

ARC Centre of Excellence for  
COHERENT X-RAY SCIENCE



THE UNIVERSITY OF  
MELBOURNE

## Arbitrarily Shaped High-Coherence Electron and Ion Bunches from Laser-Cooled Atoms

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*With thanks to:*

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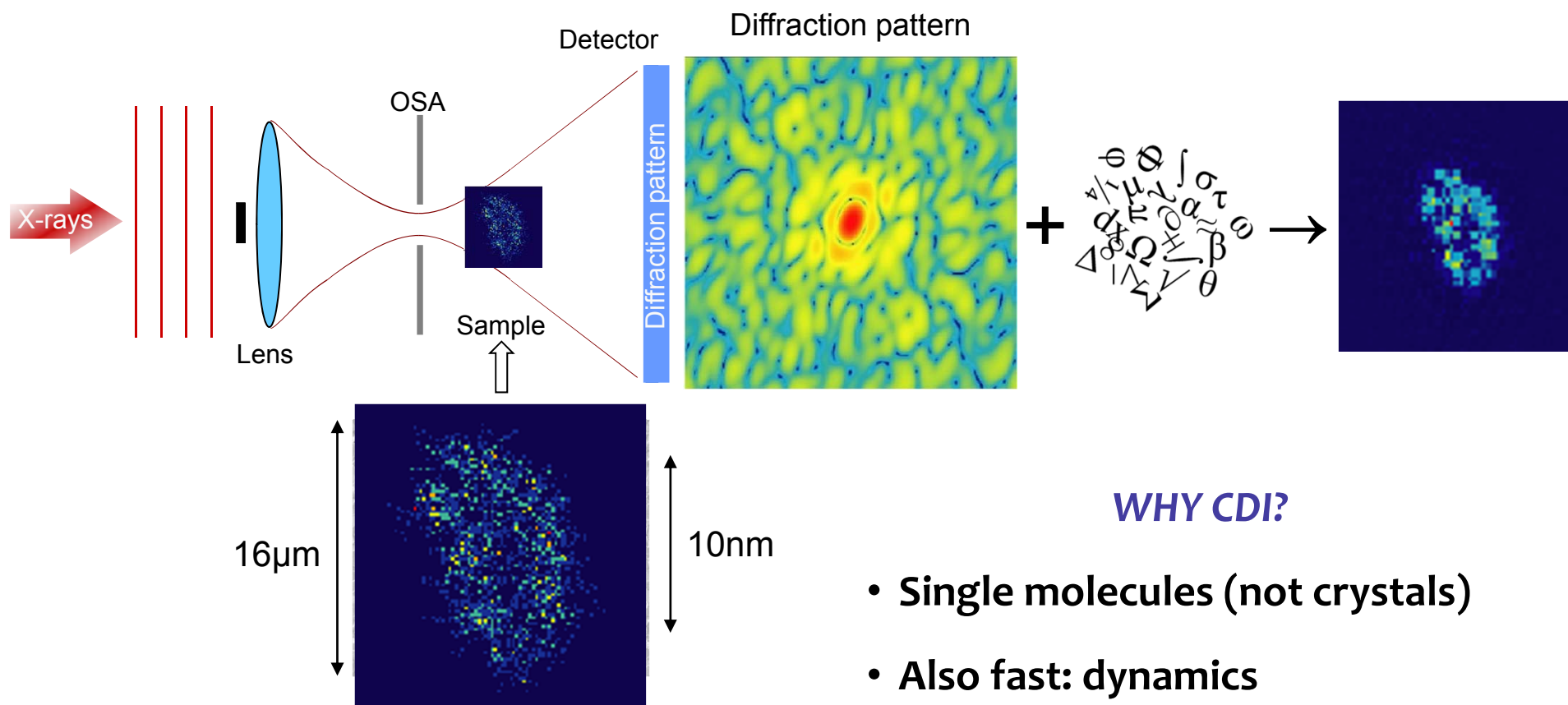


Australian Government  
Australian Research Council

# Imaging small things: diffractive (lensless) imaging

## CDI – Coherent Diffractive Imaging

- Measure diffraction pattern for (**non-crystalline**) object illuminated by x-rays
- Measure intensity, recover phase algorithmically, to determine image of object
- Need a **coherent** source



## Electrons?



- Interaction  $10^4$  to  $10^6$  times stronger than X-rays
- Lower damage for bio-molecules
- Electron optics *extremely* advanced
- Detectors have good resolution, high efficiency
- Electron microscopy is fantastic, but...

### Too slow †

- Molecules destroyed before imaged. No dynamics.
  - Space-charge repulsion limits brightness
  - Only **one electron at a time** in high-res electron microscope
  - Dynamics lost

### CDI with electrons?

- Need high **brightness**, i.e. transverse **coherence** + high **current**

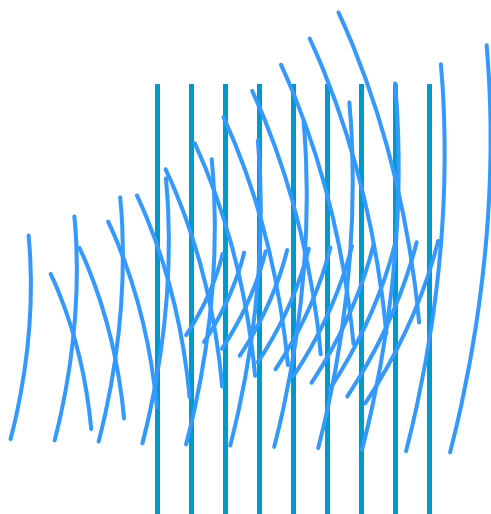
† See Bryan Reed's talk later in conference

**So is CDI possible with electrons?**

# Coherence

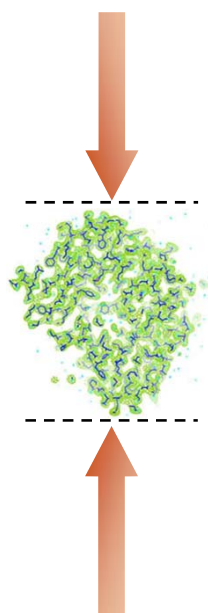
Temporal coherence

$$L = \frac{c}{\Delta f} = \frac{h}{\Delta p}$$

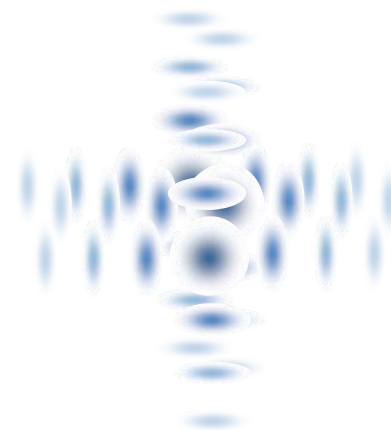
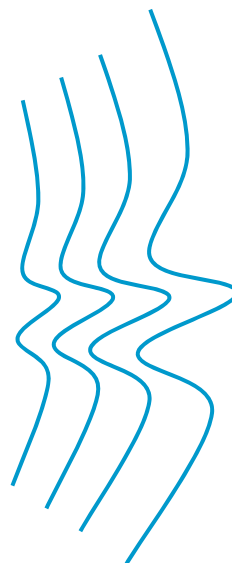


Transverse spatial coherence

$$L_c = \frac{h}{\Delta p_x} = \frac{\lambda}{2\pi \Delta\theta} = \frac{\hbar}{\sqrt{mk_B T}}$$

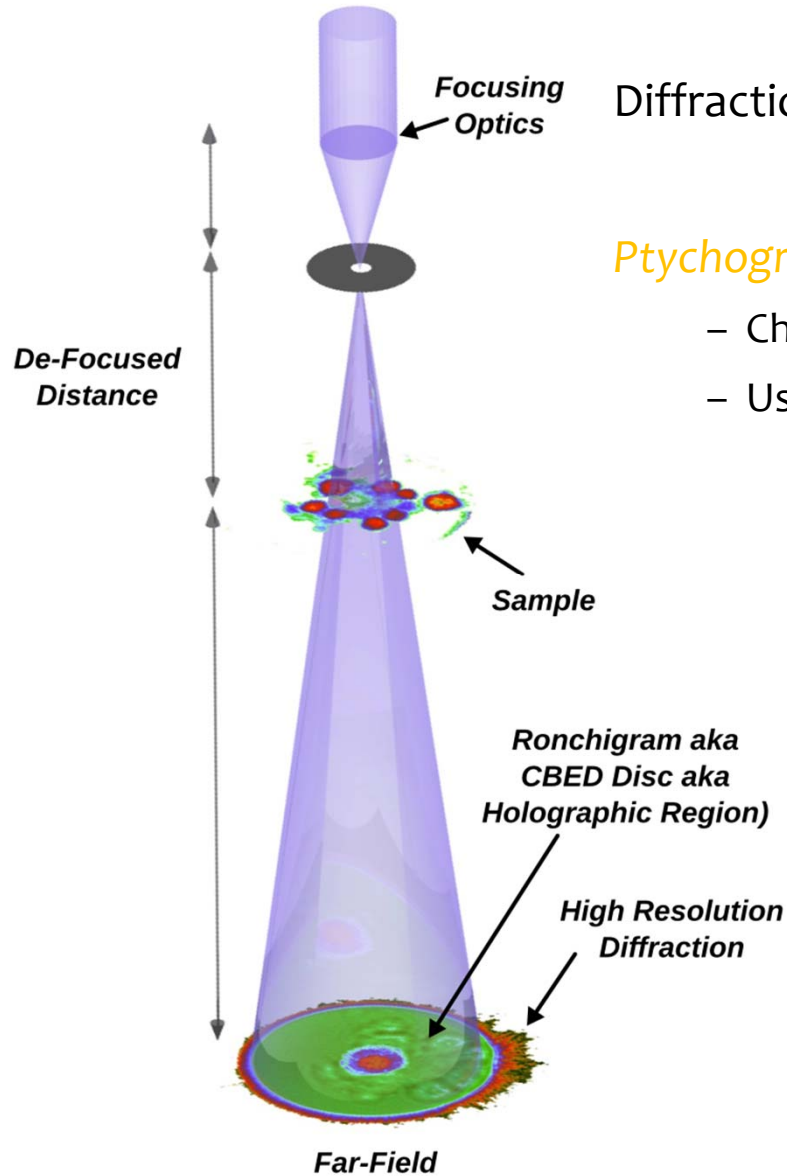


10+ nm



**So is CDI possible with electrons?**

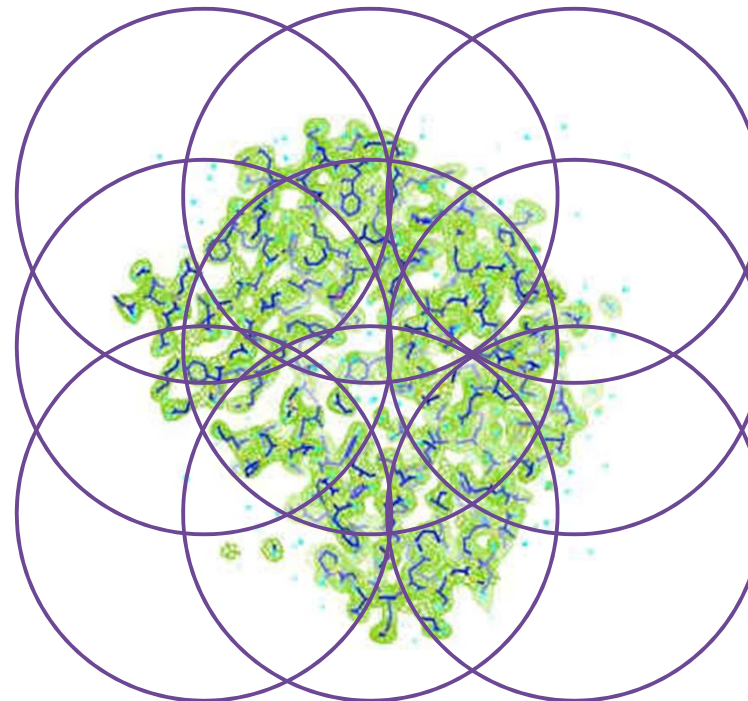
# Electron CDI



Diffraction → intensity only: *phase ambiguity*

## Ptychography

- Change phase, profile or position of illuminating beam
- Use two (or more) diffraction patterns to reconstruct



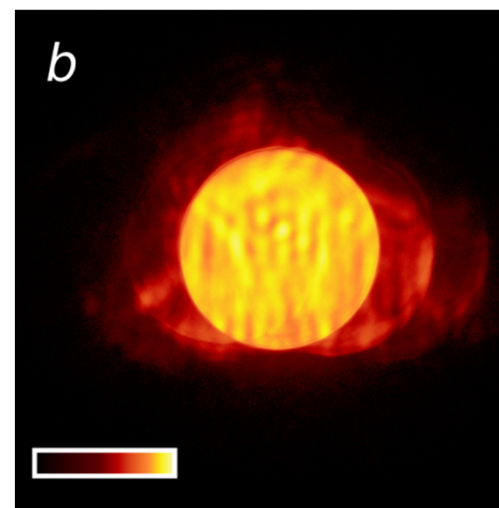
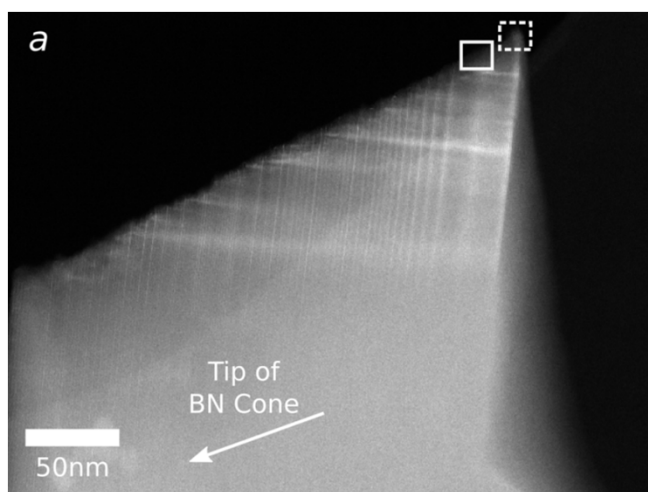
# Atomic resolution ptychographic eCDI

## Boron nitride helical cones

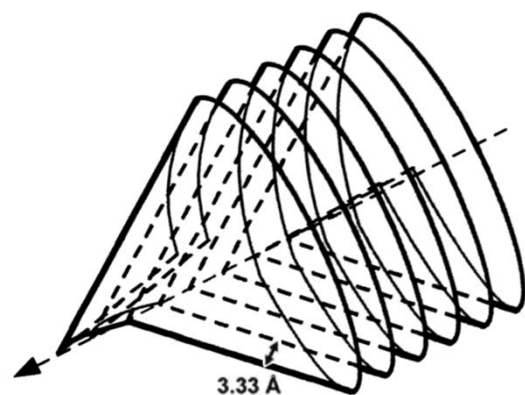
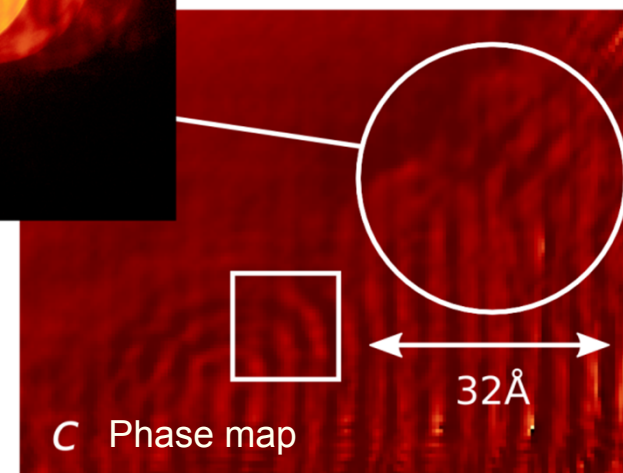
FEI Titan microscope STEM 300keV  $\lambda = 2\text{pm}$ , over-focused

For given STEM probe, peCDI  $\rightarrow$  resolution improvement

$$L_c = 0.15 \text{ nm}$$



Diffraction data  
(log scale)



**CDI works with electrons ...**

**... but very slowly**



# Too slow: coherence not enough

Coherence good, but need *lots* of electrons

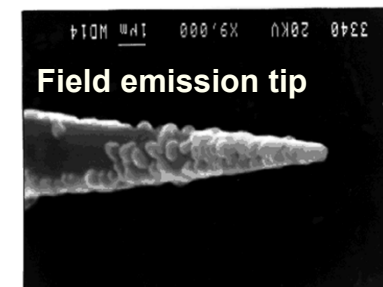
*Brightness*  $B_{\perp} = \frac{I}{4\pi \epsilon_x \epsilon_y} = \frac{mc^2 J}{\pi kT}$  (thermal source)

*Field emission tips:* coherent but not high current

*Photoemission:* high current but not coherent

*Magic:* high current and high coherence

*without space-charge explosion?*

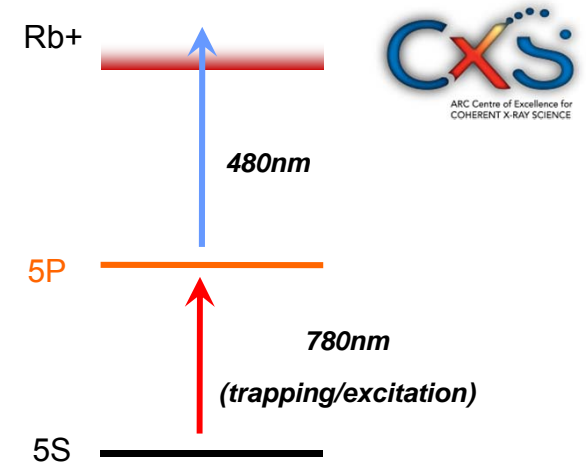
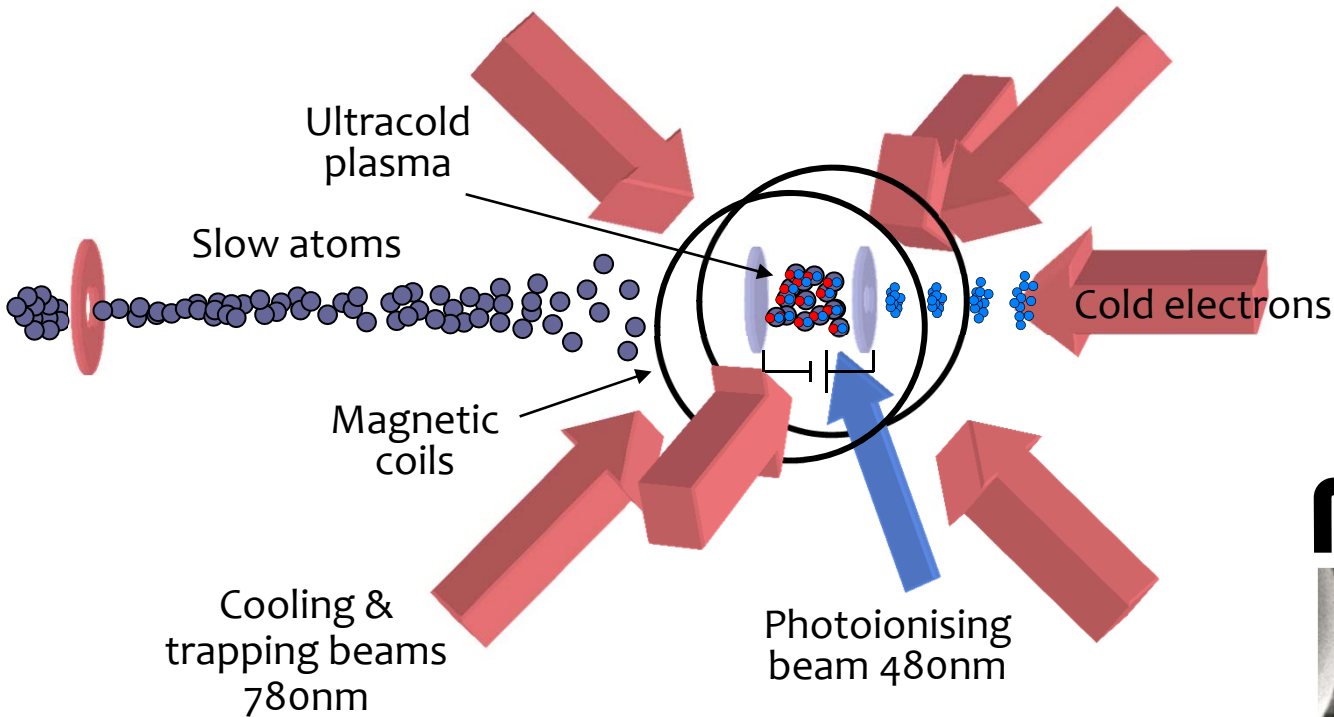


# Electrons from cold atoms

Photoionise cold atoms

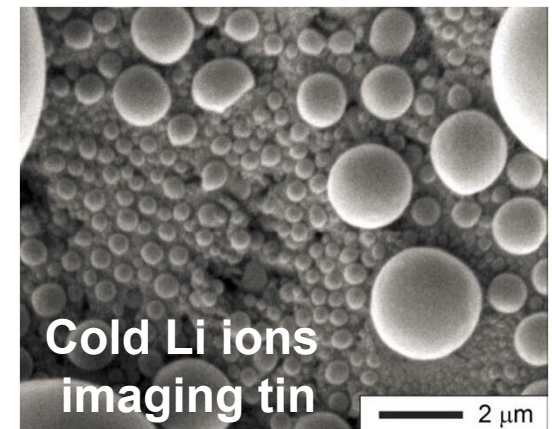
Narrow laser linewidth + Doppler-free

→ small energy uncertainty (low  $T$ )



$$T \approx 10 \text{ K}$$

$$L_c \propto \frac{1}{\sqrt{T}} \sim 8 \text{ nm}$$

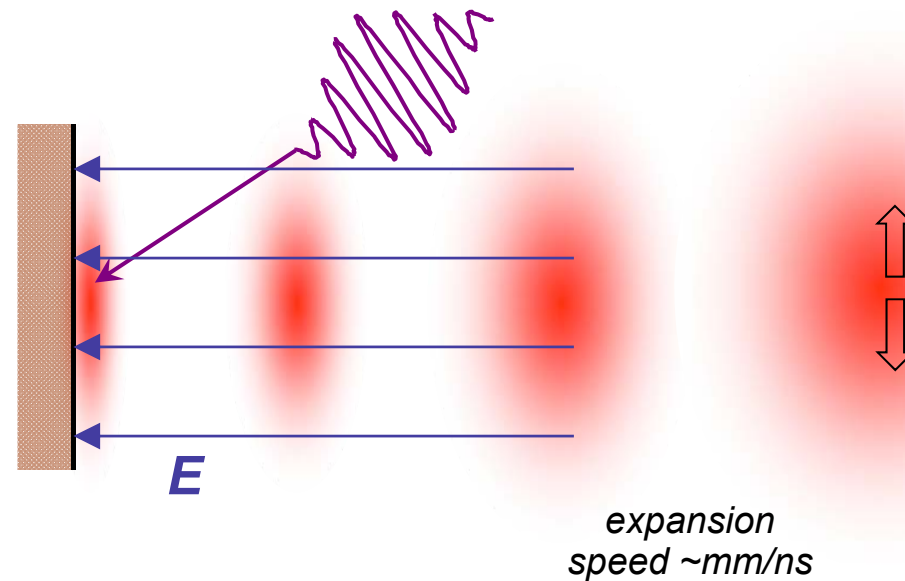
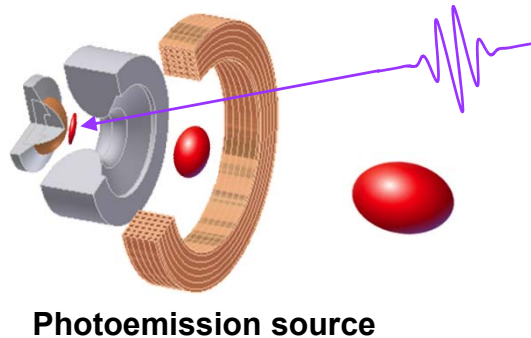
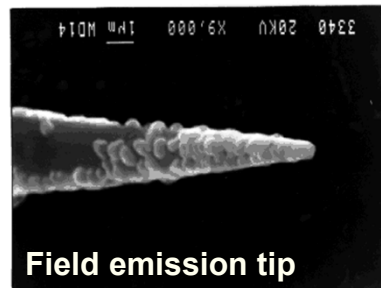


High coherence, but space-charge explosion?



# Coherence ✓ Brightness? Coulomb explosion

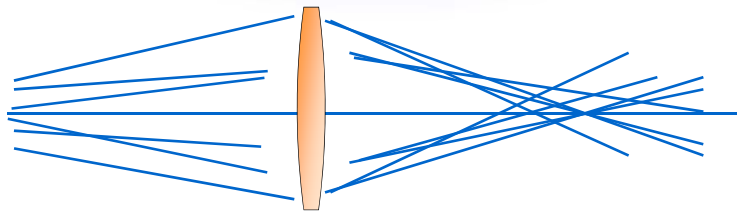
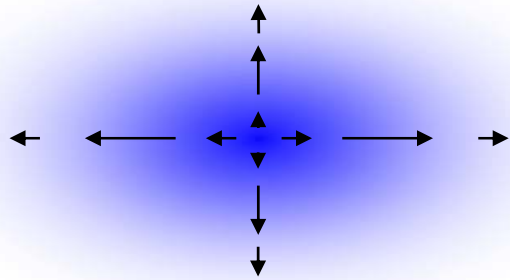
Short pulses, high electron density → Coulomb expansion



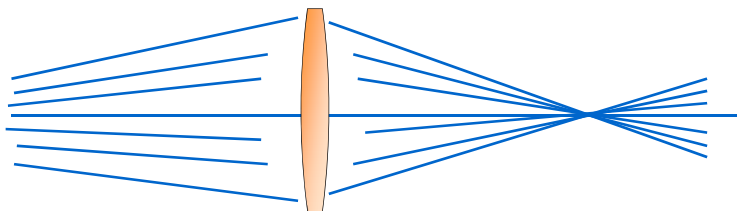
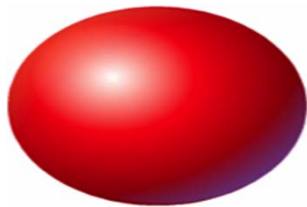
... *irreversible* space-charge explosion

# The problem and a solution

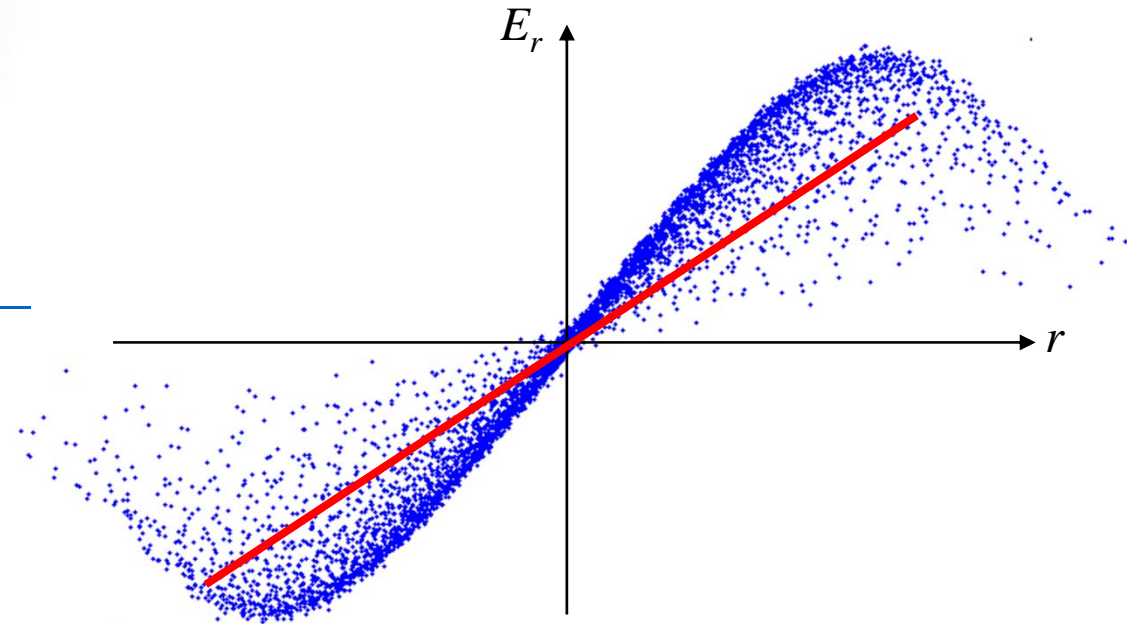
Gaussian bunch



Uniform ellipsoidal bunch



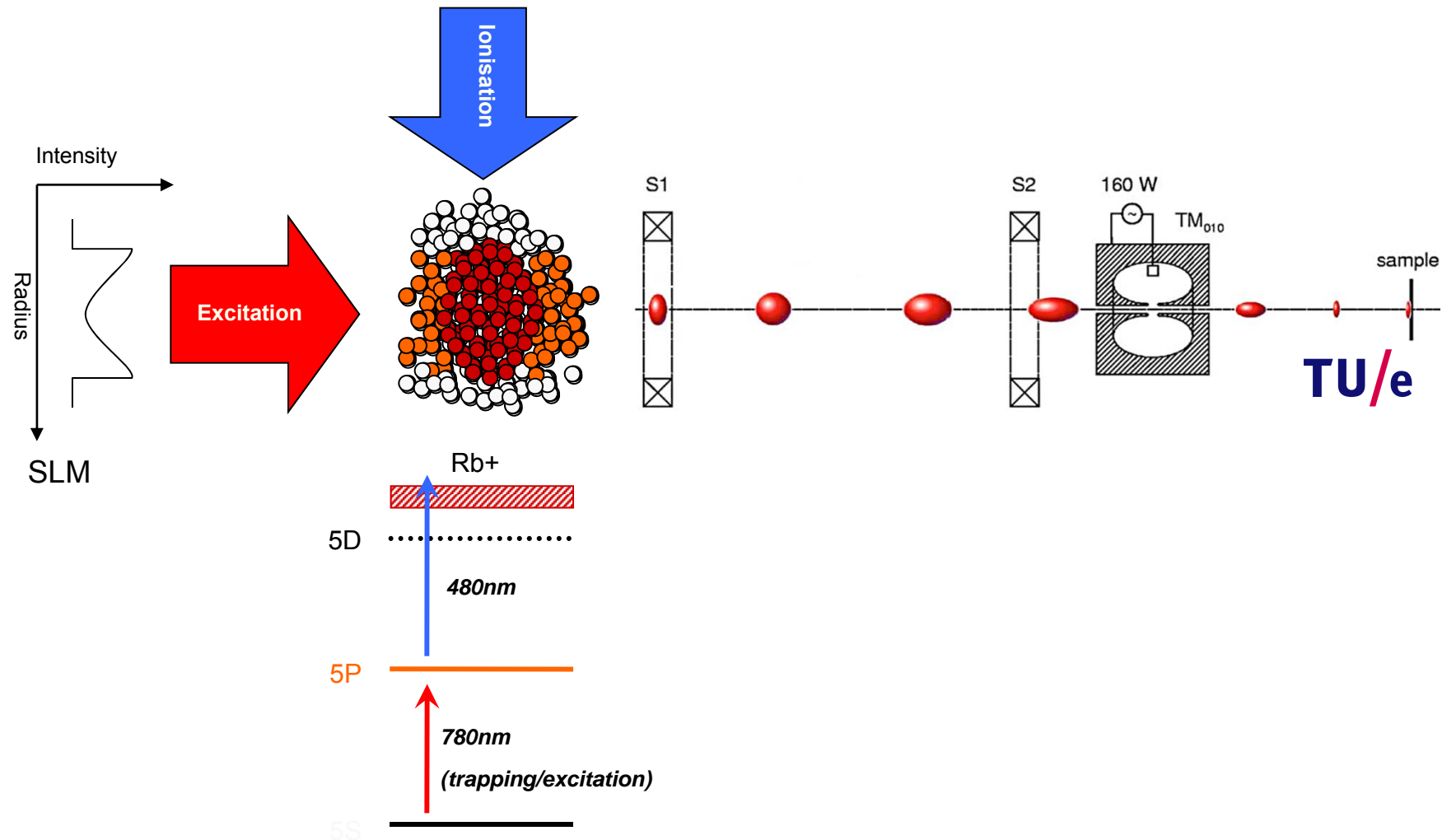
**Nonlinear** space-charge forces;  
**Irreversible** Coulomb expansion



**Linear** space-charge forces;  
**Reversible** Coulomb expansion

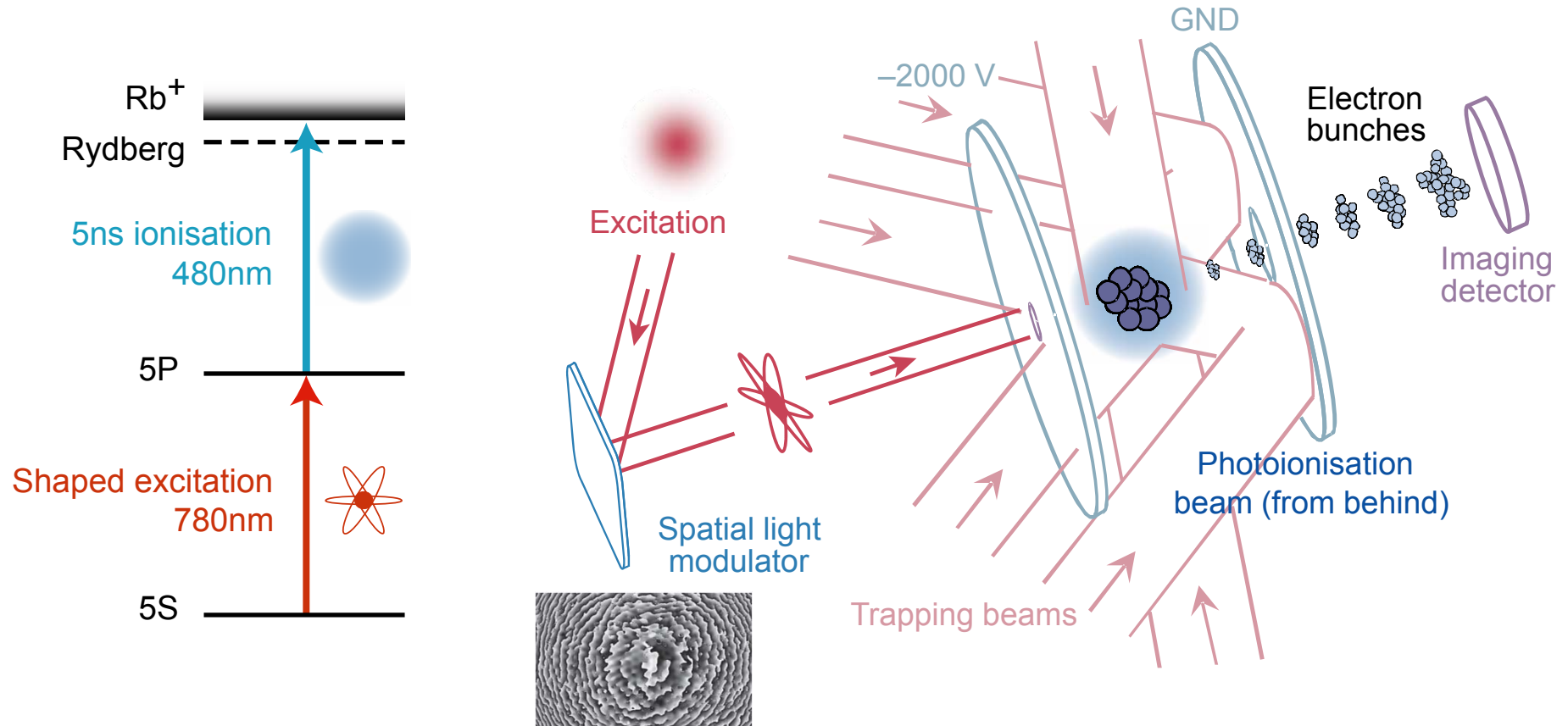
# Bunch shaping to allow explosion reversal

- Space-charge repulsion does **not** intrinsically reduce brightness
  - at least, not irreversibly
- Uniform density + elliptical bunch shape  $\Rightarrow$  can refocus bunch



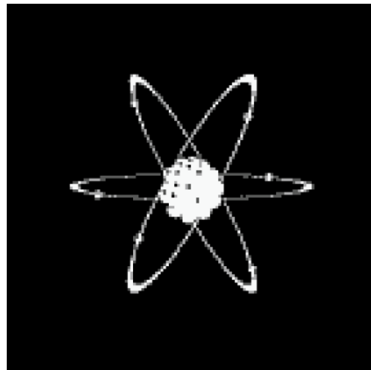
# Bunch shaping

Cold atom electron source allows spatial control

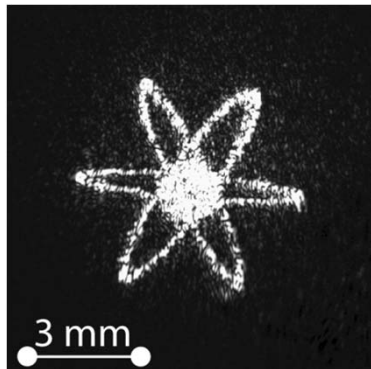


# Arbitrary bunch shaping

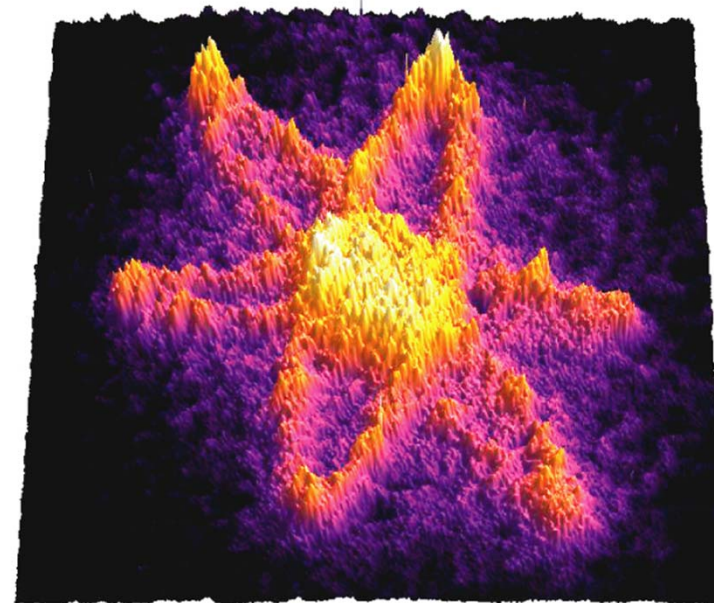
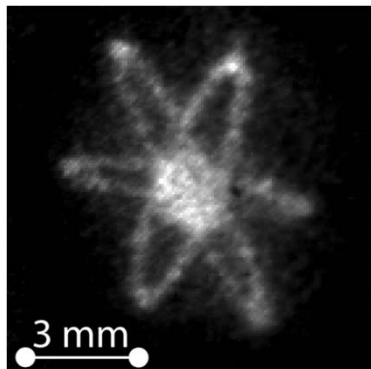
Desired



Excitation  
laser beam  
profile



Electron  
density



Propagation distance 24cm

Time of flight 14ns

Electron mass is *small*  
10K = <1 meV,  $v = 17$  km/s

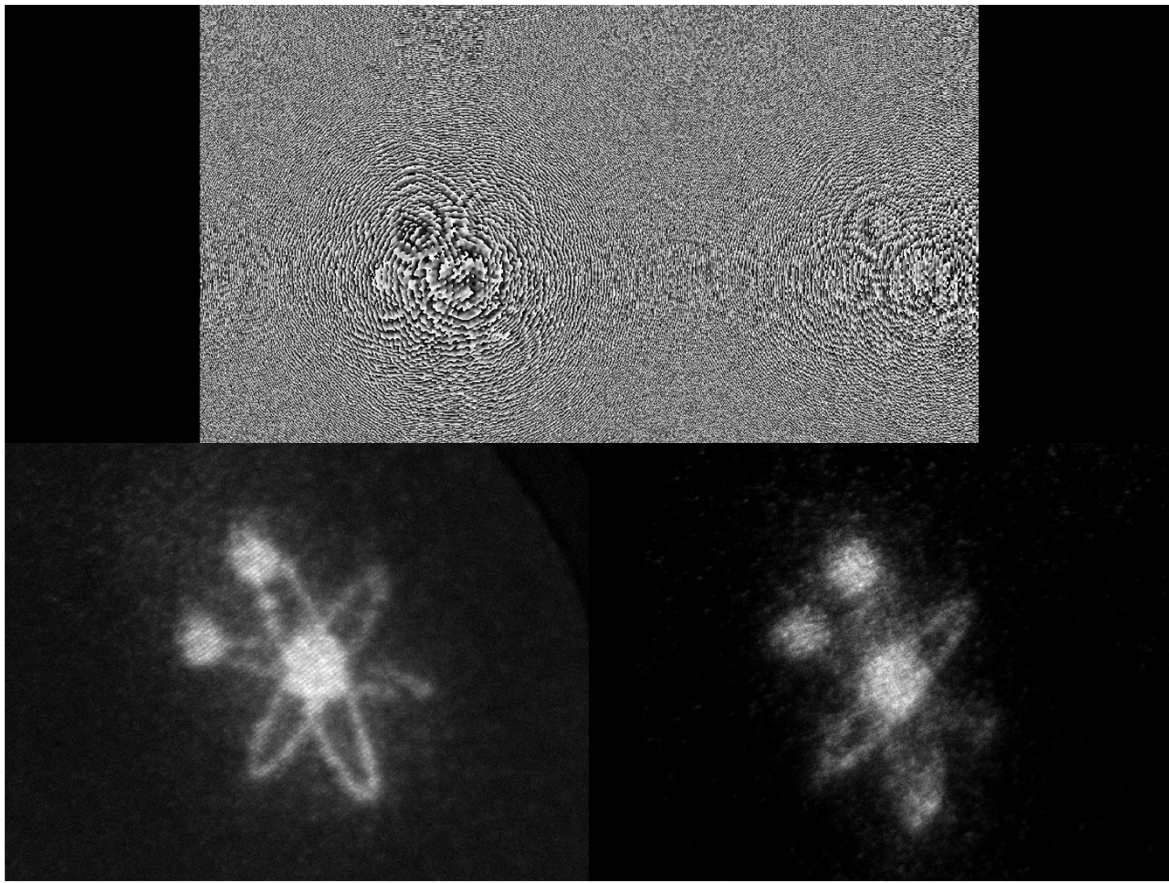


# World's most expensive TV!

Update phase mask at 10Hz: *dynamically shape electron bunch*

Phase mask on SLM

Free propagation – not reimaged



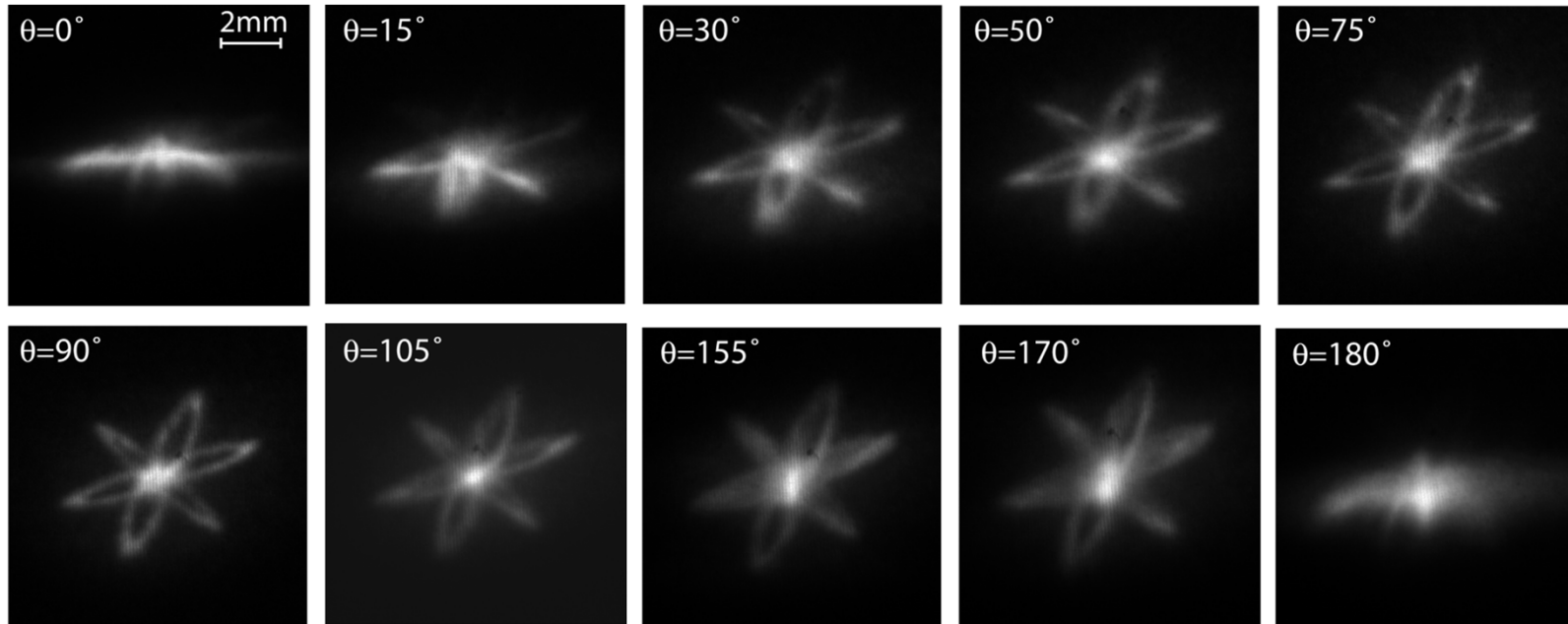
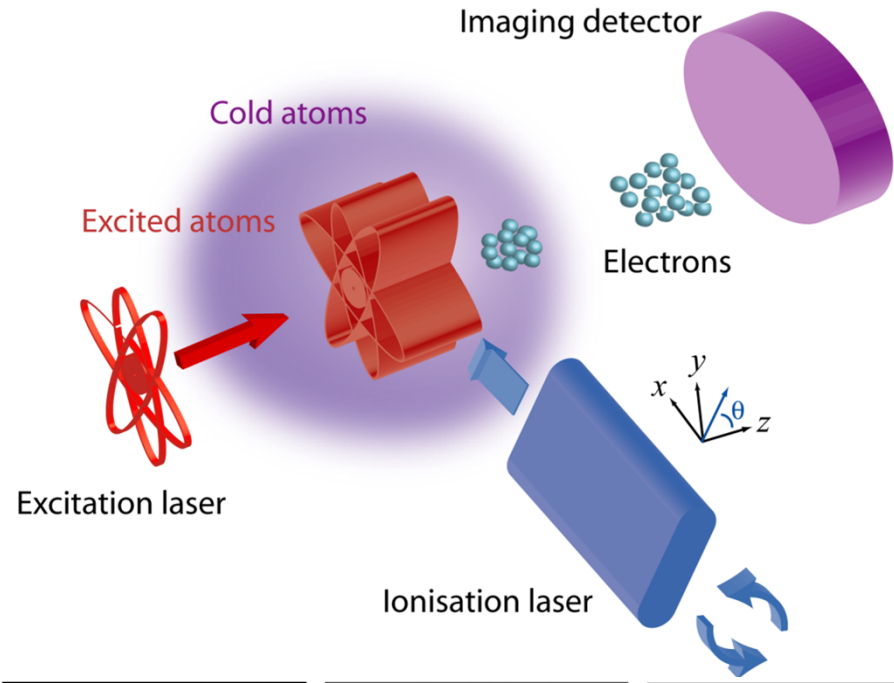
10-shot average

Electrons

Single-shot

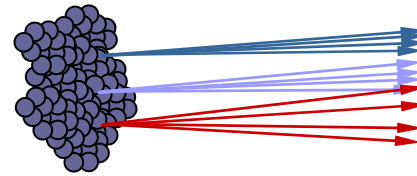
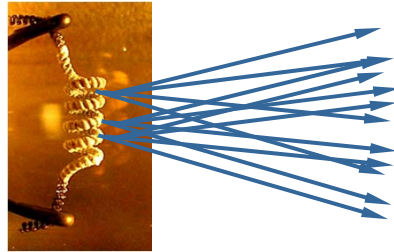


# Longitudinal shaping too



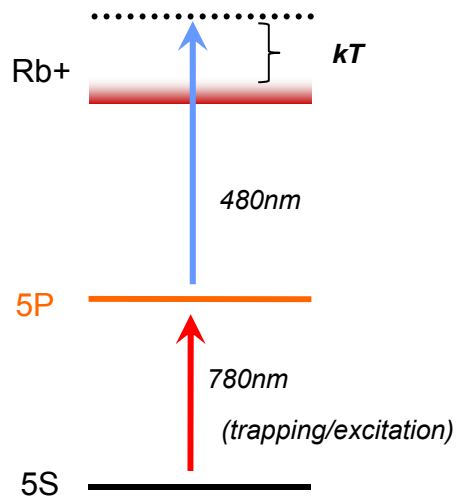
# Temperature, coherence, emittance

## Temperature and spatial coherence

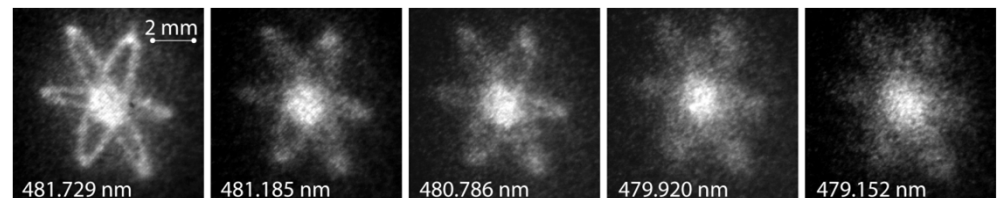
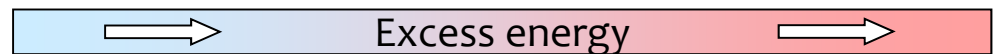


Electron mass is *small*  
 $10\text{K} = <1\text{meV}, v = 17 \text{ km/s}$

- Control excess energy via laser  $\lambda$



$$T \equiv \frac{hc}{k_B \lambda} \quad \Rightarrow \quad T = \frac{hc}{k_B} \left( \frac{1}{\lambda} - \frac{1}{\lambda_{ion}} \right) + T_0$$



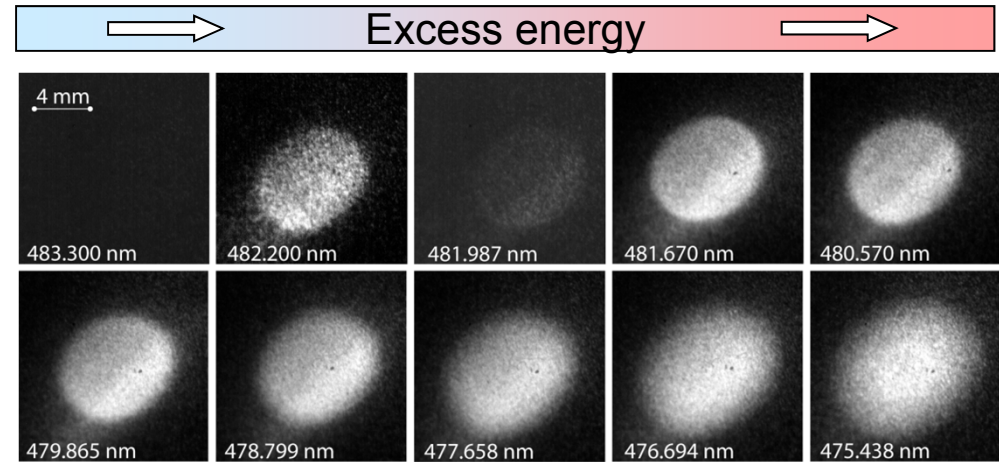
40K      70K      120K      170K (1.6nm)

# Measuring the electron coherence v1

## Quantify

- SLM → uniform density pulses
- Measure *edge resolution*

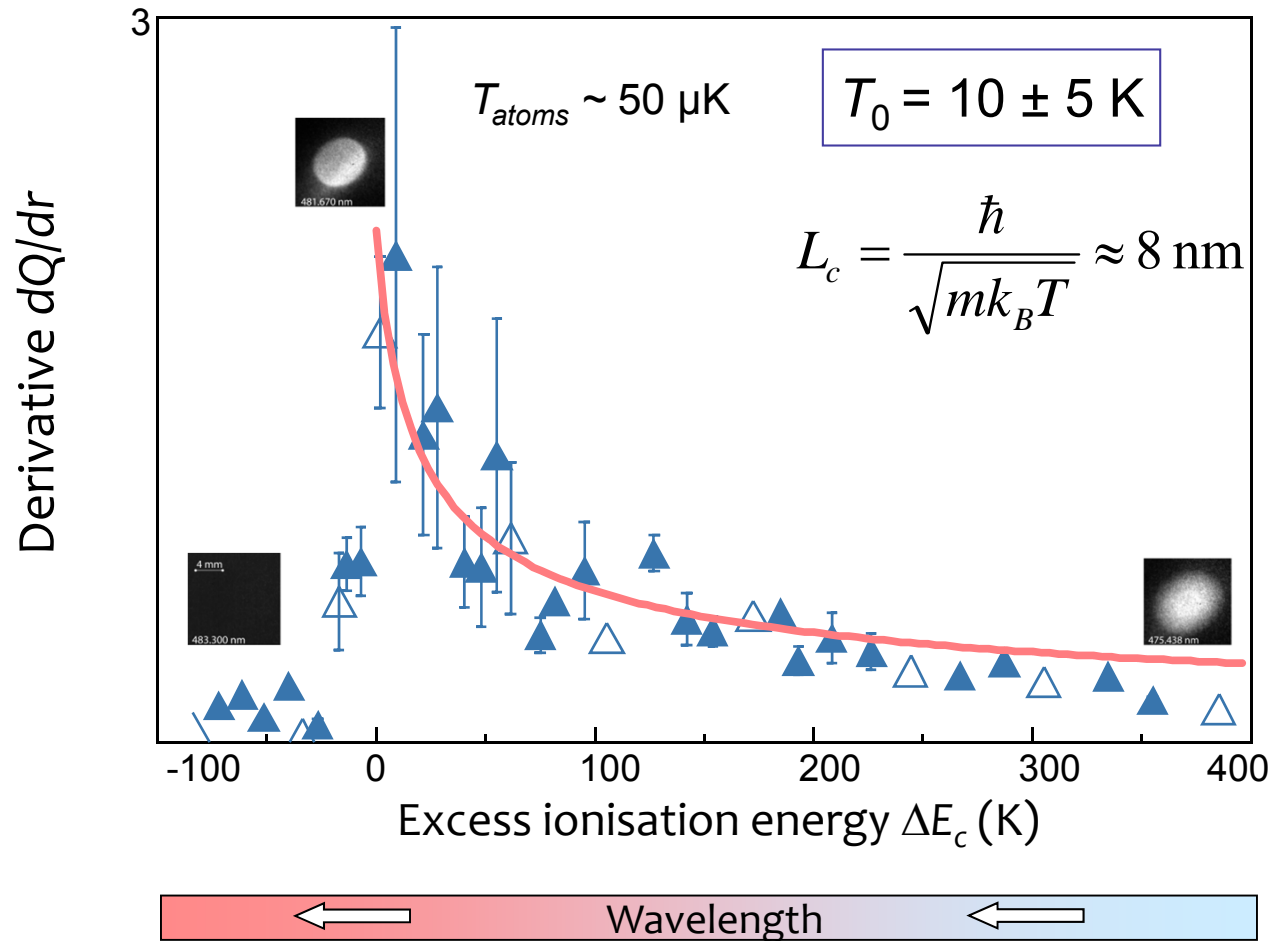
$$\frac{dQ_e}{dr} \rightarrow \text{coherence } L_c \text{ and } T$$



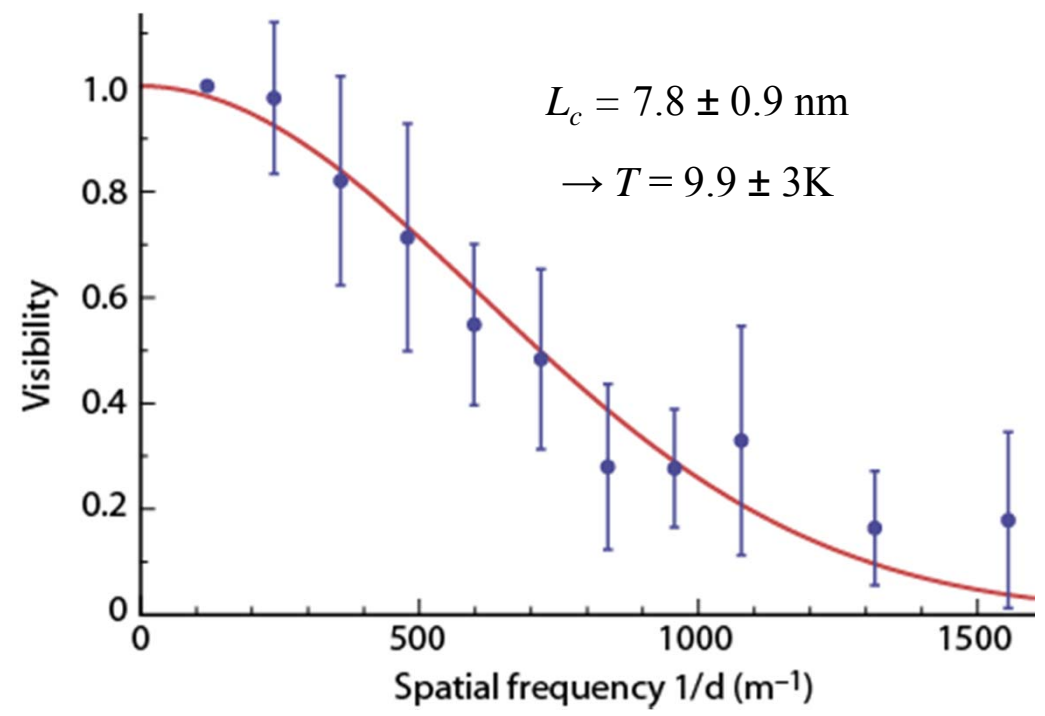
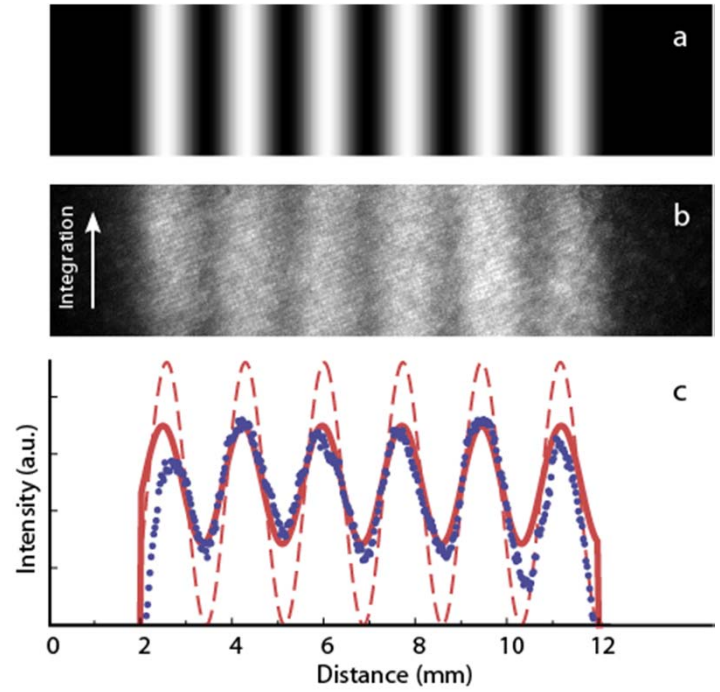
- Edge gradients related to
  - Accelerator geometry (distances  $d_1, d_2$ )
  - Excess photon energy
  - Minimum temperature  $T_0$

$$\left. \begin{array}{l} \text{– Accelerator geometry (distances } d_1, d_2) \\ \text{– Excess photon energy} \\ \text{– Minimum temperature } T_0 \end{array} \right\} \frac{dQ_e}{dr} \propto e \frac{d_1}{2d_1 + d_2} \frac{1}{\sqrt{d_1(k_B T_0 + \Delta E)}}$$

# Measuring the electron coherence v1



# Measuring the electron coherence $v_2$



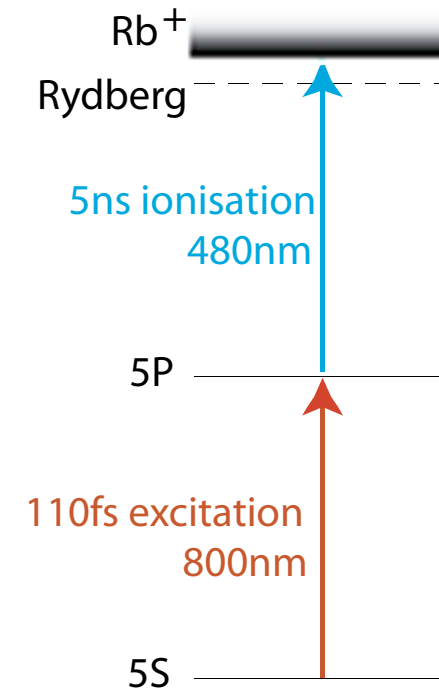
$$\mathcal{V}_{pc} = \exp\left[-\frac{2\pi^2\sigma_b^2}{d^2}\right] = \exp\left[-\frac{1/d^2}{m^2 l_c^2 / h^2 t^2}\right]$$

# Ultrafast

## Ultrafast

- Slow photoionisation (5ns) + fast excitation (110fs)
- Detector-limited sub-ns pulses
- Geometry, extraction field: predict 150ps

**Unpublished data was here**

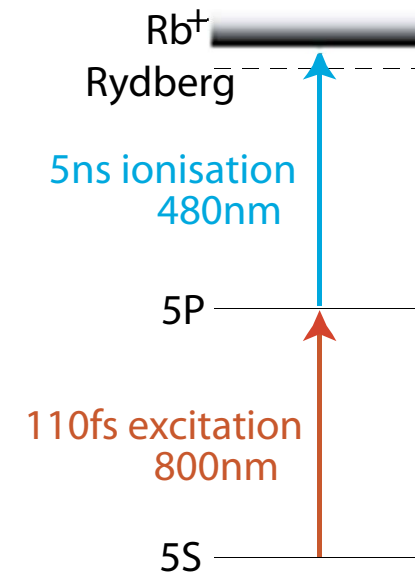


- High bandwidth (20nm): is temperature affected?



# Multiphoton and two-colour processes

- Excitation
  - Coherent: One fs photon + one 5ns photon (two-colour)
  - REMPI: Multiple fs photons
- Coherent → cold
- REMPI → hot



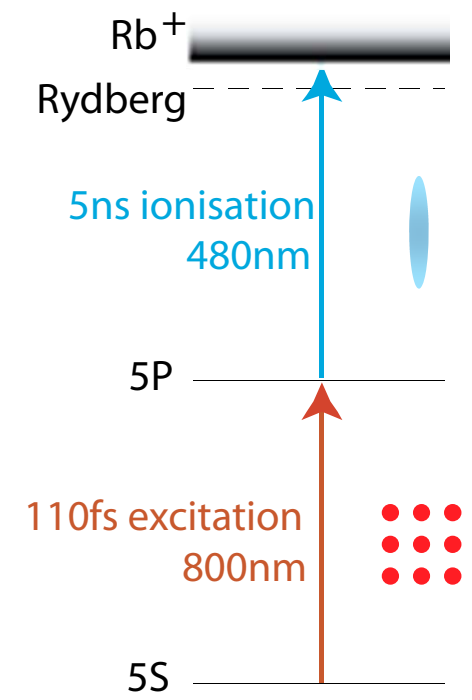
**Unpublished data was here**

# Ultrafast pepperpots



## Emittance

- Virtual pepperpot: holey mask → beamlets
- Divergence of beamlet → emittance

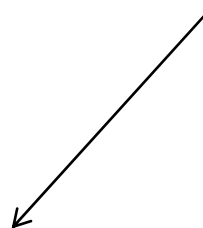


Unpublished data was here

## Short cold bunches

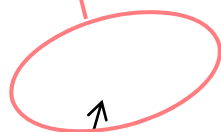
- Emittance increases, but lower than expected from fs bandwidth
- Excess energy along propagation axis

Bandwidth of fs laser  
contributes to energy spread



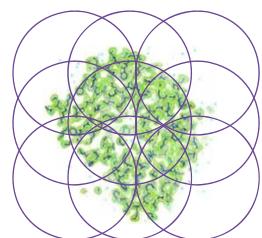
**Unpublished data was here**

**Fast and cold**



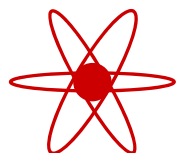
Excitation to Rydberg state  
then field-ionised (Bordas saddle)

# Main points



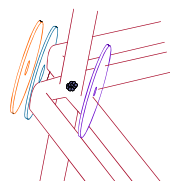
1. *Electron coherent diffractive imaging*

→ *atomic resolution with field-emission source*



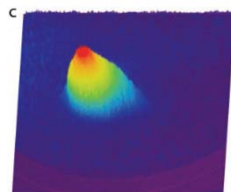
2. *Cold atoms*

→ *cold electrons, high transverse coherence*



3. *Unique capability: 3D bunch shaping*

→ *reversal of Coulomb explosion?*



4. *Ultrafast with high coherence*