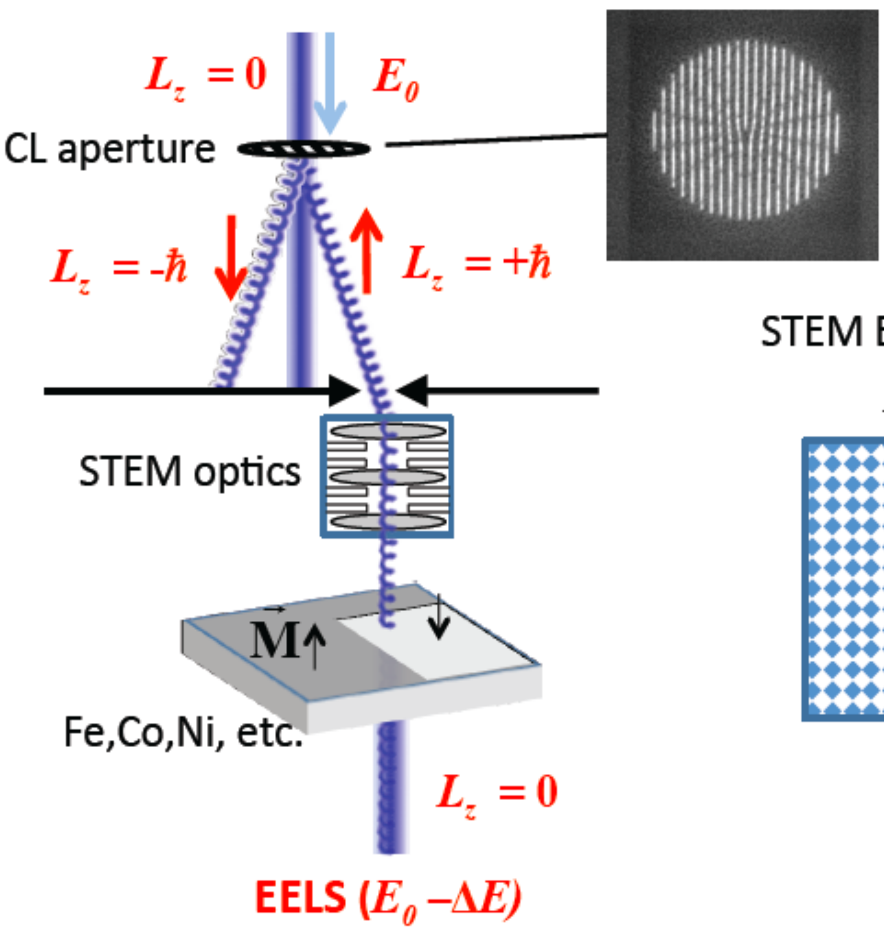


electron vortex beams

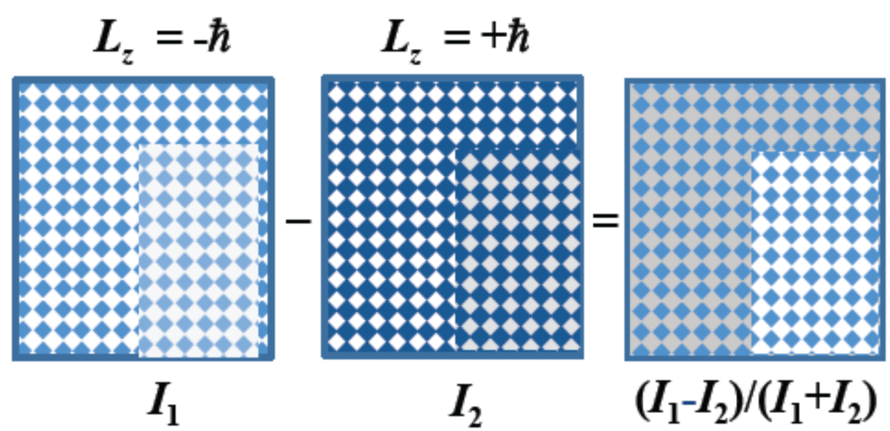
Electron Magnetic Orbital Dichroism (EMOD)

→STEM/EELS with vortex beam probe

- Pre-specimen diffraction grating, OAM beam as STEM probe
- EELS: asymmetry in $L_{2,3}$ absorption edges of left and right probe
- Challenge: vortex beam probe must have atomic size



STEM EELS images at L_3 or L_2 edge:



atomic resolution map of magnetic moments

Calculations & Optical Experiments:

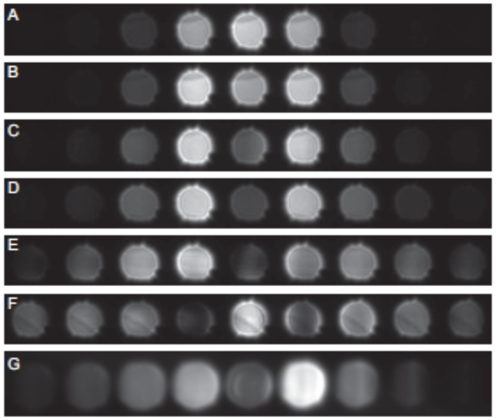
- To transfer OAM to single atoms, vortex beam needs to be atomic scale
- To use in a STEM instrument, need high current

➔ optimize diffractive optics for electrons

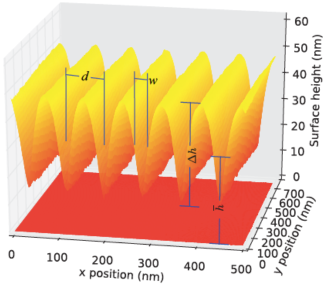
Optimizing diffraction efficiency

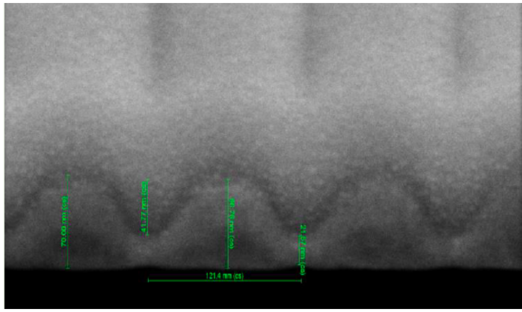
1. make lots of dose arrays (various fabrication parameters):
- groove depth (5 nm – 40 nm)
- groove width (12 nm – 60 nm)
- groove spacing (24 nm- 150 nm)
- groove shape (sinusoidal to asymmetric triangular)
- grating material (SiNx (various sources), Si, Pt)
- fabrication method (FIB milling [various], FIB-deposited Pt, EBL [various

2. measure electron diffraction efficiency
- grating in specimen holder
- TEM LAD
- measure:
  - incident beam current
  - total transmitted current
  - current in individual diffracted beams



3. measure fabricated grating surface
- AFM, FIB & SEM



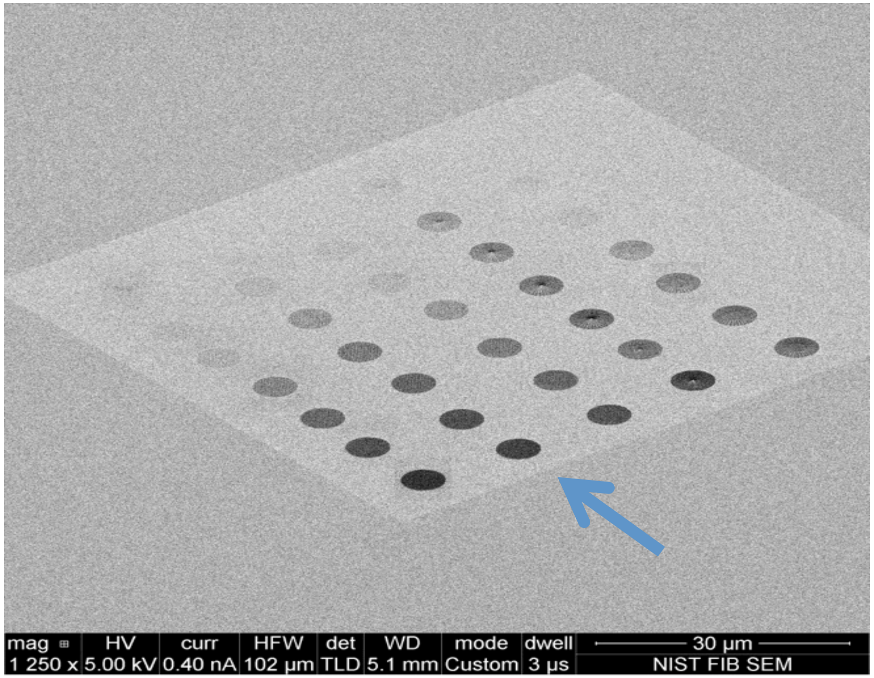


4. model

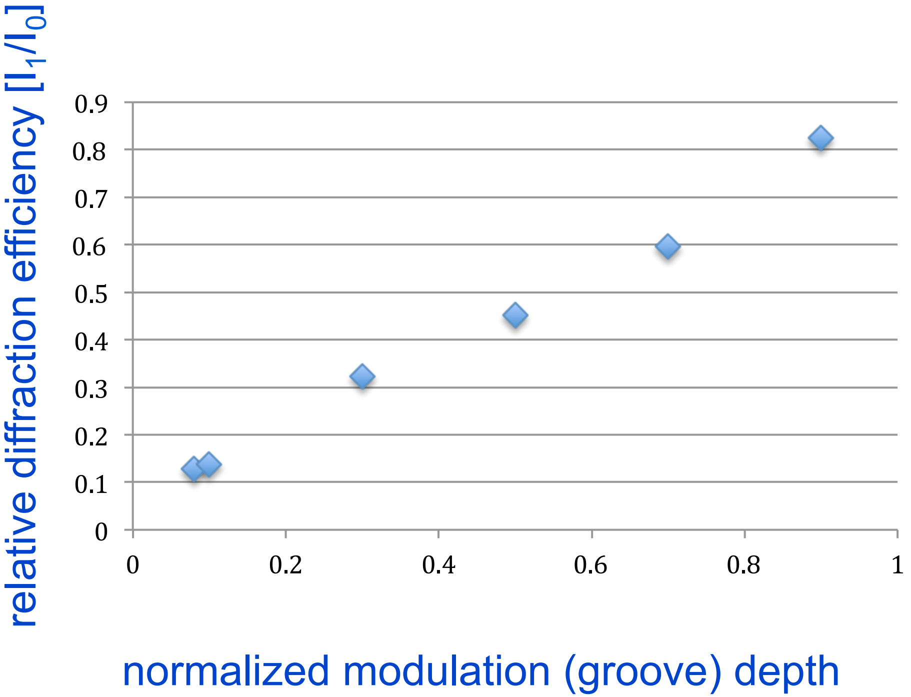
5. repeat

Maximizing diffraction efficiency of electron holograms

dose arrays (many)



High ***relative*** diffraction efficiency

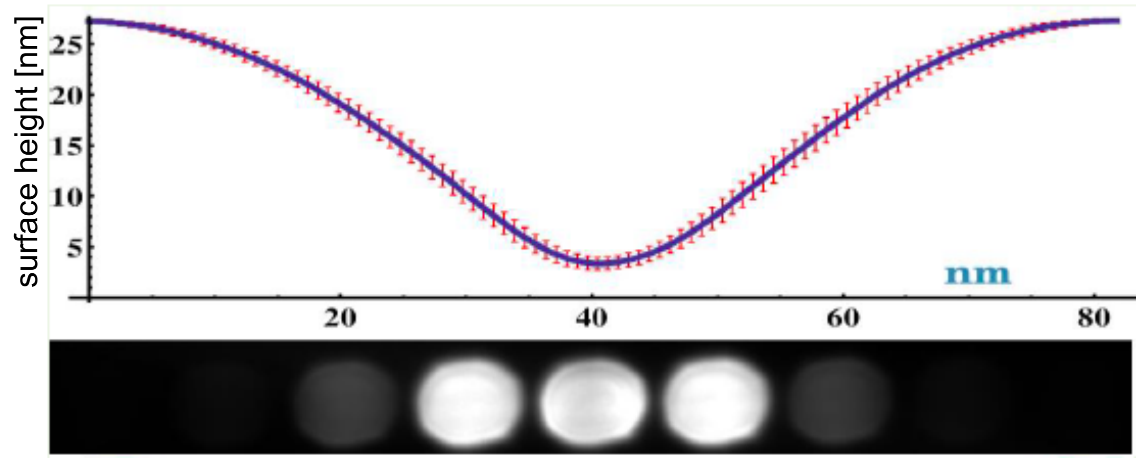


Grating groove shape influence diffraction efficiency

symmetric grating

AFM profile:

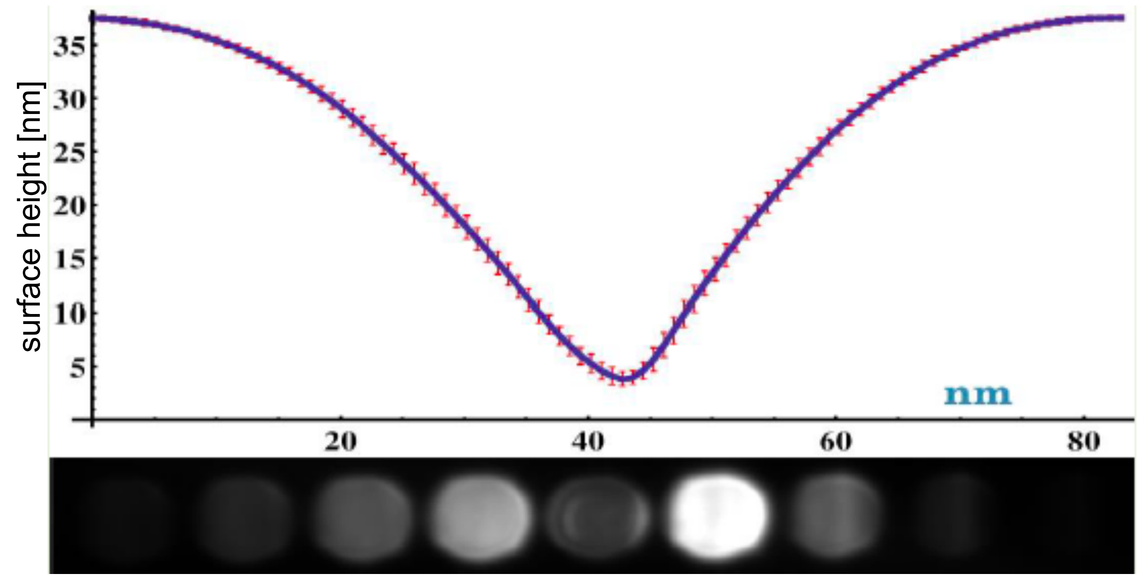
diffraction pattern:



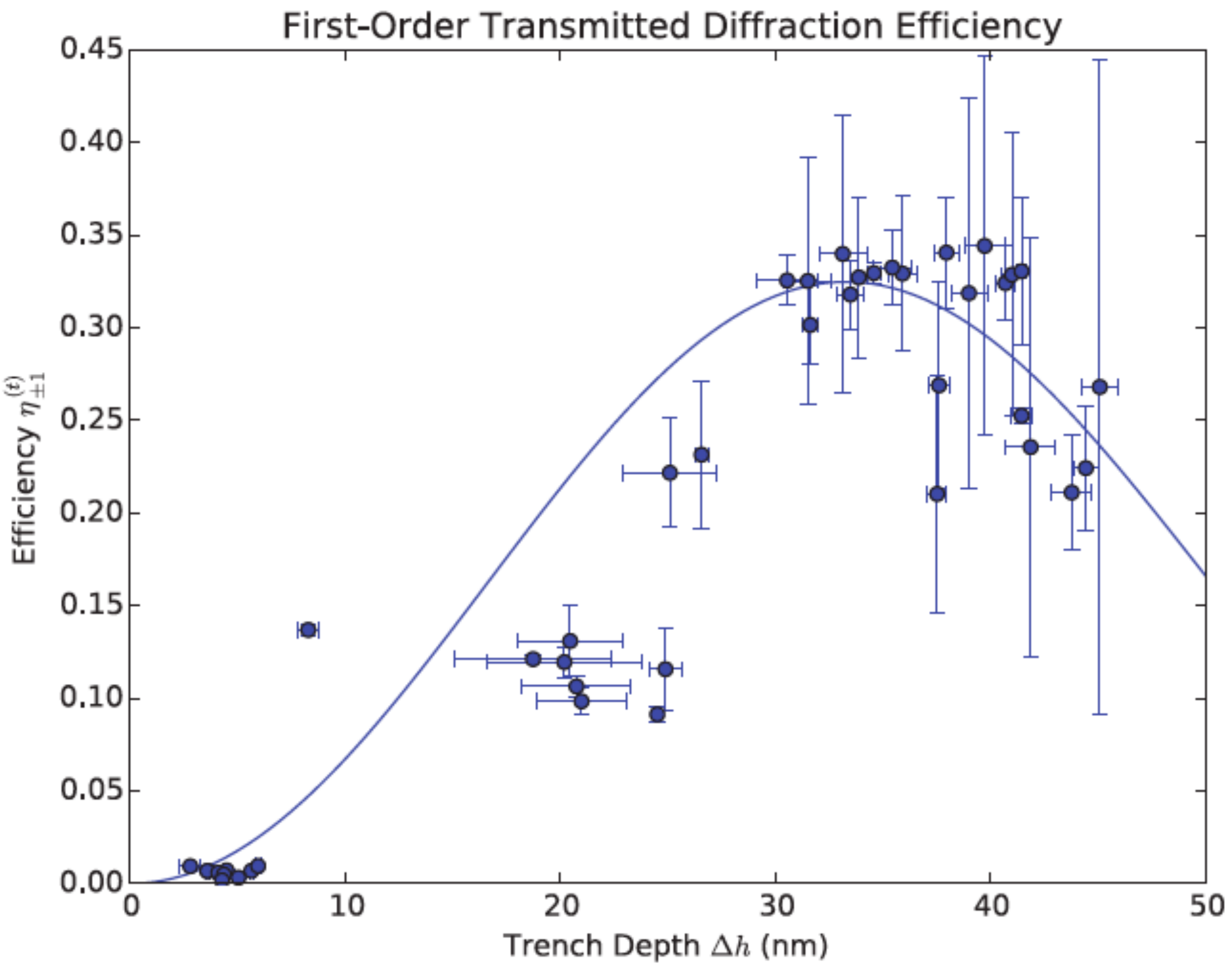
blazed grating

AFM profile:

diffraction pattern:

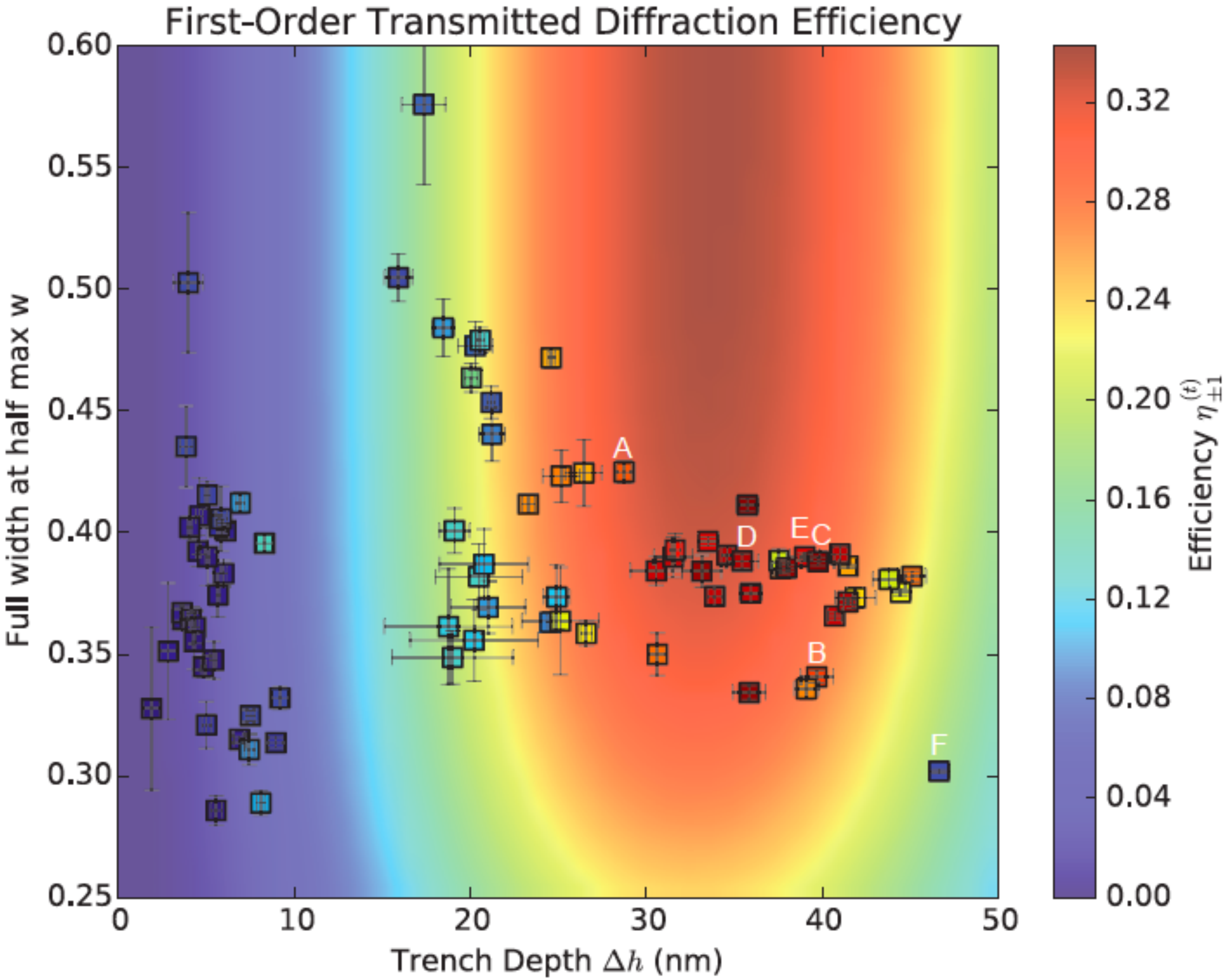


Diffractive electron optics diffraction efficiency



T. R. Harvey *et al.*, *New J. Phys.*. **16**, 093039 (2014).

Optimizing diffraction efficiency – exploration of parameter space

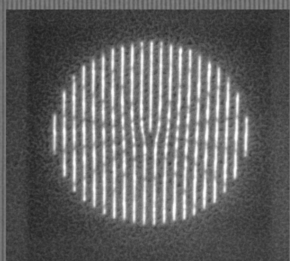


T. R. Harvey *et al.*, *New J. Phys..* **16**, 093039 (2014).

High spatial resolution
High spatial coherence
Large area

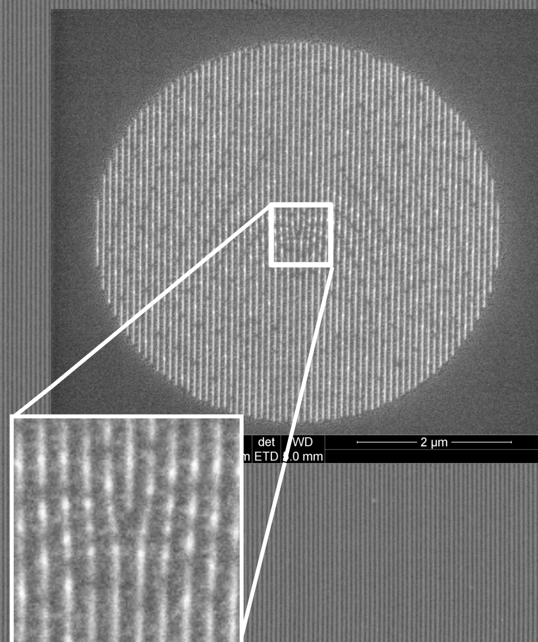
Good isolation of diffracted orders
Precisely controlled phase
Smaller probes (high resolution)

<u>2010 grating</u>



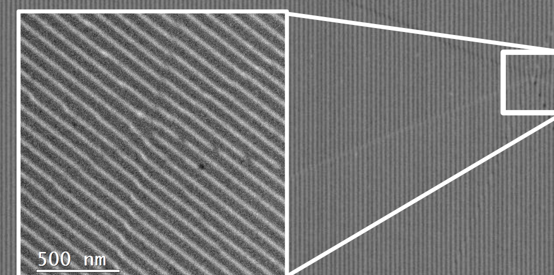
- 5 µm diameter
- 100 nm line spacing

<u>2011 grating</u>



- 10 µm diameter
- 75 nm line spacing

<u>Current gratings (background image, drawn to scale)</u>



- 80 µm diameter
- 50 nm line spacing

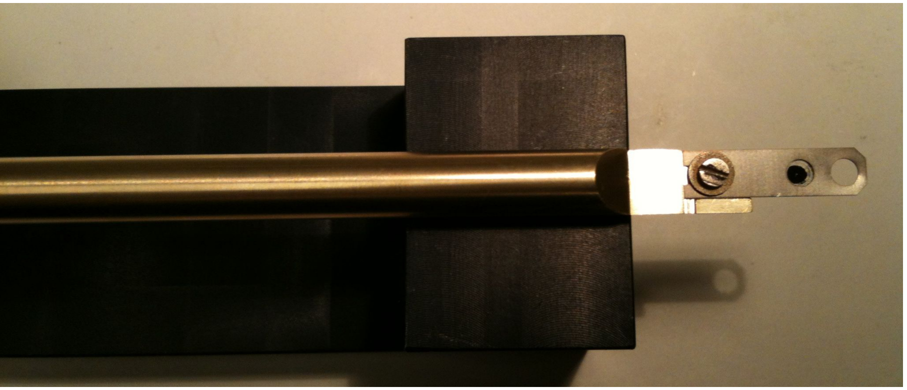
Enables:

- **atomic resolution**
- **wide field-of-view**

T. R. Harvey *et al.*, *New J. Phys.*. **16**, 093039 (2014).

Aberration-corrected electron vortex STEM probes

Modified condenser/objective apertures:

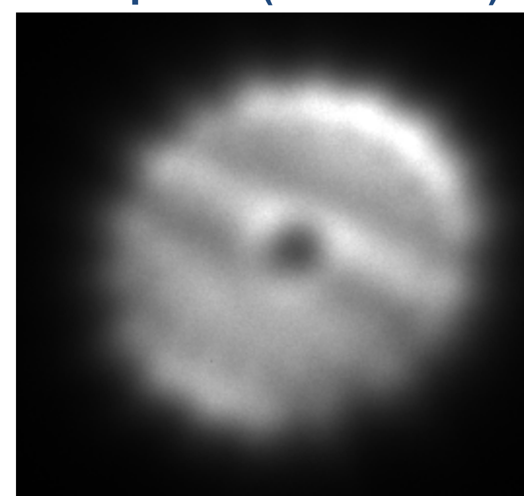


Installed in condenser lens system

- TEAM I (NCEM)
- Titan-X (NCEM)
- FEI Titan (UO-CAMCOR)
- FEI Titan (NIST-MML)
- JEOL 200CF (UIC)

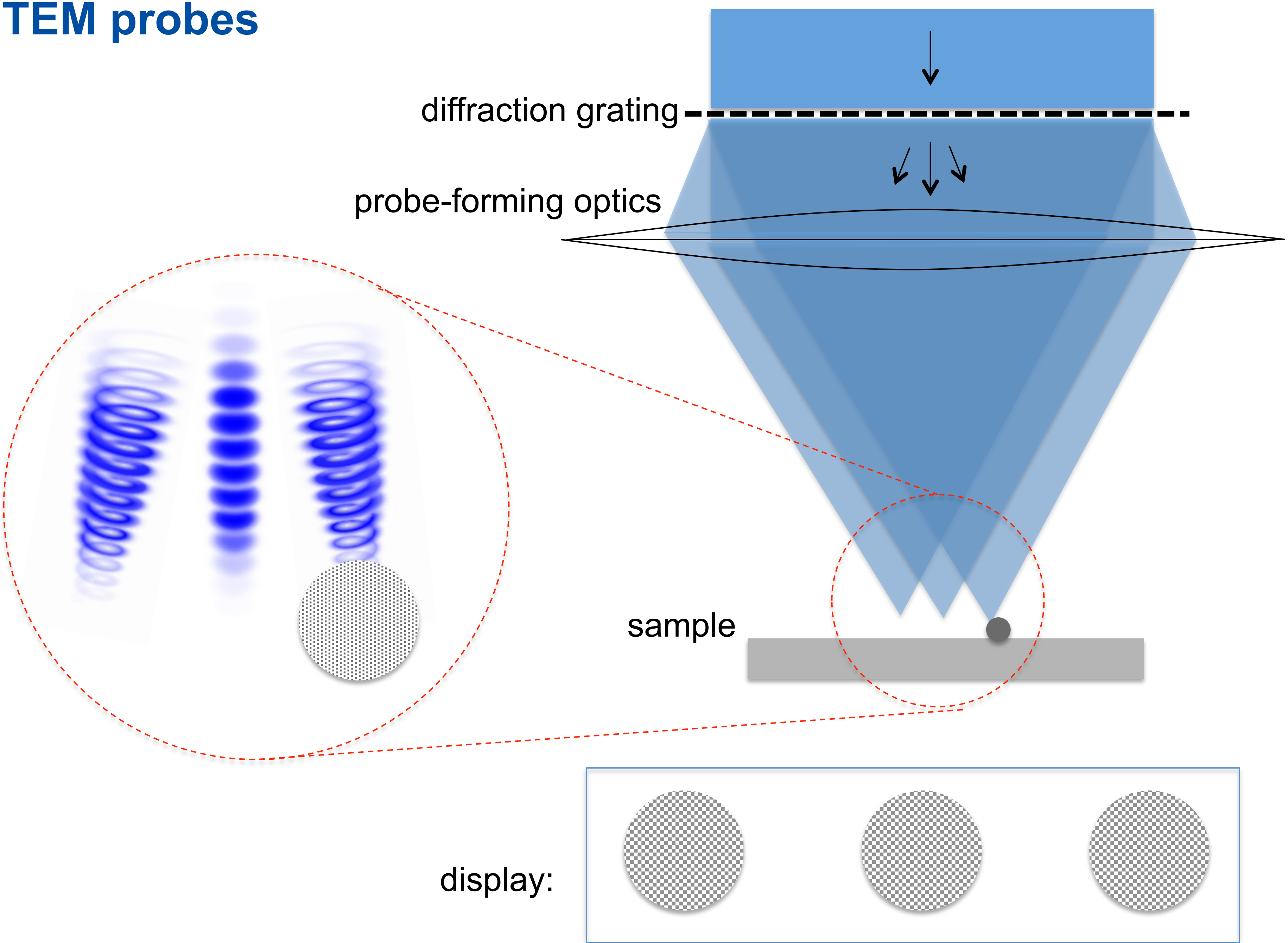
(Peter Ercius)

Aberration-corrected STEM probe (defocused):

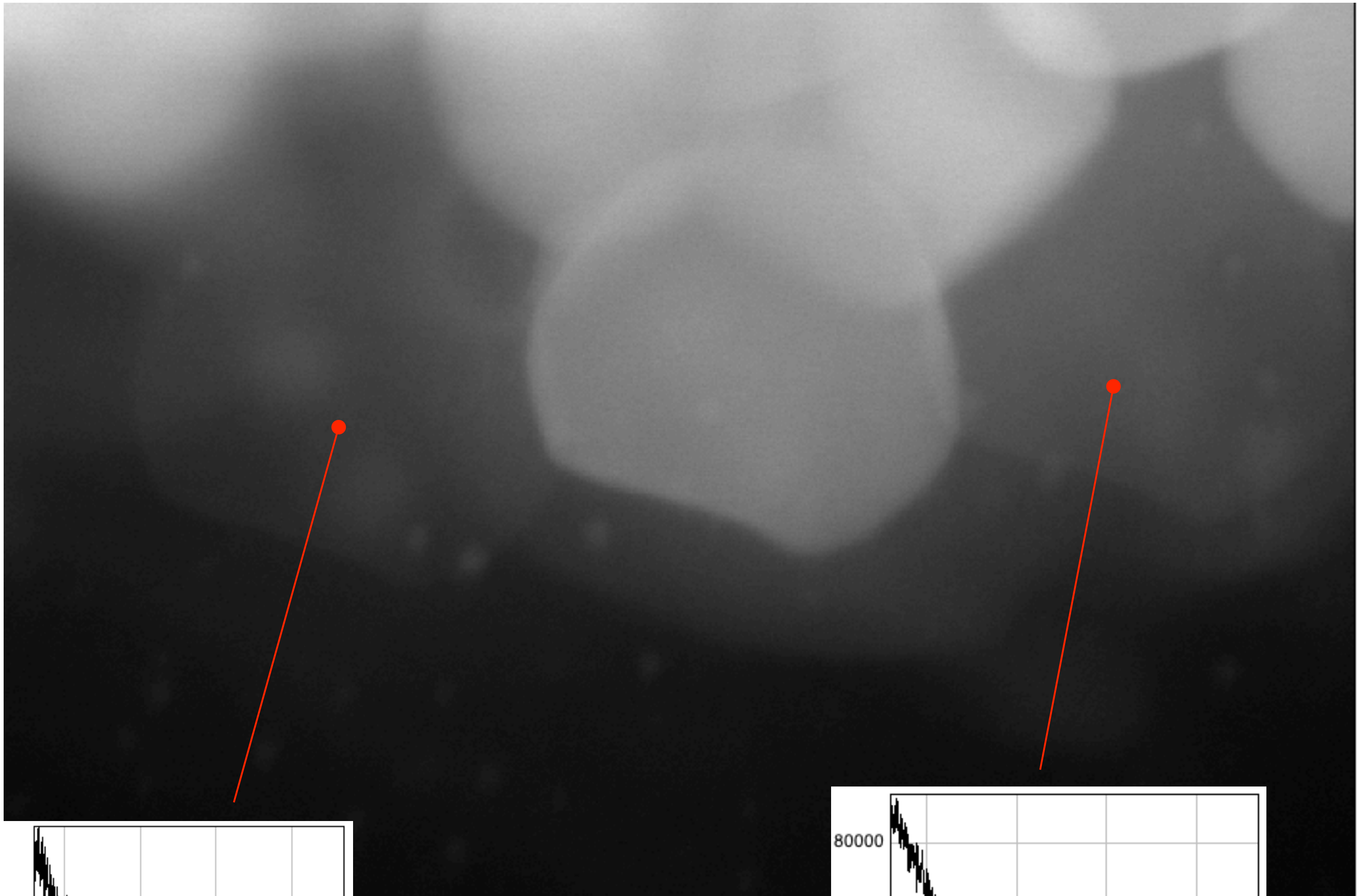


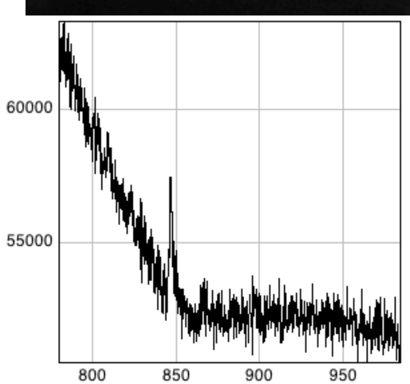
McMorran, B. United States Patent: 8680488 - *System and*

Imaging with multiple Angstrom-scale electron vortex STEM probes

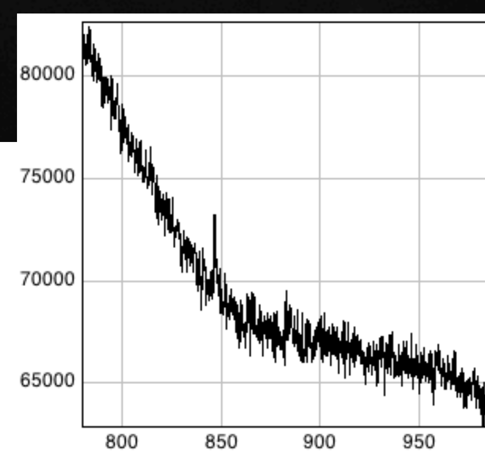


Multiple STEM probes → multiple images of the same object



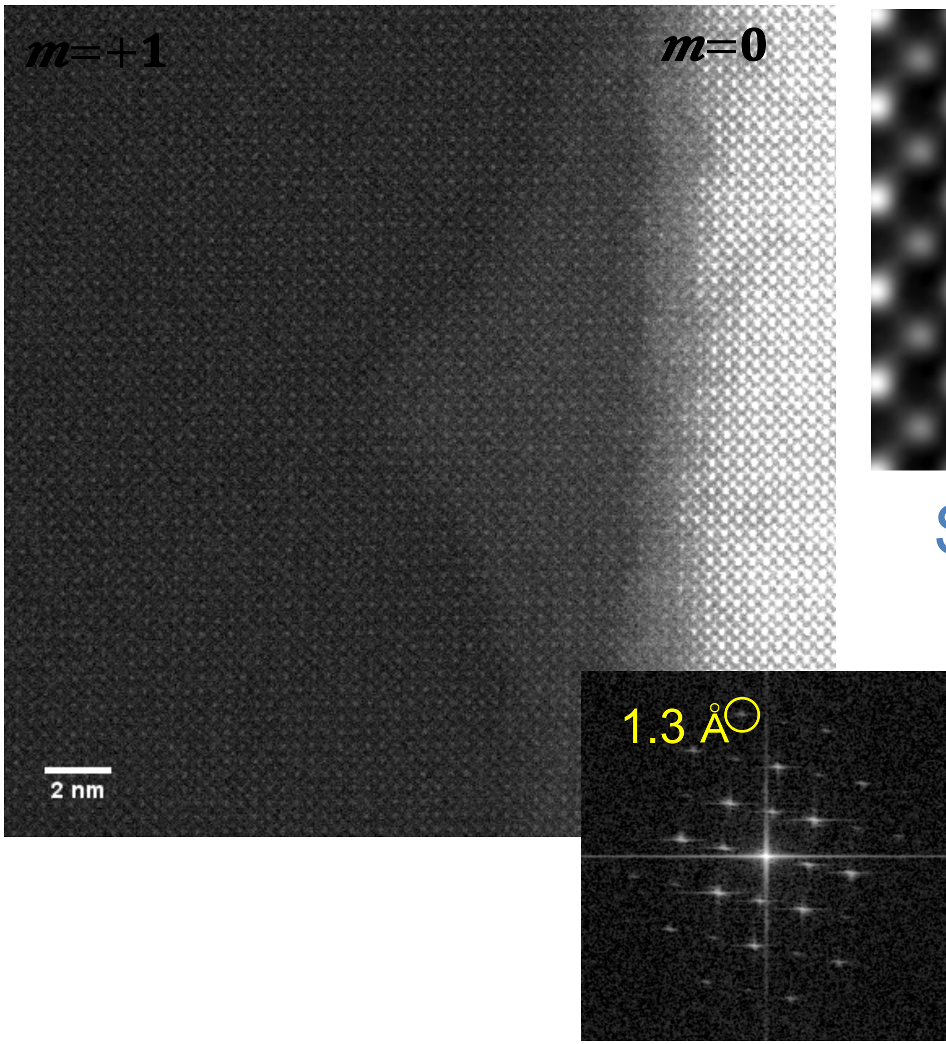


EELS using left beam
-1 vortex probe

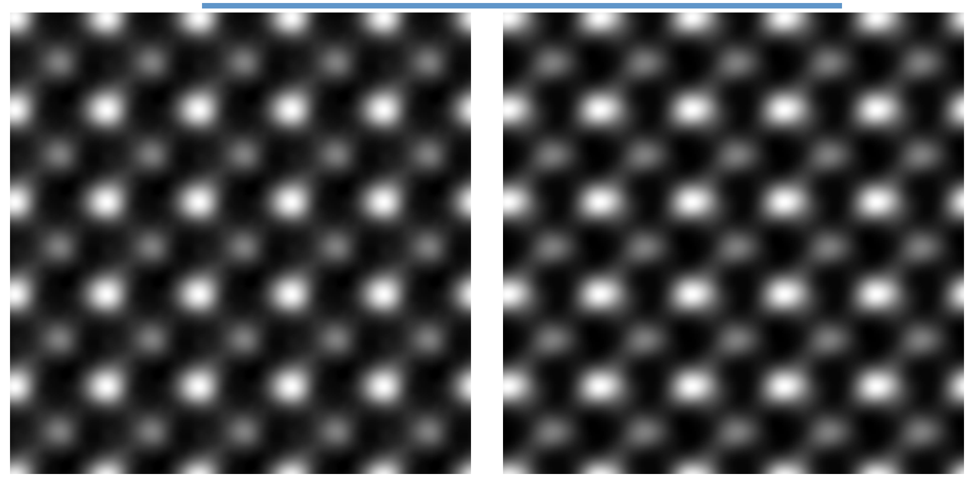


EELS using right beam
+1 vortex probe

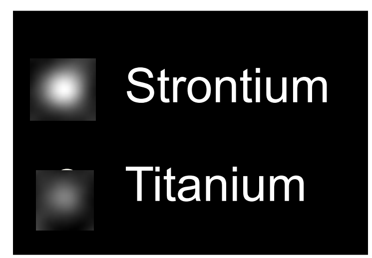
$SrTiO_3$ imaged by 0^{th} & 1^{st} order focused probes



Mean unit cells

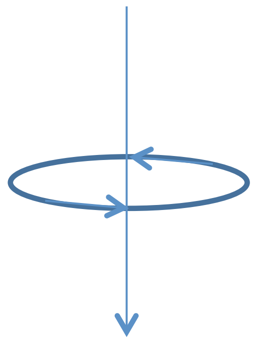


Side beam Center beam



Understanding electron vortex in external fields

Unlike optical OAM, electron OAM has a *magnetic moment*



$$\mu_z = g\,\mu_B\,\ell$$

Couples to external fields:

G. M. Gallatin and B. McMorran, Phys. Rev. A 86, 012701 (2012):

full path integral solution

magnetic field → rotating coordinate system

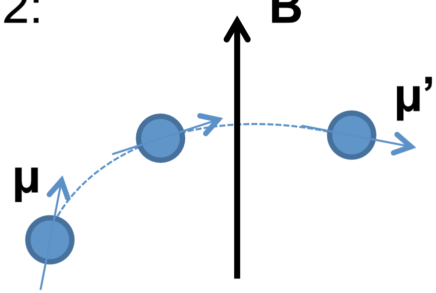
ladder operators →

propagate OAM wavefunction through field equivalent to propagiting conventional wavefunction then adding OAM

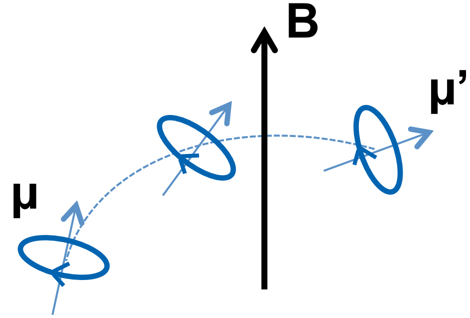
Use semiclassical ray picture to model eLG in external fields: Transverse magnetic field

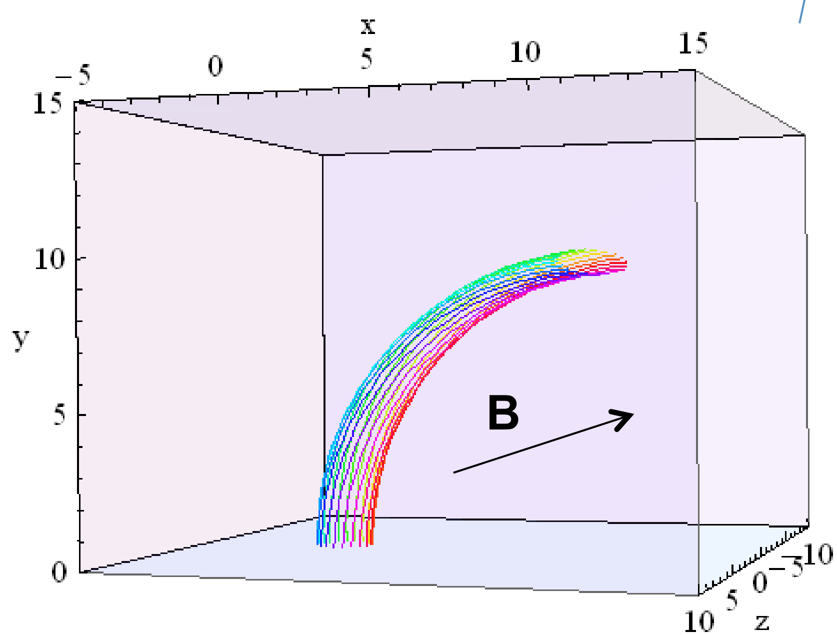
magnetic moment does not precess same as beam path
→ gyromagnetic ratio *g* is not 2 like electron spin.

Spin 1/2:



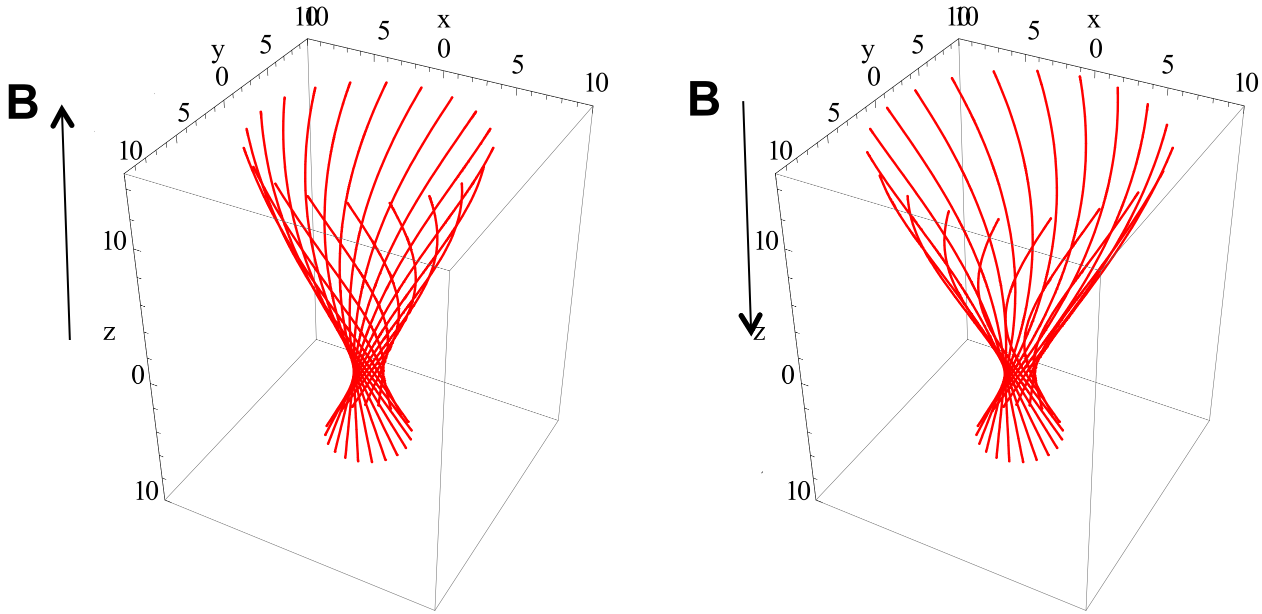
Orbital:





Use semiclassical ray picture to model eLG in external fields: Longitudinal magnetic field

Zeeman interactions with longitudinal fields
(working with G. Gallatin, NIST)



Key Points

- Nanofabricated diffractive optics

  → fine control over quantum states of free electrons

  BUT this requires a transverse spatial coherence

- Free electrons can carry quantized orbital angular momentum

  → theoretically, this can induce electronic transitions in atoms, and perhaps even nuclear transitions in atomic nuclei

  BUT angular momentum is delivered on the lengthscale of the beam

- Other possible experiments at JLab:
  - Generation of electrons with both spin *and* orbital angular momentum (electron vector beams)
  - Utilize spin-orbit coupling to induce spin polarization
  - Generation of EUV and X-ray vortex beams using electron vortex beams?

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Oregon Center for Optics

CAMCOR Oregon's High-Tech Extension Service

BERKELEY LAB Lawrence Berkeley National Laboratory

LDRD Carbon Cycle 2.0
"Electron Microscopy with Vortex Beams"

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Martin Linck (CEOS)

Juan-Carlos Idrobo (ORNL)

Ondrej Krivanek (Nion)

Phil Batson (Rutgers)

Nigel Browning (PNNL)

Rodney Herring (UVic)

Robert Klie

Preliminary Lorentz TEM results in amorphous FeGd
w/ James Lee, Sujoy Roy (ALS) Sergio Montoya, Eric Fullerton (UCSD)