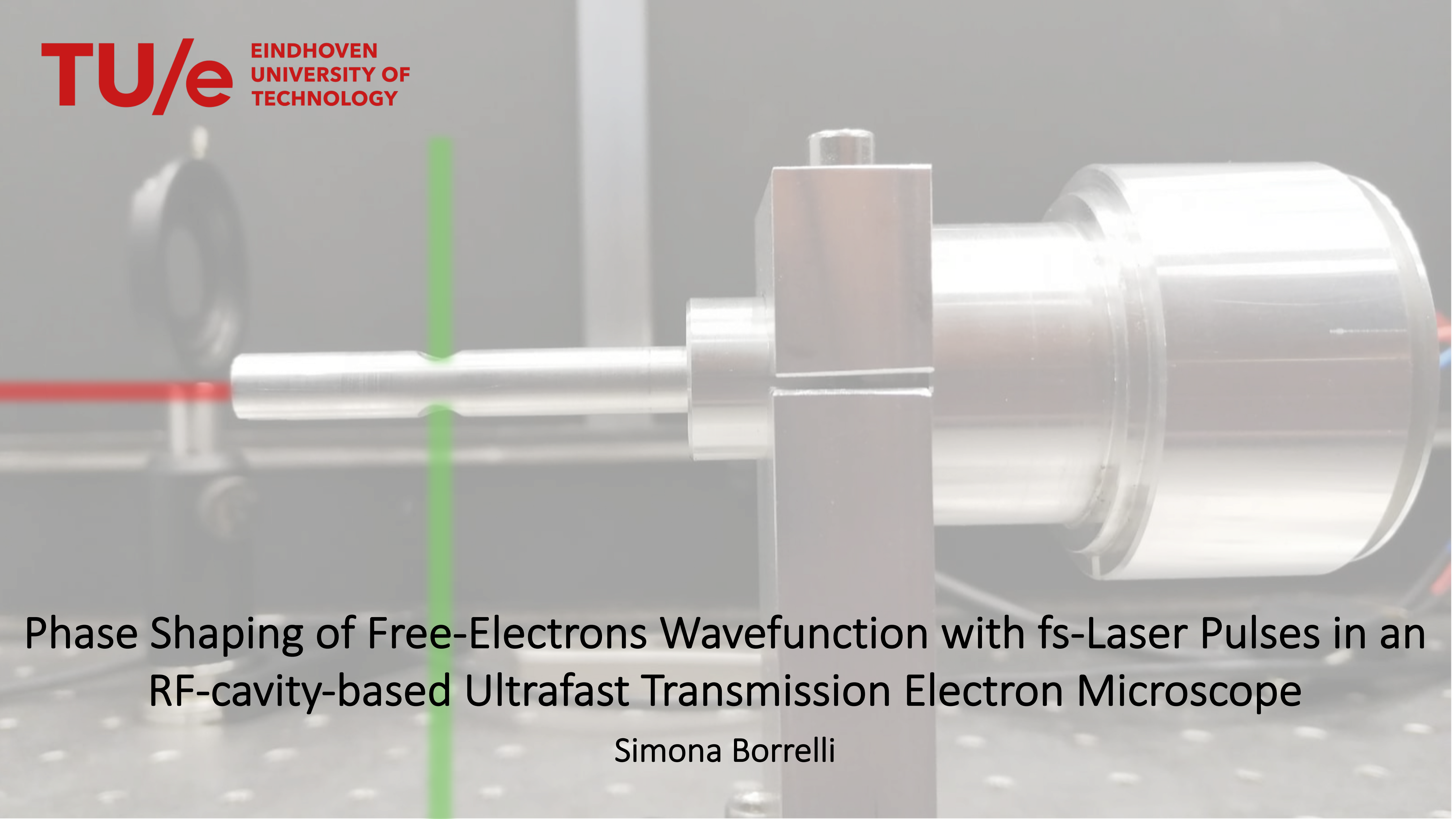
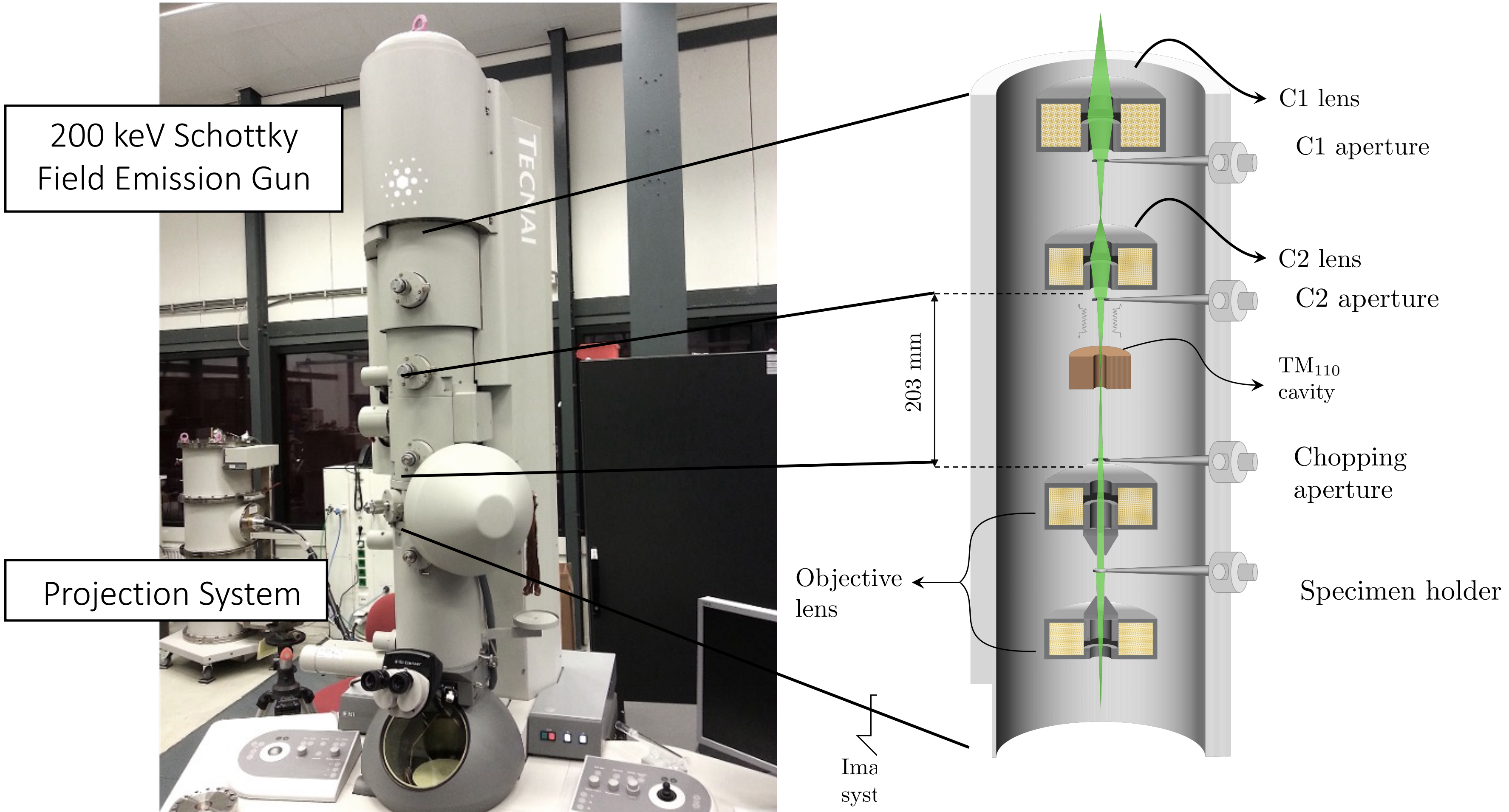


EINDHOVEN UNIVERSITY OF TECHNOLOGY

Phase Shaping of Free-Electrons Wavefunction with fs-Laser Pulses in an RF-cavity-based Ultrafast Transmission Electron Microscope

Simona Borrelli

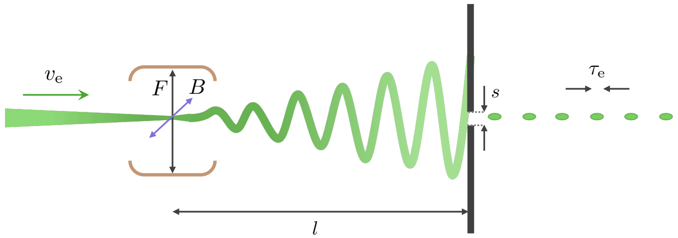
RF-Cavity-Based Ultrafast Transmission Electron Microscope



Br = (7 ± 1) x 10⁶ A/ m² sr V

~ 100 fs pulses
@ 75 MHz or 3 GHz

$\varepsilon_{n,x}$, $\varepsilon_{n,y}$ ~ pm rad



s.borrelli@tue.nl

TU/e EINDHOVEN UNIVERSITY OF TECHNOLOGY
ThermoFisher SCIENTIFIC

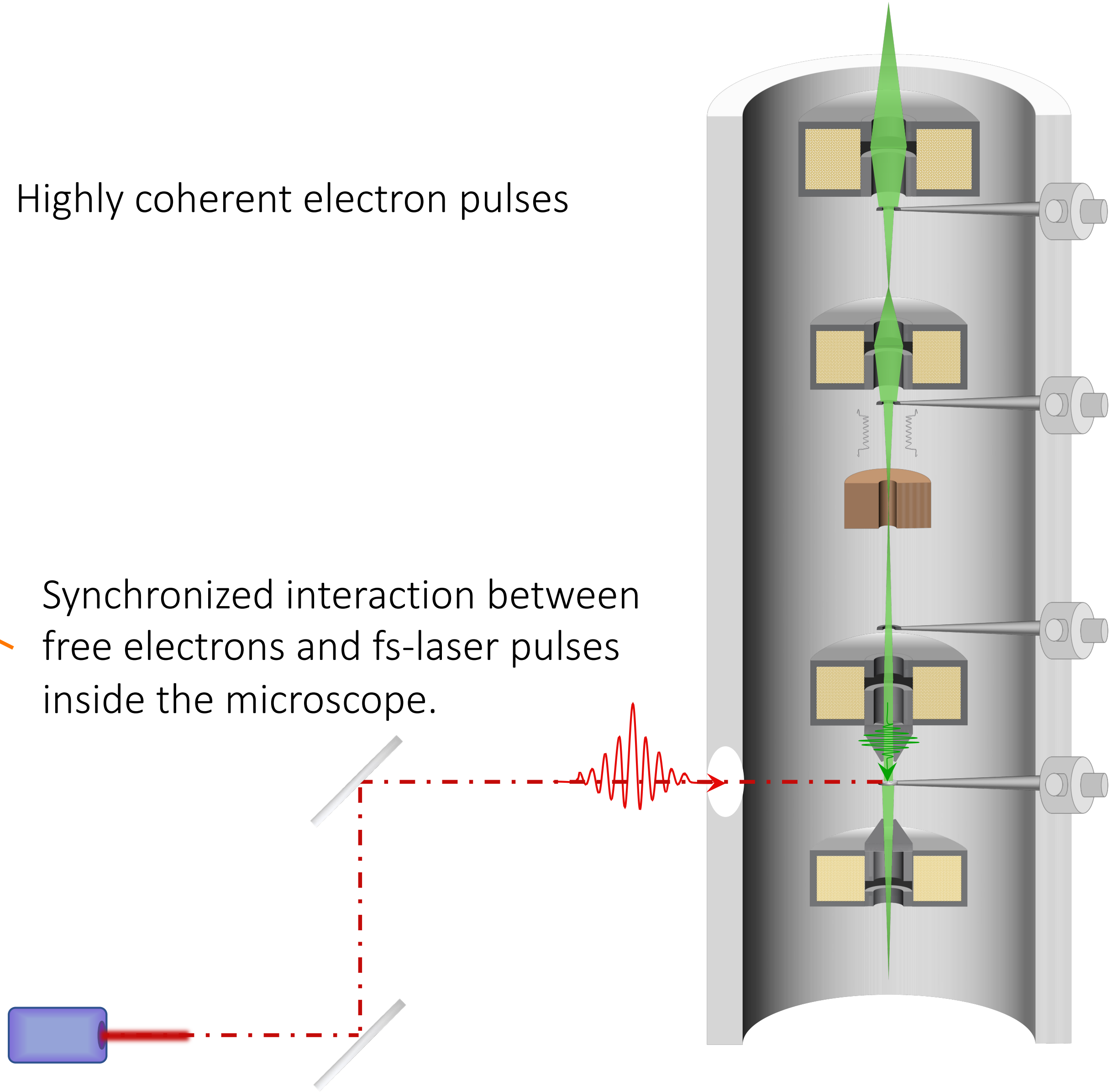
Coherent Manipulation of Electron Wavefunction with Light

Highly coherent electron pulses

Coherent manipulation and control of the electron's wavefunction with light

Synchronized interaction between free electrons and fs-laser pulses inside the microscope.

Establishing new free electron quantum optics based on the interaction of sub-ps electron pulses with the time-dependent electromagnetic fields of light.

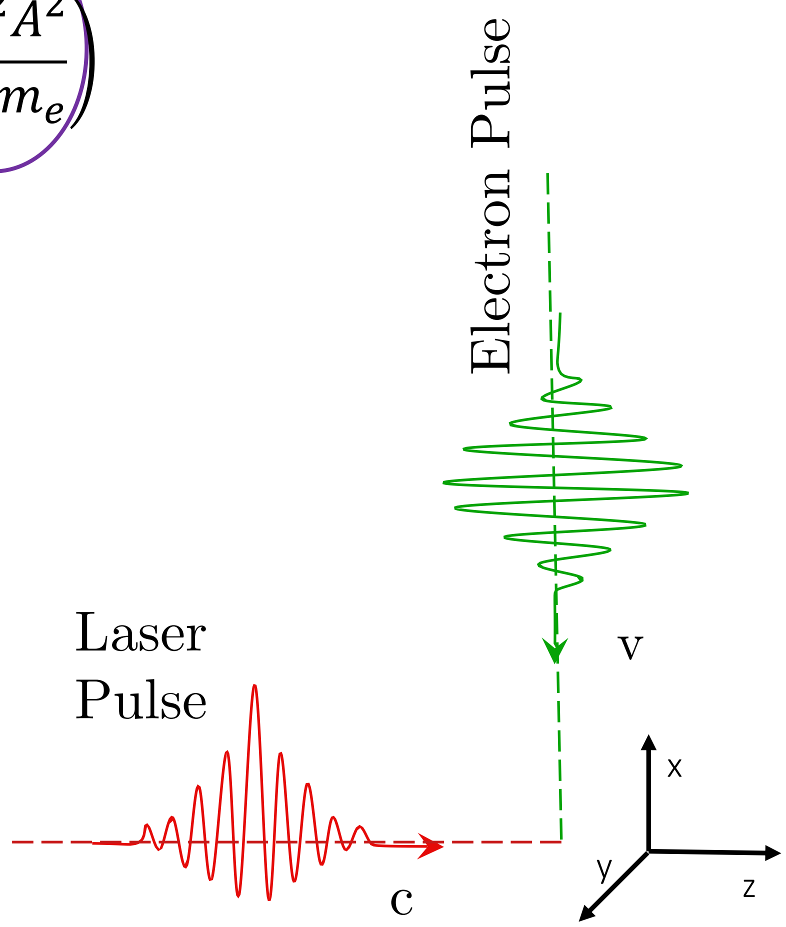


s.borrelli@tue.nl

Coherent Manipulation of Electron Wavefunction with Light

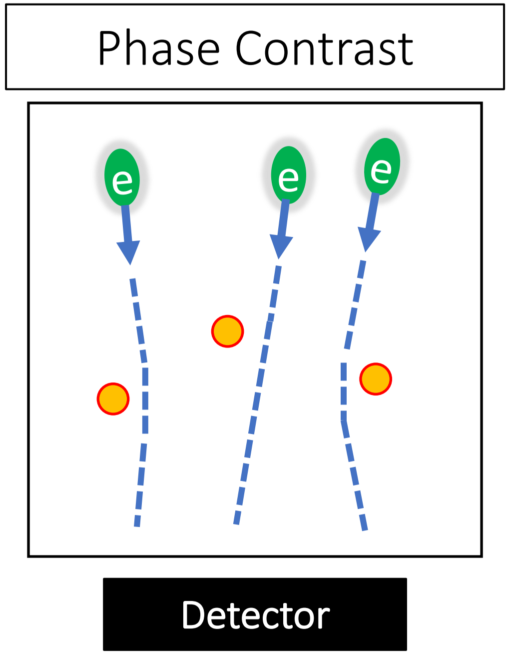
$$H_S = \left(-\frac{\hbar^2\nabla^2}{2\,m_e} - \frac{ie\hbar}{m}\bar{A}\cdot\bar{\nabla} + \frac{e^2A^2}{2\,m_e}\right)$$

Ponderomotive Phase Shaping



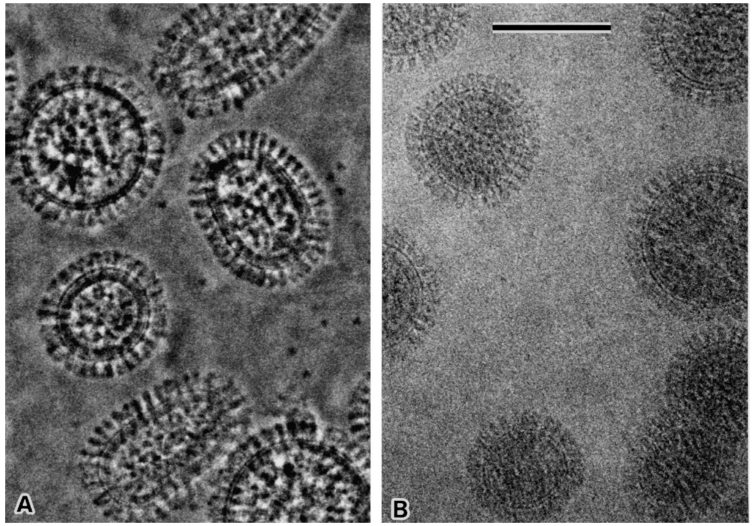
s.borrelli@tue.nl

Pulsed-Laser Zernike Phase Plate for Phase Contrast Imaging of biological samples

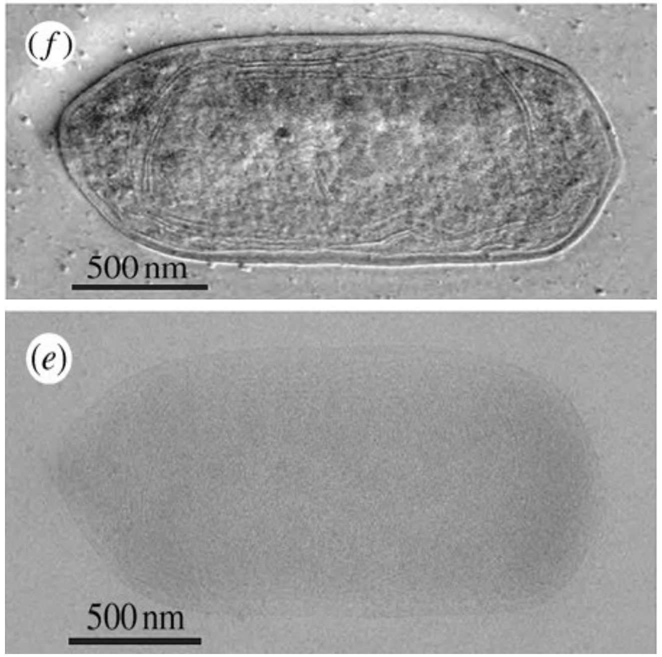


$\frac{\pi}{2}$ Phase Shift between scattered and unscattered electrons

Weakly scattering specimens



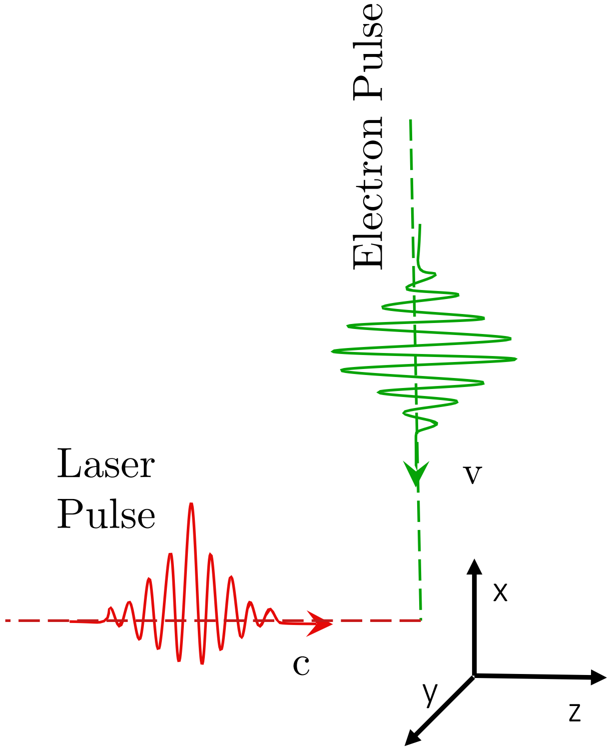
Zernike phase contrast electron micrograph (A) and a conventional electron micrograph (B) of ice-embedded influenza A viruses (*Yamaguchi Y. et al.*)



Conventional (e) and phase contrast (f)TEM image of cyanobacterial cell (*Konyuba Y. et al.*)

Phase Shaping of Electrons Wavefunction with fs-Laser Pulses

$$H_S = \left(-\frac{\hbar^2 \nabla^2}{2\, m_e} - \frac{ie\hbar}{m} \bar{A} \cdot \bar{\nabla} + \frac{e^2 A^2}{2\, m_e} \right)$$



Feasibility of a Pulsed Ponderomotive Phase Plate for Electron Beams

K.A.H. van Leeuwen[1,*], W.J. Schaap[1], B. Buijsse[2], S. Borelli[1], S.T. Kempers[1], W. Verhoeven[1,3] and O.J. Luiten[1]
[1]*Department of Applied Physics, Eindhoven University of Technology, PO Box 513, 5600MB Eindhoven, The Netherlands*
[2]*Thermo Fisher Scientific, Achtseweg Noord 5, 5651 GG Eindhoven, The Netherlands*
[3]*Present address: Thermo Fisher Scientific, Achtseweg Noord 5, 5651 GG Eindhoven, The Netherlands and*
[*]Author to whom any correspondence should be addressed.
(Dated: August 31, 2022)

We propose a scheme for constructing a phase plate for use in a Zernike-type phase contrast electron microscope, based on the interaction of the electron beam with a strongly focused, high-power femtosecond laser pulse and a pulsed electron beam. Analytical expressions for the phase shift using the time-averaged ponderomotive potential and a paraxial approximation for the focused laser beam are presented, as well as more rigorous quasiclassical simulations based on the quantum phase integral along classical, relativistic electron trajectories in an accurate, non-paraxial description of the laser beam. The results are shown to agree well unless the laser beam is focused to a waist size below a wavelength. For realistic (off-the-shelf) laser parameters the optimum phase shift of $\pi/2$ is shown to be achievable. When combined with RF-cavity based electron chopping and compression techniques to produce electron pulses, a femtosecond regime pulsed phase contrast microscope can be constructed. The feasibility and robustness of the scheme are further investigated using the simulations, leading to motivated choices for design parameters such as wavelength, focus size and polarization.

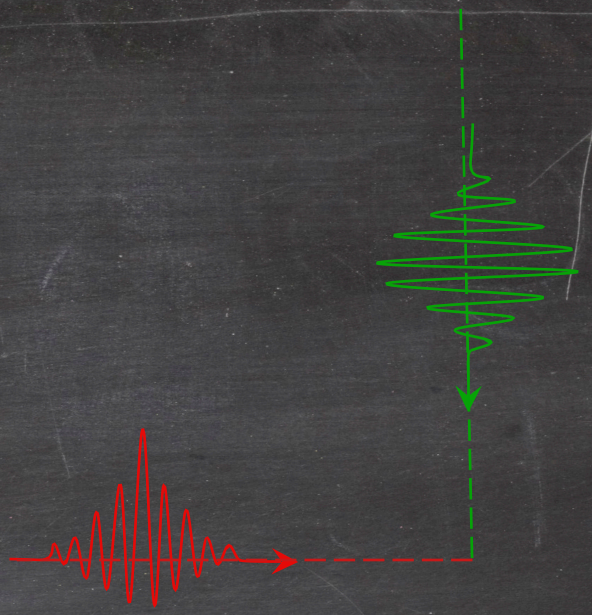
K.A.H. van Leeuwen et al. , *Feasibility of a Pulsed Ponderomotive Phase Plate for Electron Beams*, arXiv, 2022.

s.borrelli@tue.nl

Phase Shaping of Electrons Wavefunction with fs-Laser Pulses

Relativistic ponderomotive potential

$$U_p = \frac{e^2}{c\,\epsilon_0\,\gamma\,m\,\omega_0^2}\,\frac{W}{\pi^{3/2}\,w(z)^2\,\tau}\,e^{-\frac{(t-z/c)^2}{\tau^2}}\,e^{-2\frac{x^2+y^2}{w(z)^2}}$$



Phase Shift on an electron crossing the laser spot w(z) at $\bar{r} = (z, y)$ at time t_0

$$\Delta\varphi = -\frac{1}{\hbar}\int_{-\infty}^{+\infty} dt\; U_p(v(t-t_0), y, z, t)$$

Maximum Phase Shift for perfect space-time synchronization between laser and electron pulses ($t_0 = y = z = 0$)

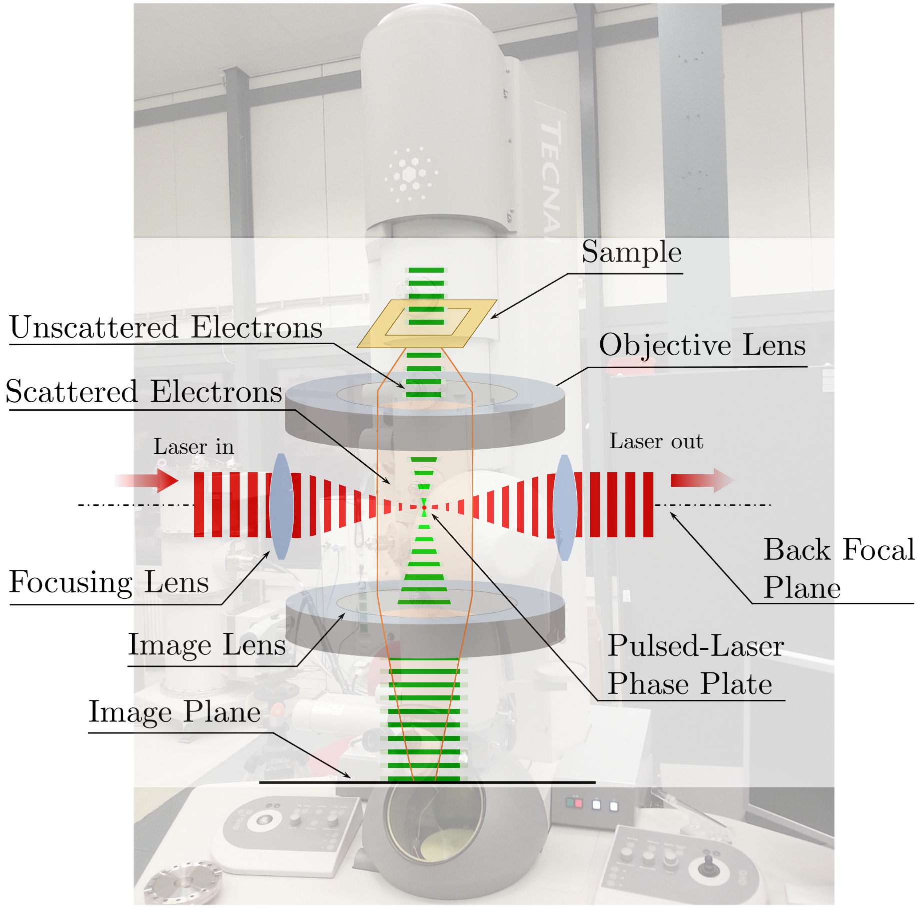
$$\Delta\varphi_{max} \sim -\frac{e^2}{\hbar\,c\,\epsilon_0\,\gamma\,m\,\omega_0^2}\,\frac{W}{\pi\,w_0^2}\,\frac{\tau_t}{\tau}$$

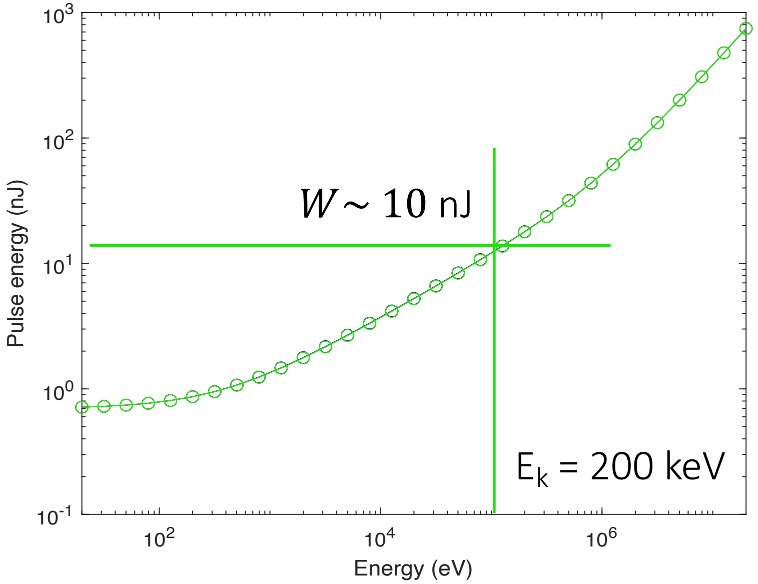
Laser and electron pulses synchronized

K.A.H. van Leeuwen et al. , *Feasibility of a Pulsed Ponderomotive Phase Plate for Electron Beams*, arXiv, 2022.

s.borrelli@tue.nl

Pulsed-Laser Zernike Phase Plate for Phase Contrast Imaging of biological samples





Phase Shift $\Delta\varphi_{max} = -\frac{\pi}{2}$

λ = 800 nm

$w_0 = 2\,\lambda$ = 1.6 µm

τ = 300 fs & $\tau_e = \frac{1}{3}\,\tau = 100$ fs

$W \sim 10$ nJ/pulse

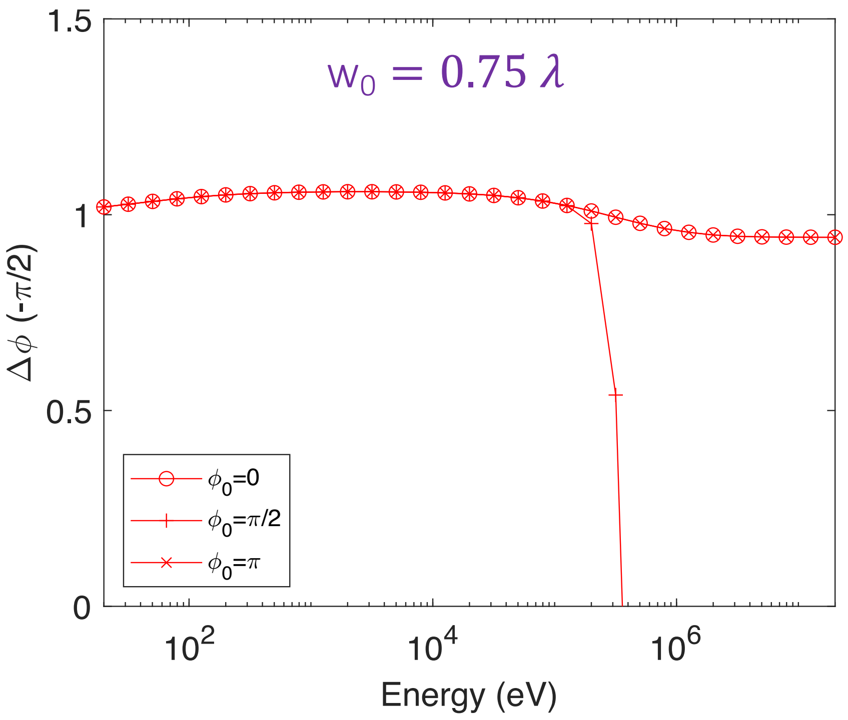
s.borrelli@tue.nl

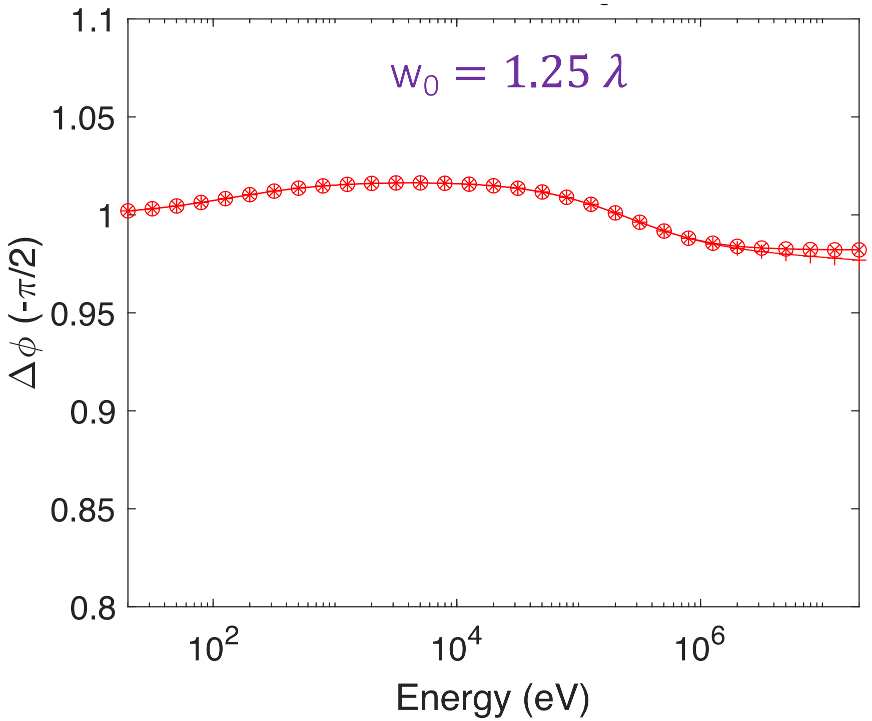
Phase Shaping of Electrons Wavefunction with fs-Laser Pulses

$$\Delta\varphi = \frac{1}{\hbar}\int_P dt\, L(x,y,z,t) = \int_P dt\left(-\frac{mc^2}{\gamma} - e\frac{\boldsymbol{p}\cdot\boldsymbol{A}}{m\gamma} - \frac{e^2\boldsymbol{A}\cdot\boldsymbol{A}}{m\gamma} + eV\right)$$

Phase shift at high electron energies depends on:

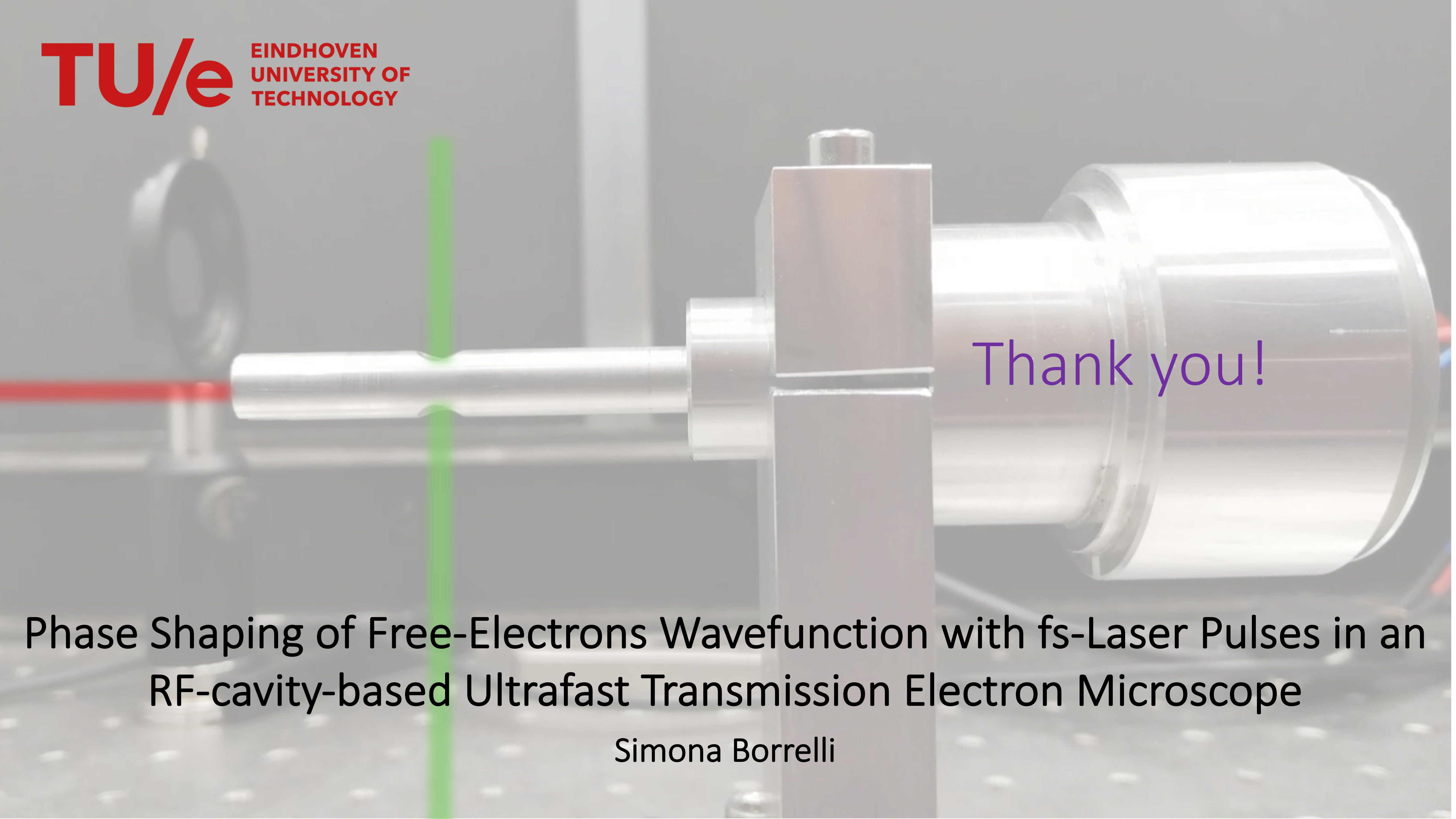
- field phase
- field polarization
- field model chosen





K.A.H. van Leeuwen et al. , *Feasibility of a Pulsed Ponderomotive Phase Plate for Electron Beams*, arXiv, 2022.

s.borrelli@tue.nl



EINDHOVEN UNIVERSITY OF TECHNOLOGY

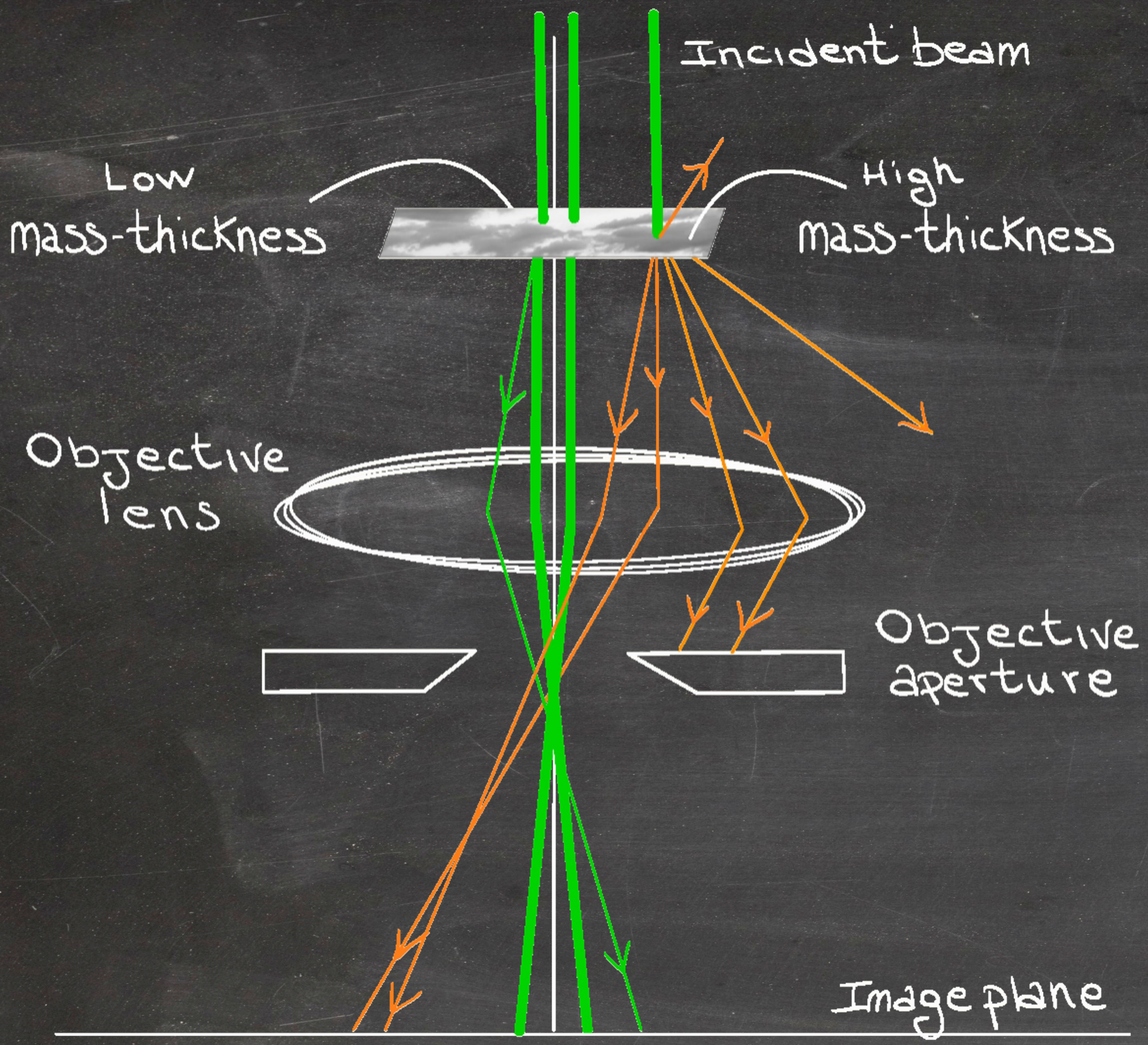
Thank you!

Phase Shaping of Free-Electrons Wavefunction with fs-Laser Pulses in an RF-cavity-based Ultrafast Transmission Electron Microscope

Simona Borrelli

Backup slides

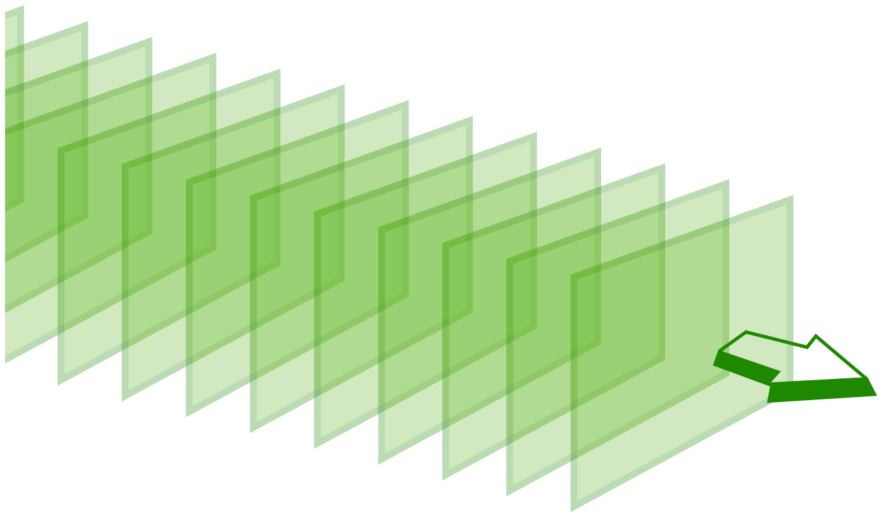
Highly scattering specimens



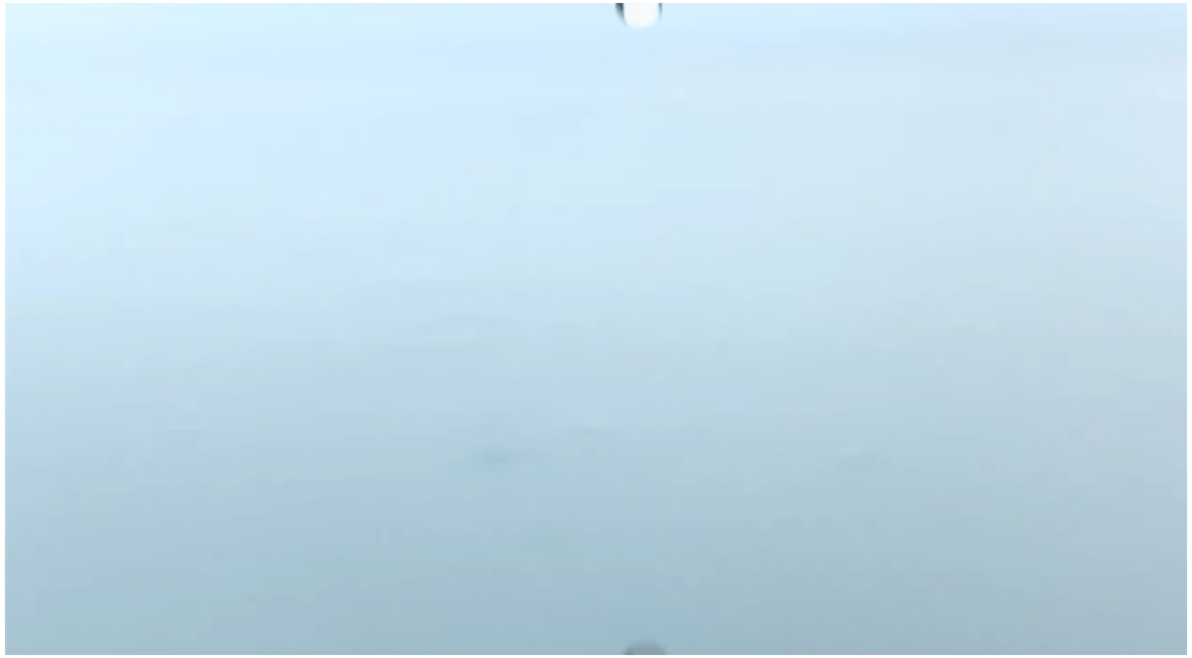
Amplitude Contrast Imaging

Diffraction Contrast Imaging

Weakly scattering specimens



$$\psi(\bar{r}, t) = A\, e^{i(2\pi \bar{k}_0 \cdot \bar{r}\, -\, \omega t)}$$



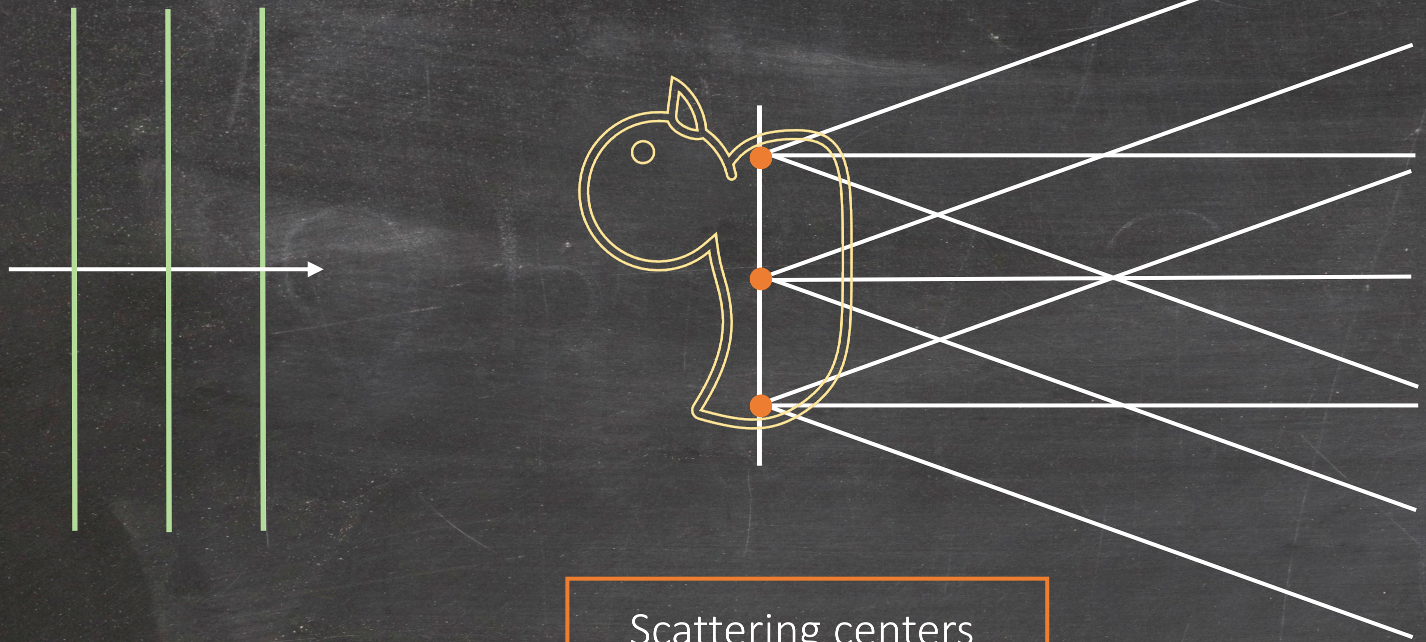
The electron planewave is modulated

Image Formation in a TEM

Electron plane wavefunction

Electron plane wave modulated by the object structure

$$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\,\phi(\bar{r})\,]}$$



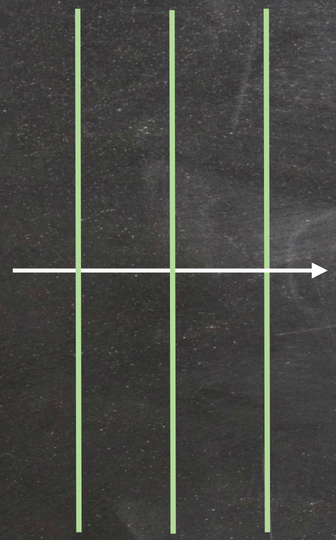
Scattering centers in the specimen

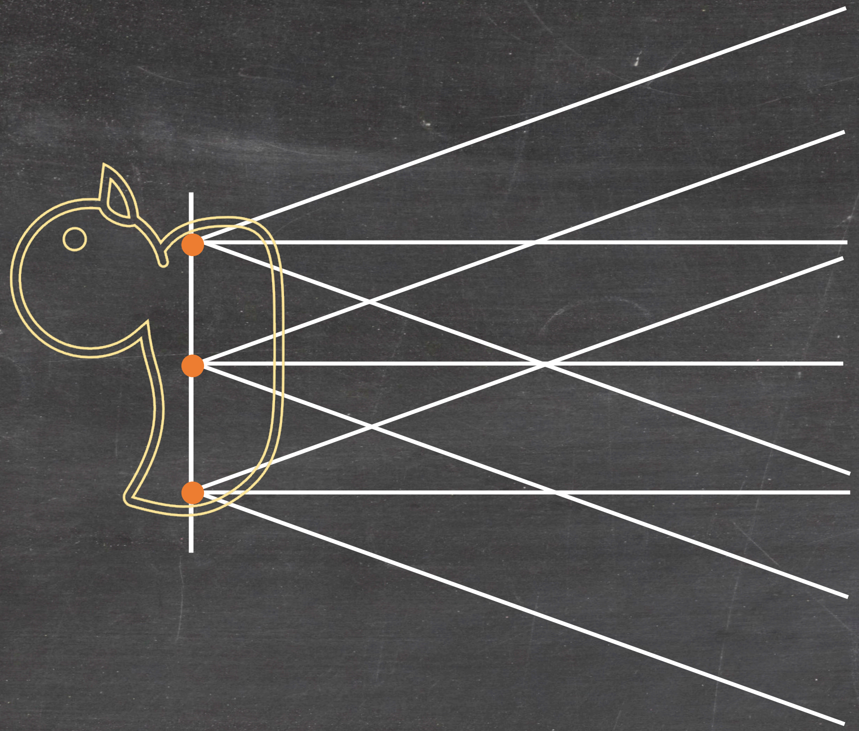
Electron plane wave modulated by the object structure

Electron plane wavefunction

... Far field...`

$$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\,\phi(\bar{r})\,]}$$





Fourier Transform of the specimen's density pattern

$$\mathrm{FT}[obj(\bar{r})\ \int_{-\infty}^{+\infty} obj(\bar{r})\ e^{-i\,2\,\pi\,\bar{q}\cdot\bar{r}}\ d\bar{r}\ \ ]$$

Scattering Angle

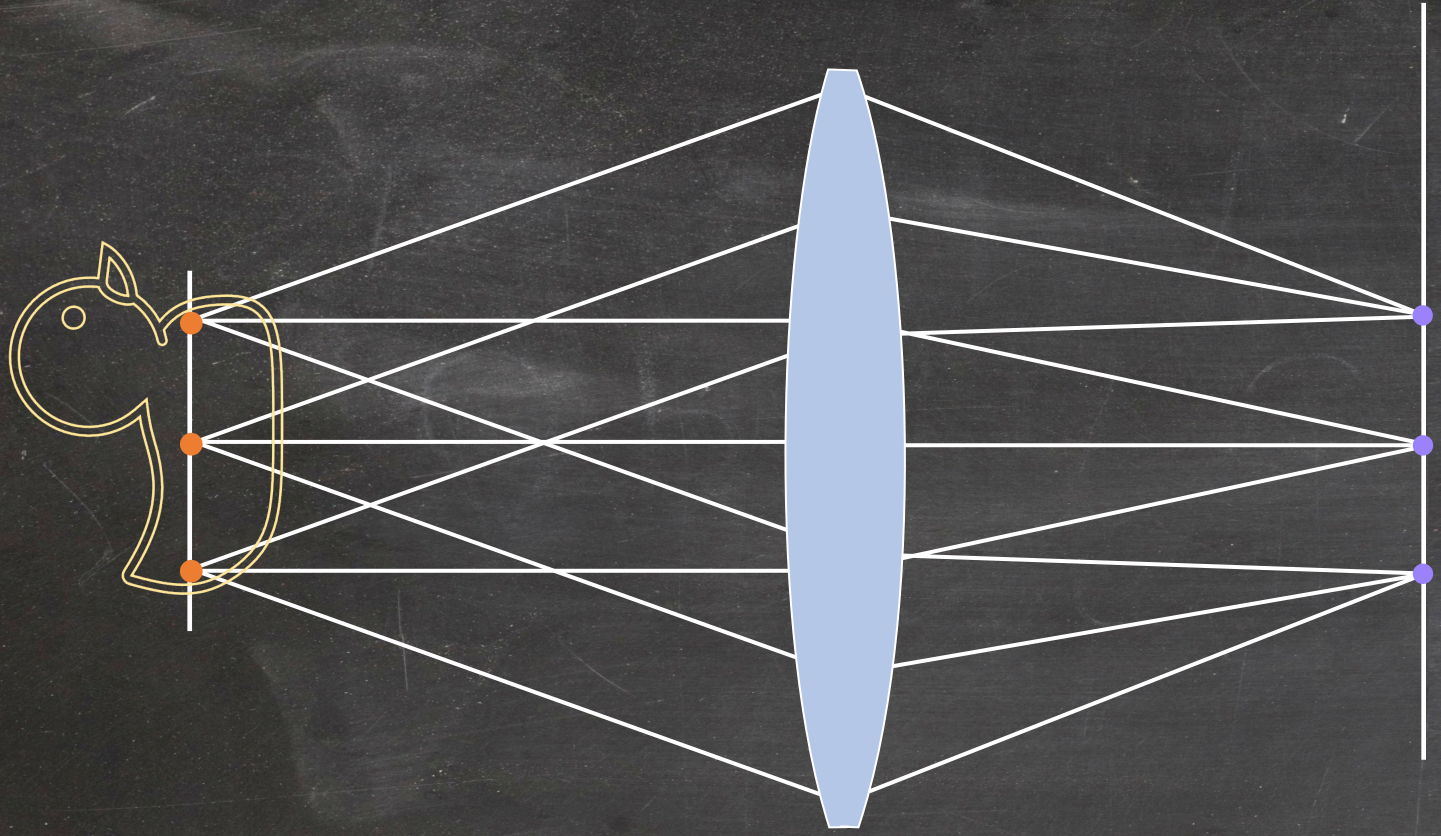
Spatial Frequency $\bar{q} = k\ \bar{\vartheta}$

Wavenumber

Electron plane wave modulated by the object structure

$$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\,\phi(\bar{r})\,]}$$

Fourier Transform of the specimen’s density pattern

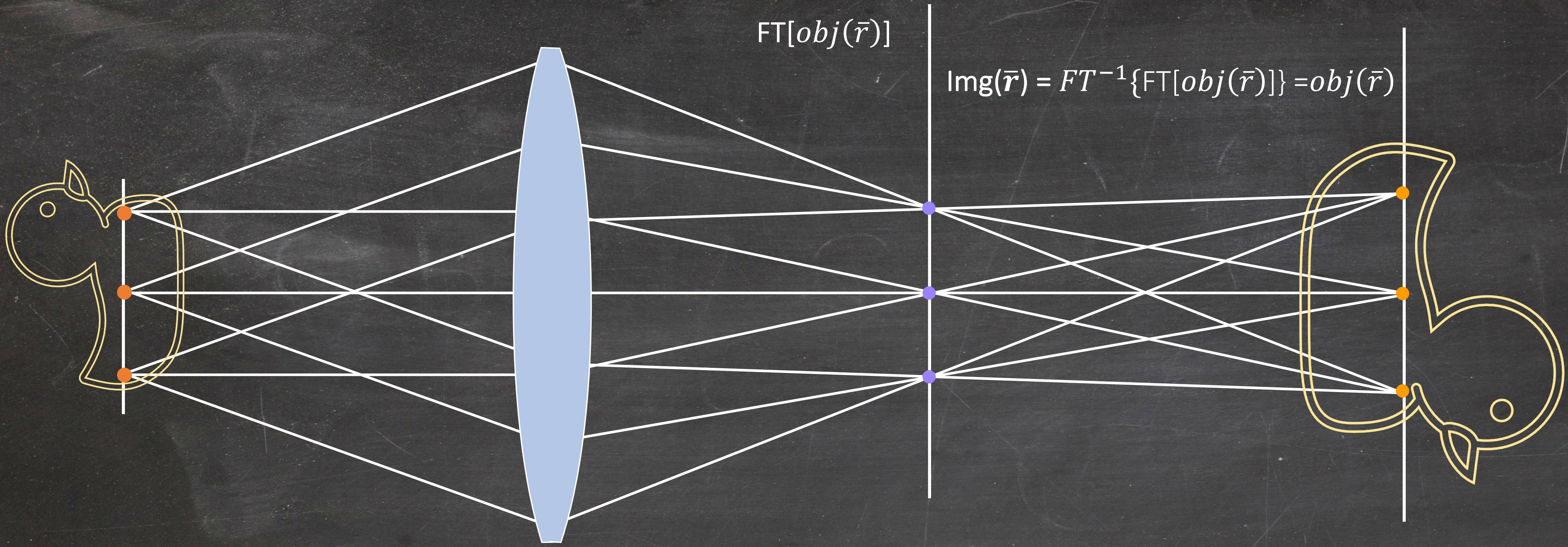


Electron plane wave modulated by the object structure

Image wave

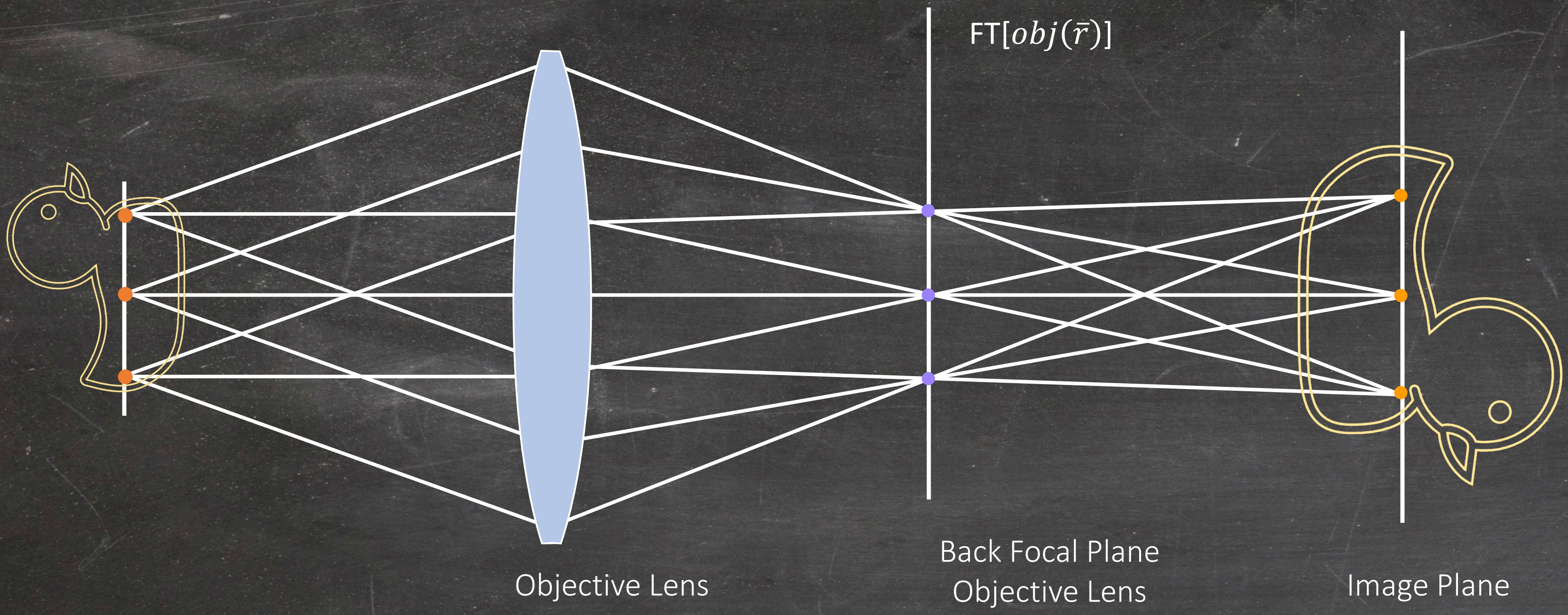
$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\,\phi(\bar{r})\,]}$

$\text{Img}\ (\bar{r}) = A(\bar{r})\ e^{i\,[\phi(\bar{r})\,]}$



$$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\phi(\bar{r})]}$$

$$\text{Img}\ (\bar{r}) = A(\bar{r})\ e^{i\,[\phi(\bar{r})]}$$



Detector

$$I = |Img\ (\bar{r})|^2 = A(\bar{r})^2$$

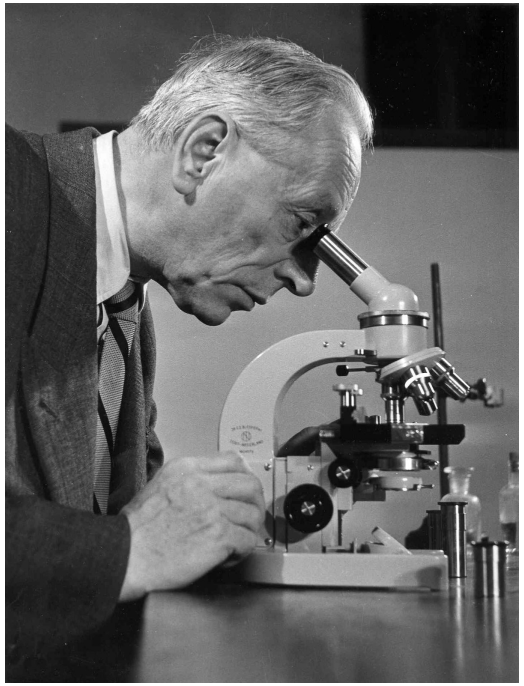
Amplitude Contrast

In the image only the amplitude information is retrieved.
The phase information is lost.

HOW RETRIEVE PHASE INFORMATION???

Phase Contrast Imaging….

… With …

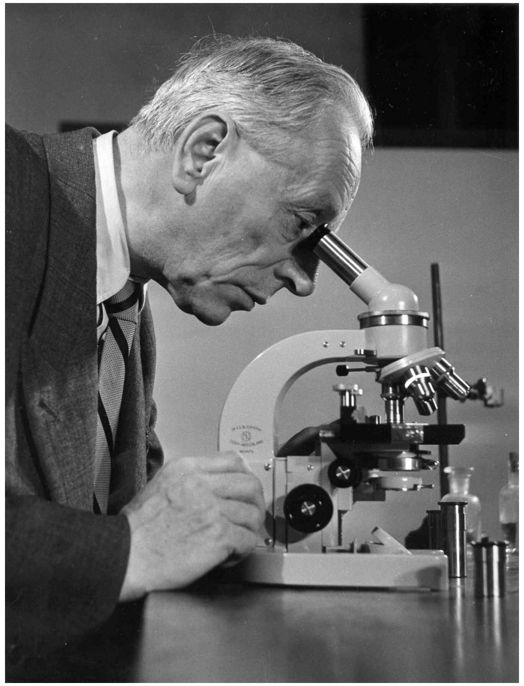


Zernike Phase Plate

HOW RETRIEVE PHASE INFORMATION???

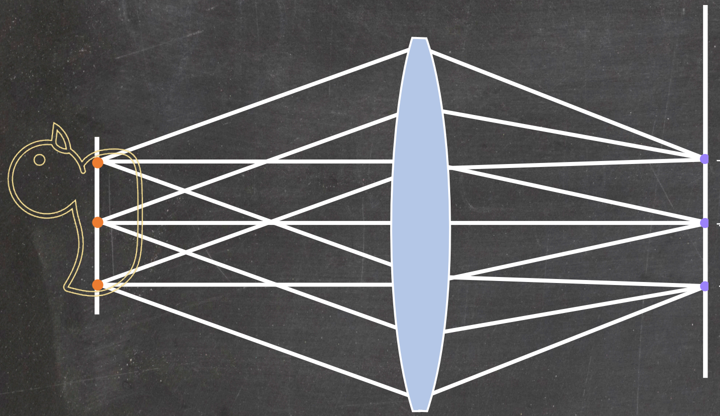
Phase Contrast Imaging....

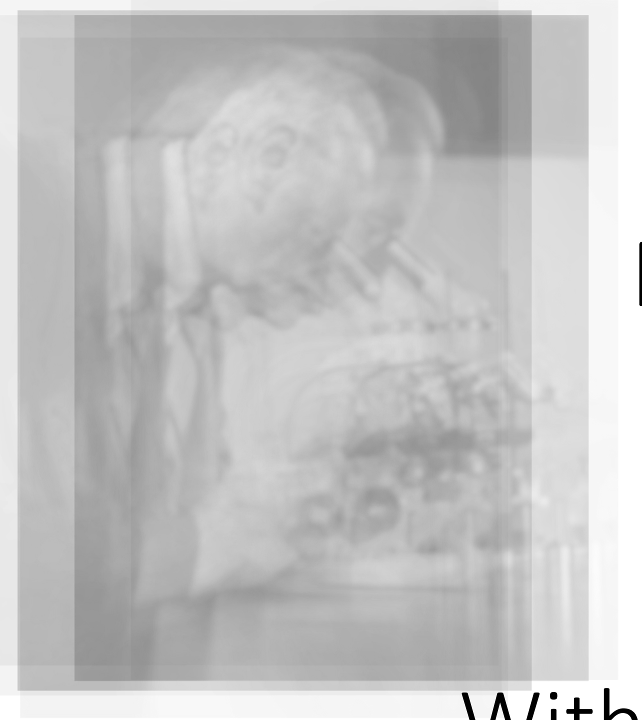
... With ...



Zernike Phase Plate

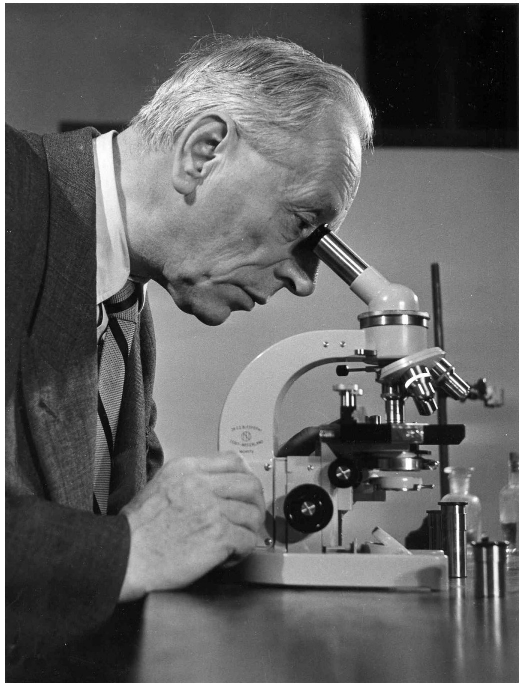
Phase informtaion can be retrieved by inserting into the Fourier space a $\lambda/4$-phase plate that introduces an additional $\frac{\pi}{2}$ −**phase shift** between the zero-order beam (q = 0) and the high-order diffracted beams (q ≠ 0)





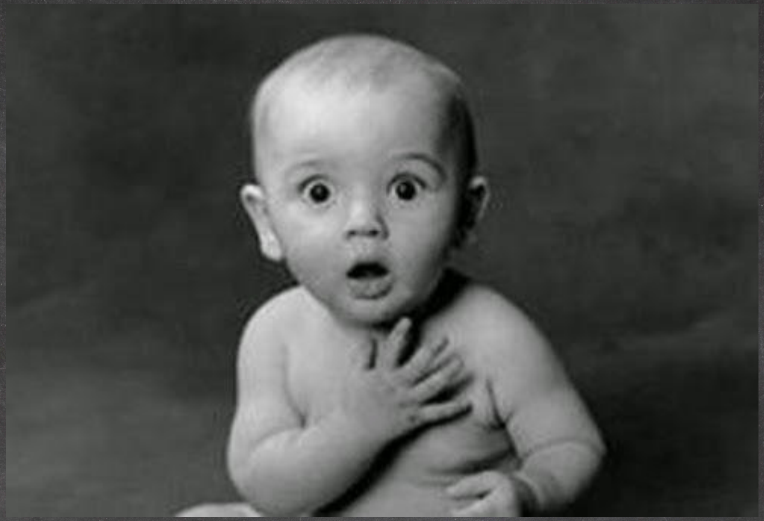
Phase Contrast Imaging….

… With …



Zernike Phase Plate

$$i = e^{i\frac{\pi}{2}}$$

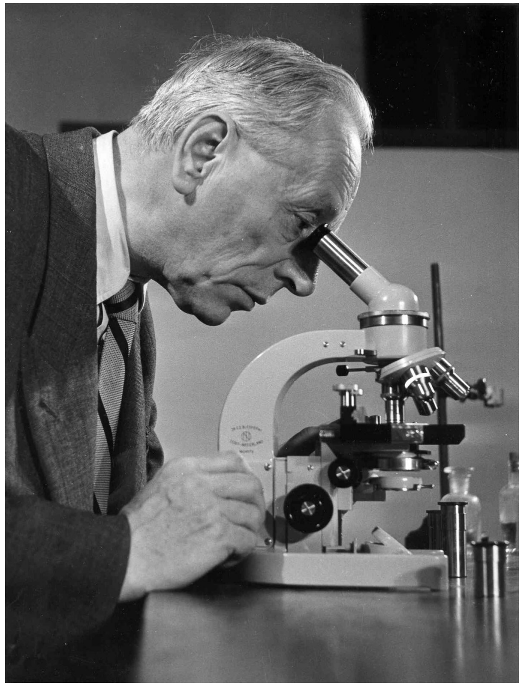


Phase information can be retrieved by inserting into the Fourier space a $\lambda/4$-phase plate that introduces an additional $\frac{\pi}{2}$ −phase shift between the zero-order beam (q = 0) and the high-order diffracted beams (q ≠ 0)

Phase Contrast Imaging….

… With …

Zernike Phase Plate



Weakly scattering specimens

$$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\,\phi(\bar{r})\,]}$$

$A(\bar{r}) = 1 - t(\bar{r})$

$\phi(\bar{r}) \leq 2\pi/10$

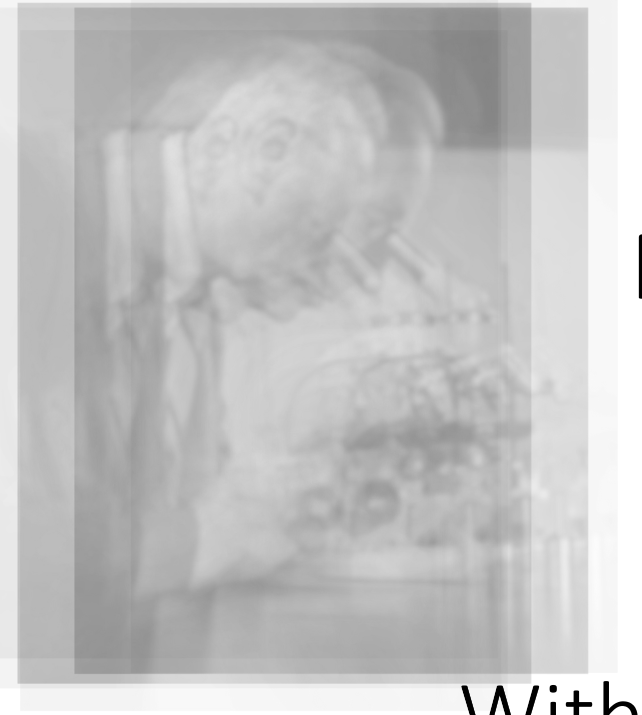
Spatial frequency q_0

$t \ll 2\pi/10$

$$obj\ (x) \approx 1 - (\,t - i\phi)\cos(2\,\pi q_0 \cdot x + \varepsilon\,) = Img(x)$$

Amplitude Contrast

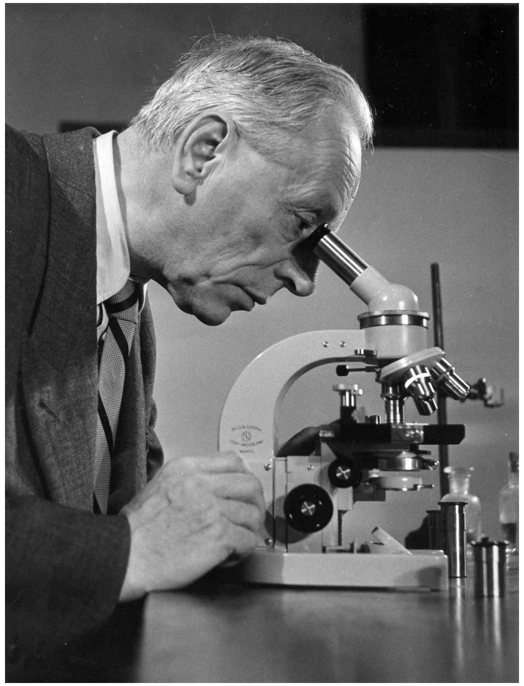
$$I = |Img(x)|^2 \approx 1 - \mathbf{2}\,\boldsymbol{t}\cos(2\,\pi\,q_0 \cdot x + \varepsilon\,)$$



Phase Contrast Imaging….

… With …

Zernike Phase Plate



Weakly scattering specimens

$$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\,\phi(\bar{r})\,]}$$

$\mathrm{A}(\bar{r}) = 1 - t(\bar{r})$

$\phi(\bar{r}) \le 2\pi/10$

Spatial frequency q_0

$\mathrm{t} \ll 2\pi/10$

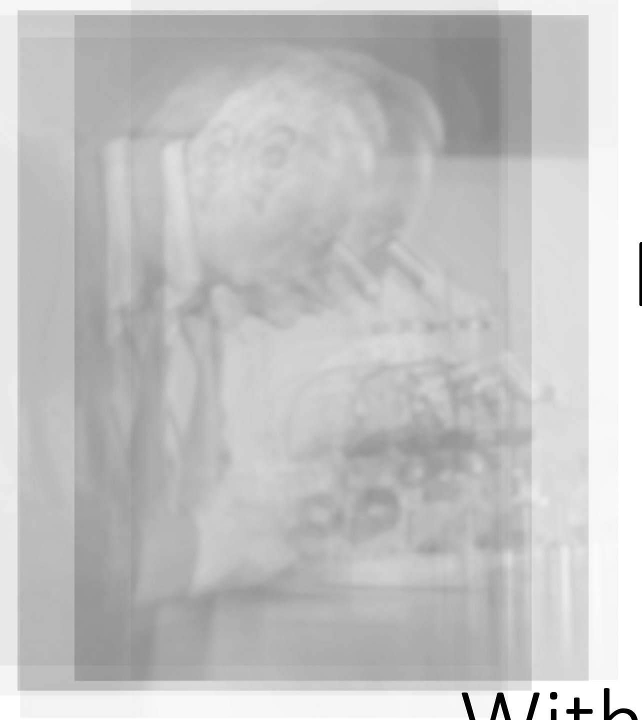
$$obj\ (x) \approx 1 - (\,t - i\phi)\cos(2\,\pi q_0 \cdot x + \varepsilon\,) = Img(x)$$

Amplitude Contrast

$$|Img(x)|^2 \approx 1 - \mathbf{2}\,\boldsymbol{t}\cos(2\,\pi\, q_0 \cdot x + \varepsilon\,)$$

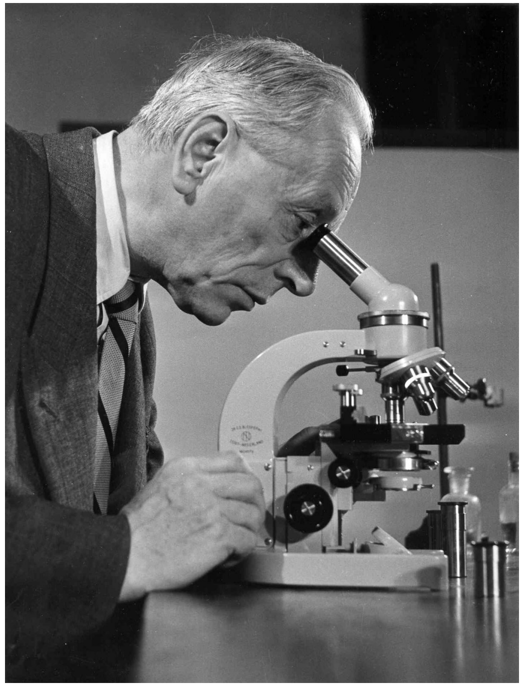
λ/4-Phase Plate

$$e^{i\frac{\pi}{2}} \Rightarrow \mathrm{I}mg(x) \approx 1 - (i\,t + \phi)\cos(2\,\pi q_0 \cdot x + \varepsilon\,)$$



Phase Contrast Imaging….

… With …



Zernike Phase Plate

Weakly scattering specimens

$$obj\ (\bar{r}) = A(\bar{r})\ e^{i\,[\,\phi(\bar{r})\,]}$$

$$A(\bar{r}) = 1 - t(\bar{r}) \qquad \phi(\bar{r}) \le \frac{2\,\pi}{10}$$

At a certain spatial frequency q_0

$$obj\ (x) \approx 1 - (\,t - i\phi)\cos(2\,\pi q_0 \cdot x + \varepsilon\,) = Img(x)$$

Amplitude Contrast

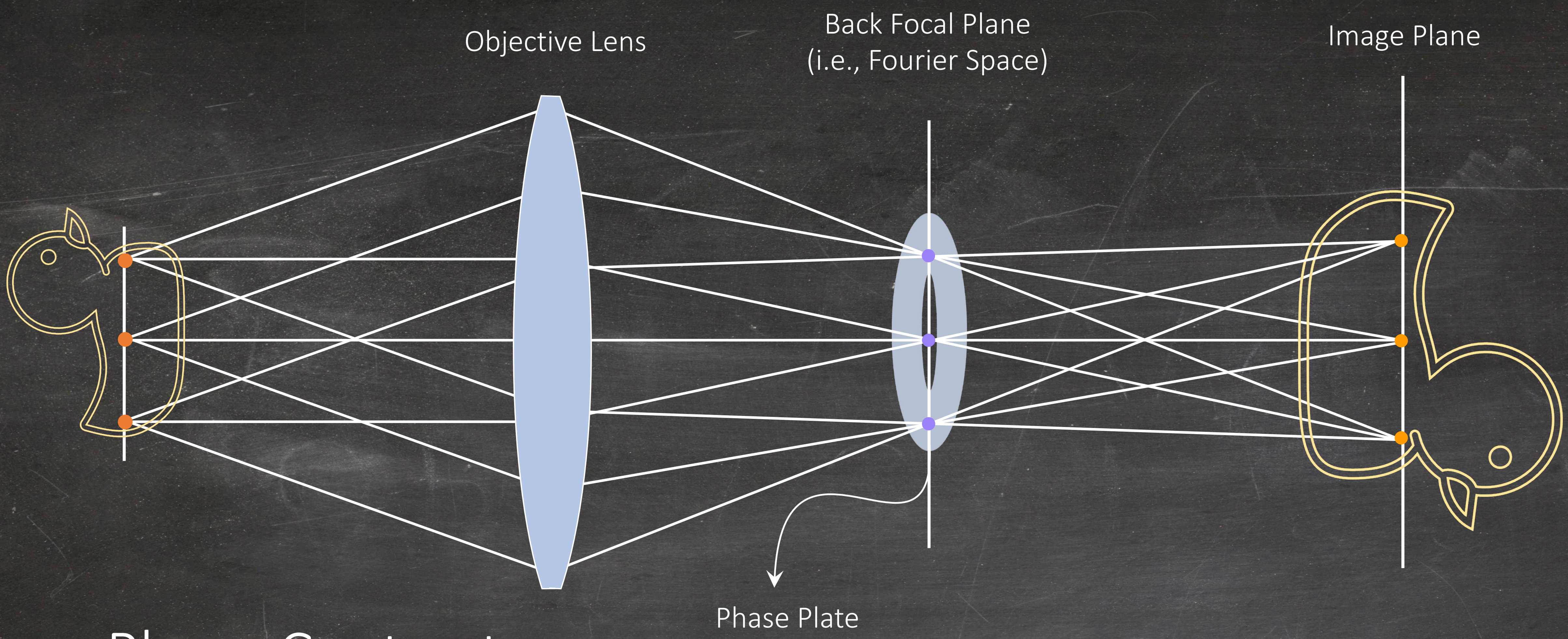
$$I = |Img(x)|^2 \approx 1 - \mathbf{2}\,\mathbf{t}\cos(2\,\pi\, q_0 \cdot x + \varepsilon\,)$$

λ/4-Phase Plate

$$Img(x) \approx 1 - (i\,t + \phi)\cos(2\,\pi q_0 \cdot x + \varepsilon\,)$$

Phase Contrast

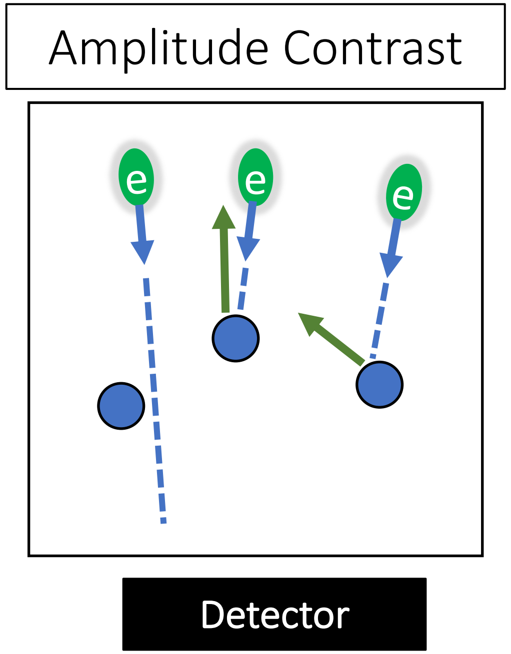
$$I = |Img(x)|^2 \approx 1 - 2\ \phi(\bar{r})\cos(2\,\pi\, q_0 \cdot x + \varepsilon\,)$$



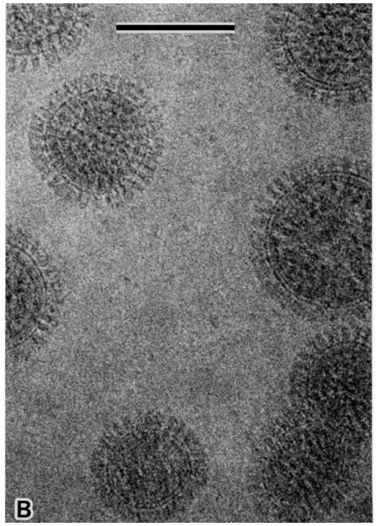
Phase Contrast Imaging with Zernike Phase Plates

The phase modulation induced by the specimen is imaged by the detector.

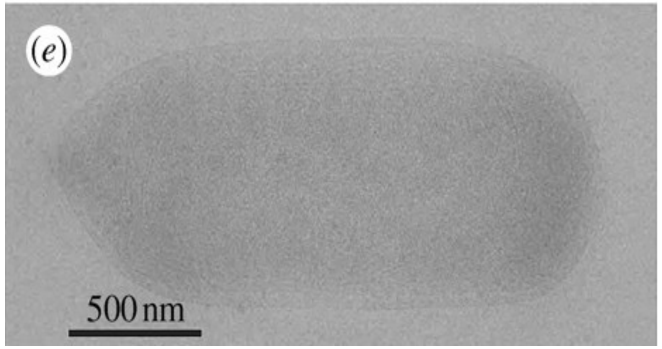
Ponderomotive Phase Shaping



Weakly scattering specimens

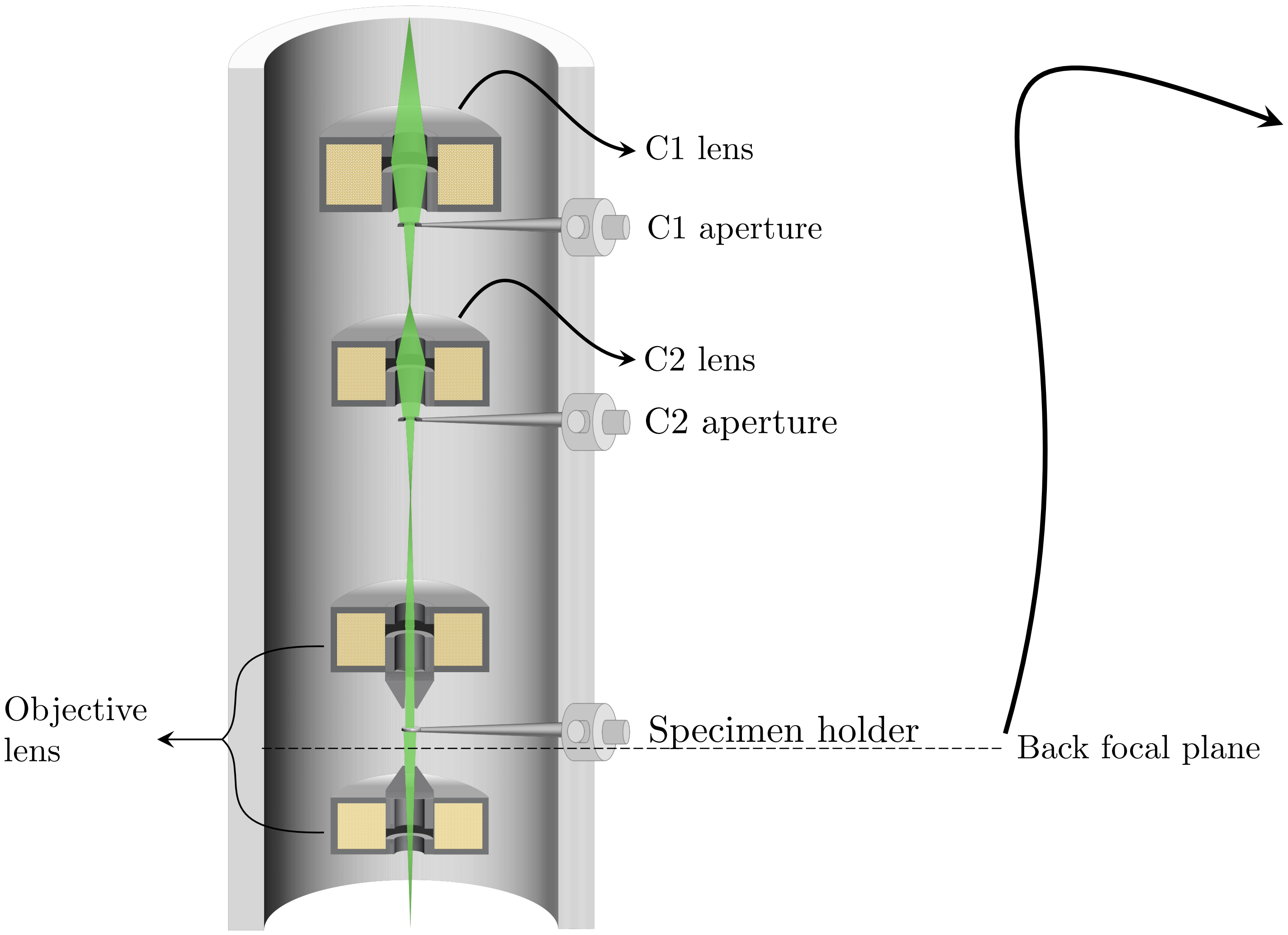


Conventional electron micrograph (B) of ice-embedded influenza A viruses (*Yamaguchi Y. et al.*)

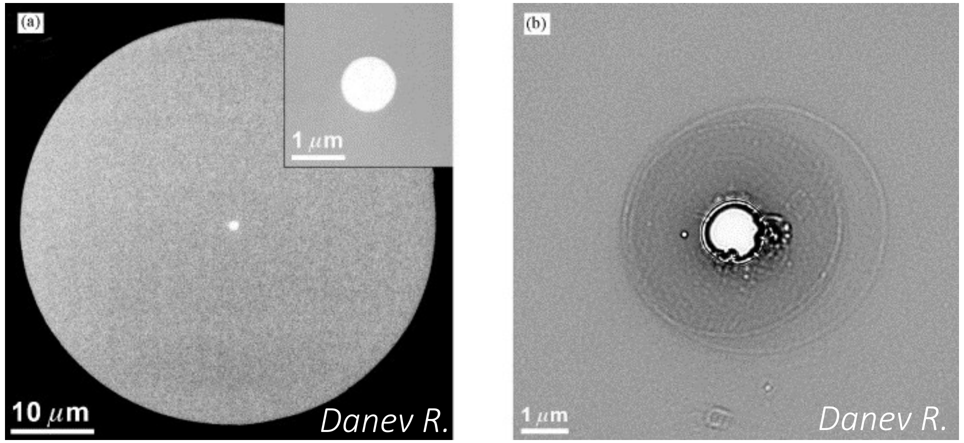


Conventional (e) TEM image of cyanobacterial cell (*Konyuba Y. et al.*)

Zernike Phase Plates for Phase Contrast Imaging



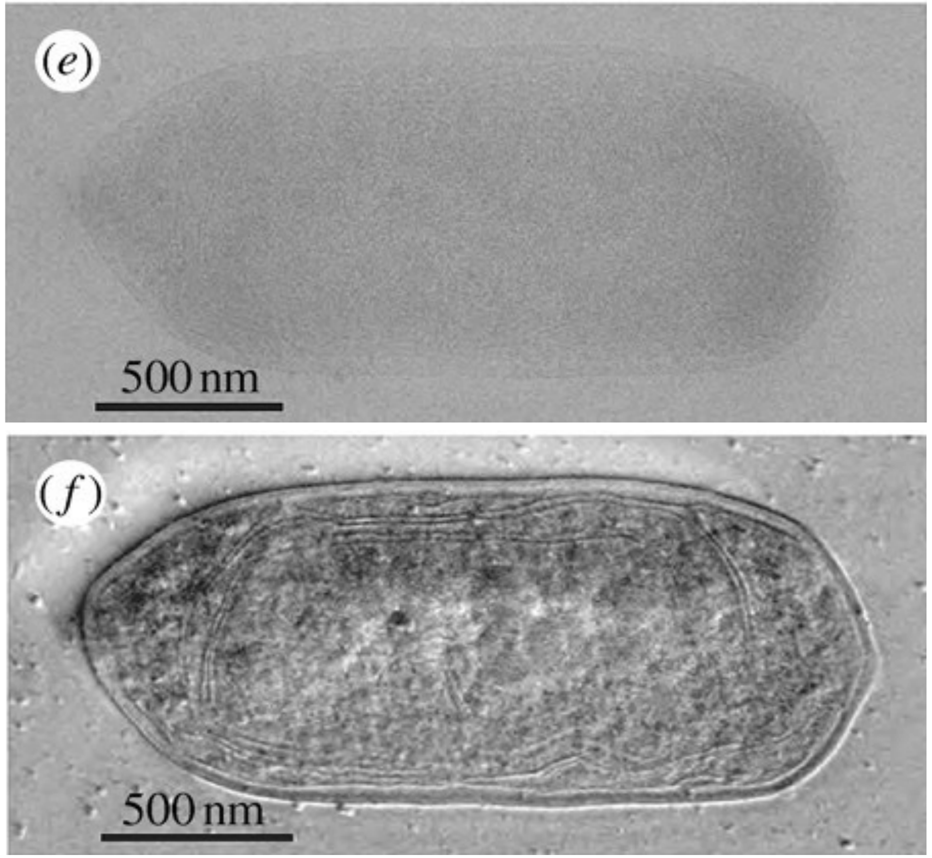
SEM image Zernike phase plates



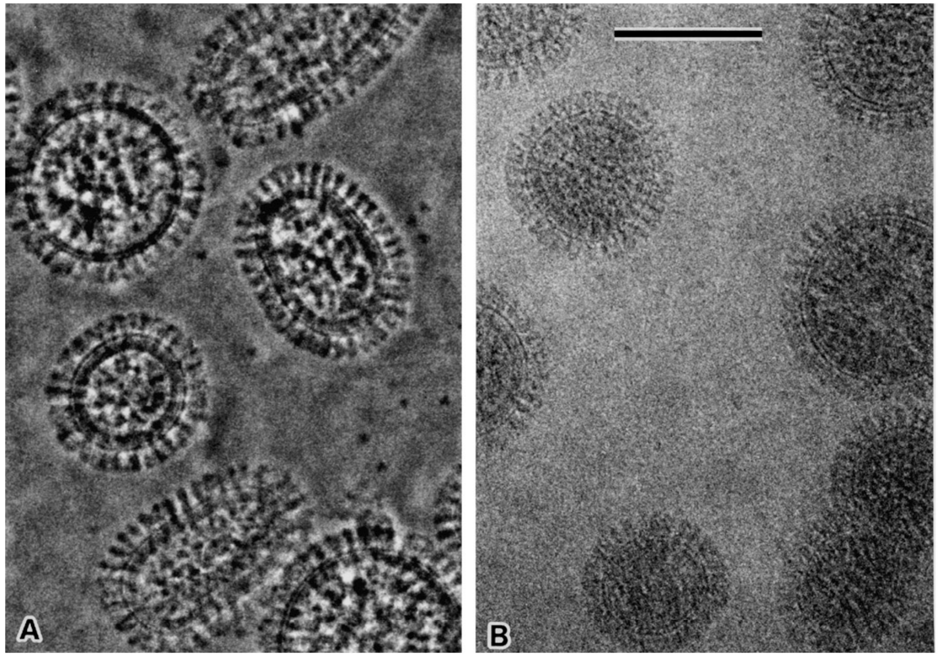
Limitations inherently related to the presence of matter:

- Image distortion due to charging
- Loss of electrons and decoherence
- Limited Spectroscopic capabilities
- Contamination from the electron beam
- Non-tunable and time-varying phase shift

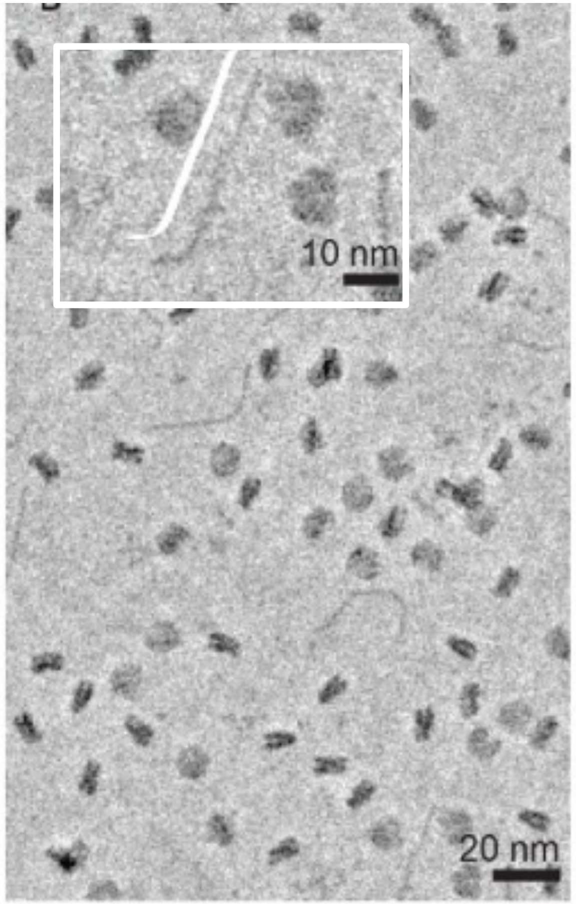
Phase Contrast Imaging with Zernike Phase Plates



Conventional (e) and phase-Contrast (f) TEM image of cyanobacterial cell (*Konyuba Y. et al.*)

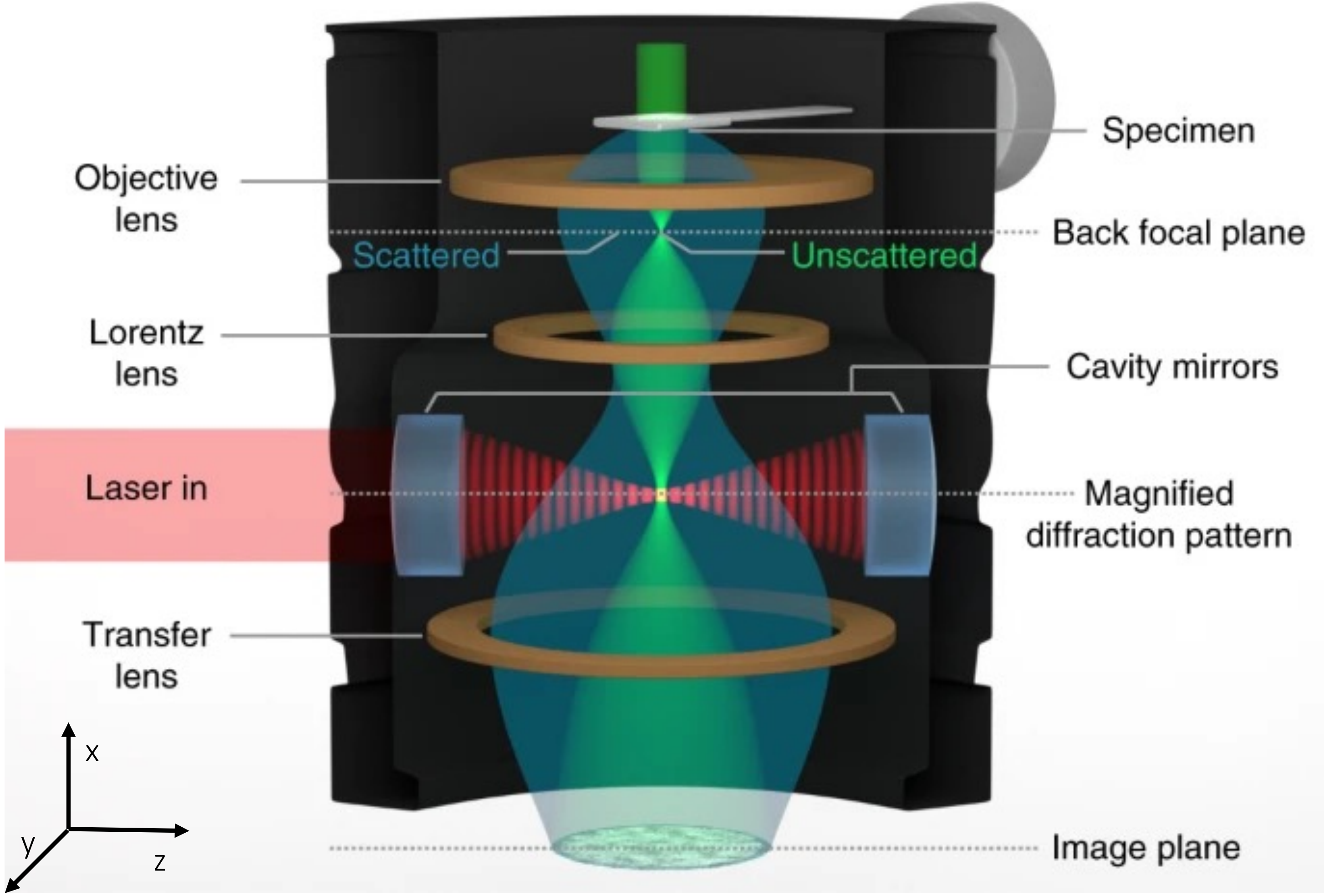


Zernike phase contrast electron micrograph (A) and a conventional electron micrograph (B) of ice-embedded influenza A viruses (*Yamaguchi Y. et al.*)



Phase plate cryo-EM image of nucleosome core particles and DNA *(E.Y.D. Chua et al.)*

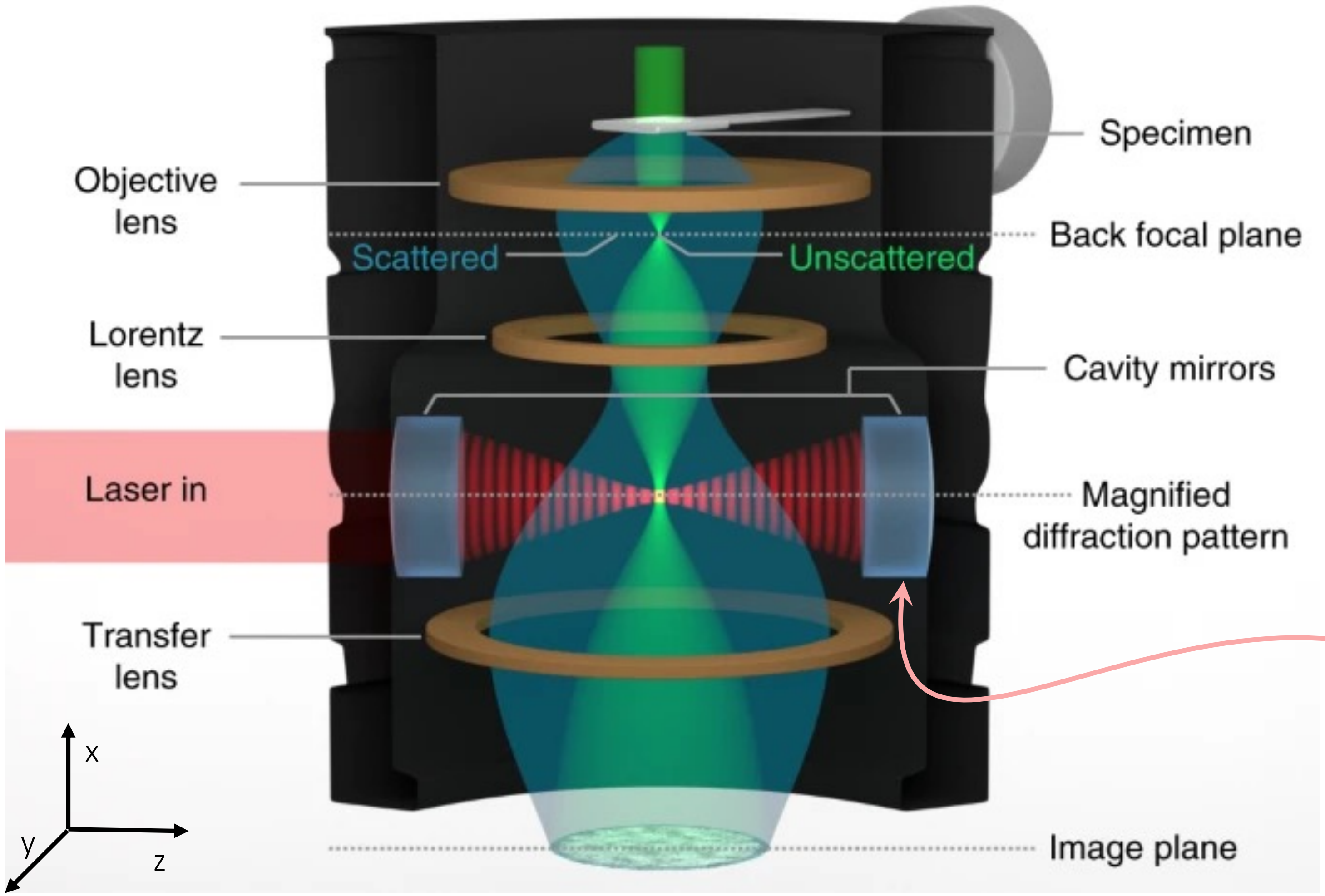
Laser-Based Zernike Phase Plate



(Schwartz *O. et al., 2019*)

Phase Plate for TEM based on the ponderomotive interaction between a tightly focused CW standing laser wave and the continuous zero-order electron beam transmitted by the sample.

Laser-Based Zernike Phase Plate



(Schwartz *O. et al., 2019*)

Short electron-light interaction time in a micron-scale laser focus

Phase Shift

$$\Delta\varphi = \frac{\pi}{2} \Rightarrow P_{laser} \sim 10 \text{ GW/cm}^2$$

Fabry-Pérot Cavity into TEM to magnify optical power

$$P_{laser} = 43 \text{ GW/cm}^2$$

Pulsed Laser Phase Plate

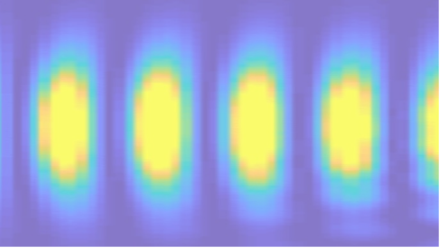
To develop a Pulsed Laser Phase Plate

- Laser beam tightly focused at the interaction point inside the TEM
- Dedicated fs-laser with ad-hoc operational parameters
- Electron pulse compression
- Space-Time synchronization between laser and electron pulses
- Laser and electron beam diagnostics

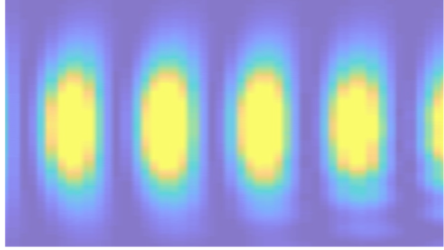
Pulsed Laser Phase Plate

To develop a Pulsed Laser Phase Plate

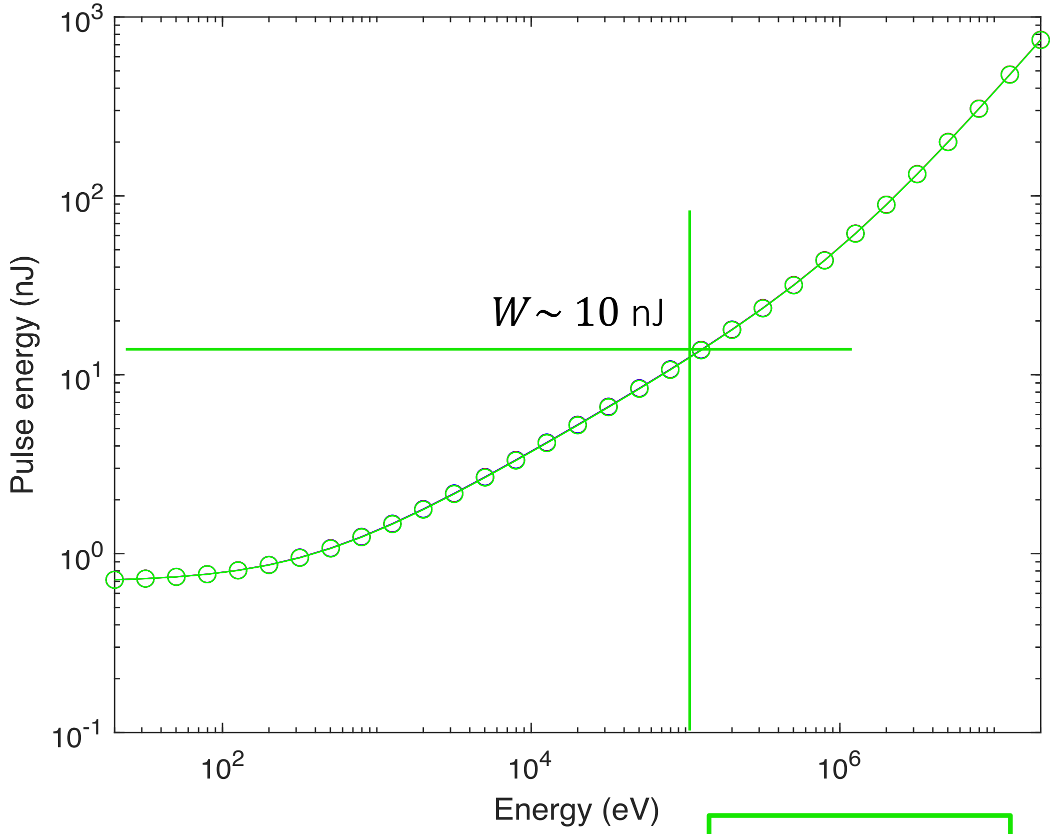
- Laser beam tightly focused at the interaction point inside the TEM
- Dedicated fs-laser with ad-hoc operational parameters
- Electron pulse compression
- Space-Time synchronization between laser and electron pulses
- Laser and electron beam diagnostics



Pulsed-Laser-Based Zernike Phase Plate



$$\Delta\varphi_{max} \sim \frac{e^2}{\hbar\, c\, \epsilon_0\, \gamma\, m\, \omega_0^2} \frac{W}{\pi\, w_0^2} \frac{\tau_t}{\tau} = \frac{\pi}{2}$$



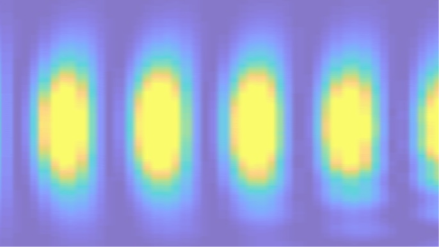
E_k = 200 keV

Experimental Parameters

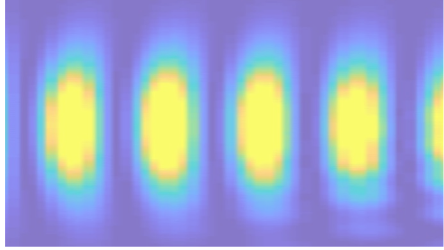
At first glance

- λ = 800 nm
- $w_0 = 2\,\lambda$
- τ = 100 fs ⇨ τ_e = 0.3 $\tau \sim 33$ fs
- $W \sim 10$ nJ/pulse

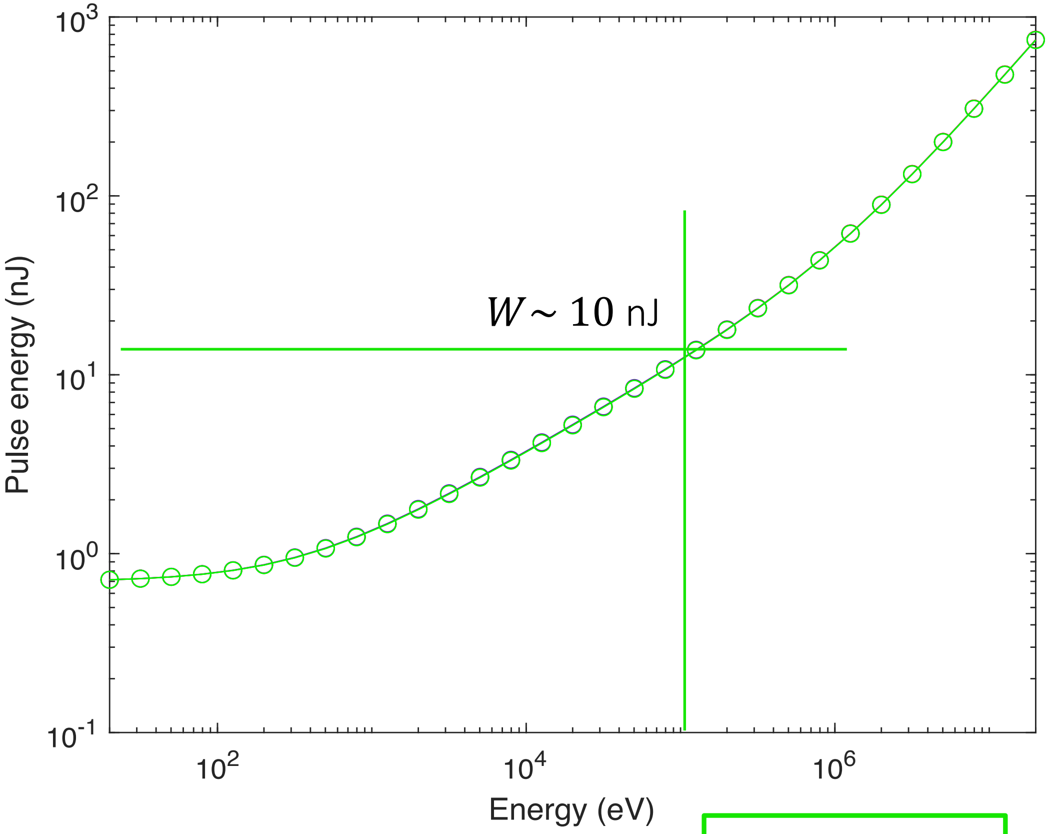
T. van Leeuwen, W. Schaap



Pulsed-Laser-Based Zernike Phase Plate



$$\Delta\varphi_{max} \sim \frac{e^2}{\hbar\, c\, \epsilon_0\, \gamma\, m\, \omega_0^2}\, \frac{W}{\pi\, w_0^2}\, \frac{\tau_t}{\tau} = \frac{\pi}{2}$$



E_k = 200 keV

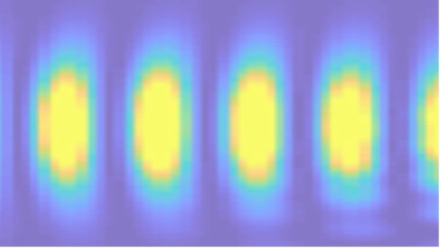
Experimental Parameters

At first glance

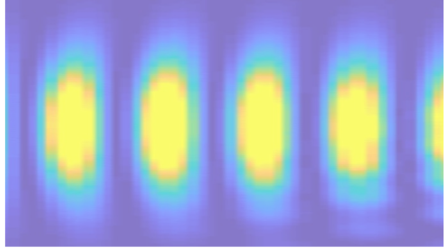
- λ = 800 nm
- $w_0 = 2\,\lambda$
- τ = 100 fs ⇨ τ_e = 0.3 $\tau \sim 33$ fs
- $W \sim 10$ nJ/pulse

Compression & Syncronization < 10 fs

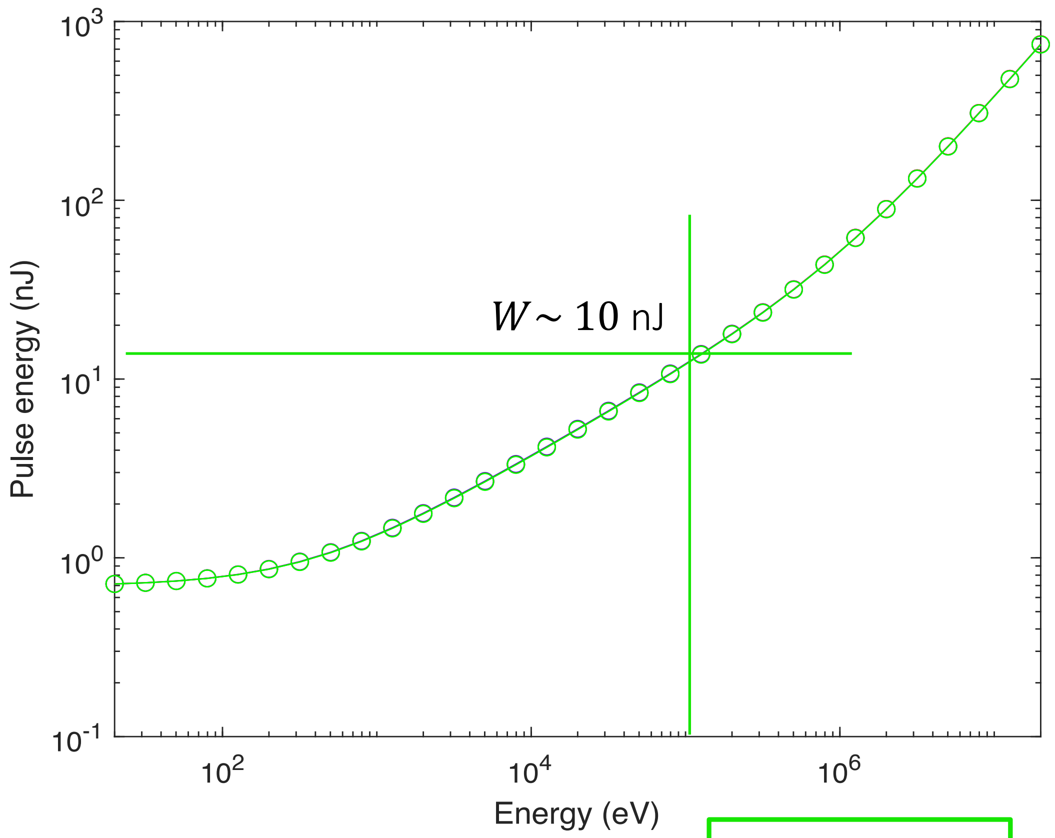
T. van Leeuwen, W. Schaap



Pulsed-Laser-Based Zernike Phase Plate



$$\Delta\varphi_{max} \sim \frac{e^2}{\hbar\, c\, \epsilon_0\, \gamma\, m\, \omega_0^2}\, \frac{W}{\pi\, w_0^2}\, \frac{\tau_t}{\tau} = \frac{\pi}{2}$$



E_k = 200 keV

Experimental Parameters

At first glance

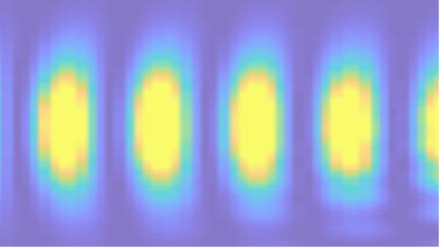
- λ = 800 nm
- $w_0 = 2\,\lambda$
- τ = 100 fs ⇨ τ_e = 0.3 $\tau \sim 33$ fs
- W ~ 10 nJ/pulse

Average Power

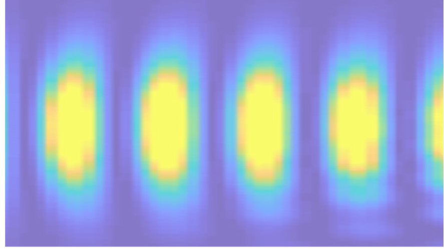
$$P_{avg} = \frac{W(\lambda)\ rep\ rate}{\sqrt{\pi}}$$

Rep Rate (MHz)	P_{avg} (W)
75	0.2
250	0.8
$3 * 10^3$	9.2

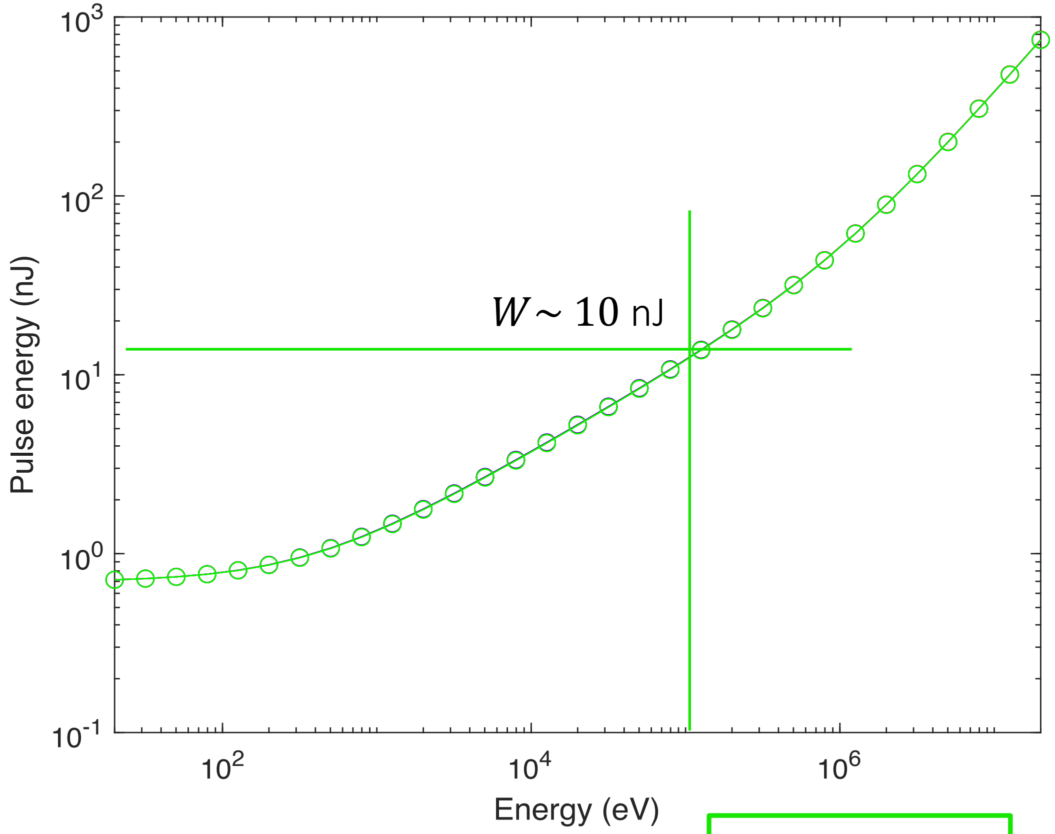
T. van Leeuwen, W. Schaap



Pulsed-Laser-Based Zernike Phase Plate



$$\Delta\varphi_{max} \sim \frac{e^2}{\hbar\, c\, \epsilon_0\, \gamma\, m\, \omega_0^2} \frac{W}{\pi\, w_0^2} \frac{\tau_t}{\tau} = \frac{\pi}{2}$$



E_k = 200 keV

Experimental Parameters

At first glance

- λ = 800 nm
- $w_0 = 2\,\lambda$
- τ = 100 fs ⇨ $\tau_e = 0.3\,\tau \sim 33$ fs
- W ~ 10 nJ/pulse

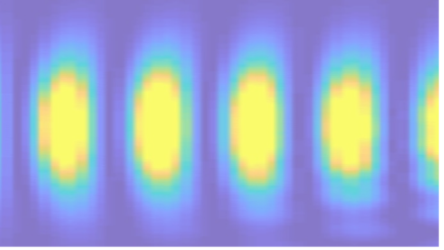
Average Power

$$P_{avg} = \frac{W(\lambda)\ rep\ rate}{\sqrt{\pi}}$$

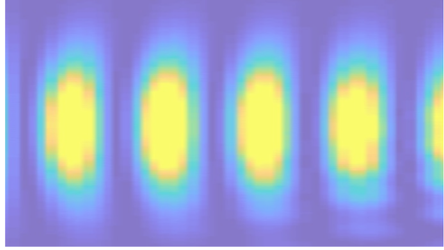
Rep Rate (MHz)	P_{avg} (W)
75	0.2
250	0.8
$3*10^3$	9.2

Jom says: “ Let’s think big!”

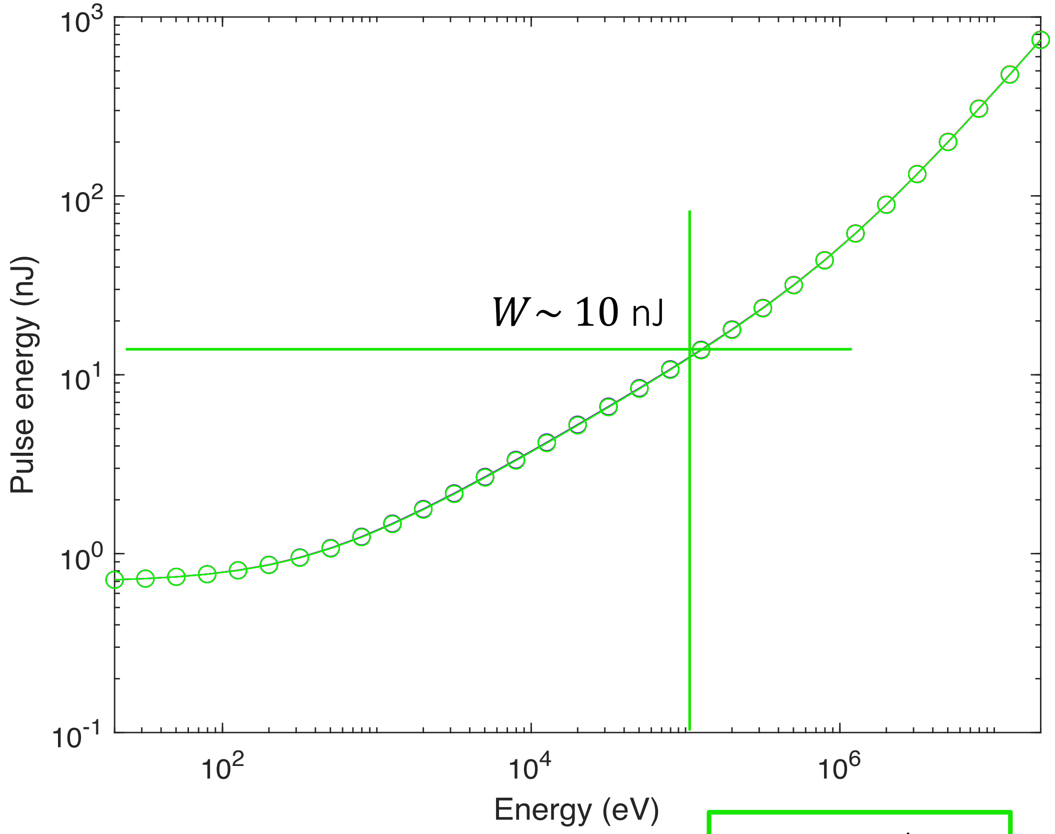
T. van Leeuwen, W. Schaap



Pulsed-Laser-Based Zernike Phase Plate



$$\Delta\varphi_{max} \sim \frac{e^2}{\hbar\, c\, \epsilon_0\, \gamma\, m\, \omega_0^2}\, \frac{W}{\pi\, w_0^2}\, \frac{\tau_t}{\tau} = \frac{\pi}{2}$$



E_k = 200 keV

Experimental Parameters

At first glance

- λ = 800 nm
- $w_0 = 2\,\lambda$
- τ = 100 fs ⇒ τ_e = 33 fs
- W ~ 10 nJ nJ/pulse

- τ = 30 fs ⇒ τ_e ~ 10 fs
- W ~ 5 nJ/pulse

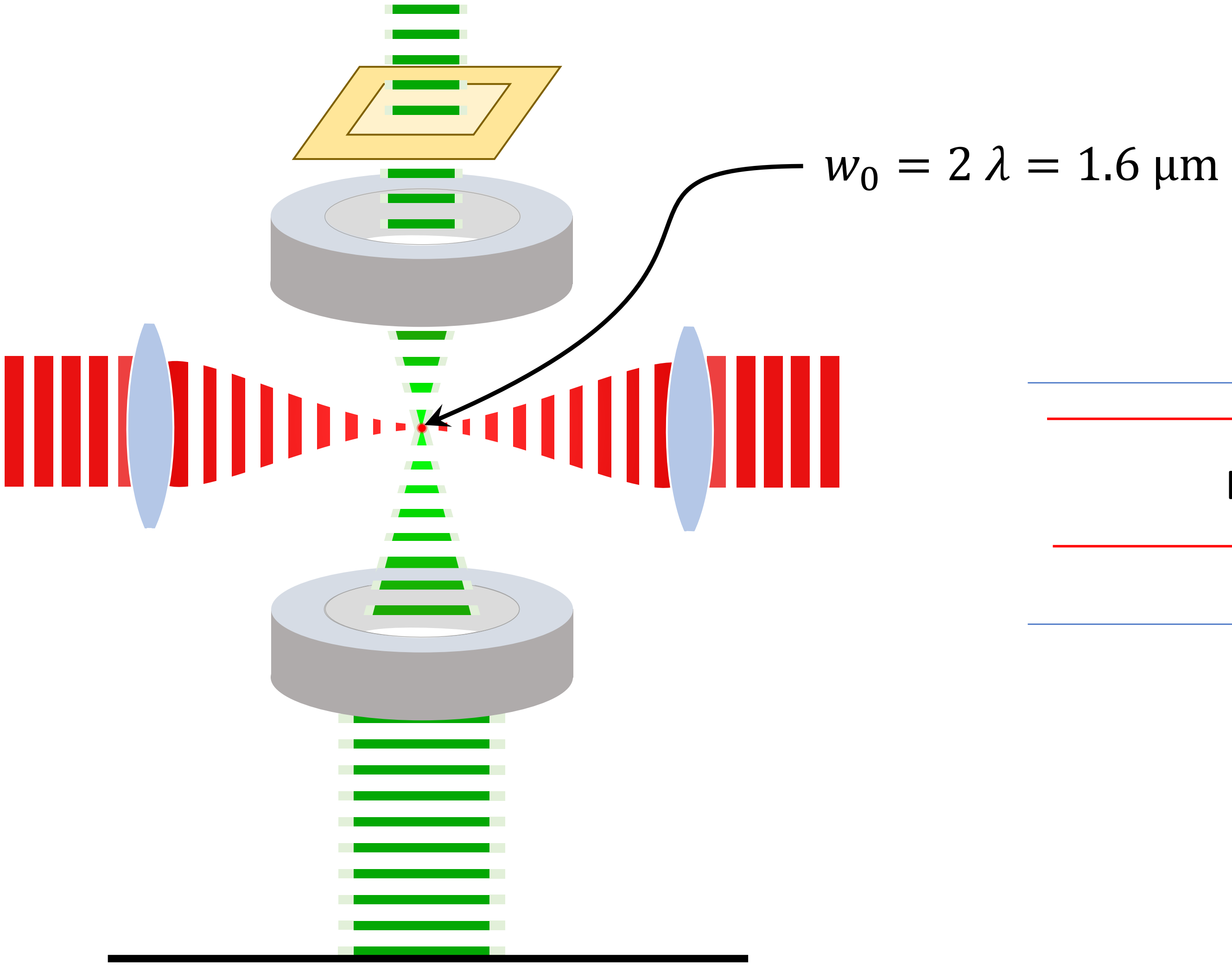
T. van Leeuwen, W. Schaap

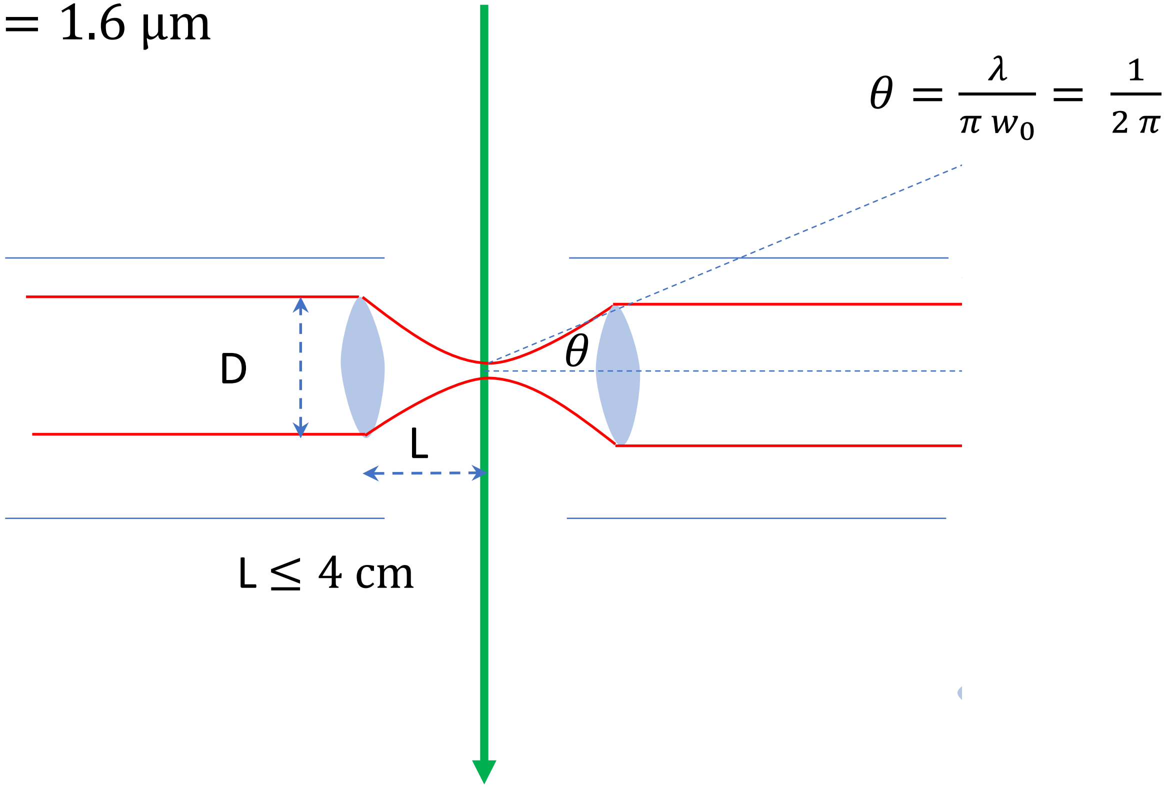
Pulsed Laser Phase Plate

To develop a Pulsed Laser Phase Plate

- Laser beam tightly focused at the interaction point inside the TEM
- Dedicated fs-laser with ad hoc operational parameters
- Electron pulse compression
- Space-Time synchronization between laser and electron pulses
- Laser and electron beam diagnostics

Pulsed-Laser-Based Zernike Phase Plate

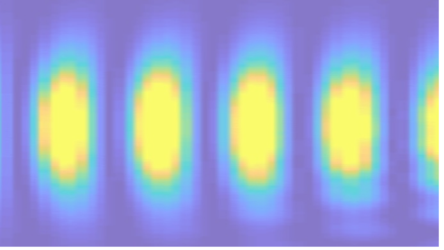




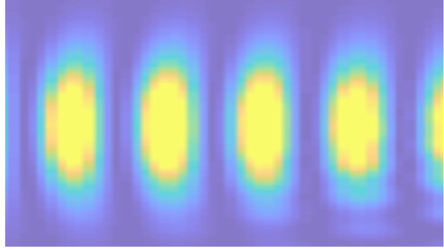
Pulsed Laser Phase Plate

To develop a Pulsed Laser Phase Plate

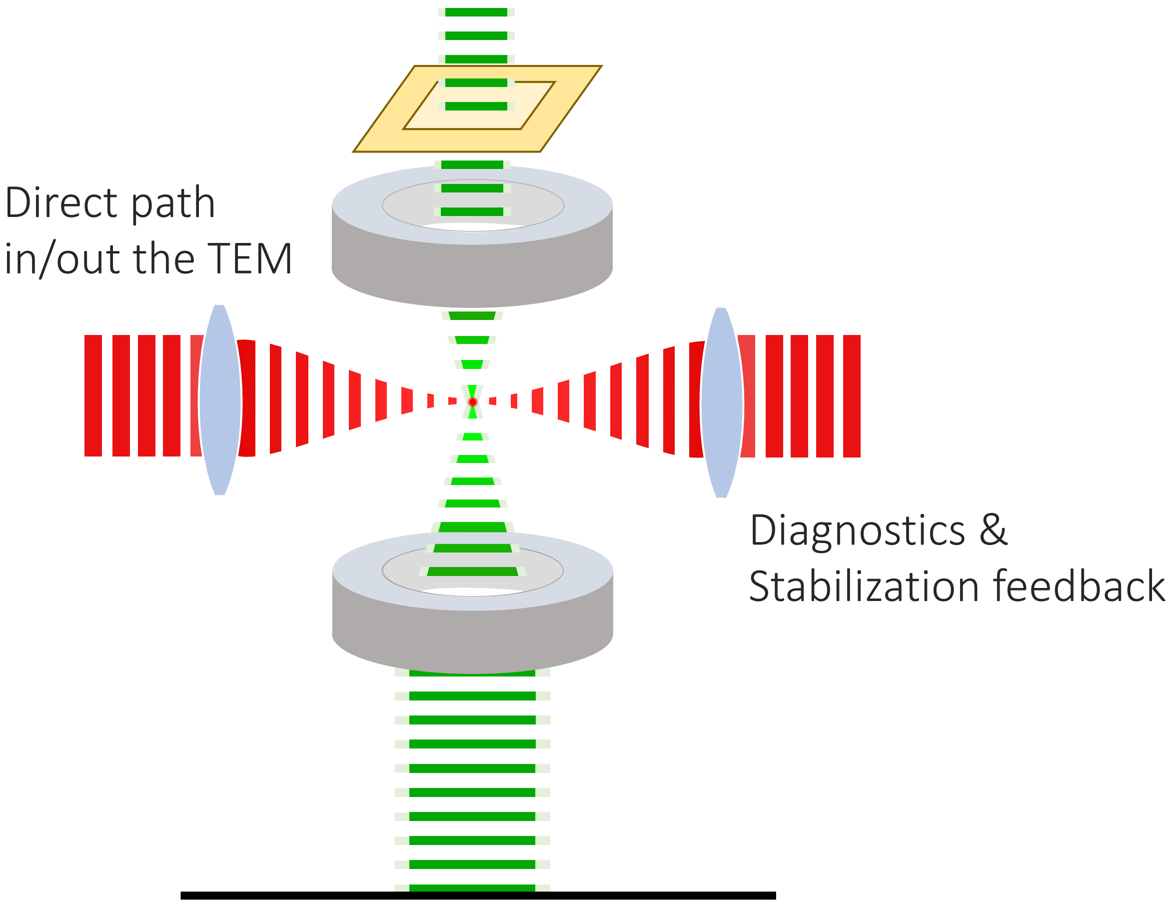
- Laser beam tightly focused at the interaction point inside the TEM
- Dedicated fs-laser with ad hoc operational parameters
- Electron pulse compression
- Space-Time synchronization between laser and electron pulses
- Laser and electron beam diagnostics



Pulsed-Laser-Based Zernike Phase Plate



Spatial Alignment and Diagnostics



Temporal Alignment