Electron Beam Physics and the Limits of Instrument Performance

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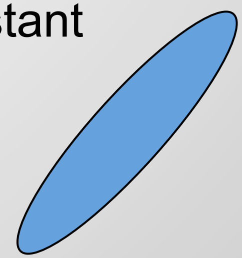
Time-resolved electron probes divide roughly into three categories

	Stroboscopic	Pure Diffraction	Single Shot Imaging
Example	UFTEM (4-D UEM)	UED (keV-MeV)	DTEM
Electrons/Pulse	~1-10^3	~10^3-10^6	~10^7-10^{10}
Pulses/Acquisition	10^5+	usually 1	1
Can do Imaging?	Yes (multi-shot)	No	Yes (single-shot)
Time Resolution	Sub-ps	Sub-ps to few ps	5-1000 ns
Spatial Resolution	Å	Many µm	few nm
Energy Spread	~1 eV	~100 eV	~10 eV
Coherent Fluence	<<1	<<1	~1 or more
Spectroscopy	Yes	Not demonstrated	Not demonstrated
Main Strengths	Spatiotemporal resolution	Compatible with pulse compression etc.	Unique, irreversible events in real space. Movie mode.
Main Weakness	Extremely repeatable processes only	Imaging is difficult. Large minimum spot size.	Resolution

Key concept: Phase space



Phase Space:
A 6D space (x, y, z, p_x, p_y, p_z) that defines the state of an electron at any instant

N electrons become a cloud of *N* points in this space
    Typically ~a 6D ellipsoid

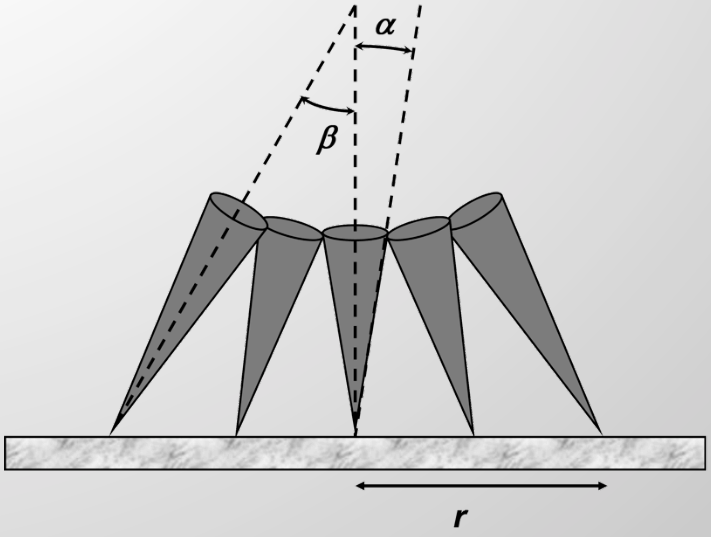
Define a 6-dimensional density $\rho(x, y, z, p_x, p_y, p_z)$.

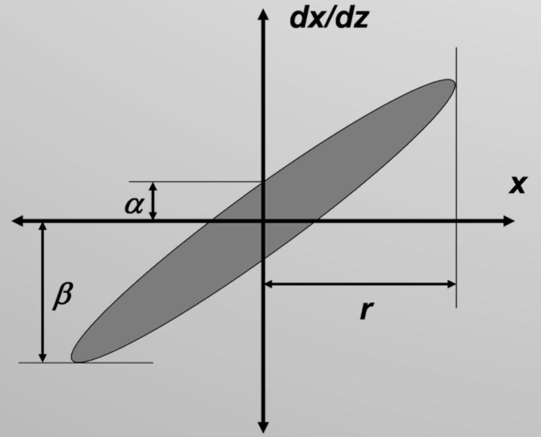
Liouville's Theorem: If we follow some electron's path over time, then the density ρ in its neighborhood is constant

Implies important conservation laws:
- **(Normalized) Brightness**
- **(Normalized) Emittance**
- **Coherent Fluence**

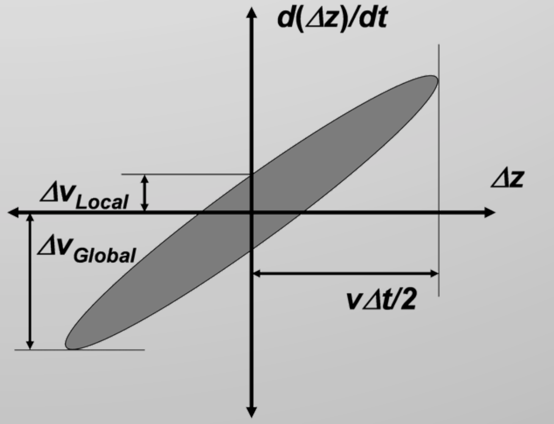
Brightness is related to the transverse phase space profile

- Relevant angle is α, the local convergence
- α is inversely related to spatial coherence, which impacts spatial resolution
- Emittance $\varepsilon \sim r\alpha \sim$ area of phase space ellipse
- Brightness ~ current/ε^2





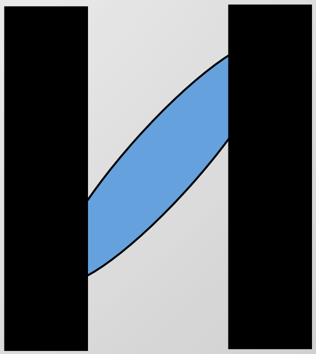
Transverse Phase Space



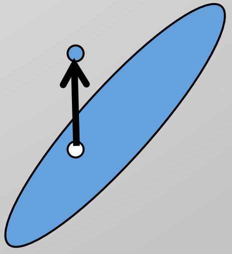
Longitudinal Phase Space

The single-particle Liouville theorem has important exceptions

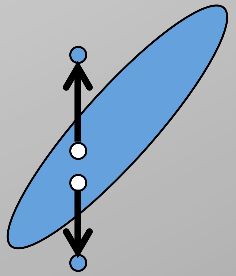
Exception 1: Apertures



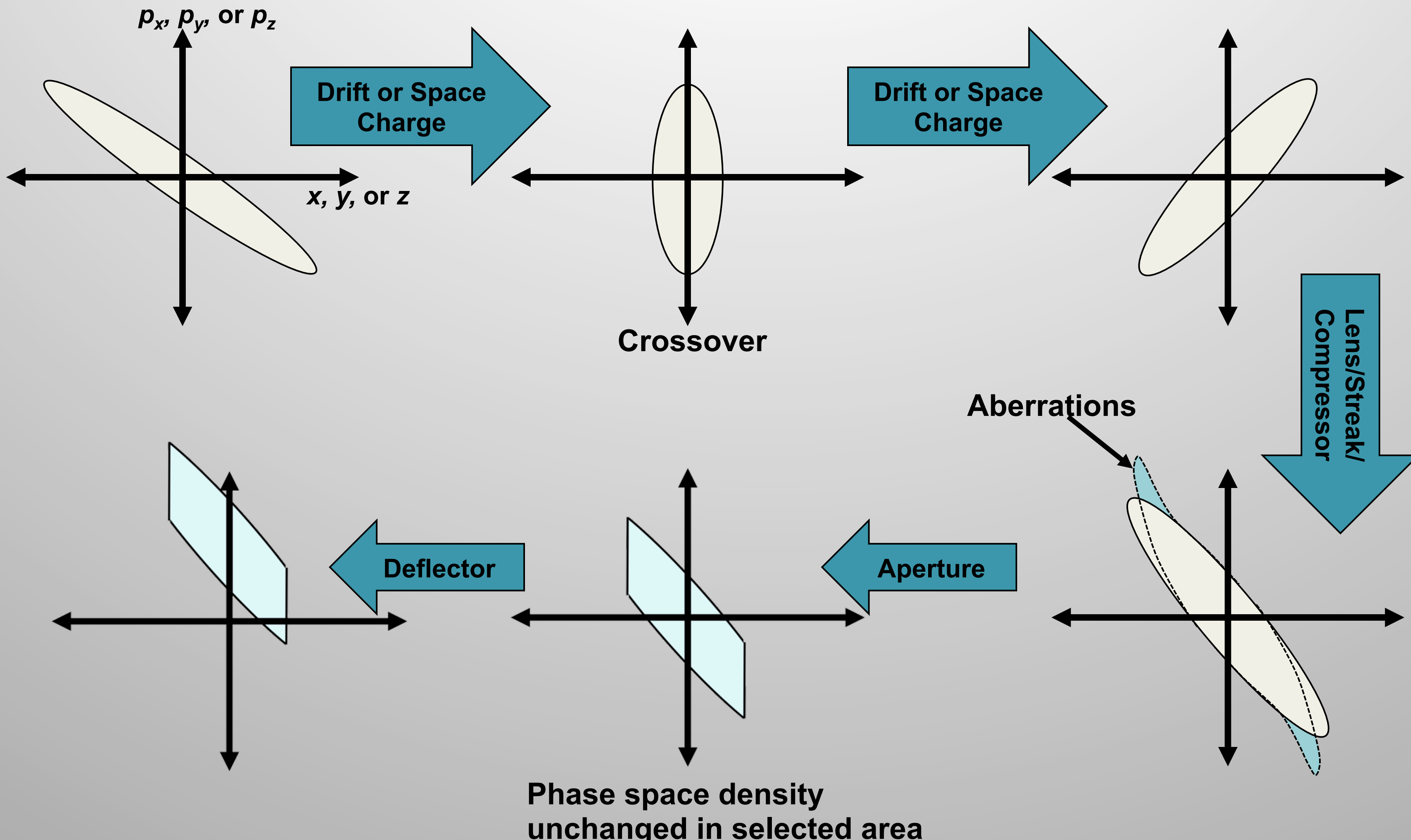
Exception 2: Electron-Sample Interactions (desirable)



Exception 3: Discrete Electron-Electron Interactions (undesirable)



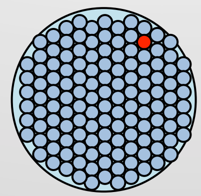
All the operations of electron optical elements are just phase space transformations



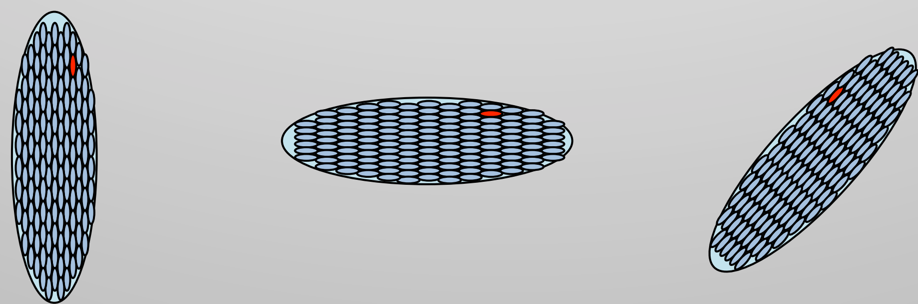
Because of Liouville's theorem, we can think of quantized phase space blocks as physical objects to be manipulated

By "Phase Space Block" I mean a volume h^3 in 6D space that, because of quantum mechanics, can contain at most 2 electrons.

The electron gun launches a whole bunch of these blocks and randomly fills about 10^{-4} of them with electrons.

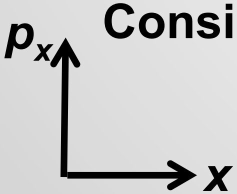


As the beam propagates, the *shape* of each block can change, but not its *volume*.

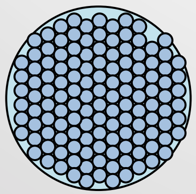


Because these blocks generally move together, if the neighboring region starts out with a 10^{-4} fill factor, it will always have a 10^{-4} fill factor.

The condenser lens system trades off current density against spatial coherence

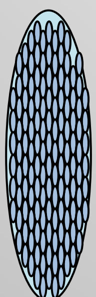


Consider 2 of the 6 phase space dimensions

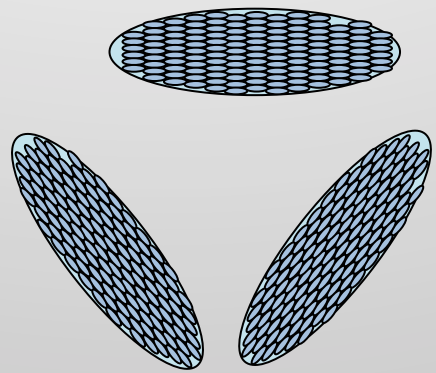


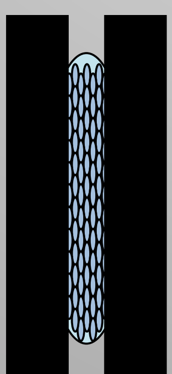
A blob of *x*-p_x space broken up into blocks of area *h*

(x width of blocks)/(number of blocks in p_x direction) = transverse coherence length



Condenser lenses can change the shape but not the area or number of blocks

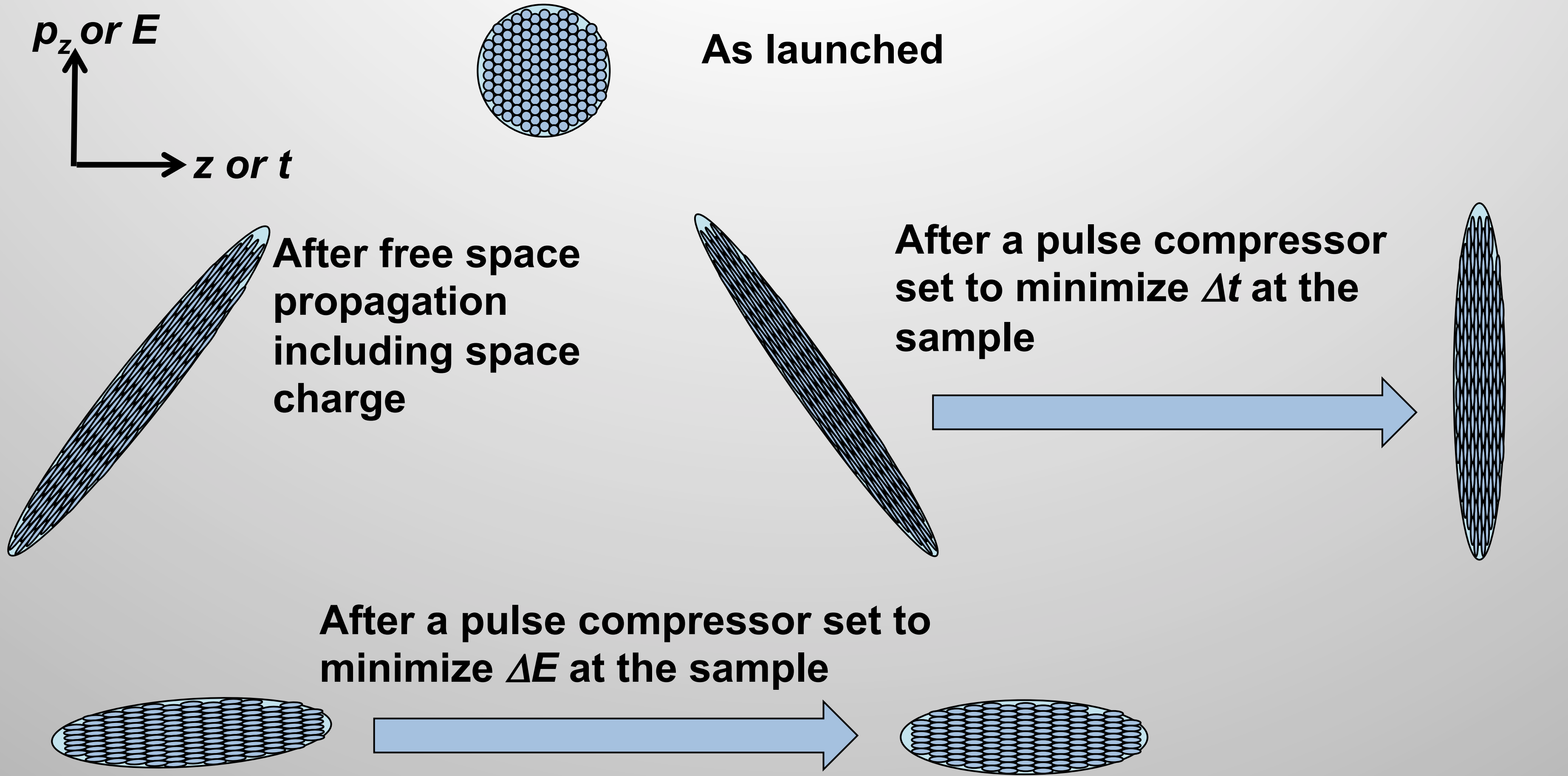




Blocking part of the beam with an aperture reduces the *total* current but has no effect on the fill fraction or the brightness

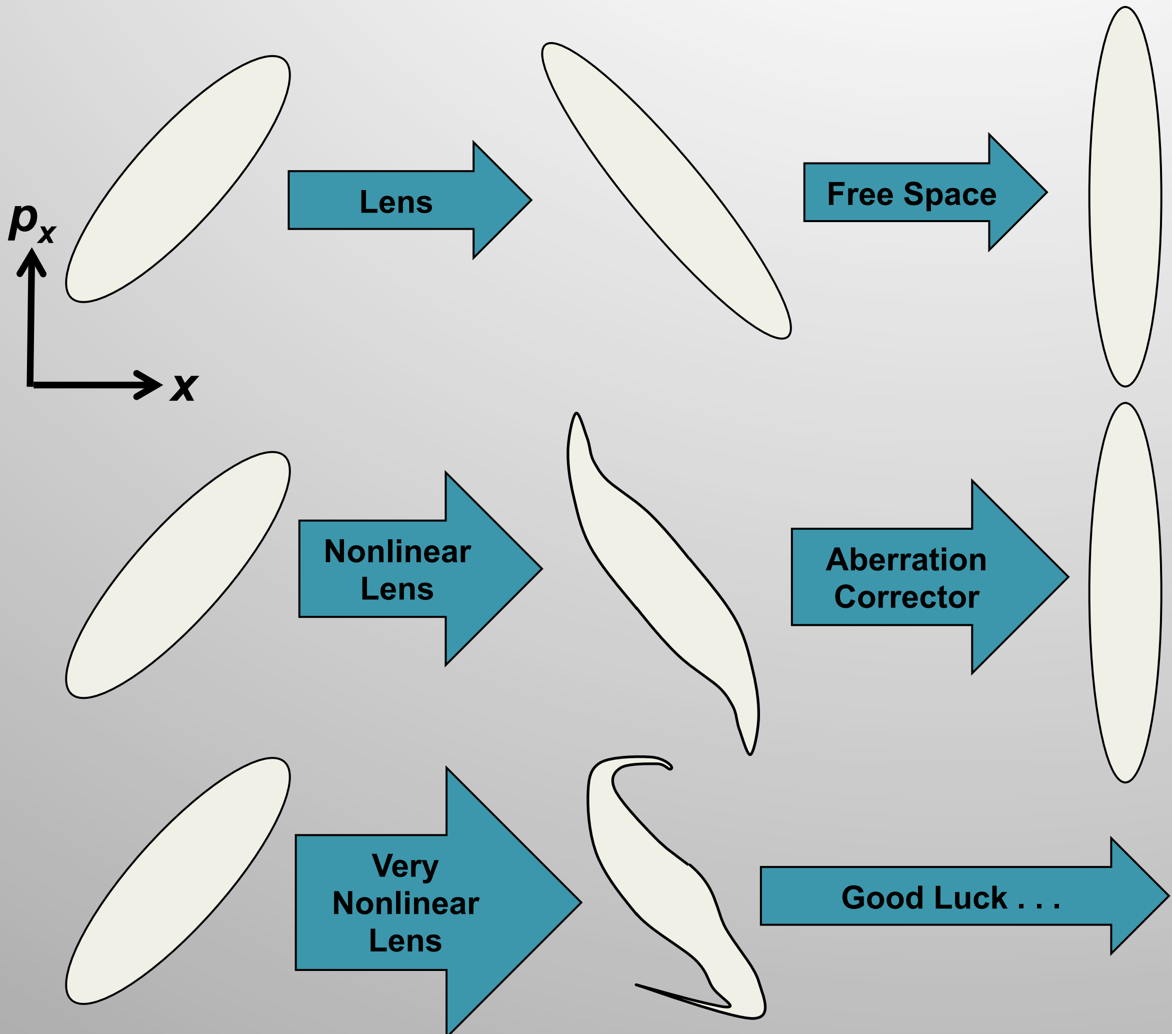
No matter what you do, the coherent fluence (electrons per transverse coherence area per pulse) remains the same.

Exactly the same thing happens with a pulse compressor



As before, we can change the shape all we want, but not the area or the number of blocks. There's always a tradeoff.

Aberrations act as nonlinear transformations in phase space



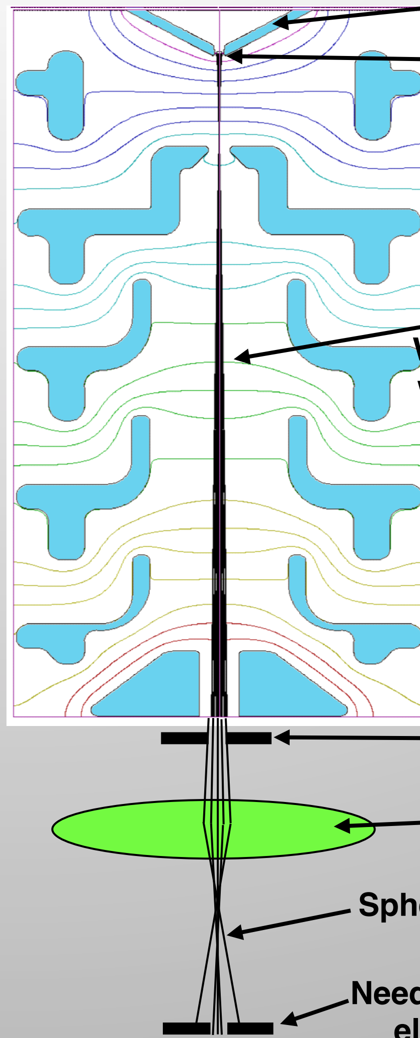
This works the same for spatial and temporal lenses

This has become a common feature in newer TEMs

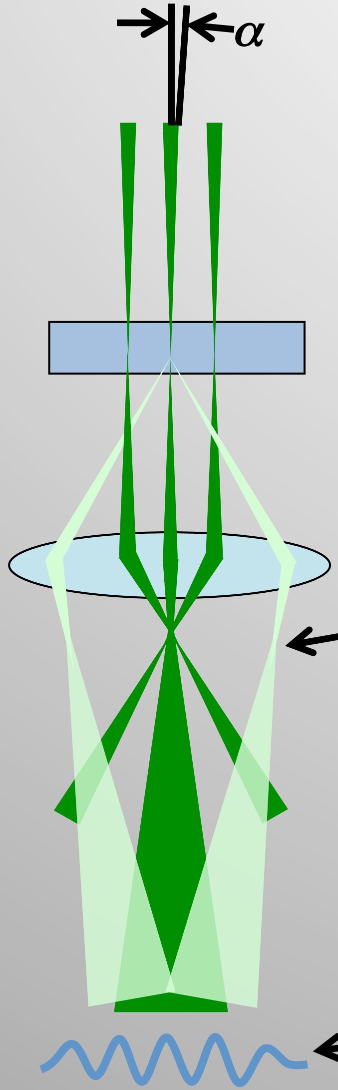
In principle, such a transformation is reversible, but don't get your hopes up!

Once you generate electrons, you want to keep them: Low aberrations and large apertures

- Normal TEM throws away most electrons to improve coherence.
  - DTEM can't afford to do this
- Large diameter pulse beam is blocked by fixed apertures.
  - 97% of electrons blocked
  - Highly aberrated electrons



High local convergences angles at the sample degrade performance for all operating modes



$$r_{coherence} \sim \lambda/\alpha$$

Condenser lens lets you increase current density by increasing α, but there's a price to pay for this.

Energy spread has similar effects.

Diffraction:
Bragg scatter is convolved with initial angular distribution

Incoherent Imaging: High angles couple with lens aberrations to scramble real-space information

Partially Coherent Imaging (e.g. diffraction contrast): Everything is washed out; very weak contrast and blurry

Coherent Imaging (e.g. atomic resolution bright field TEM): Large convergence angle severely reduces fringe visibility

The coherent fluence N_C is an invariant figure of merit for a pulsed electron gun

- N_C = number of electrons per transverse coherence area per pulse
- N_C is conserved with respect to focusing, aperturing, acceleration, pulse compression, and space charge effects
- Aberrations and incoherent scattering can degrade it
- No passive system can ever improve it (few exceptions)
- **N_C must be > 10 (preferably 100 or 1000) to do ANY coherent imaging**

$$N_C = \frac{N\lambda^2}{r^2\alpha^2} = \frac{\pi\lambda^2 B\Delta t}{e}$$

N = Electrons per pulse

λ = Electron wavelength

r = Transverse radius

α = Local convergence angle

B = Brightness (current/area/ solid angle)

Δt = Duration of pulse

	Previous DTEM	Movie mode DTEM	UED and MeV-UEM
N_C	~1 (15 ns)	1-200 (5-1000 ns)	usually << 1

The punch line: If you want to do *any kind of single-shot coherent imaging at all*, if your gun doesn't produce a coherent fluence >> 1, it won't work.

This includes diffractive image reconstruction.

No matter what you do, you can't get a statistically significant number of electrons into a coherence area unless they start out there!

Practical experience on DTEM indicates that $N_c \sim 1$ is needed to produce good diffraction-contrast images of microstructure.

Low-resolution shadow imaging is not limited by this.

When evaluating electron sources for single-shot imaging, calculate the coherent fluence or risk wasting a lot of time!

This is a function of *the gun*, and it has nothing to do with lenses, apertures, voltage, pulse compressors, etc.

Coherent fluence is also relevant for diffraction

Gun produces a coherent fluence N_C.

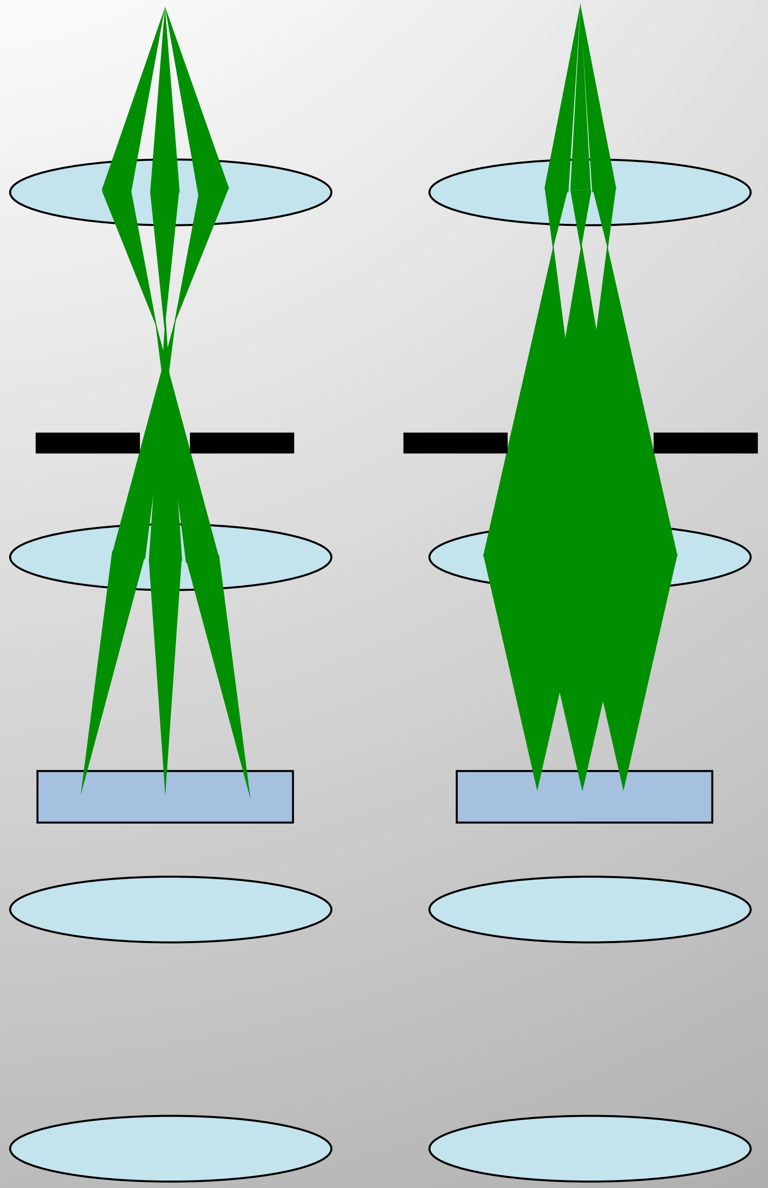
Experiment that needs reciprocal space resolution Δk.

You want to have N electrons in your pattern.

What spot size r or sample area $A = \pi r^2$ should you use?

Answer:

$$A \sim \frac{N}{N_C\left(\Delta k\right)^2}$$

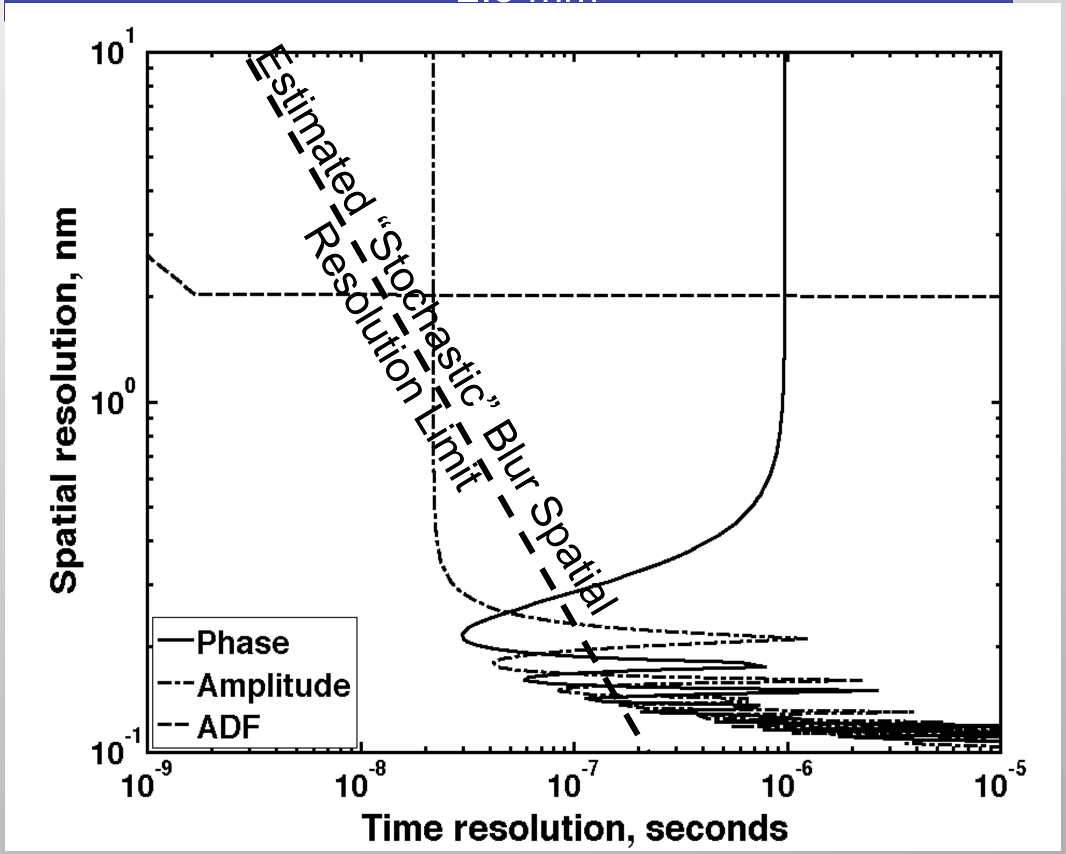


TEMs have multiple condenser lenses, apertures, and post-sample lenses for a reason!

Spatial and temporal resolution can be traded off against each other, depending on the contrast mechanisms

- Variable pulse width allows experimental trade-offs of
  - Signal
  - Coherence
  - Resolution
  - Space charge mitigation
- 1μs pulses should give 0.3nm spatial resolution.
  - 500 ns pulse already gives near "CW" image quality
  - Voltage droop is not a problem

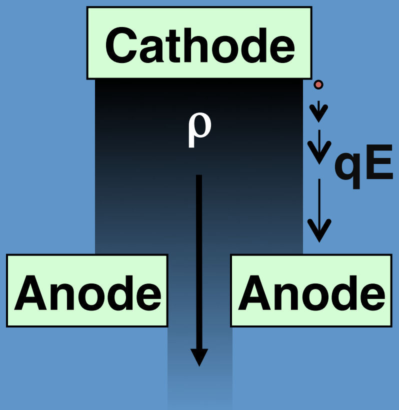
JEOL 2000FX DTEM:
Energy Spread = 3 eV, α = 5 mrad, Cs = 2 mm, Cc = 2.3 mm



B W. Reed et al. Microscopy and Microanalysis, (2008)

Electron-electron interactions become important at high currents

Longitudinal Global Space Charge

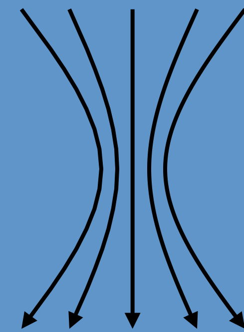


Child's Law:

- Field from recent electrons retards emission
- Fundamentally limits current density:

$$j \propto V^{3/2} / d^2$$

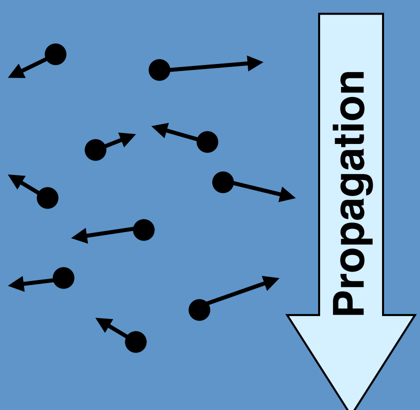
Lateral Global Space Charge



Defocusing:

- To first order, it's just a diverging lens
- Higher orders create spherical aberration

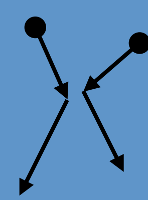
Longitudinal Inhomogeneous



Boersch Effect:

- Pulse viewed in rest frame
- Most velocity is lateral
- Equilibration increases ΔE

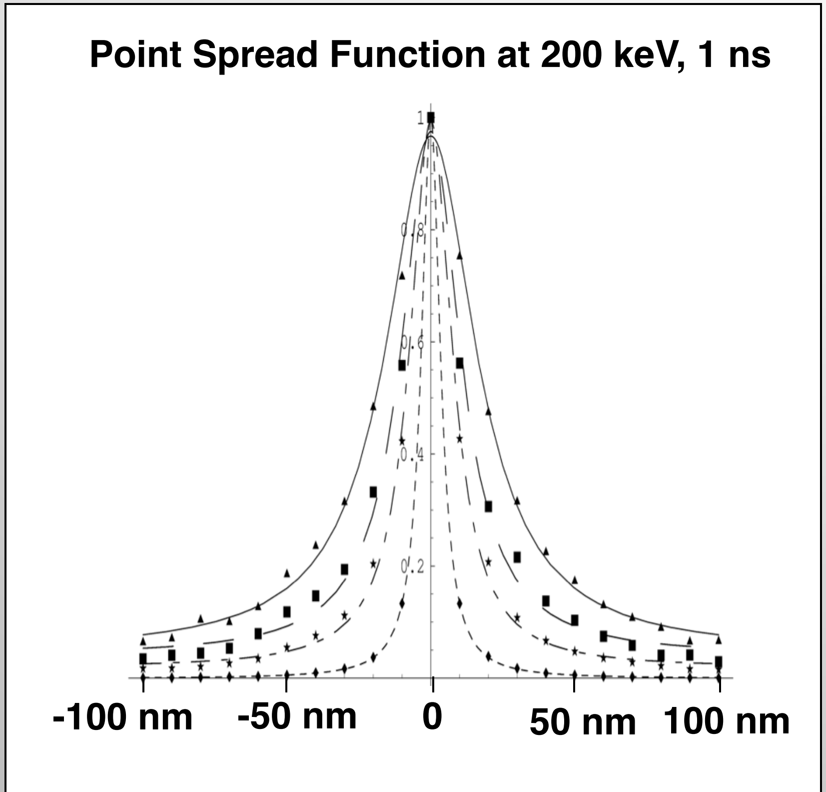
Lateral Inhomogeneous

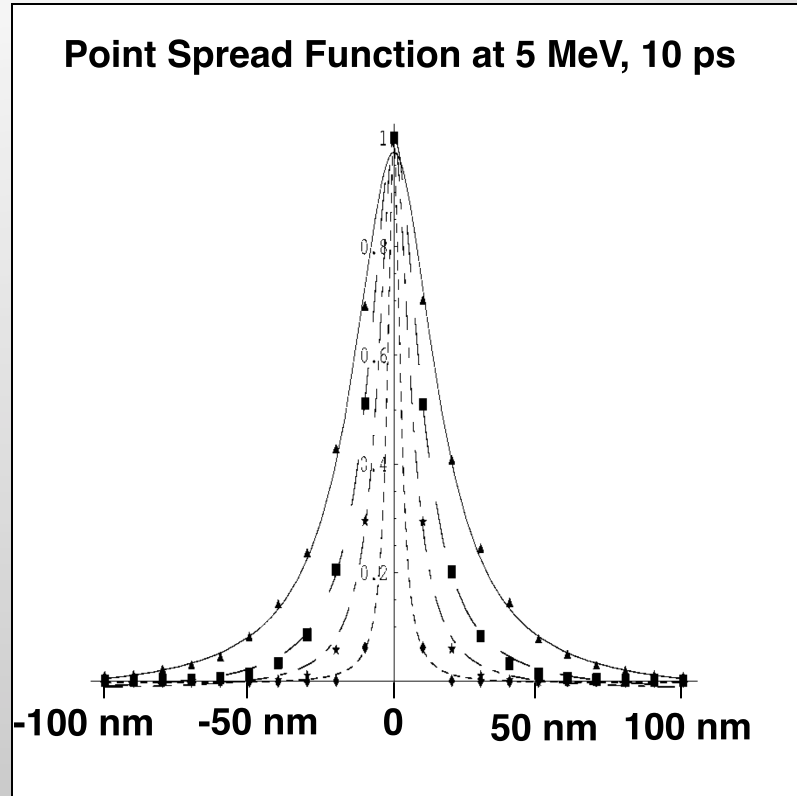


Trajectory Displacement:

- Random electron-electron scatter
- High-resolution information stored in lateral velocities is gradually destroyed

Ultrafast (< 10 ps) imaging may be possible at higher voltages

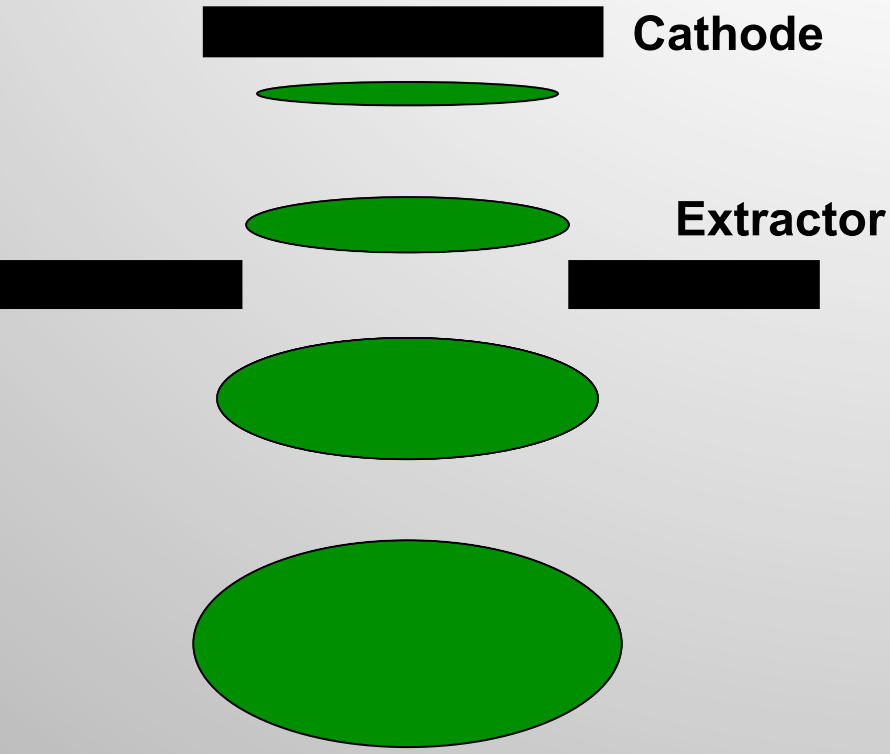


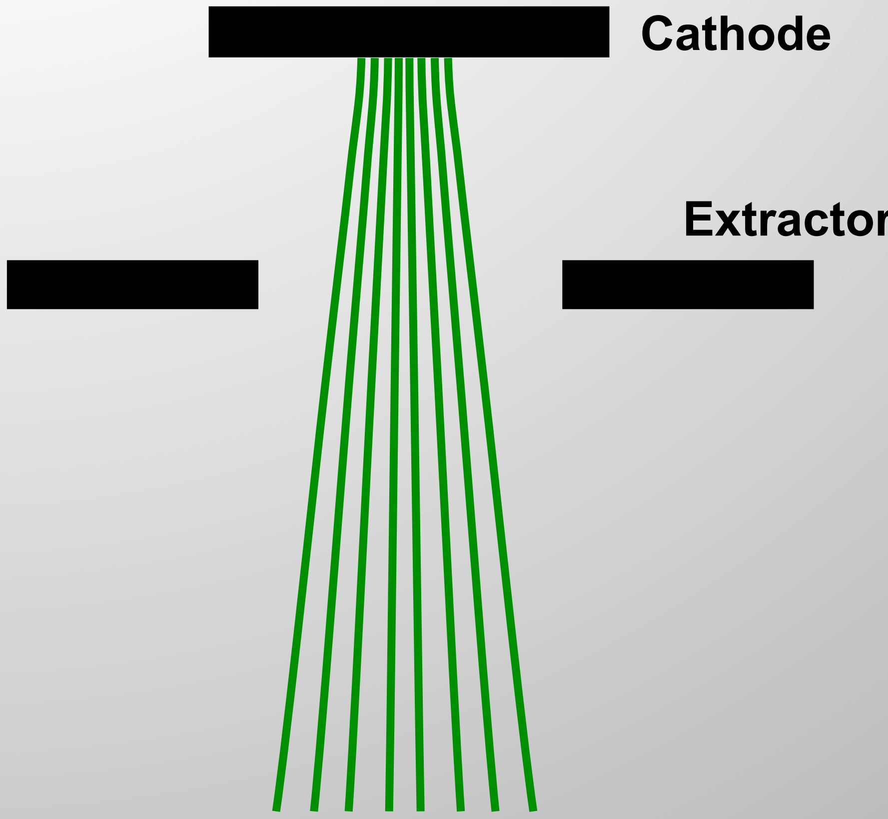


For picosecond resolution imaging, relativistic energies may be the only way to obtain high spatial resolution.

M. R. Armstrong, B. W. Reed, B. R. Torralva, and N. D. Browning, *Appl. Phys. Lett.* 90, 114101 (2007).

Space charge has very different effects in femtosecond and nanosecond systems



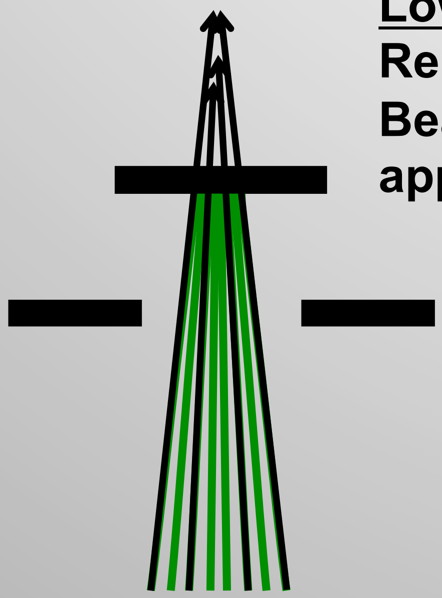


Femtosecond:
"Pancake" aspect ratio
Charge/area limited by electric field
Longitudinal expansion worsens Δt and ΔE
Can be undone with pulse compressors

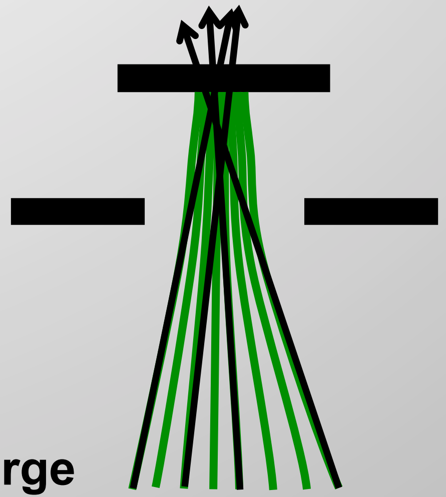
Nanosecond:
"Pencil" aspect ratio
Steady state current/area limited
Transverse expansion
Can be undone with lenses

Good performance of nanosecond guns requires local capacitance and flat spatiotemporal profiles

(100 mA) (100 ns) = 10 nC needs 10 nF local capacitance to keep $\Delta E < 1$ eV
but everything on the high-voltage cable within 50 ns (around 10 meters of coax) can potentially contribute



Low Current:
Relatively little space charge
Beams extrapolate back to a small apparent source

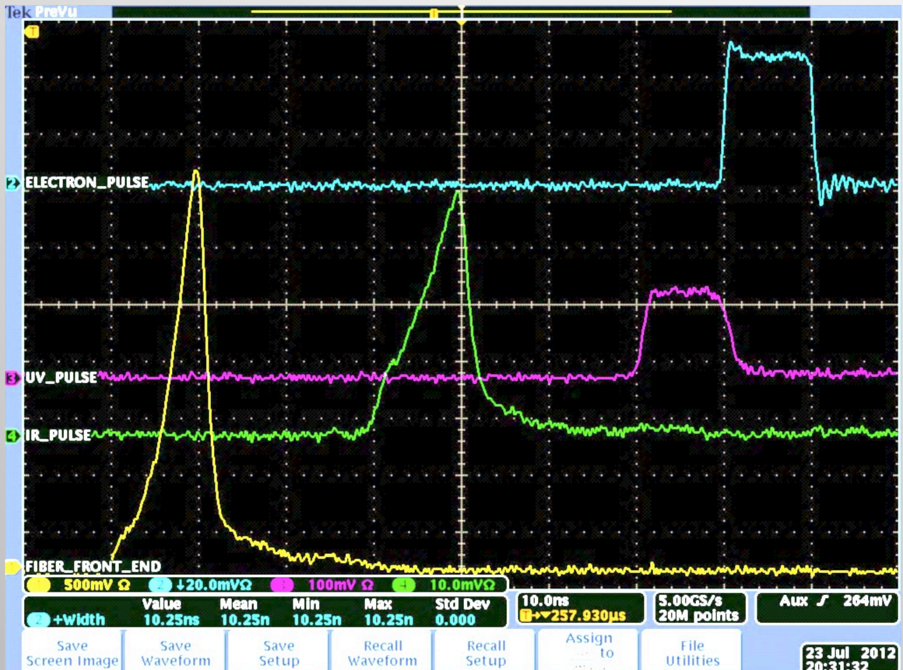


High Current:
Stronger space charge
Apparent cathode position shifts
Hot spots and asymmetries exaggerated
Effective brightness can be degraded

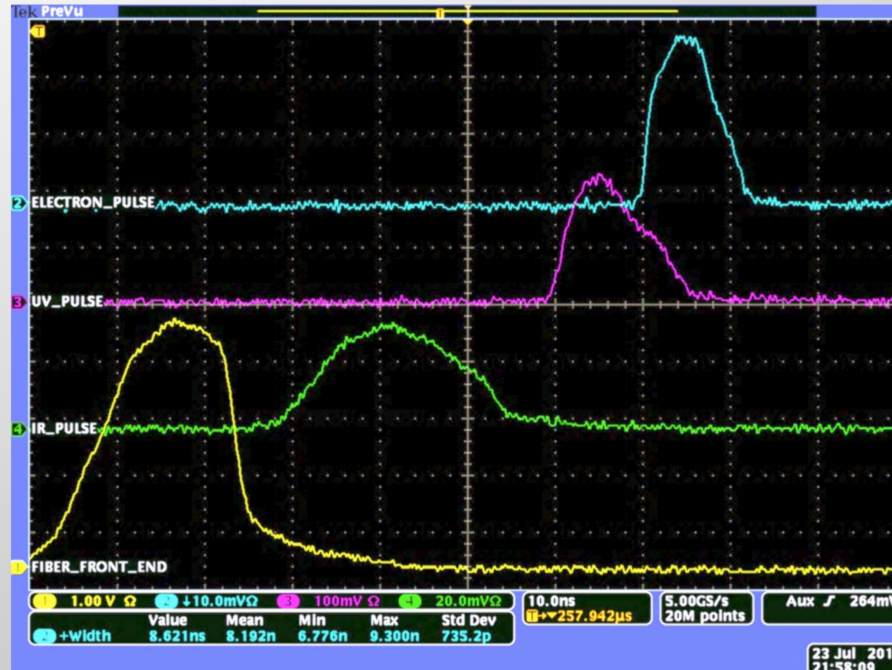
If the current changes during the pulse, the apparent cathode position moves and the effective brightness is degraded!

Sophisticated pulse-shaping is required to get a temporal flat-top in a multi-nanosecond pulsed laser

10ns square electron pulse

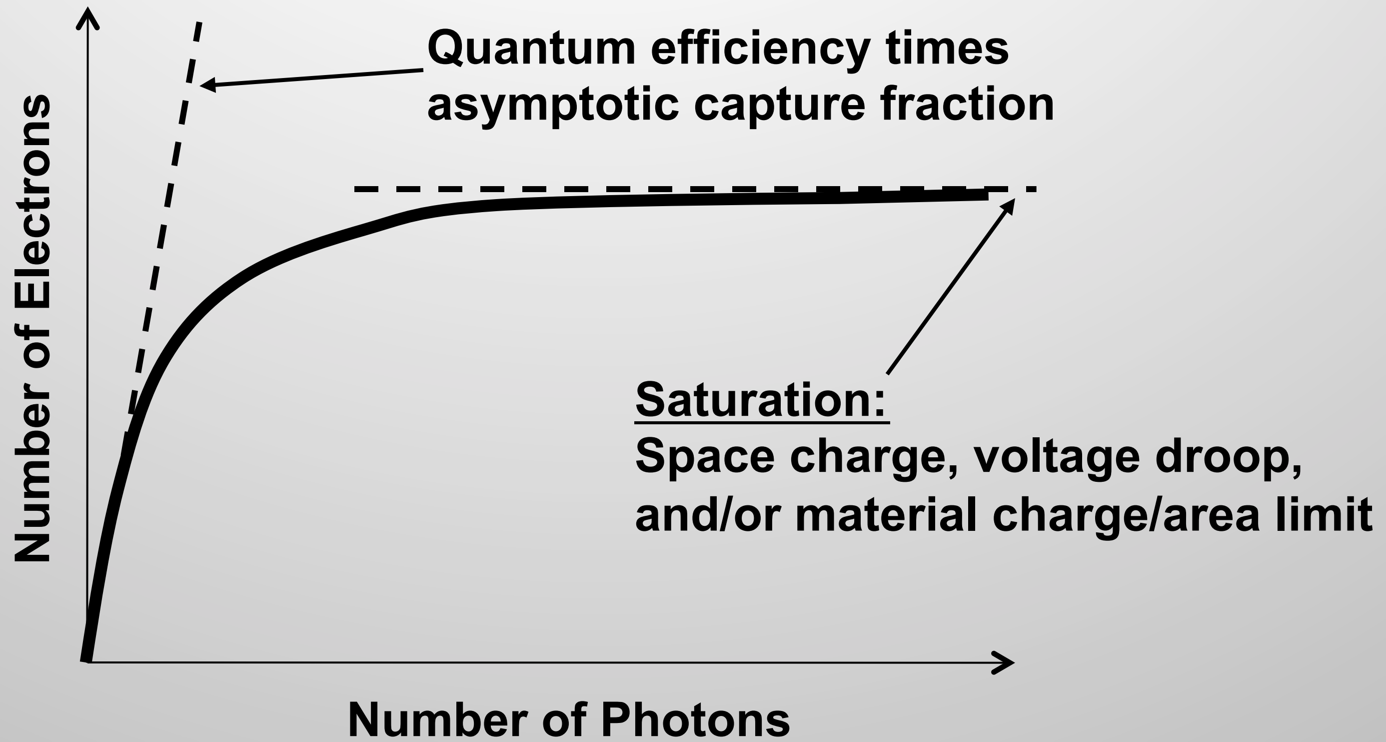


10ns FWHM Gaussian electron pulse



Q-switched pulsed lasers do not provide optimal temporal profiles for single-shot DTEM applications

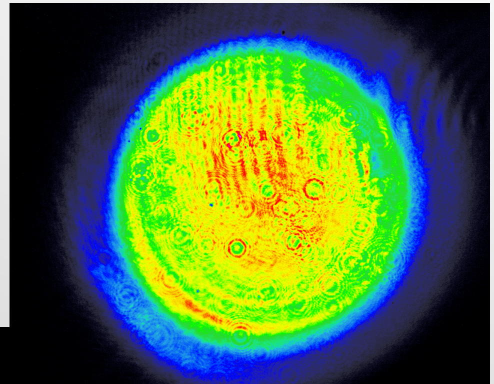
Electron yield saturates as the cathode laser energy increases

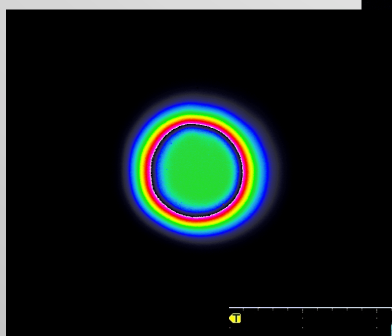


The deficit between the two dashed asymptotes represents electrons generated but not used

The type and quality of the cathode laser determine the spatial and temporal resolution that can be achieved in the DTEM!

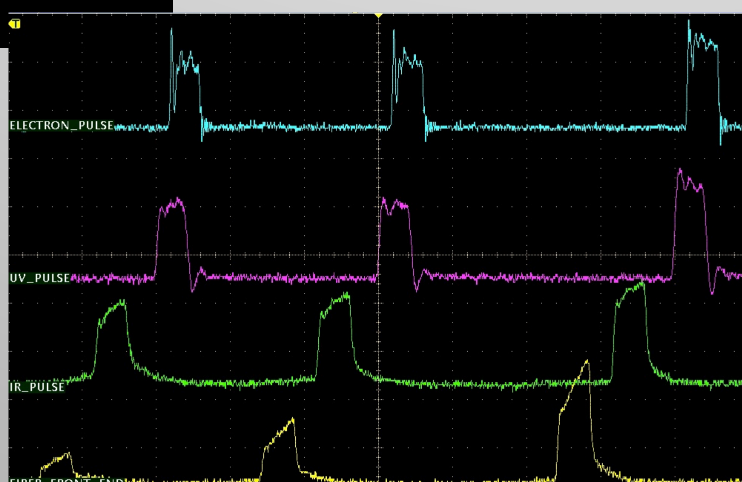
IR beam profile after final Amplifier





Focused UV beam profile on photocathode

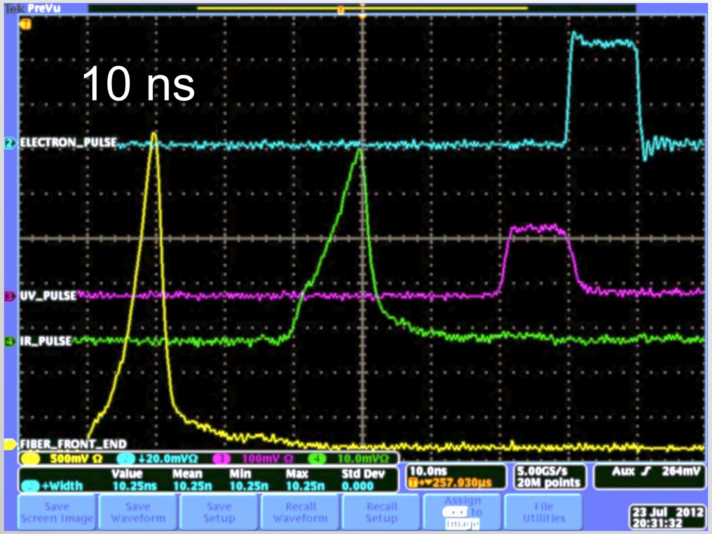
Temporal profiles of a 10 ns pulse train from AWG laser

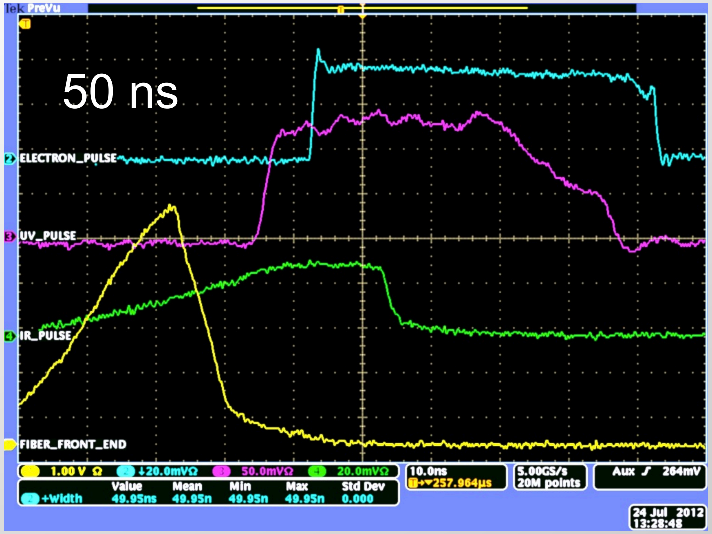


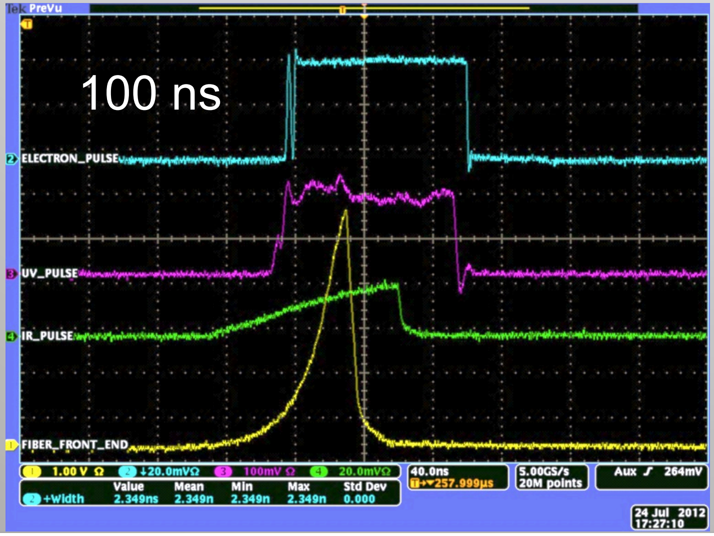
To have high spatial resolution, the laser must have:

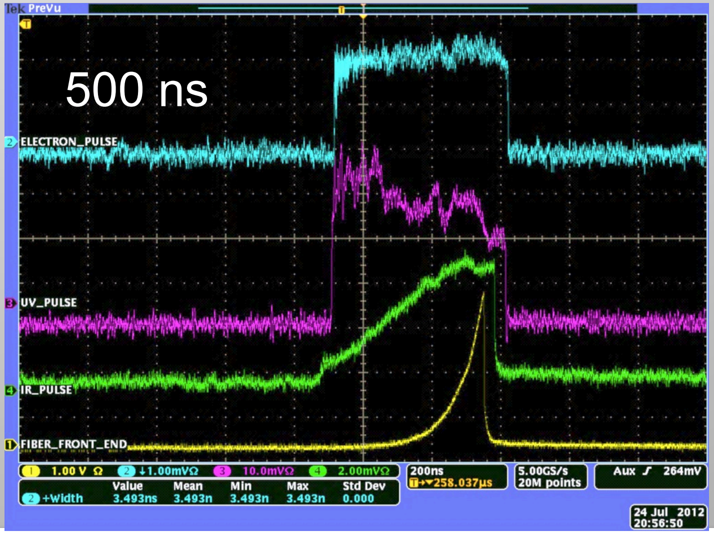
- Excellent pointing stability
- Good beam quality (focusability)
- Requisite energy and wavelength to produce enough electrons
- Spatial profile and size that balances space-charge with preferred pulsed electron beam distributions for the TEM optics
- Temporal profiles optimized to reduce space charge and electron-electron interactions

We are able to generate electron pulses with arbitrary widths tailored for experimental need

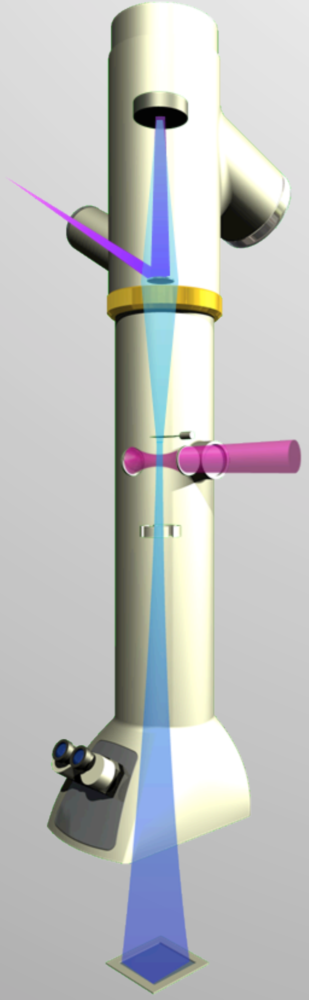


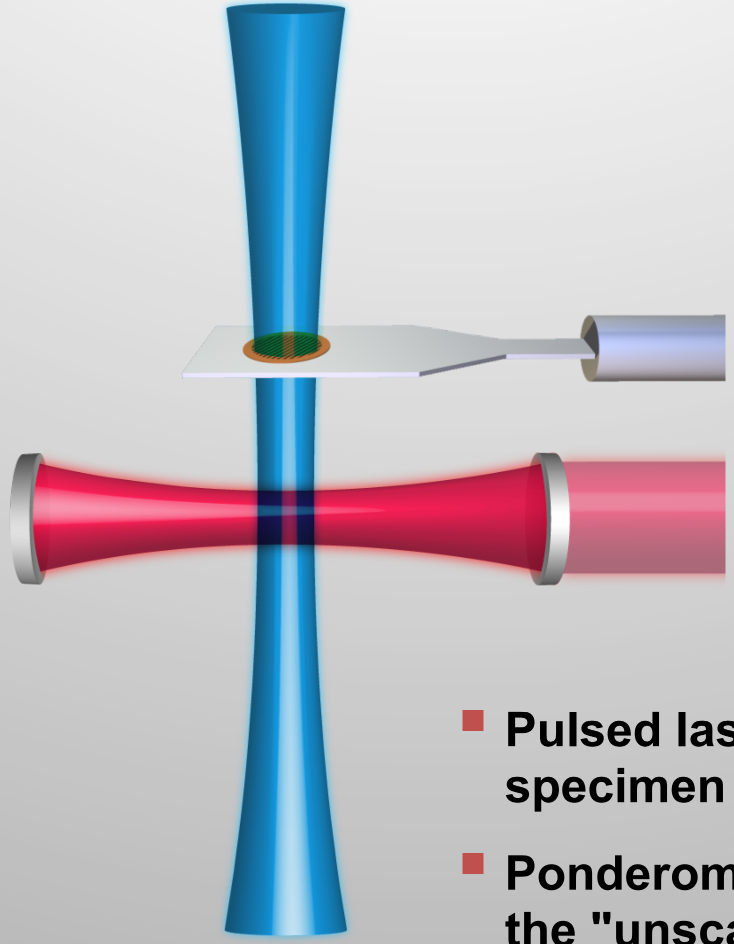


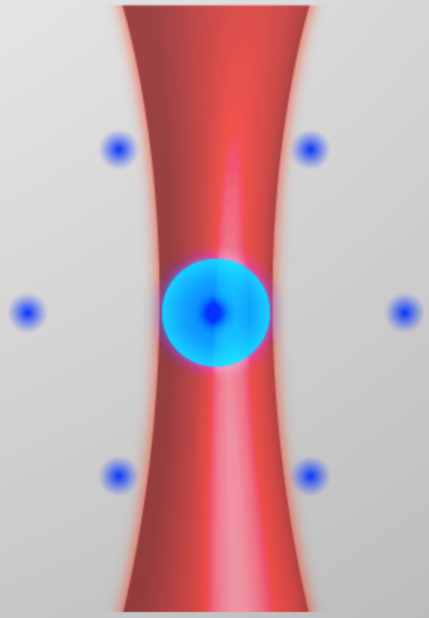




DTEM is compatible with a Pulsed Ponderomotive Phase Plate which would use a pulsed laser to enhance phase contrast imaging







- Pulsed laser focused onto a post-specimen crossover
- Ponderomotive effect phase-shifts the "unscattered"electrons

IDES

The basics of quantum electrodynamics in one slide

This is the nonrelativistic Hamiltonian (the function that governs how things change in time) for a charged particle in a field:

$$H = \frac{\left(\vec{p} - q\vec{A}\right)^2}{2m} + qV = \frac{p^2}{2m} - \frac{q}{2m}\left(\vec{p}\cdot\vec{A} + \vec{A}\cdot\vec{p}\right) + \frac{q^2A^2}{2m} + qV = \frac{p^2}{2m} - \frac{q}{m}\vec{p}\cdot\vec{A} + \frac{q^2A^2}{2m}$$

If we pick a convenient gauge

What you need to know about $\vec{A}$:

- It's the electromagnetic vector potential
- It's polarized like the electric field
- Usually the terms involving it are "small"
- Every time it appears, it represents creating or annihilating a single photon

Relativity adds some complications:

- Need both spin states for electron and positrons (four total components)
- Correction factors that involve β's and γ's
- Higher-order terms (O(A^2), O(A^3), etc.)
- Spin-field and spin-orbit interactions

So what does that QED stuff mean for us?

Electron going about its business

Interaction term 2:
Compton scattering and ponderomotive effect

$$H = \frac{p^2}{2m} - \frac{q}{m}\vec{p}\cdot\vec{A} + \frac{q^2A^2}{2m}$$

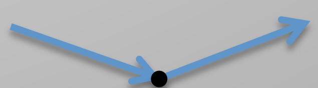
Interaction term 1:
Absorption or emission of a single photon

About term 1:

- To first order, it has no effect in free space (conservation laws)
- *In the presence of another object*, though, there's no problem
  - XRF, EELS (especially aloof), PINEM, microwave cavities, static lenses, etc.
- Stimulated emission: It's easier to copy a photon that's already there

About term 2:

- You're creating one photon while annihilating another one
- If they're different photon energies, it's Compton scattering

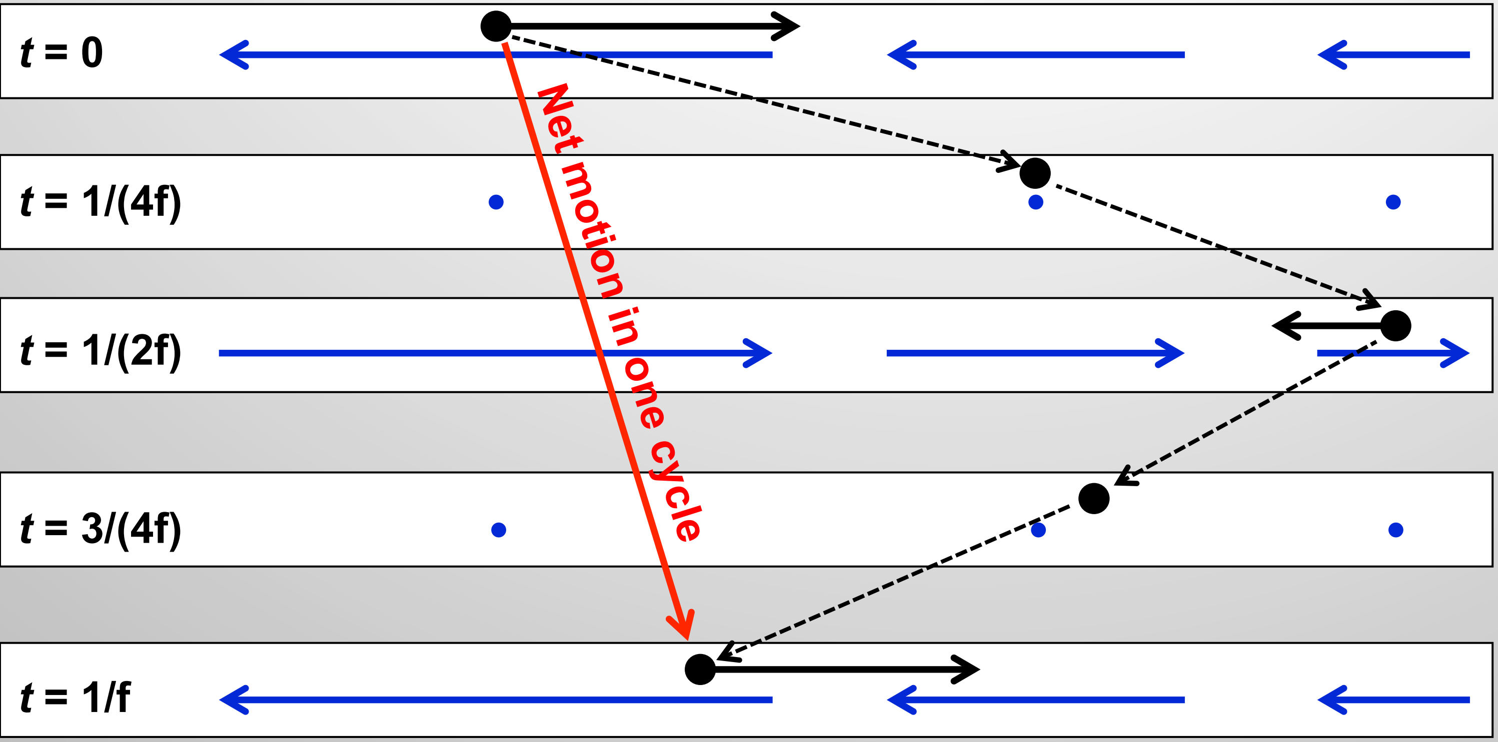


- If they're the same photon energy, it's a ponderomotive phase shift

Ponderomotive Effect: Basic Classical Version

Imagine an electron in a field of nonuniform light intensity I:

Blue Arrow = Electric Field
Black Arrow = Electron Acceleration



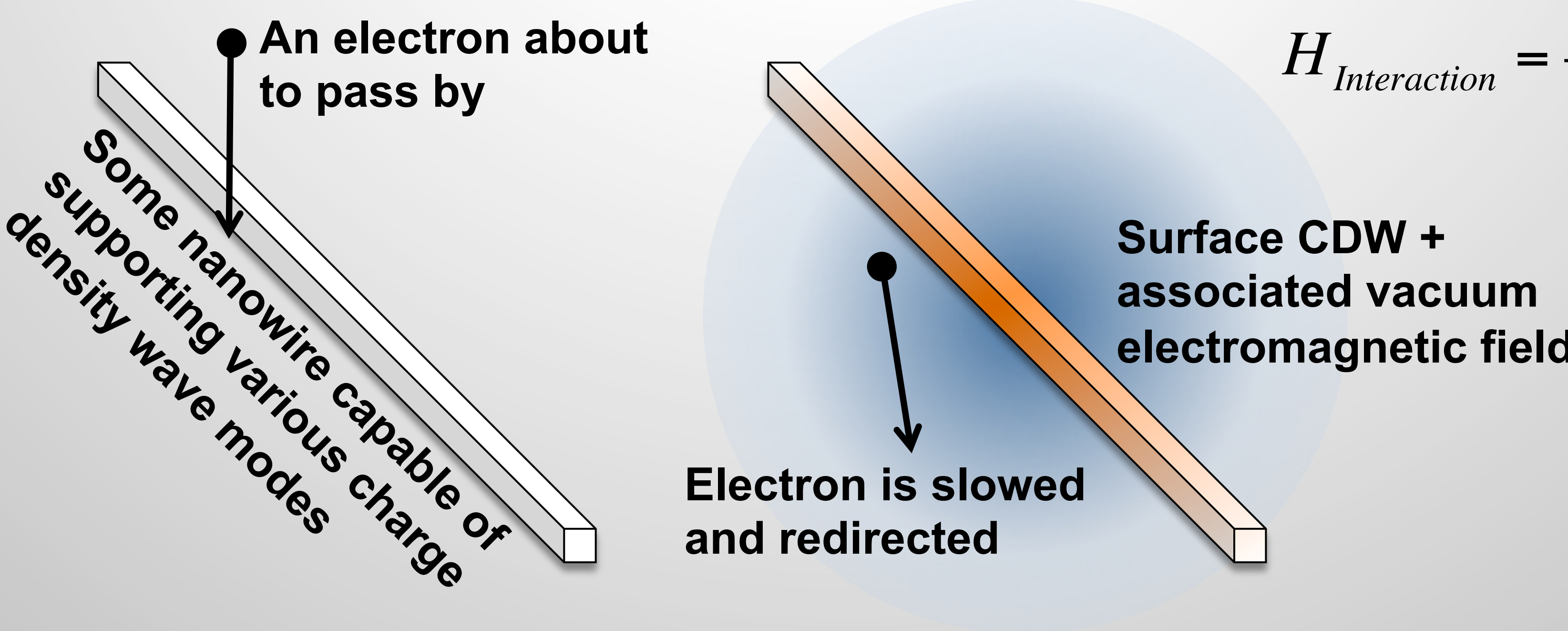
Ponderomotive effect is a net drift of charged particles away from regions of high field intensity, as if under the influence of a potential:

$$U_{ponderomotive} = \frac{q^2\lambda^2}{8\pi^2\varepsilon_0\gamma mc^3} I(\vec{r},t)$$

Warning: This is a simplified version. It gets much more complex in standing waves, focused spots, etc. Smorenburg et al., Proc. SPIE 8079, 80790Z (2011)

Quantum mechanics gives the same result but (1) reduces the need to assume slow intensity variation, and (2) clarifies that the trajectories shouldn't be taken too literally.

EELS, PINEM, and other photon creation/annihilation events



$$H_{Interaction} = \frac{q}{m}\vec{p}\cdot\vec{A}$$

Now the *A* term creates and annihilates *polaritons* rather than pure photons. This changes the momentum conservation rules.

Microwave cavity acceleration/deceleration is based on the same fundamental interaction but on a very different scale.

Quantum electrodynamics summary

- The "correct" model is the Dirac equation with a quantized photon field
  - All particle-field interactions have to be describable in these terms
  - For engineering purposes it's really complicated and annoying to work with
- A nonrelativistic version that still includes photon quantization captures the essence of the physics that we use in our systems
  - Ponderomotive effect is much more general than purely classical models would suggest
  - EELS and PINEM arise naturally and can be handled by a semiclassical treatment that hides most of the quantum mechanics in the material response (e.g. $\varepsilon(k,\omega)$) and $E = \hbar\omega$
  - EELS and microwave cavities have more in common than you might think
- Sometimes the classical-trajectory model works just fine, but we should always remember that it's just an approximation
  - Sufficient to provide relativistic and polarization corrections to ponderomotive effect
  - Pretty much always works in microwave regime
  - Erroneously suggests that ponderomotive effect can cause random energy gain/loss
  - Gives a misleading picture of the state of an electron after sample interaction

Summary

- The electron column is just a tool for manipulating phase space
- The gun plays the crucial role of *defining* and *filling* the phase space
- Coherent fluence is an important figure of merit
- Single-shot resolution limits ultimately derive from quantum mechanics and irreversible stochastic blur effects
- Given the properties of the electron source and column, the current DTEM is not far from its ultimate resolution limits
- There are several avenues for improving those limits
  - Variable pulse length
  - ADF-DTEM
  - Higher voltages
  - Pulse compression
  - Brighter electron gun
  - Reduced emission energy spread
  - Aberration correction (C_s *and* C_c)
  - Phase plates
- Good single-shot performance depends crucially on having the right laser
- Quantum electrodynamics provides a unifying framework for ponderomotive effects, EELS, PINEM, and microwave cavities