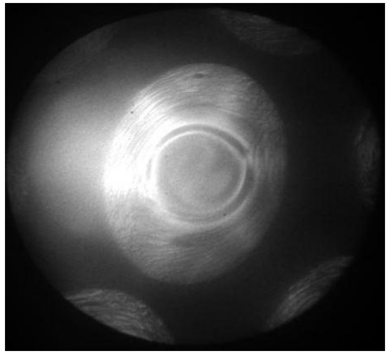
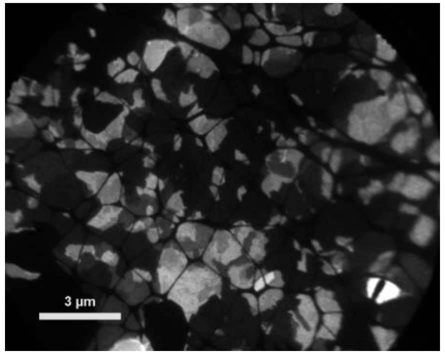
Computation and measurement of aberrations in low energy electron microscopy

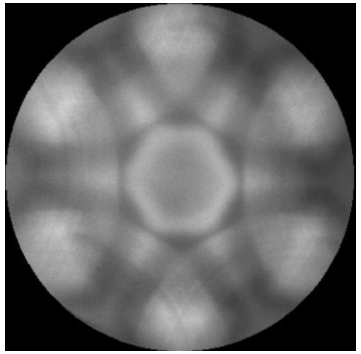
Rudolf M Tromp

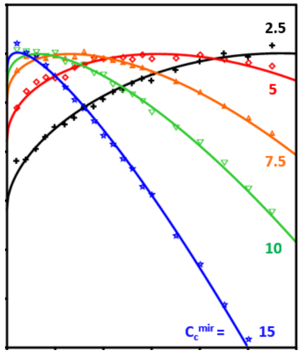
IBM T.J. Watson Research Center, Yorktown Heights, NY 10598

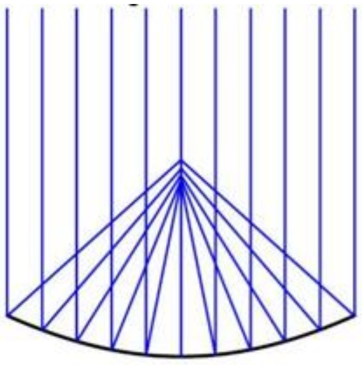
Kamerlingh Onnes Laboratory, Leiden University, The Netherlands

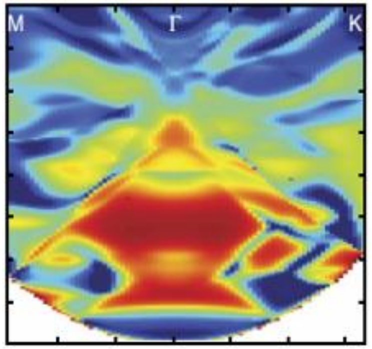




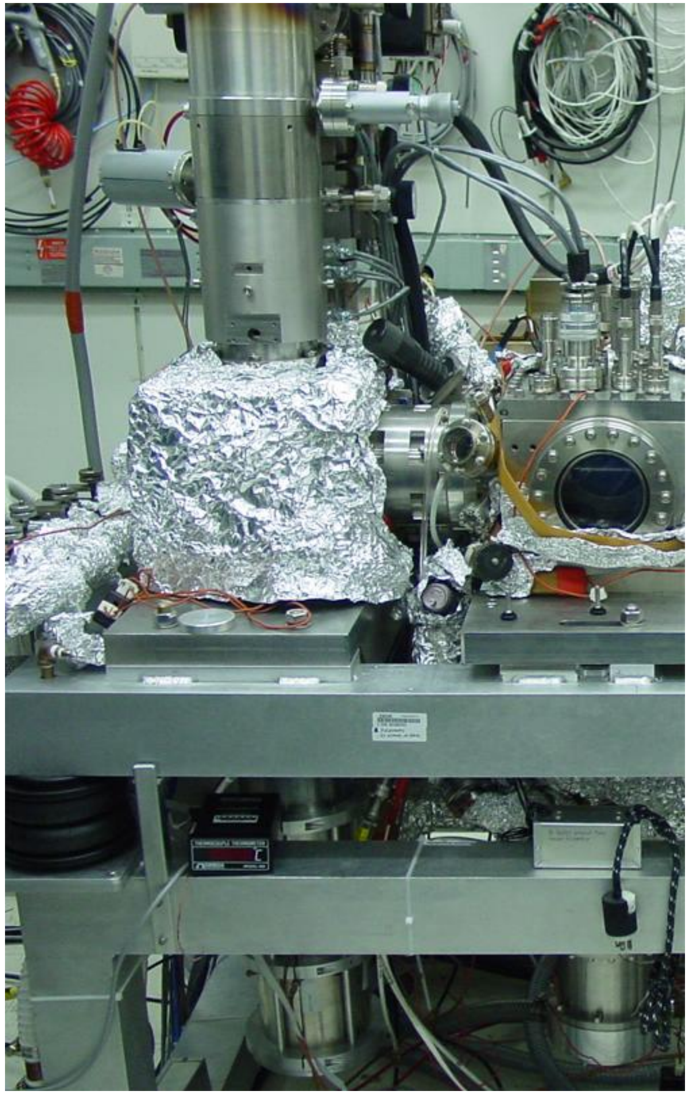


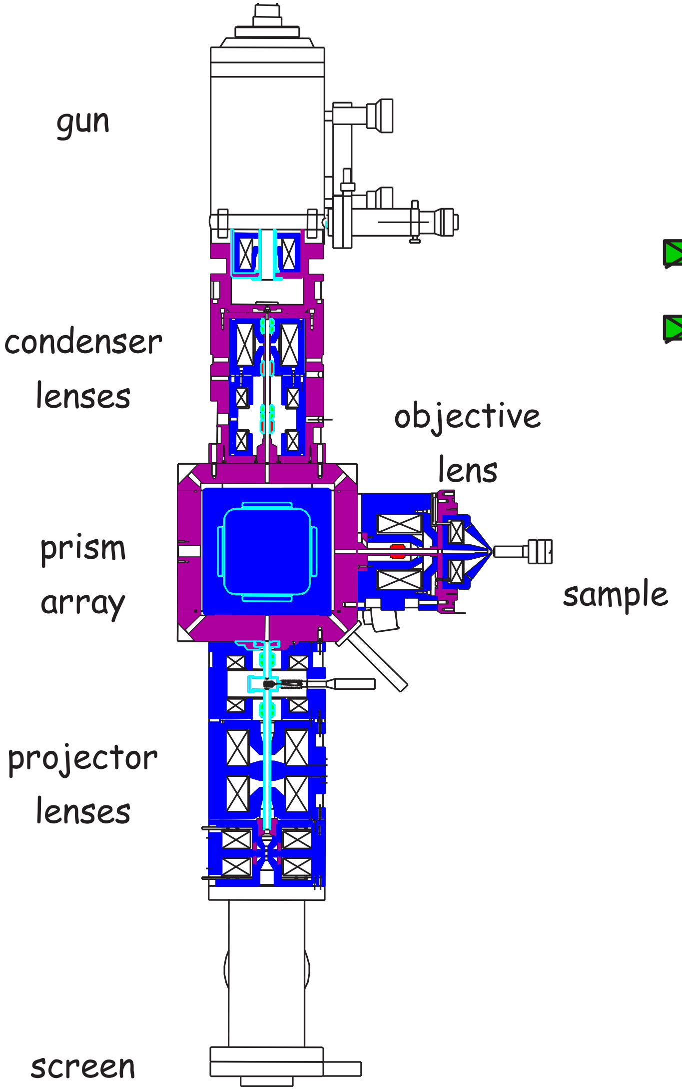


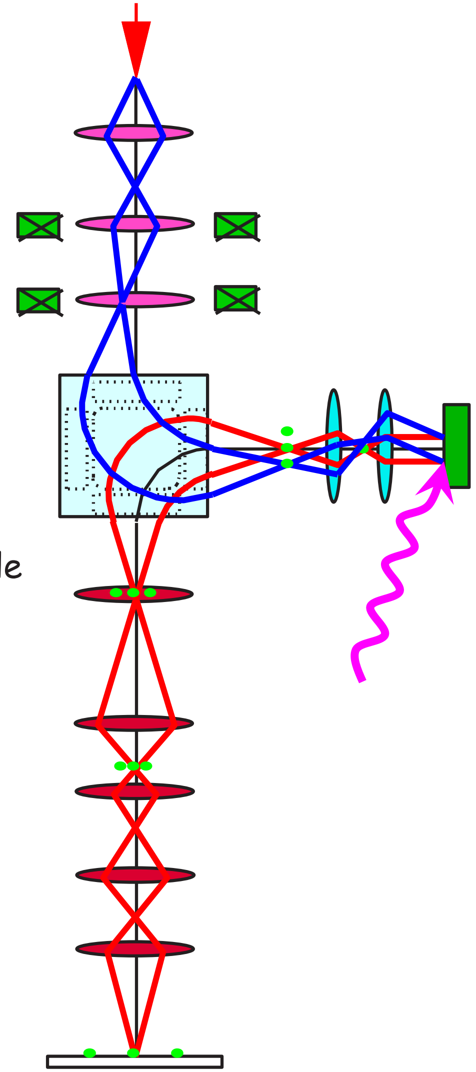




IBM/SPEC (AC)-LEEM/PEEM Design: ~30 instruments sold

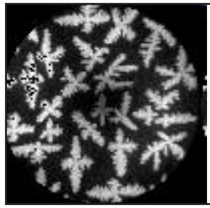






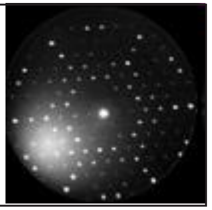
R.M.Tromp, M. Mankos, M.C. Reuter, A.W. Ellis, M. Copel
Surface Review and Letters 5 , 1189 (1998)

Lab-based LEEM/PEEM imaging modes



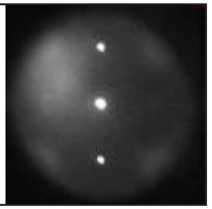
PEEM imaging
Hg light source

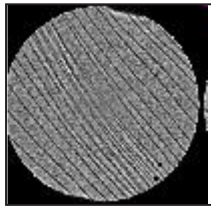
LEED
Atomic Structure



Mirror Microscopy
Topography
Work Function

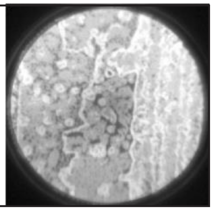
Selected Area LEED
Local Atomic Structure
200 nm

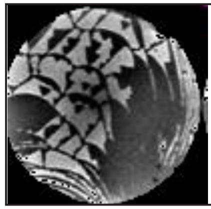




Bright field LEEM
Phase contrast

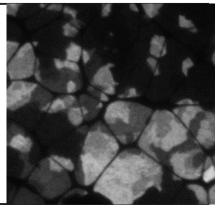
LEEM-EELS
Local Electronic Structure
Spectroscopy + Imaging





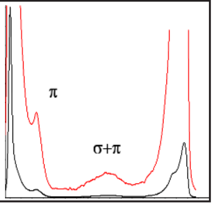
Bright field LEEM
Reflectivity
Structure factor

eV-TEM
Transmission imaging
without damage

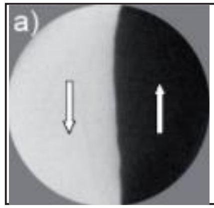


Dark field LEEM
Structure symmetry

Transmission EELS
Low energy loss on
the nanoscale

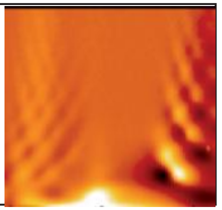


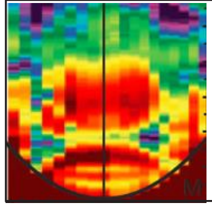
Lab-based LEEM/PEEM imaging modes



SPLEEM
Magnetic domain imaging

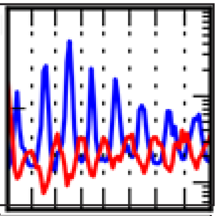
IV-SPLEEM
Magnetic quantum Well asymmetry

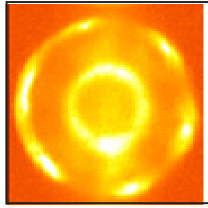




ARRES
Empty state band structure

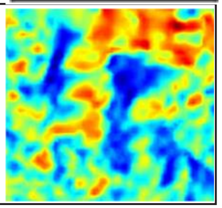
SPA-LEED-PLD
Atomic Layer Oscillations during PLD growth

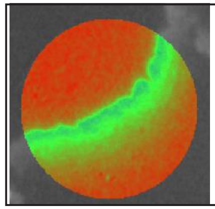




PEEM-ARPES
Filled state band structure

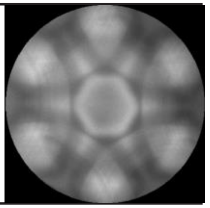
SPA-LEED
Local strain measurement

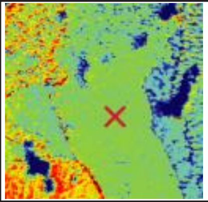




LEEM-IV Imaging
Local Atomic Structure 2-5 nm

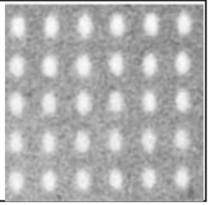
CBED
Local Atomic and Electronic structure



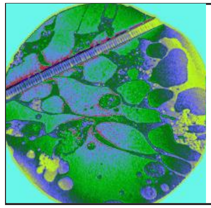
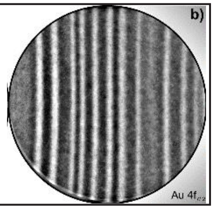
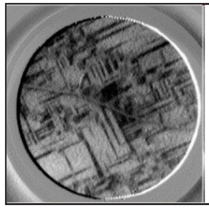
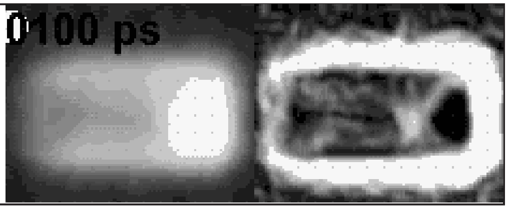
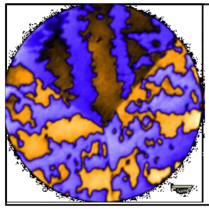
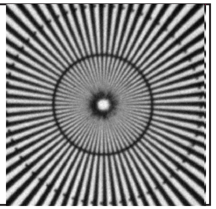
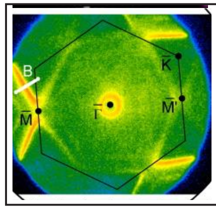
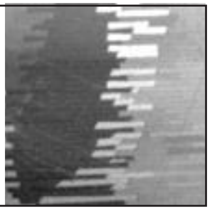
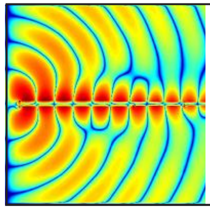
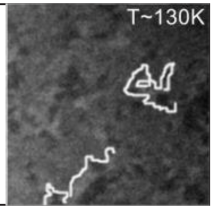


LEEM potentiometry
Contact-less nanoscale device measurements

LEEM lithography
Structure fabrication with few eV electrons



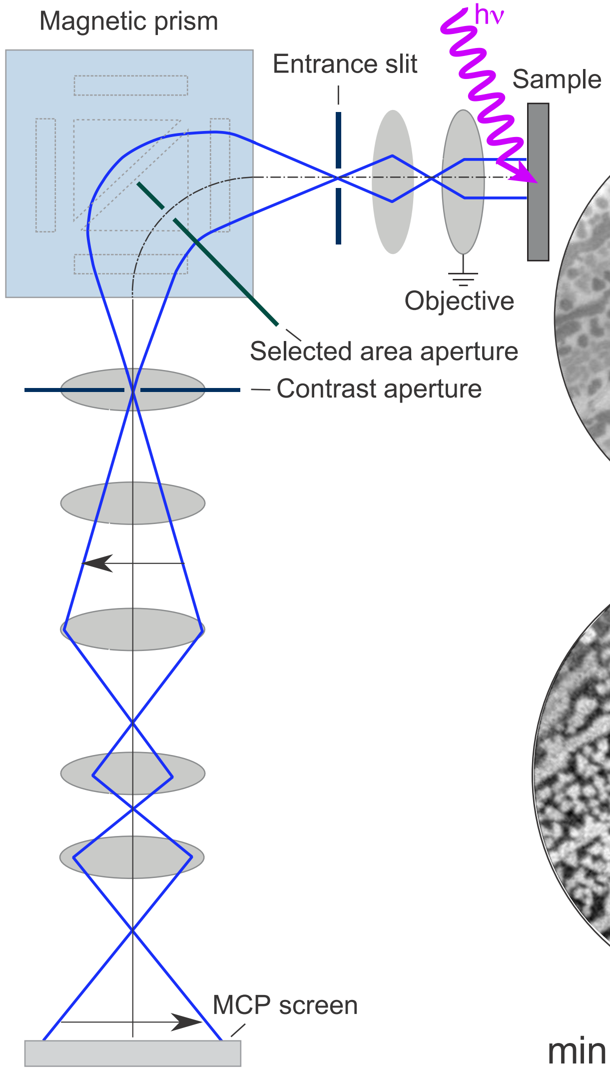
Synchrotron-based PEEM imaging modes

 PEEM-IV Imaging 20 nm resolution Local chemistry	Dynamic Imaging In-situ processing Elemental/chemical 
 Linear Magnetic Dichroism Antiferromagnetism	Picosecond Resolution Imaging 
 Circular Magnetic Dichroism Ferromagnetism	Localized Spectroscopy Elemental, chemical Magnetic, Valence 
 Valence Band Imaging Surface, bulk Topological	Biological Imaging Organic, Inorganic Elemental, chemical 
 Plasmonics Dynamics, geometry	Cryo-PEEM Solid State Bio, soft matter 

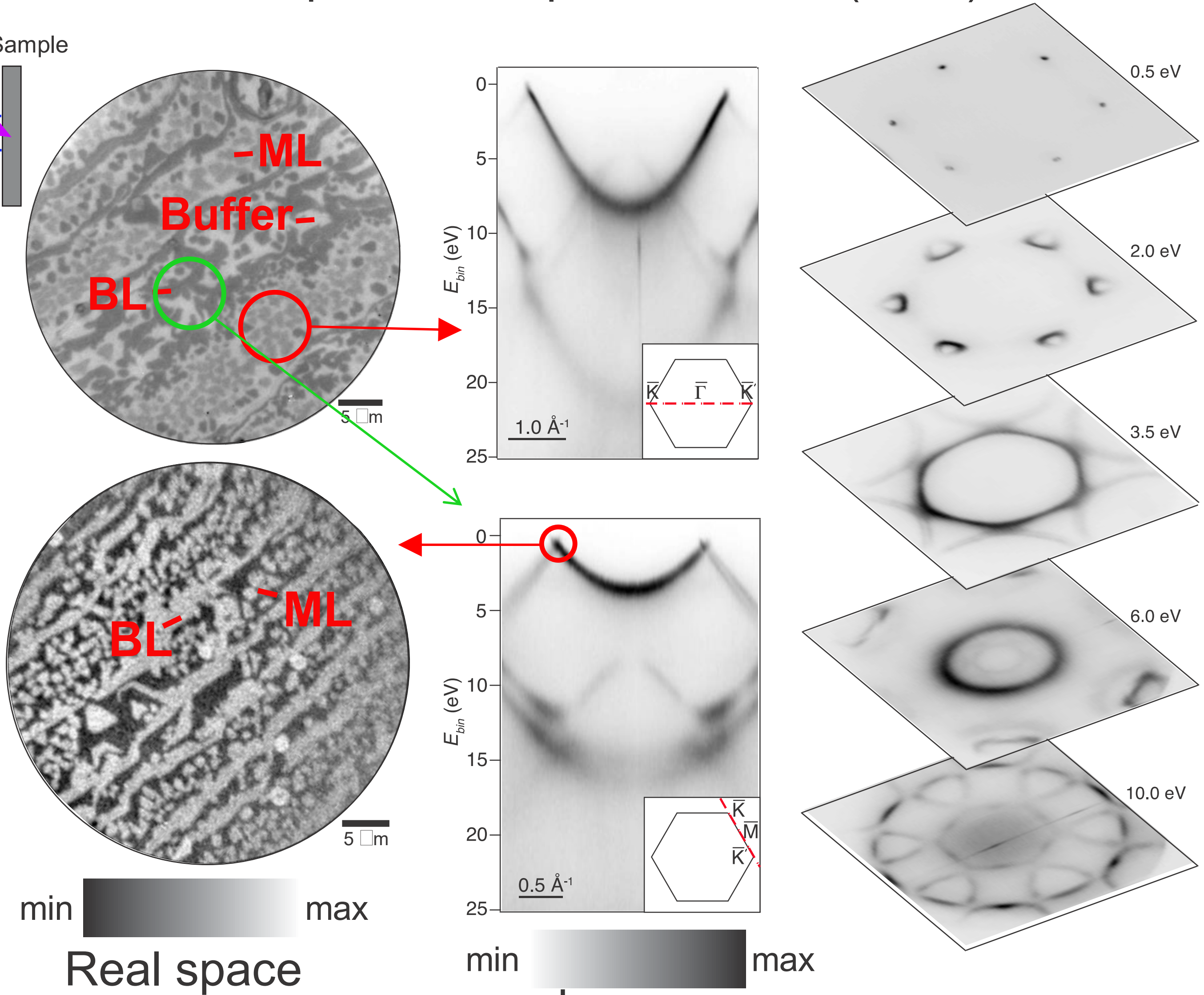
IBM/SPECS PEEM with an integrated imaging energy analyser: ARPES

Søren Ulstrup

Eli Rotenberg



Epitaxial Graphene/6H-SiC(0001)



k-space

Dark-field imaging with bands

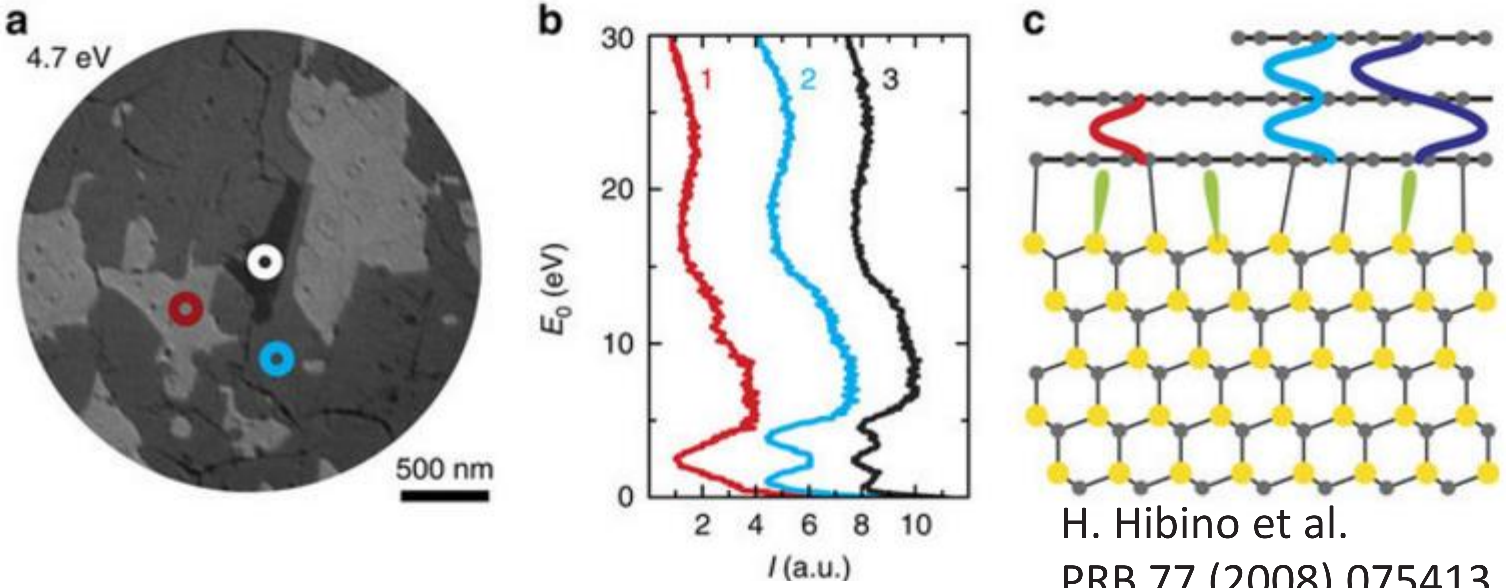
ALS ADVANCED LIGHT SOURCE

SPECS™

R. M. Tromp *et al.*, J. Phys. Cond. Matter **21** 314007 (2009)

ARRES

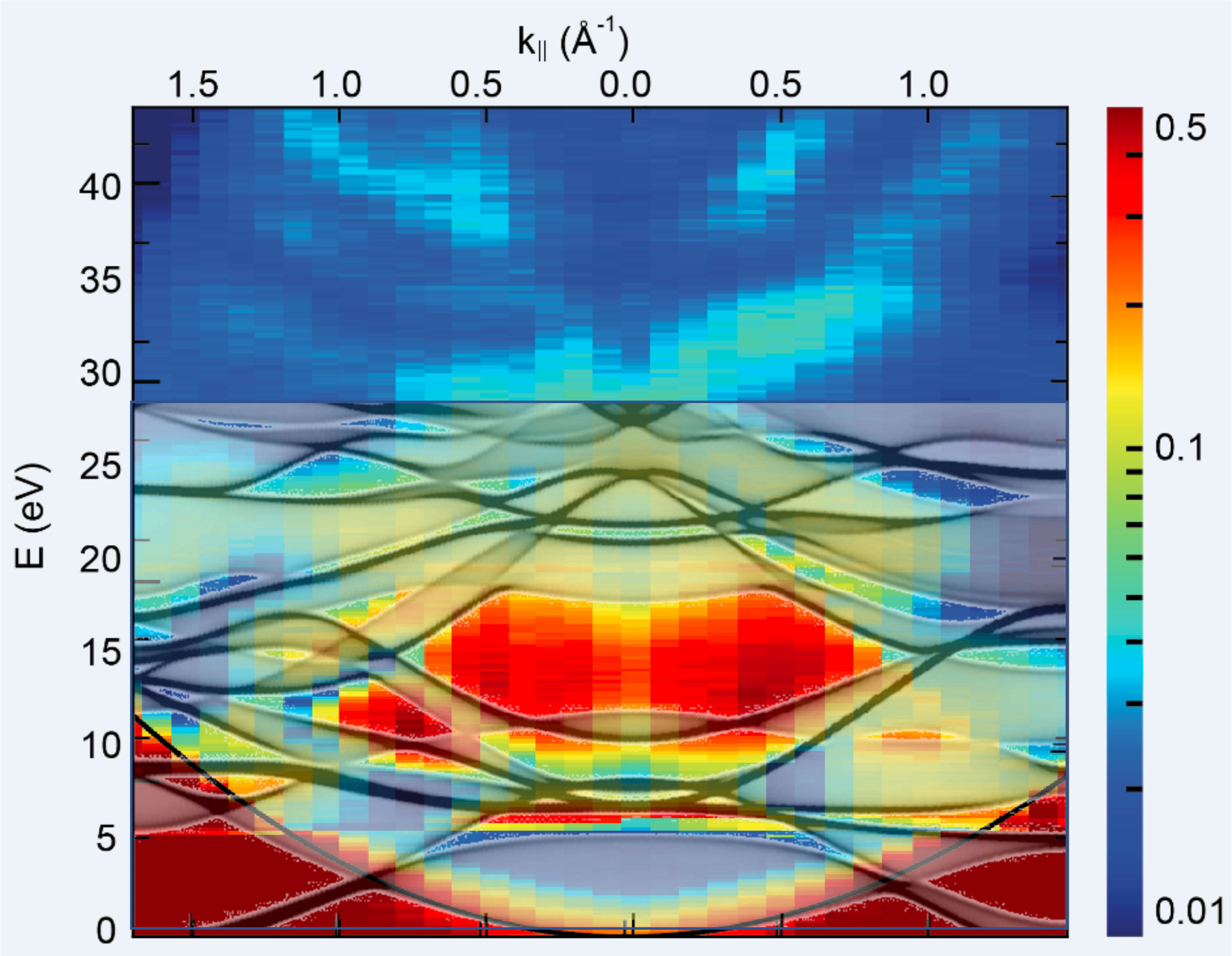
Angle
Resolved
Reflected
Electron
Spectroscopy



H. Hibino et al.
PRB 77 (2008) 075413

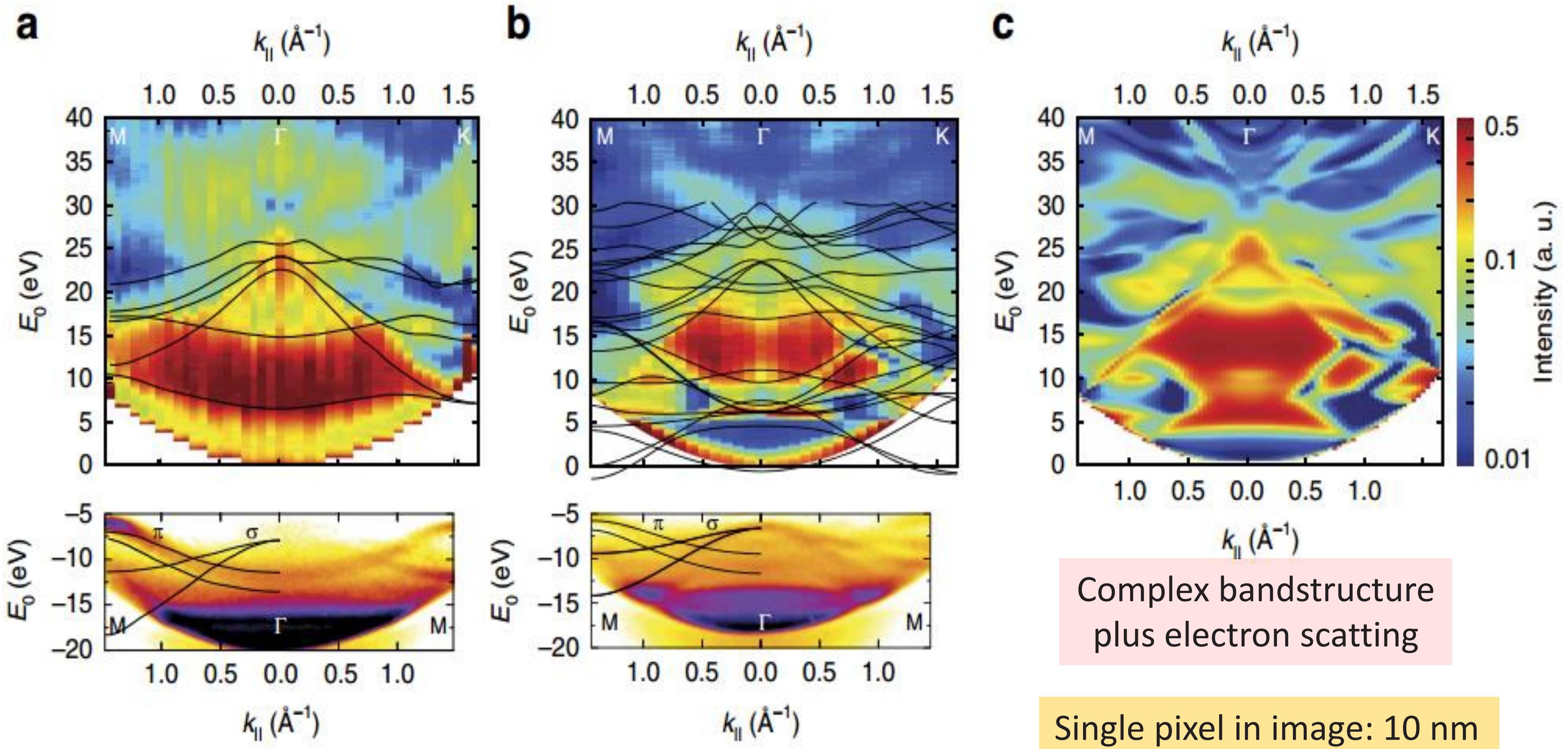
J. Jobst, J. Kautz, D. Geelen, R.M. Tromp, S.J. van der Molen
Nature Comm. 6, 8926 (2015)

Empty State Bands Bulk hBN



J. Jobst, A. v.d. Torren, E. Krasovski, J. Bagley, C.R. Dean, R.M. Tromp, S.J. v.d. Molen

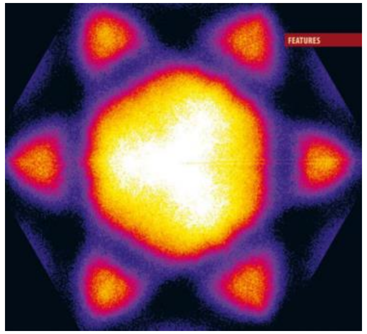
ARPES and ARRES for graphite and hBN



Complex bandstructure plus electron scatting

Single pixel in image: 10 nm

hBN 12 eV



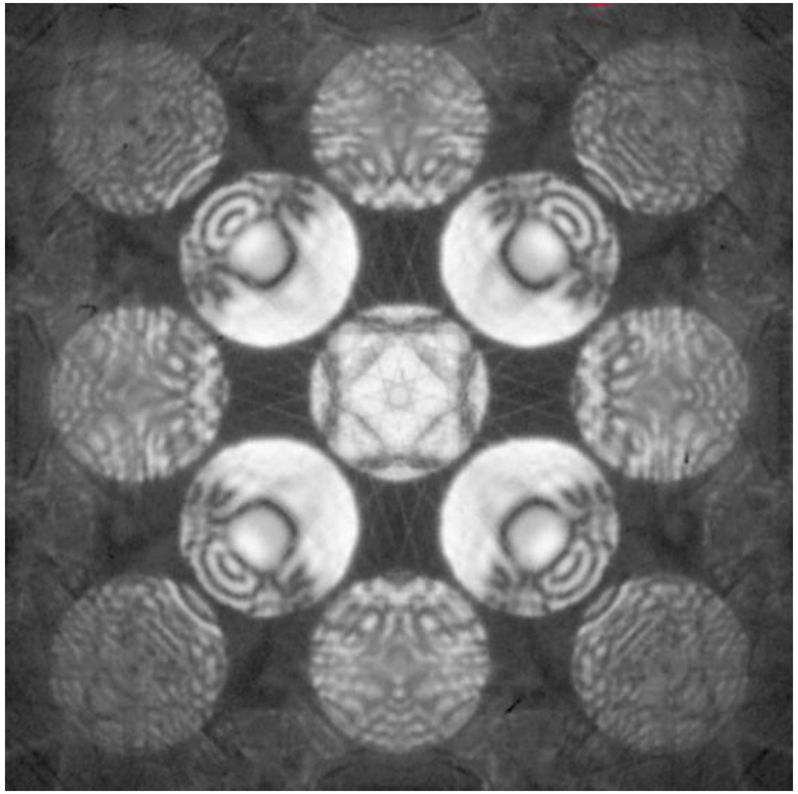
ARRES

ARPES

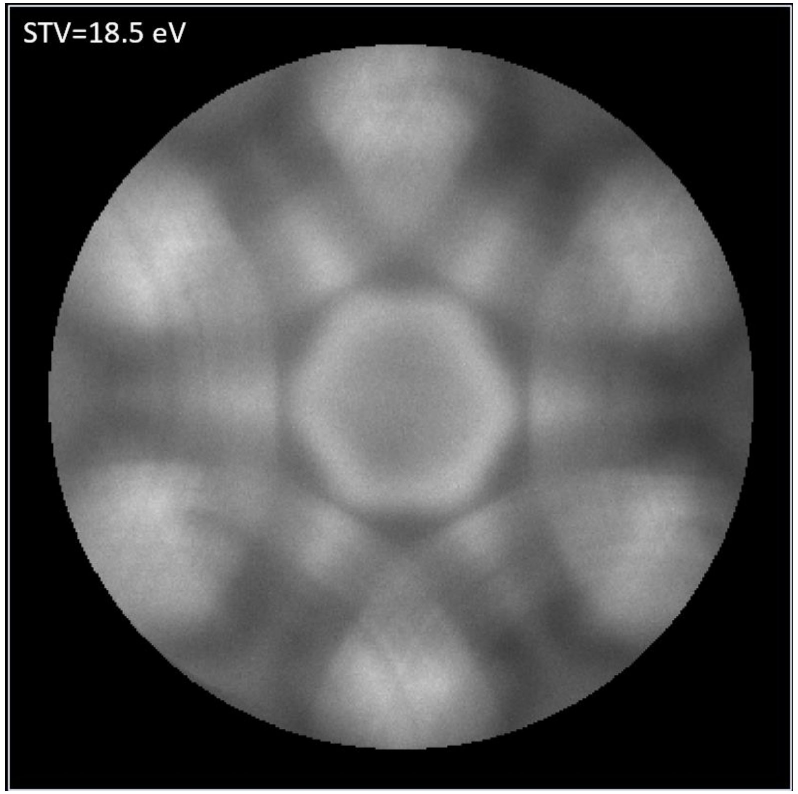
J. Jobst, A. v.d. Torren, E. Krasovskii, J. Bagley, C.R. Dean, R.M. Tromp, S.J. v.d. Molen, Nature communications **7** (2016) 13621

Eugene Krasovskii
(Trieste, 2008)

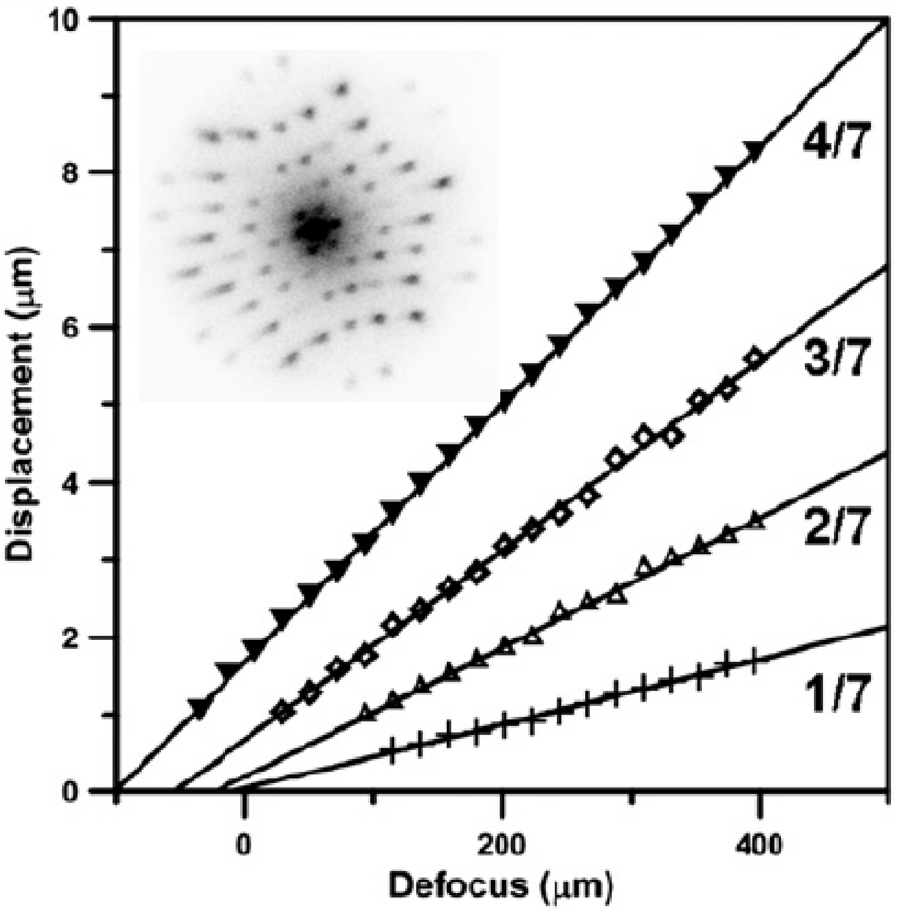
'Instead of angular scan of a parallel beam, why not use a convergent beam?' asked Frank Meyer zu Heringdorf



Jian-Min Zuo
CBED 100 keV

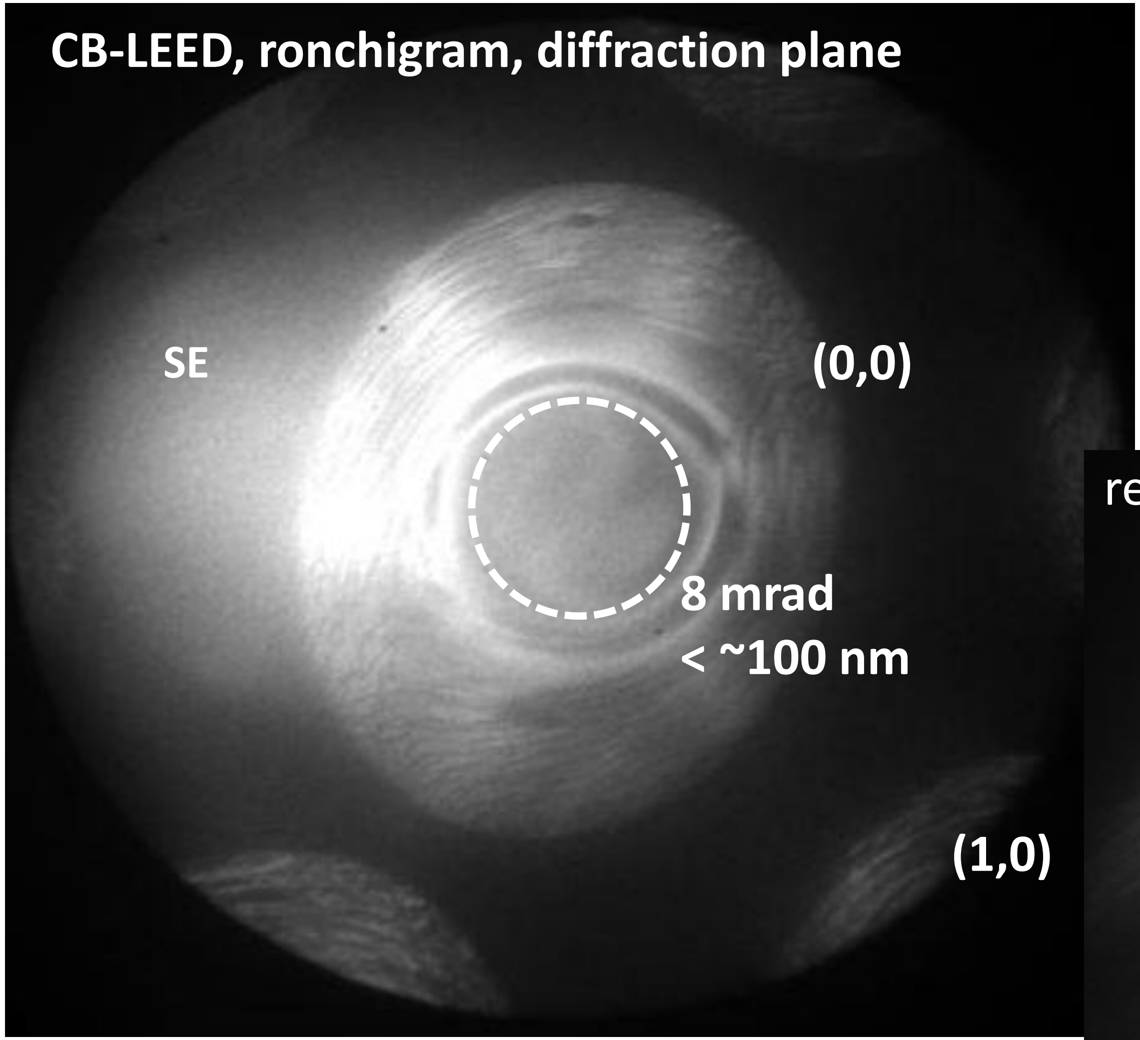


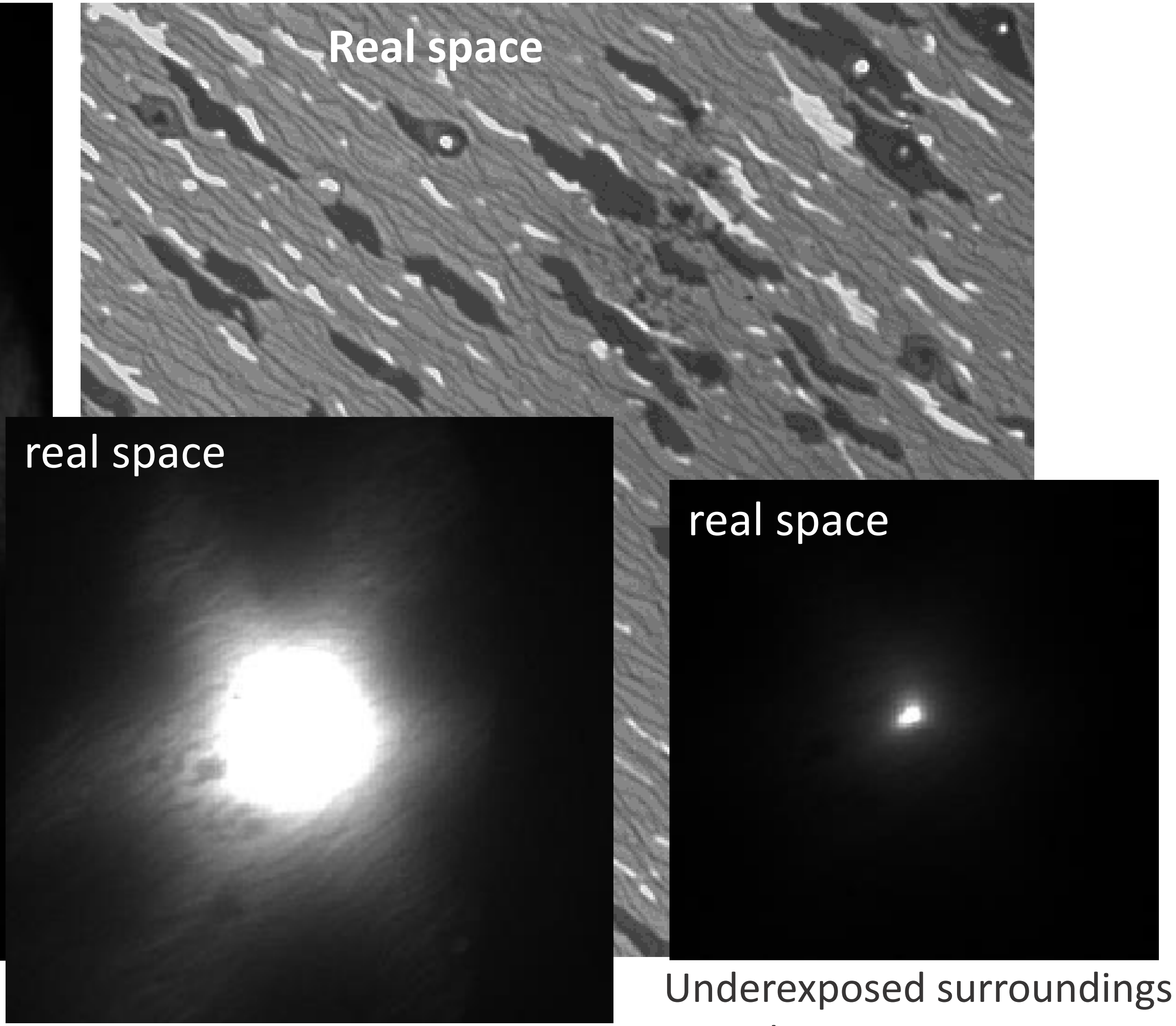
Frank Meyer zu Heringdorf et al.
CB-LEED for Graphene on Pt(111)



Spherical aberration limits spot size

Ronchigrams in LEEM

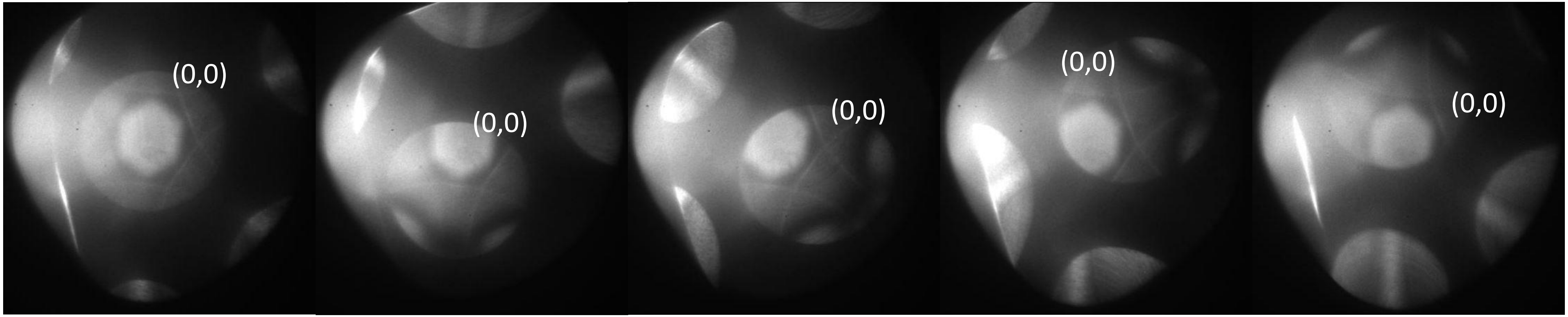




Overexposed central spot

Underexposed surroundings
Spot diameter < 100 nm

Graphene/SiC, several tilts



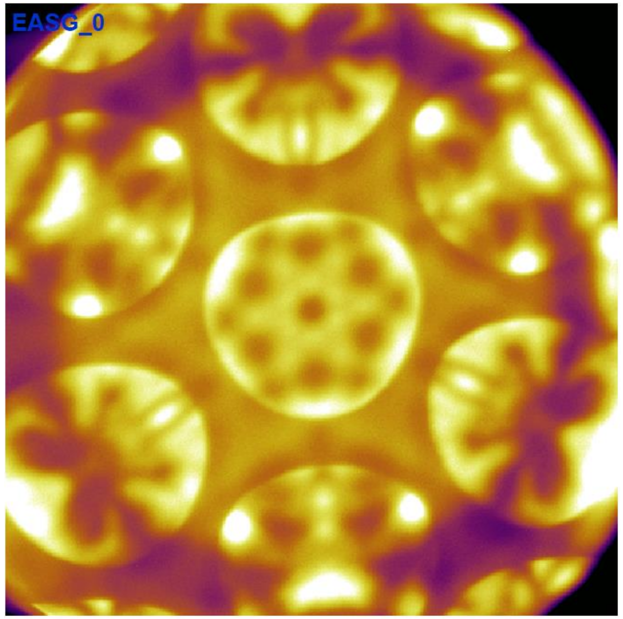
R.M. Tromp, unpublished

Tilt (0,0) disk to see different parts of Brillouin zone.
Avoids beam overlap which is undesirable for spectroscopy.
But: diffracted beams may have additional information

Very promising:

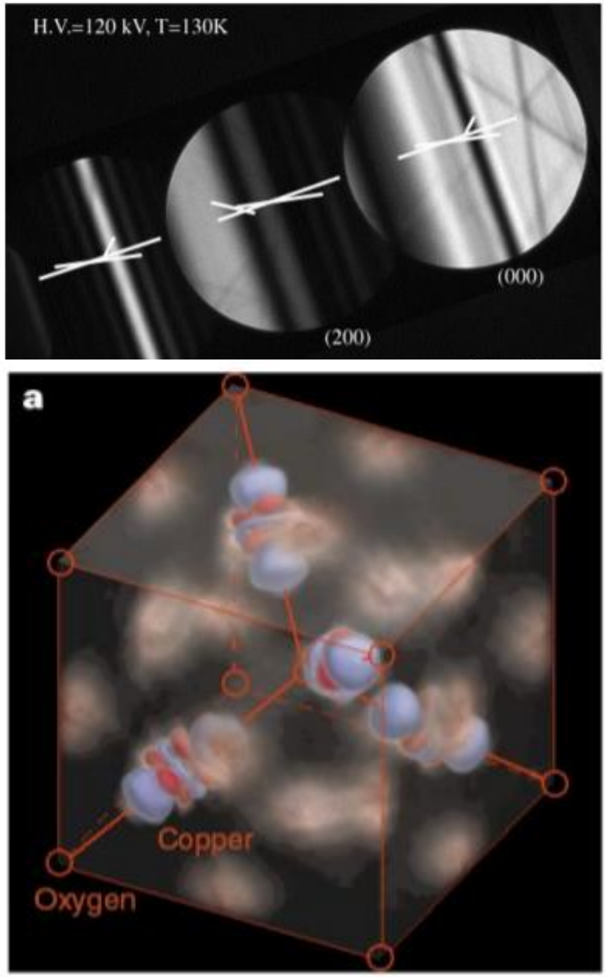
- **High momentum resolution – fine structure visible**
- **Parallel data acquisition (large k-range)**
- **High energy resolution possible**
- **But spatial resolution only ~100 nm**
- **Parallel beam scan resolution ~10 nm**

Frank Meyer zu Heringdorf

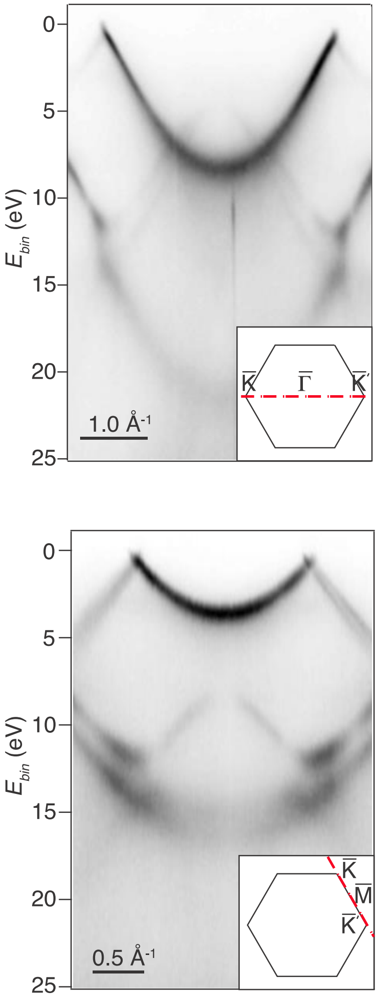


CBED/ARPES/ARRES

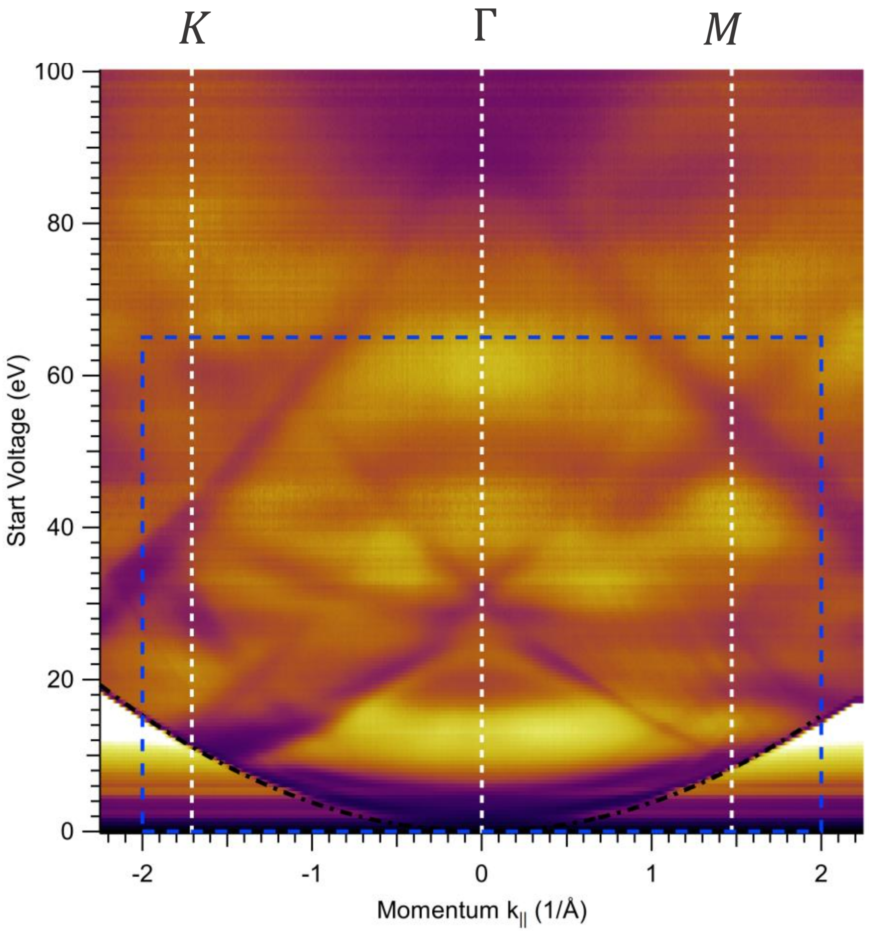
J.M. Zuo, M. Kim, M. O'Keeffe, J. Spence
Direct observation of d-orbital holes in Cu-Cu bonding in Cu_2O
NATURE 401 (1999) 49-52



CBED:
Valence-electron density



ARPES:
Valence-electron
Band structure



CB-LEED/ARRES:
Conduction-electron
Band structure

Aberration Correction in Electron Optics: Walter Henneberg / Alfred Recknagel 1937

June 27, 1939. W. HENNEBERG ET AL 2,163,787

ELECTRON DEVICE

Filed April 26, 1937

Fig. 1

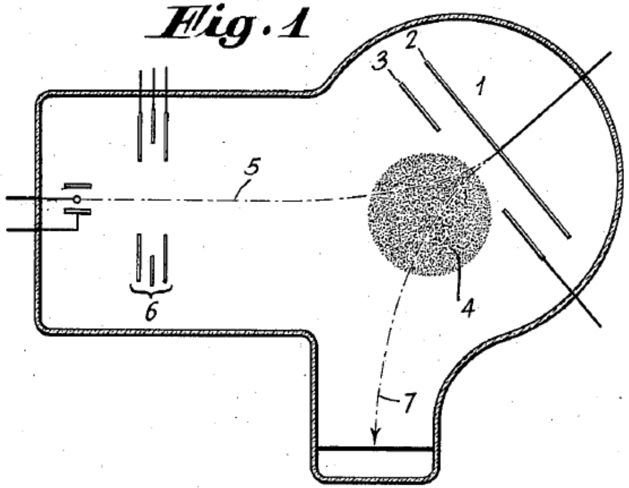
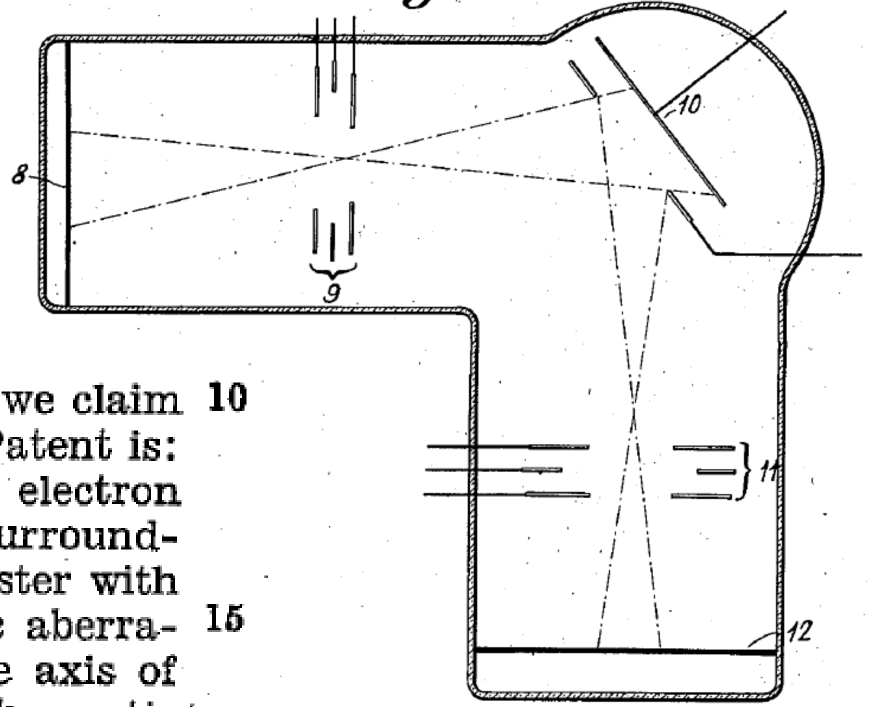


Fig. 2

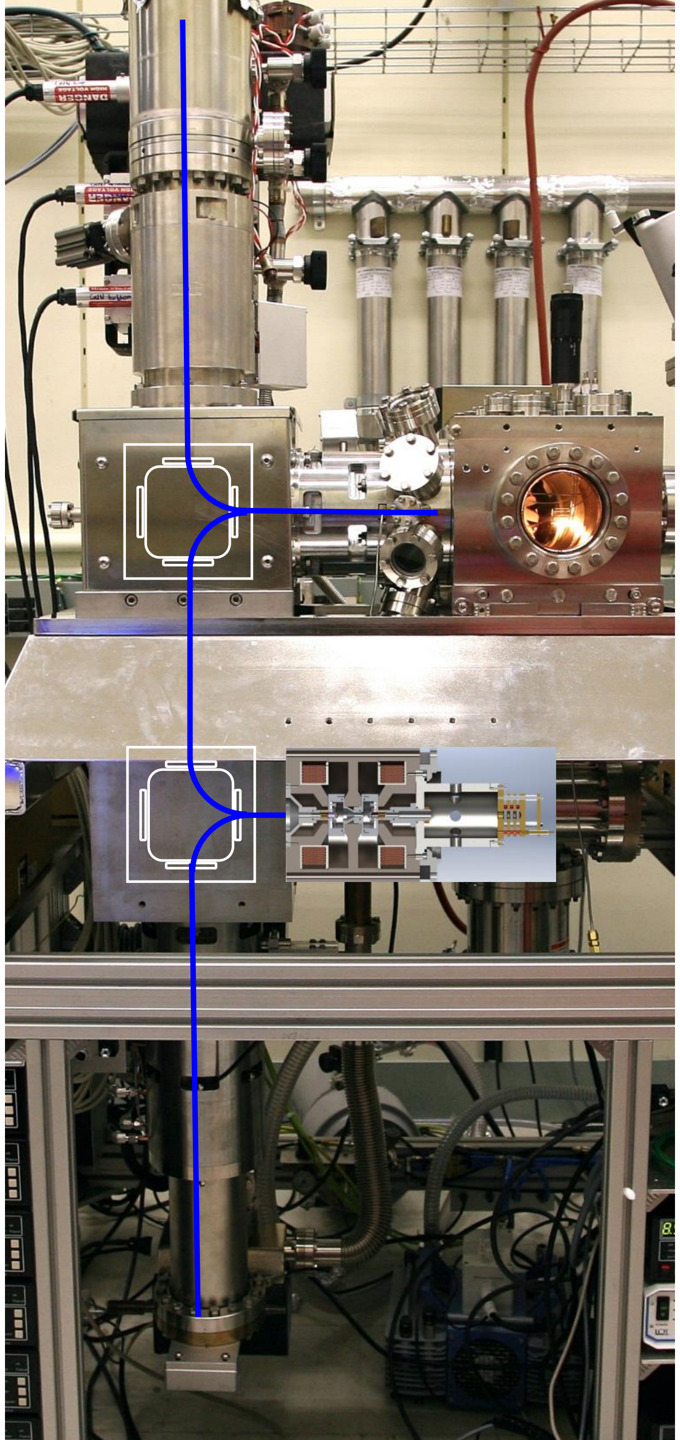


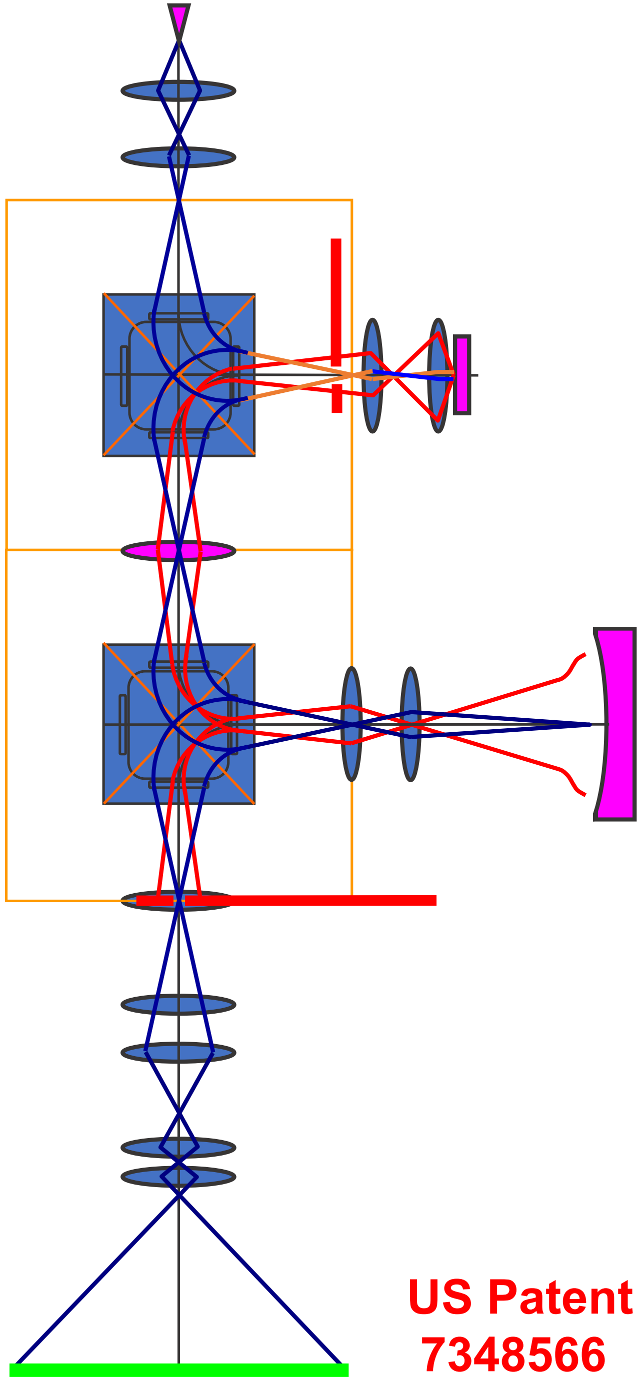
Having described our invention, what we claim as new and desire to secure by Letters Patent is:

1. A cathode ray tube comprising an electron emitting cathode, a control electrode surrounding the cathode, an electron lens in register with the cathode, said lens having chromatic aberration, an electron mirror inclined to the axis of the electron lens, said mirror having chromatic aberration complementary to the aberration of said lens and an impact surface having its axis at right angles to the axis of the electron lens.

INVENTORS
WALTER HENNEBERG
ALFRED RECKNAGEL
BY H. S. Grover
ATTORNEY

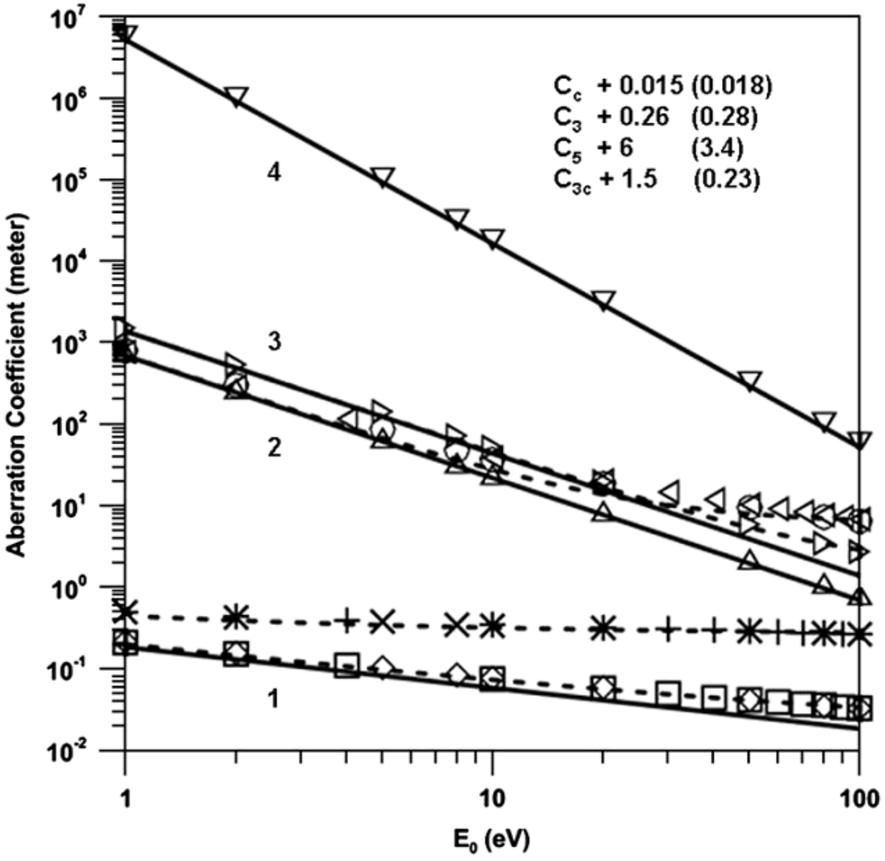
R.M. Tromp, J.B. Hannon, A.W. Ellis, W. Wan, A. Berghaus, O. Schaff, Ultram. **110** (2010) 852-861





US Patent 7348566

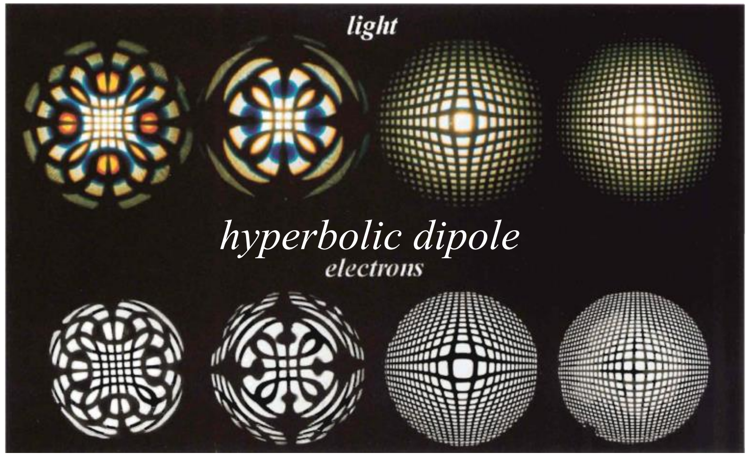
Aberrations strongly energy dependent



$$C_3 = -C_c = L\sqrt{E/E_0}$$

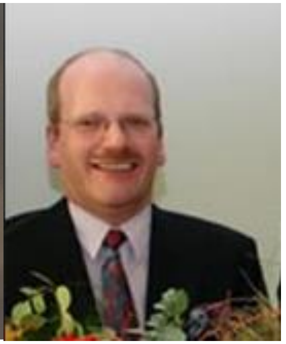
4-element electron mirror: f, C_c, C_3 adjustable

Gertrude Rempfer

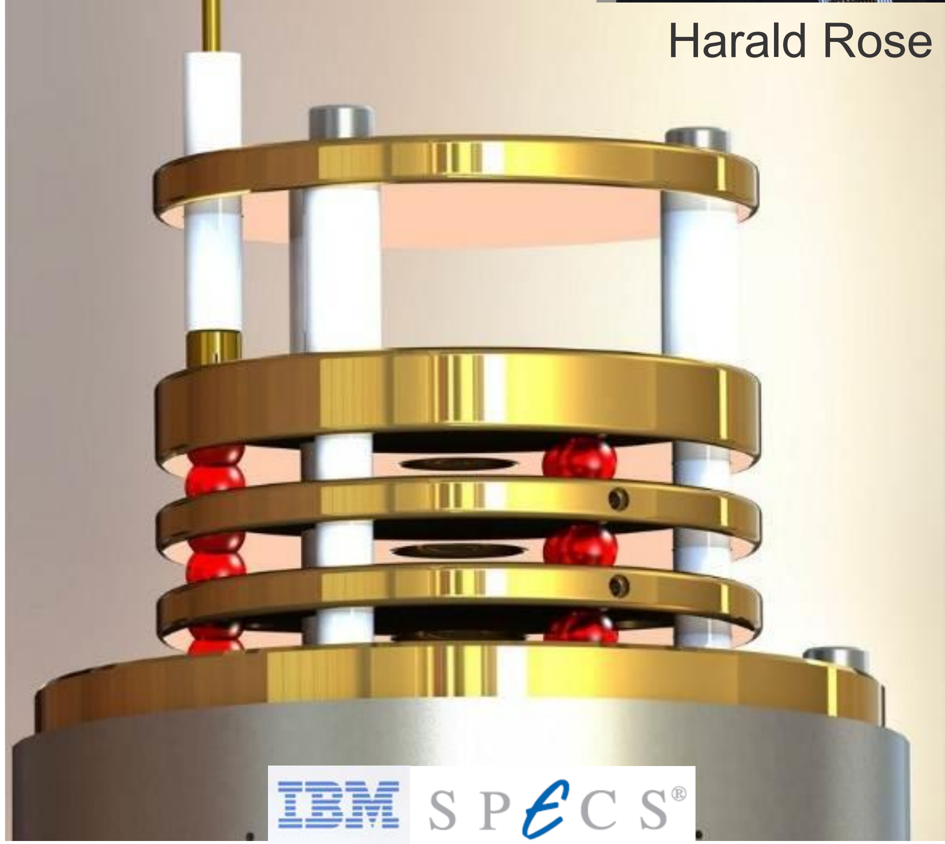


G. Rempfer et al. Microsc. Microanal. 3, 14-27, 1997

Harald Rose



Dirk Preikszas



ground plane

mirror plane, V_1

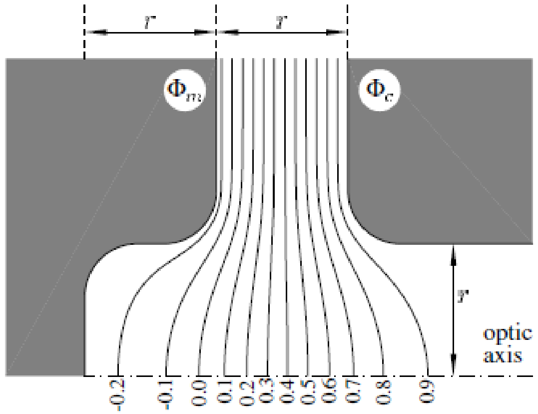
lens element, V_2

lens element, V_3

ground plane

Three power supplies
18 kV
1ppm stability

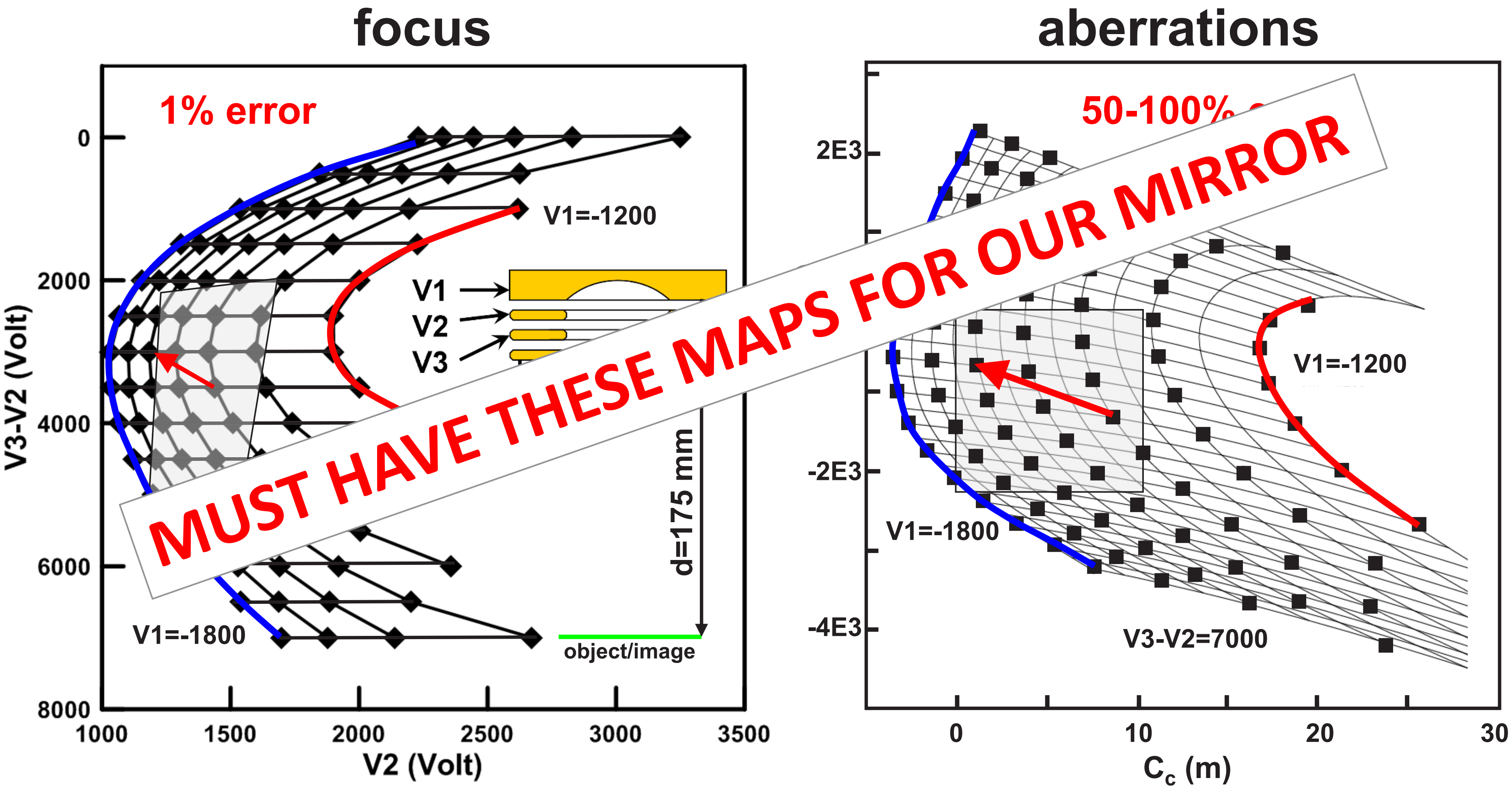
Boundary Element vs. Finite Difference



	Author(s)	Voltage	image Z	C_3	C_5	C_c
BEM	CPO 'benchmark'	-3.198593	-0.119994(2)	-532.37(6)	-8.90(3)E5	-9.623(4)
	D Preikszas	-3.198593	-0.120000	-532.9	-8.743E5	-9.62
	R Tromp (Munro)	-3.198612	-0.120000	-532.89	-8.737E5	-9.624
Finite Difference	D Preikszas	-3.1936	-0.11646	-531.8	-8.747E5	-9.62
	B Lencova	-3.1936	-0.12008	-495	-1.18E6	-9.60
	B Lencova/J Zlamal	-3.1936		-487	-1.29E6	
	E Munro et al 1995	-3.1936		-667	-28.7E6	-8.40

Source: CPO user manual, xmpl2d17, mirror with negative aberrations, Frank Read

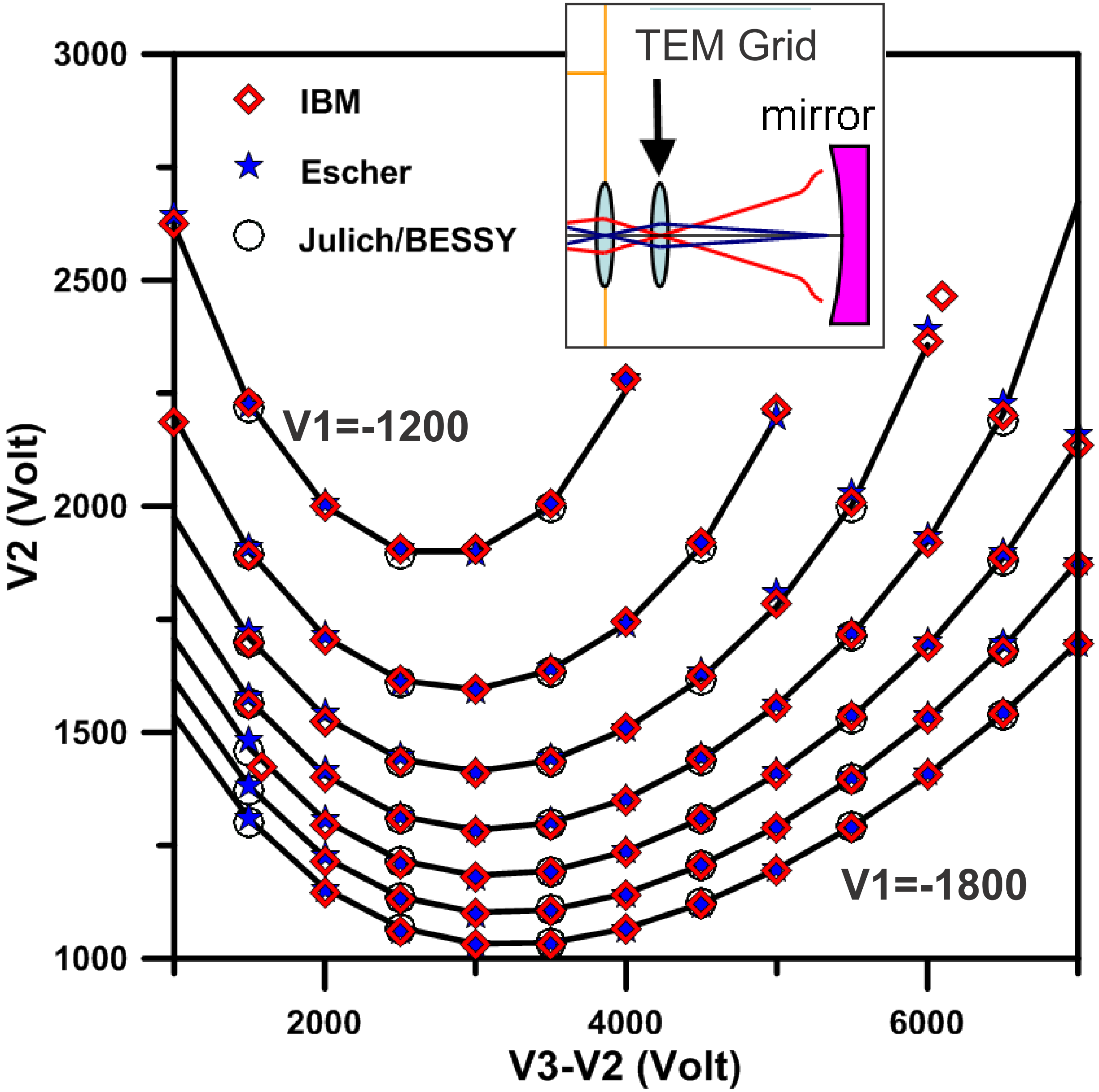
Correlation of first and higher order properties

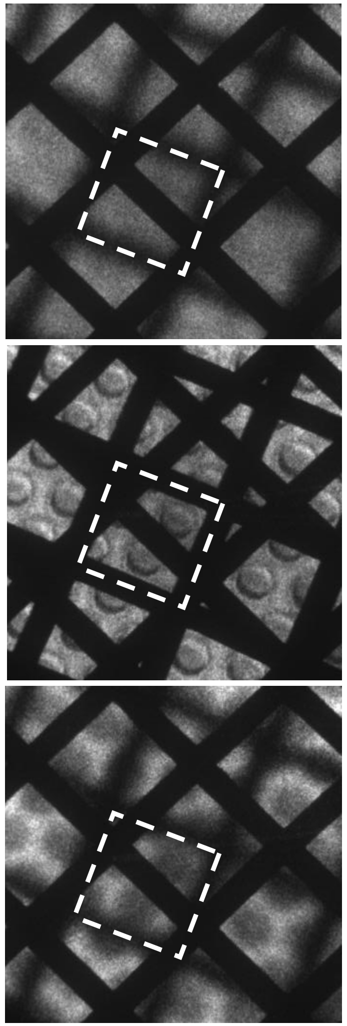


R.M. Tromp, J.B. Hannon, A.W. Ellis, W. Wan, A. Berghaus, O. Schaff, Ultram. **110** (2010) 852

IBM

First order properties, first order of business

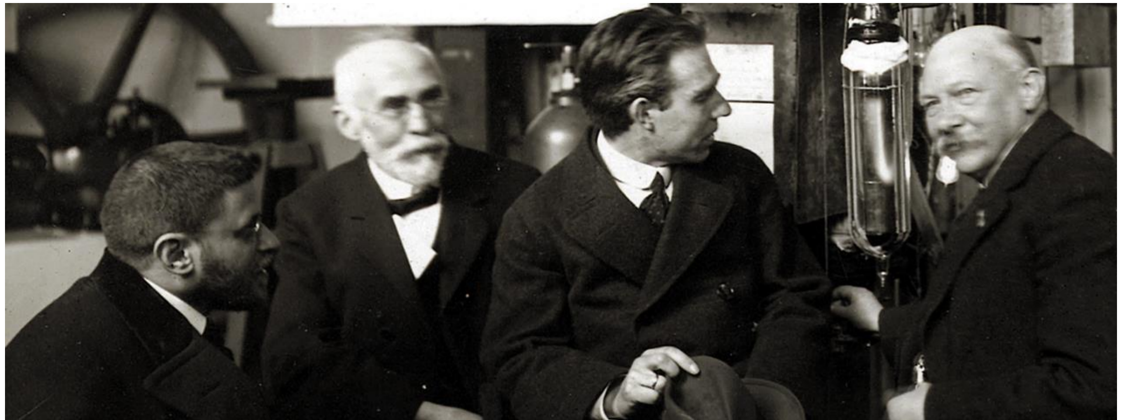




Adjustable parameter:
V1 offset of **10 V**
(out of 16500, i.e. **0.06%**)

R.M. Tromp, J.B. Hannon, W. Wan, A. Berghaus, O. Schaff, Ultramicroscopy http://dx.doi.org/10.1016/j.ultramic.2012.07.016

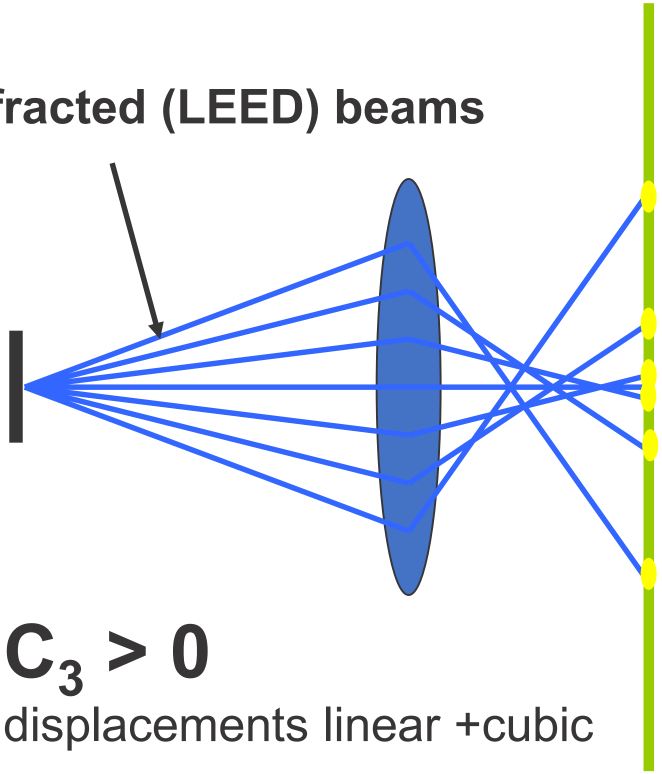
To correct we must measure



Heike Kamerlingh Onnes:
‘Door meten tot weten’

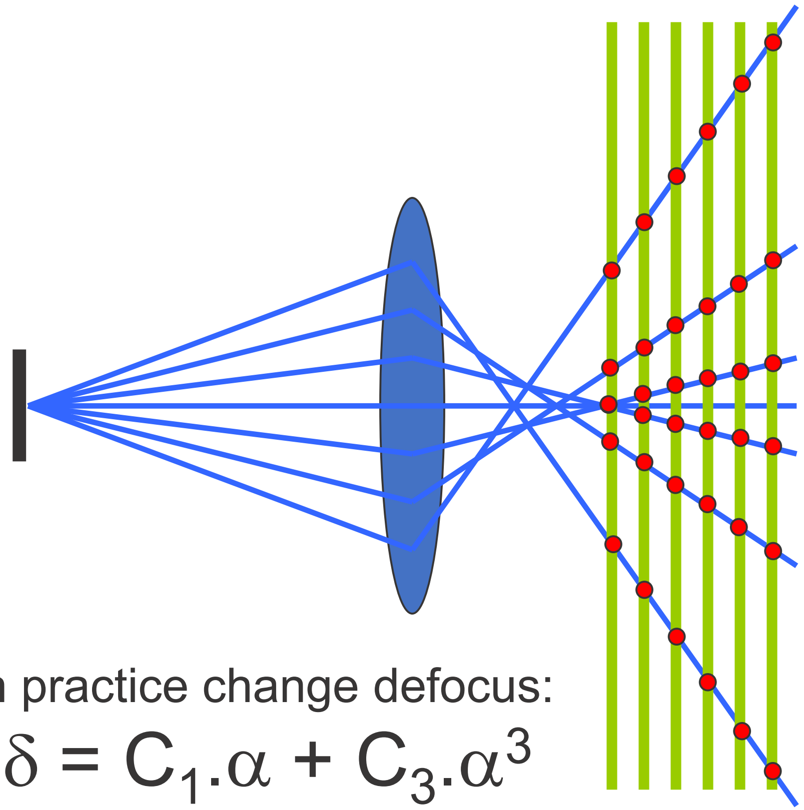
Direct visualization of C_3 using micro-illumination

Diffracted (LEED) beams



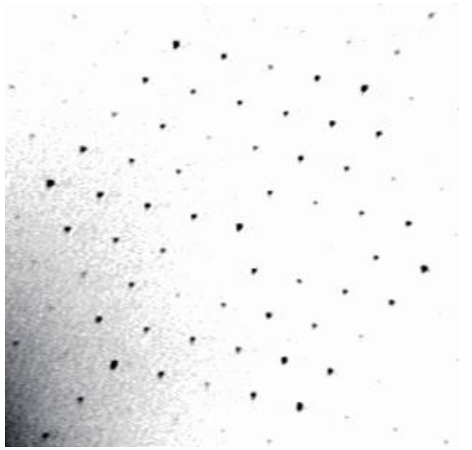
$C_3 > 0$

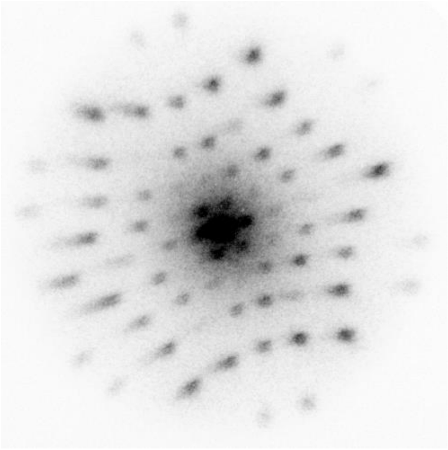
displacements linear +cubic



In practice change defocus:

$$\delta = C_1.\alpha + C_3.\alpha^3$$



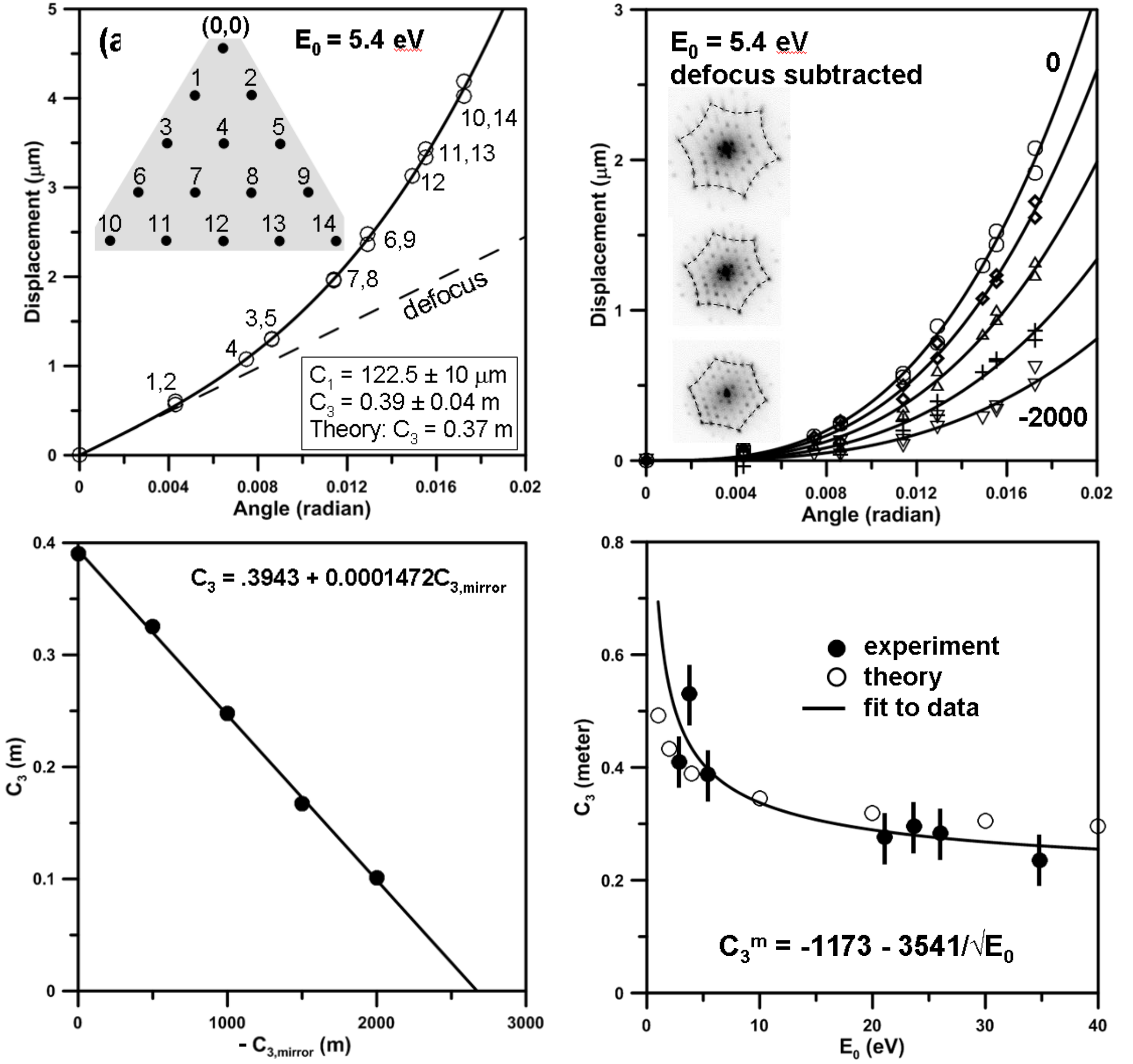


Si(111)(7x7) 5.4 eV

A. Berghaus O. Schaff J. Hannon

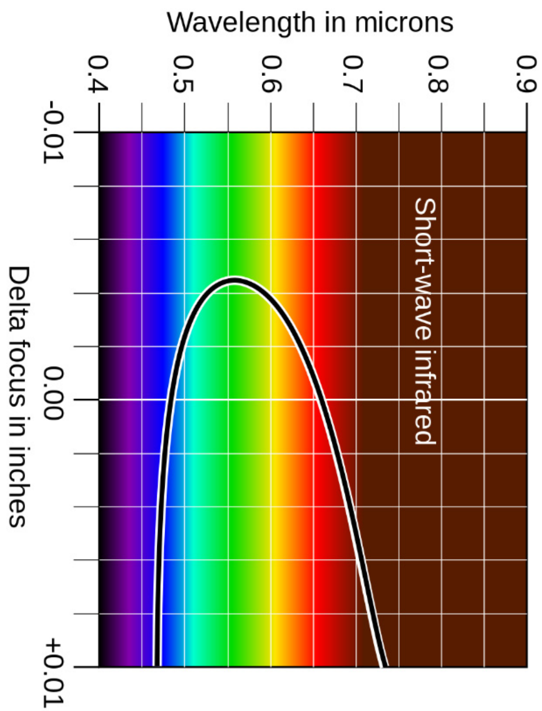
R.M. Tromp, J.B. Hannon, W. Wan, A. Berghaus, O. Schaff, Ultramicroscopy http://dx.doi.org/10.1016/j.ultramic.2012.07.016

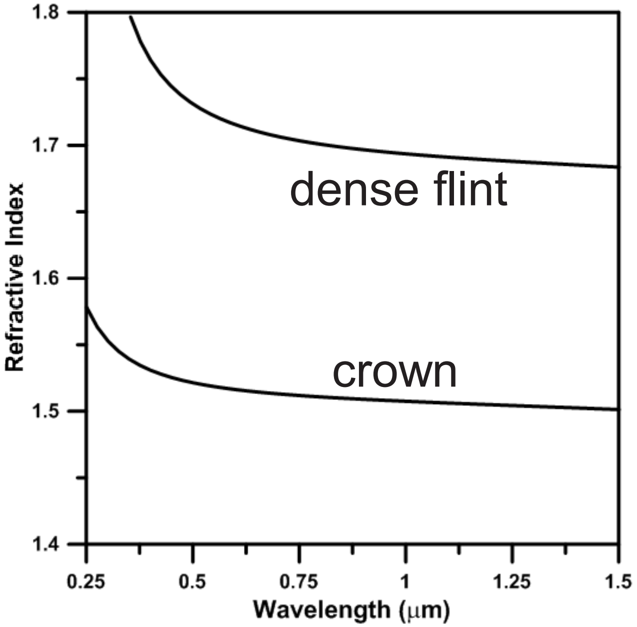
Measurement and Correction of C_3

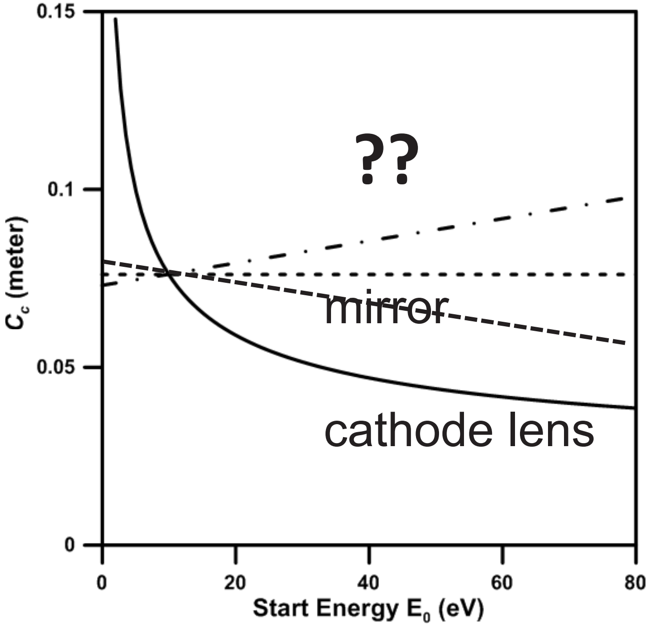


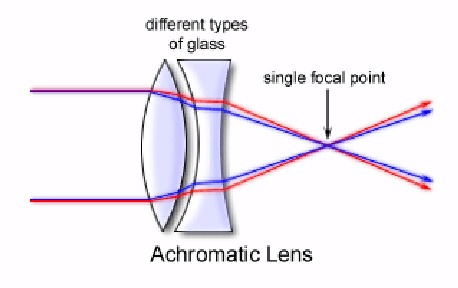
R.M. Tromp, J.B. Hannon, W. Wan, A. Berghaus, O. Schaff,
Ultramicroscopy http://dx.doi.org/10.1016/j.ultramic.2012.07.016

An achromat for photons…… and electrons?



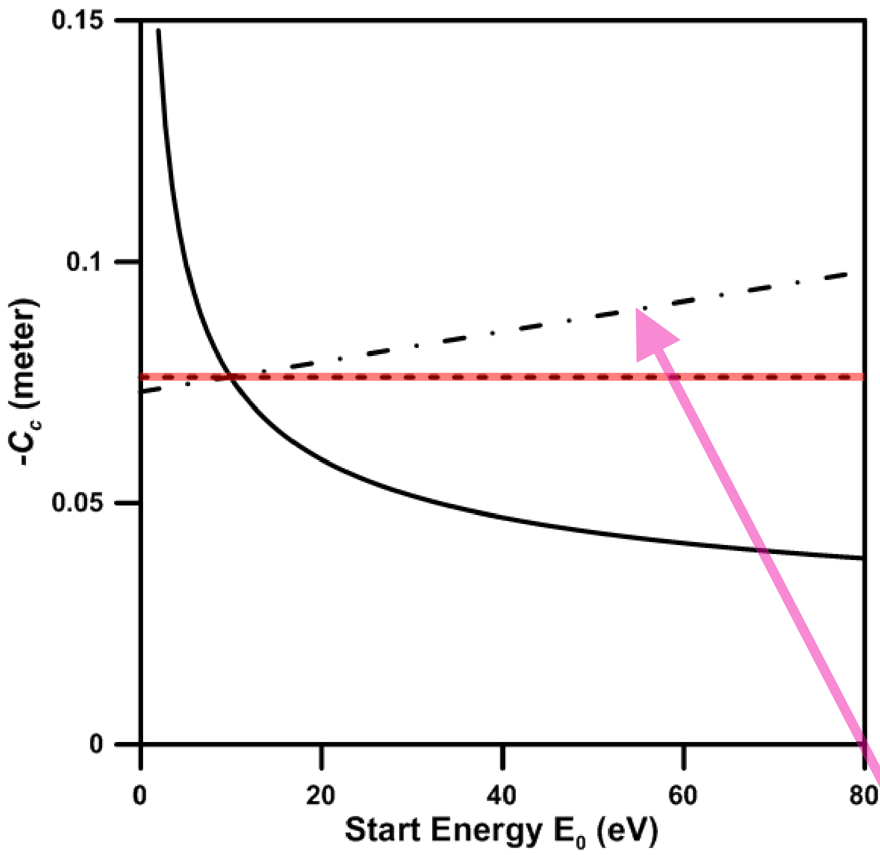


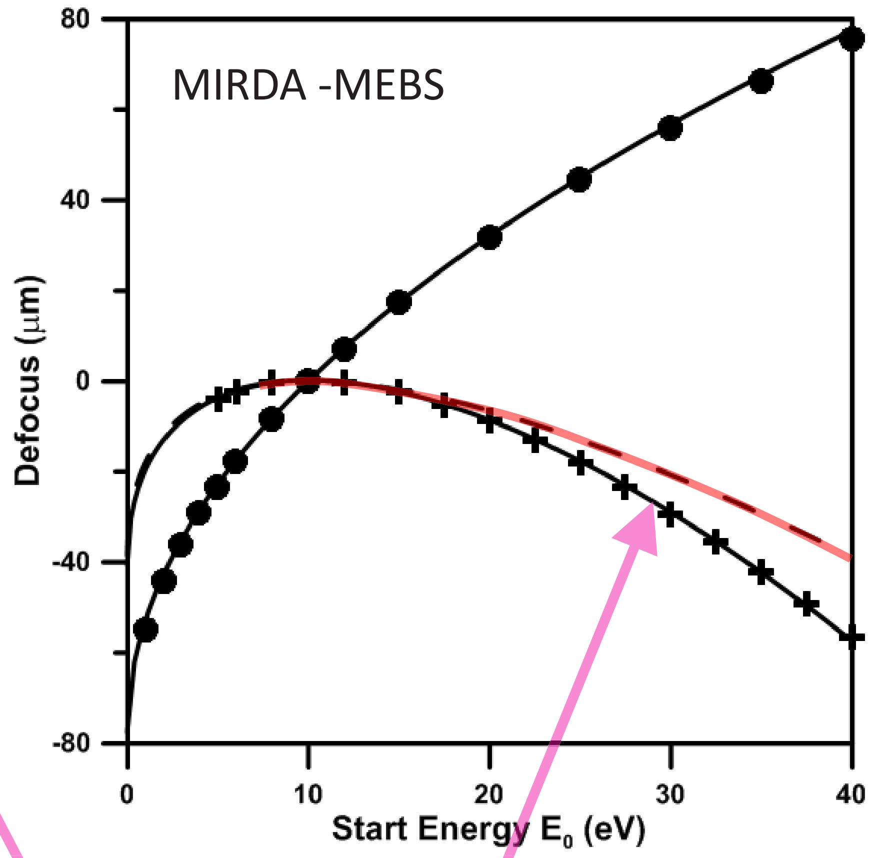




$$C_c = -L\sqrt{(E/E_0)}$$

Chromatic aberration of the mirror is not constant



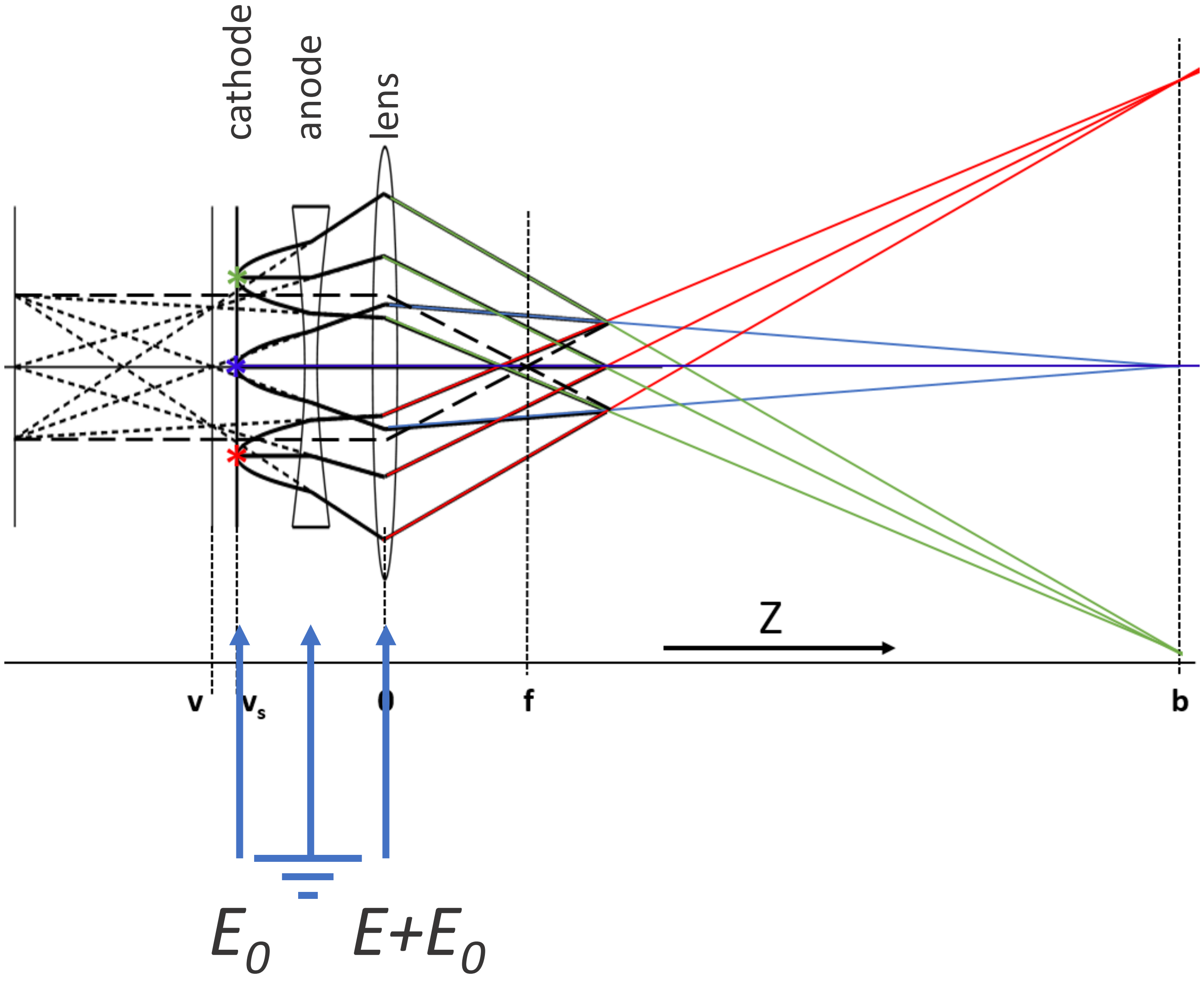


$$\Delta f = -L\frac{\left(\sqrt{E_C}-\sqrt{E_0}\right)^2}{\sqrt{E.E_C}} - \beta(E_c - E_0)^2/E^2$$

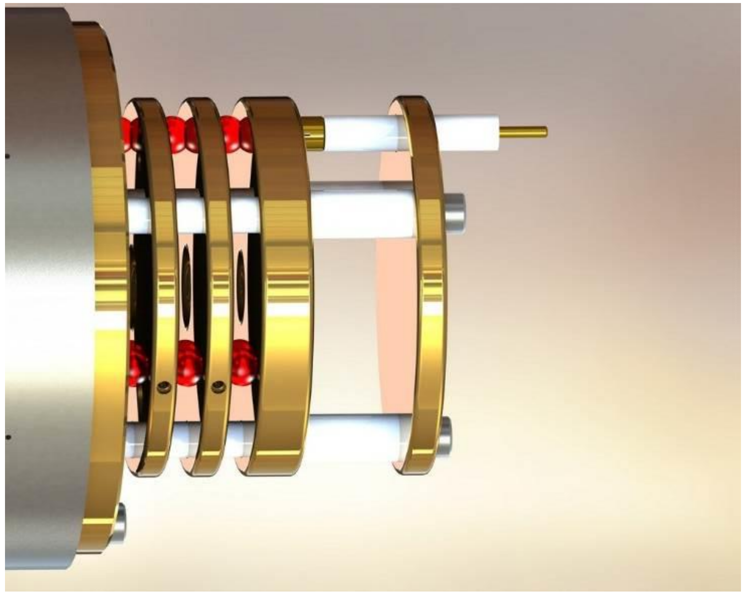
mirror dispersion (C_{cc})

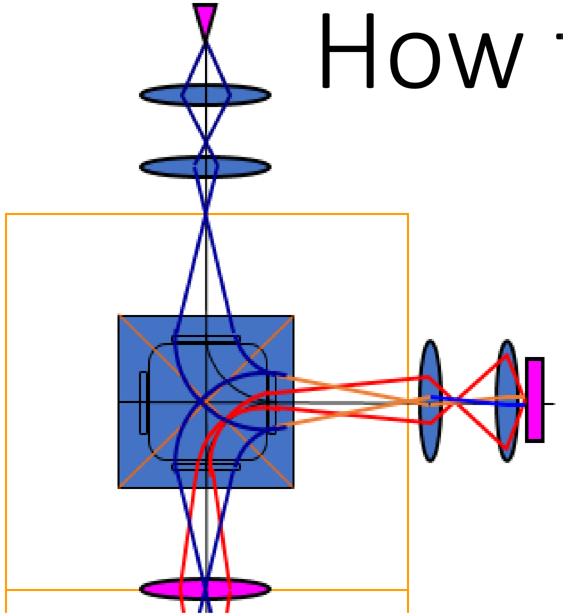
How to measure chromatic aberration?

Defocus changes with take-off energy E_0, and with final energy $E+E_0$, both.



+





How to measure chromatic aberration?

Three options:

1. Gun voltage ($E+E_0$) constant, change sample voltage: changes E_0, but not $E+E_0$
   Measures uniform field aberration only, not magnetic lens, not electron mirror
2. Use photoelectrons, change sample bias: changes E, but not E_0
   Measures magnetic lens and mirror, but not uniform field
3. Change gun voltage, sample bias constant: changes both E_0 and $E+E_0$
   Measures all three
   BUT: also changes deflection angles through prism arrays and sample illumination
   MUST adjust illumination for each data point + **large off-axis** aberrations in mirror path

Measure C_c without changing electron energy?

Electron energy changes

Reference energy constant

$$\frac{E+dE}{E} = 1 + \frac{dE}{E}$$

$$\frac{E+dE}{E} \cong \frac{E}{E-dE}$$

Electron energy constant

Reference energy changes

$$dC_1 = C_c \frac{dE}{E}.$$

Measure C_c without changing electron energy?

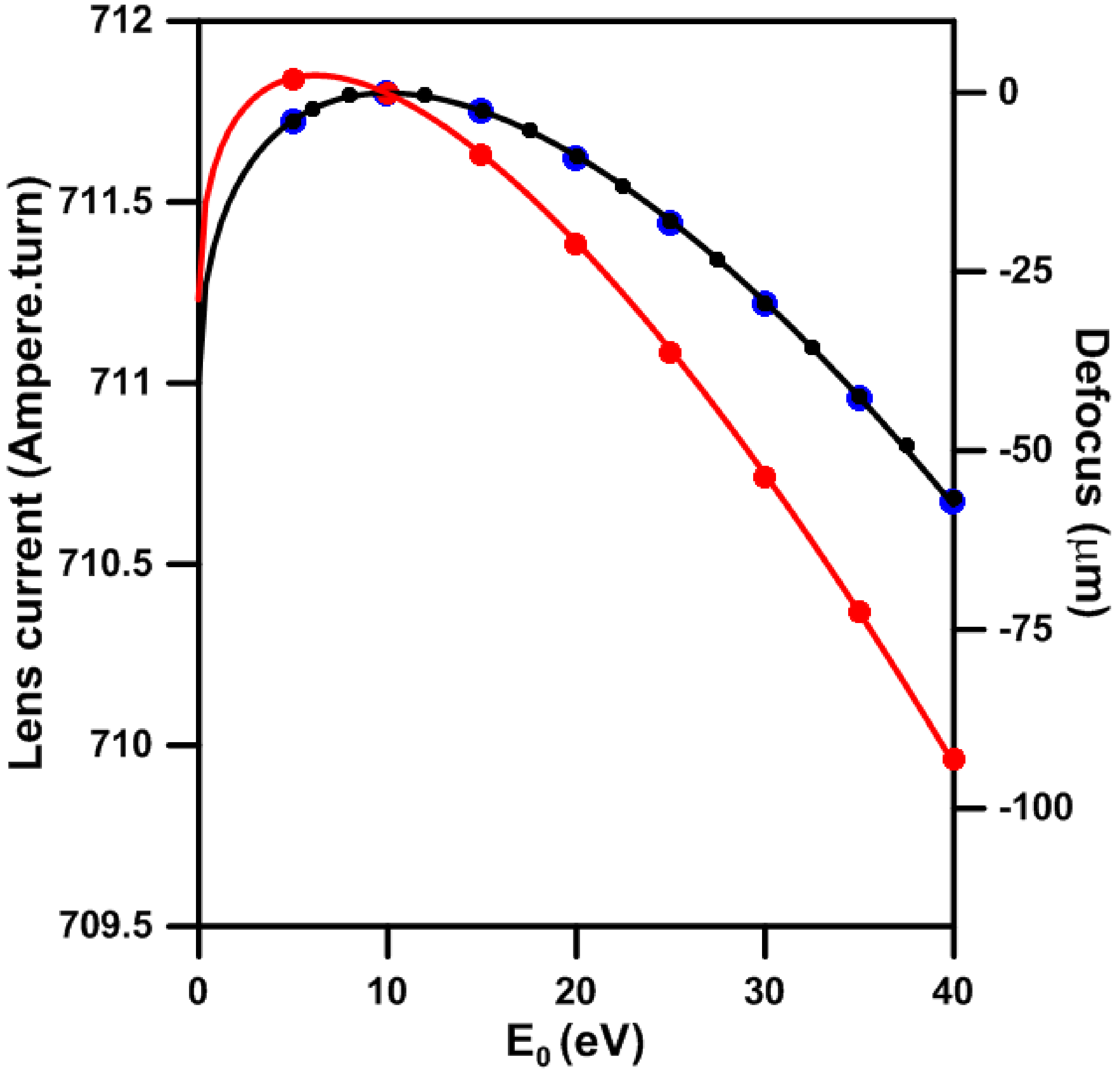
Reference energy = nominal column energy
All focal lengths are fixed relative to the nominal column energy.

Now:
- Keep electron energy fixed (no problems with alignment)
- Change reference energy, but keep all focal lengths constant *for the reference energy*.

$$\frac{dV}{V} = \frac{dE}{E}$$ (electrostatic lenses and electron mirror elements)

$$\frac{dI}{I} = \frac{1}{2}\frac{dE}{E}$$ (magnetic lenses)

A little trick to measure C_c with fixed electron energy

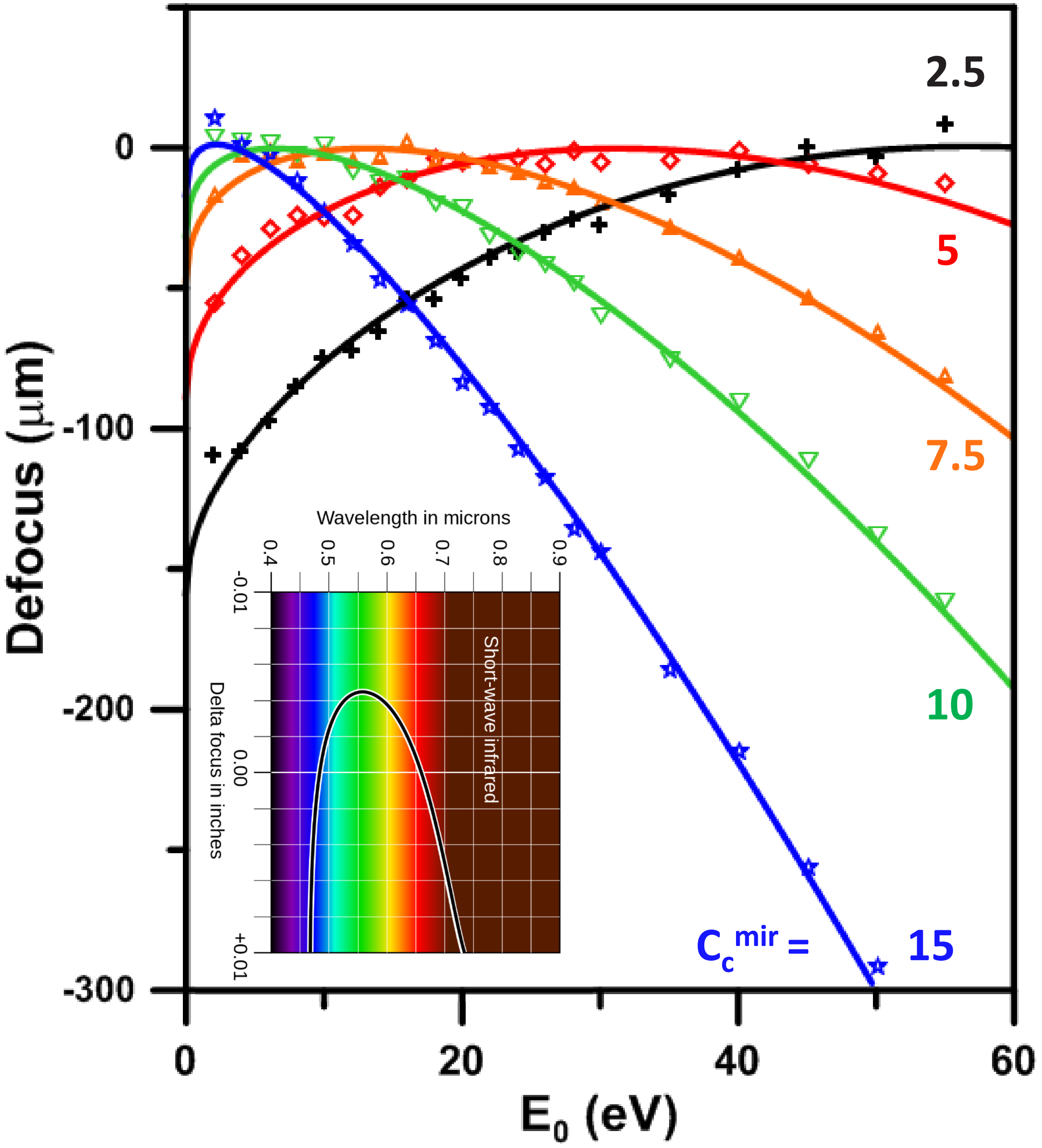


Raytracing (MEBS):

- ● Change electron energy
- ● Change reference energy
- ○ Correct for objective lens reference excitation

● = ○ **to better than 1:10^5**

$$\Delta C_1 = -L\frac{\left(\sqrt{E_c}-\sqrt{E_0}\right)^2}{\sqrt{E_c E_0}} - C_{cc}^{mir}(E_c - E_0)^2/E^2$$



Experiments on graphene/SiO_2

Measurement time is minutes, not hours

Small macro plugin adjusts reference energy (magnetic and electrostatic settings) automatically with sample bias.

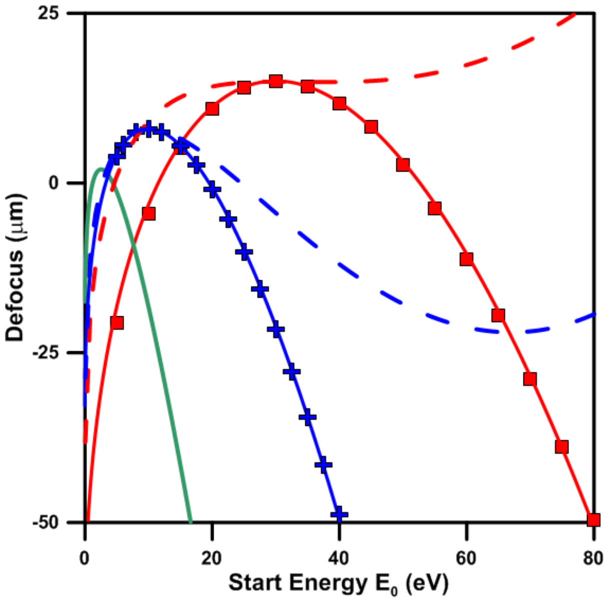
Lines: fits with E_c as only parameter

$$\Delta C_1 = -L\frac{\left(\sqrt{E_c}-\sqrt{E_0}\right)^2}{\sqrt{E_c E_0}} - C_{cc}^{mir}(E_c - E_0)^2/E^2$$

Mirrors :

- Have been around a long time (Recknagel)
- Can correct C_c, C_3, (C_5)
- Are very compact compared to multipole optics
- Are used successfully in LEEM/PEEM, SEM
- Have promise for LV-TEM
- Are relatively poorly understood
- But raytracing has excellent predictive value

If we learn how to control dispersion (C_{cc}), then we can make a PEEM apochromat (resolution 2x, transmission 10x).

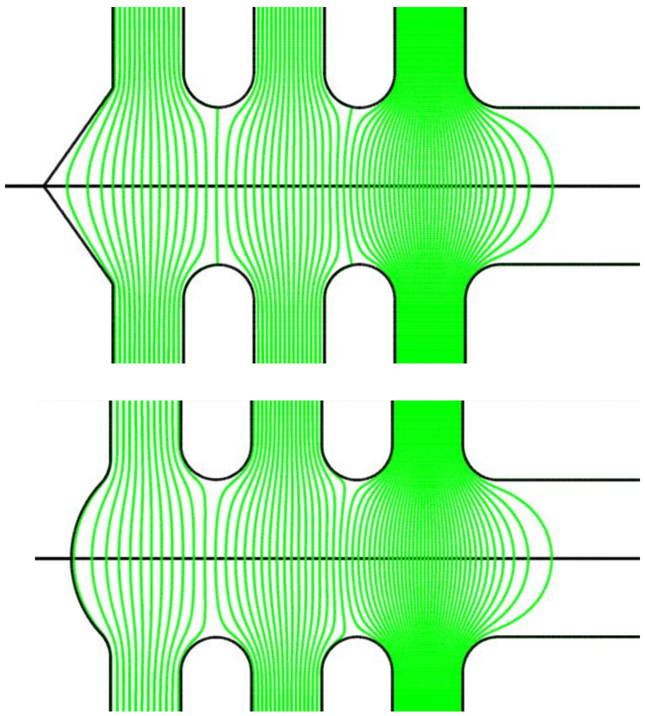


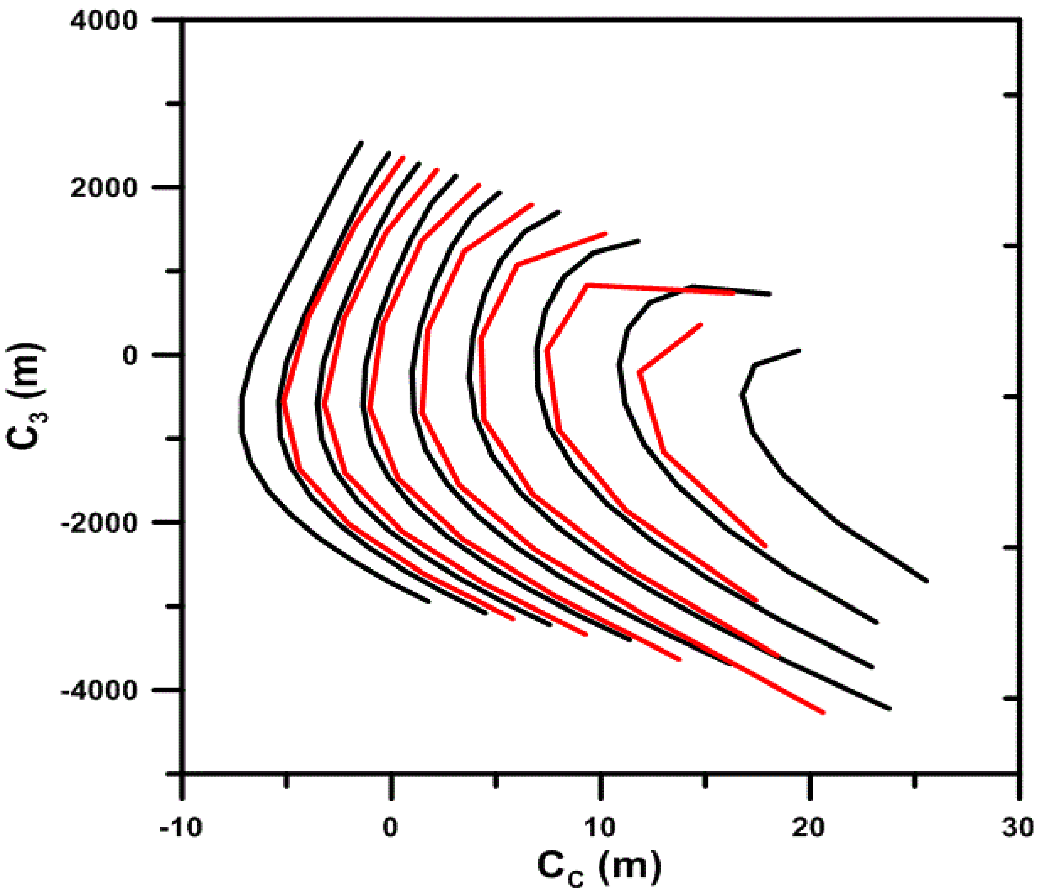
Maybe the most poorly understood component in electron optics

$$\Delta f = -L\frac{\left(\sqrt{E_c}-\sqrt{E_0}\right)^2}{\sqrt{E.E_c}} - \beta(E_c - E_0)^2/E^2$$

CHANGE SIGN

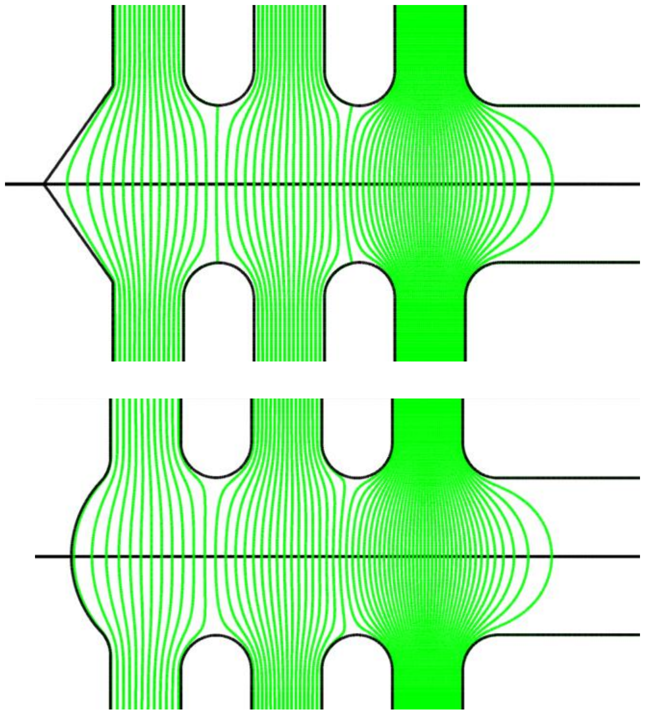
Does the shape of the mirror matter?

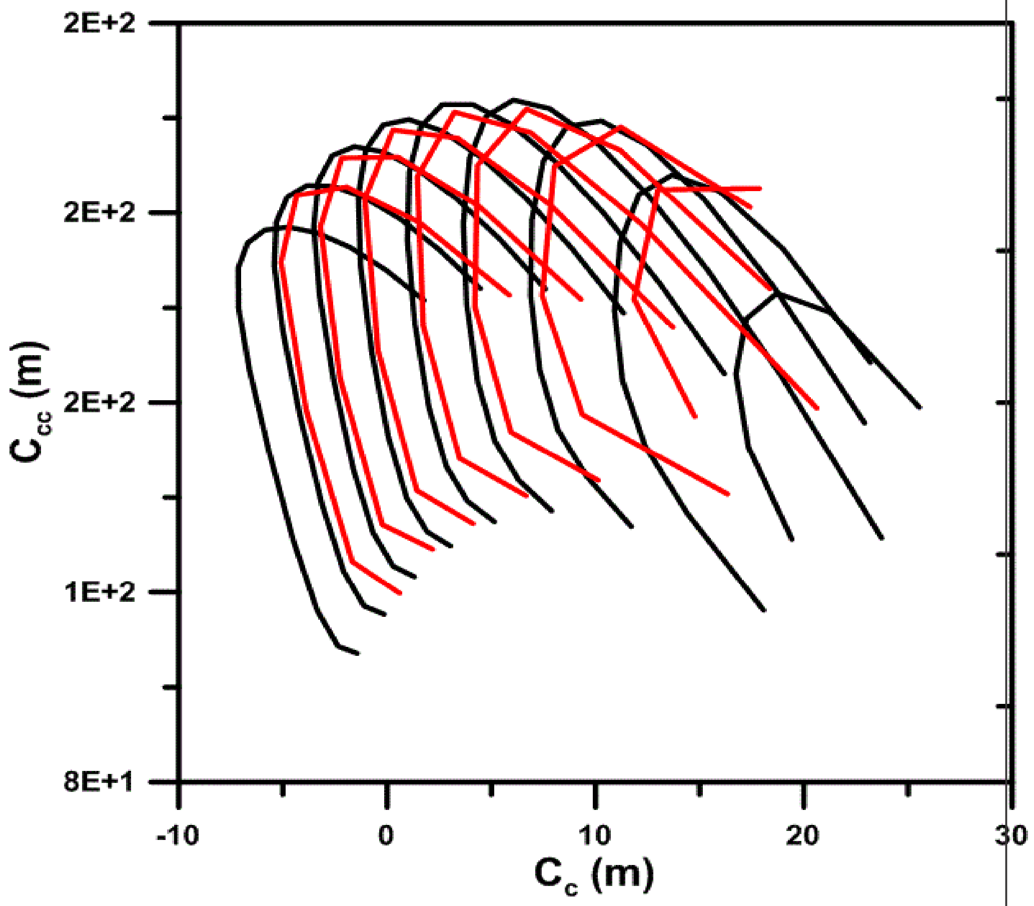




Not here….

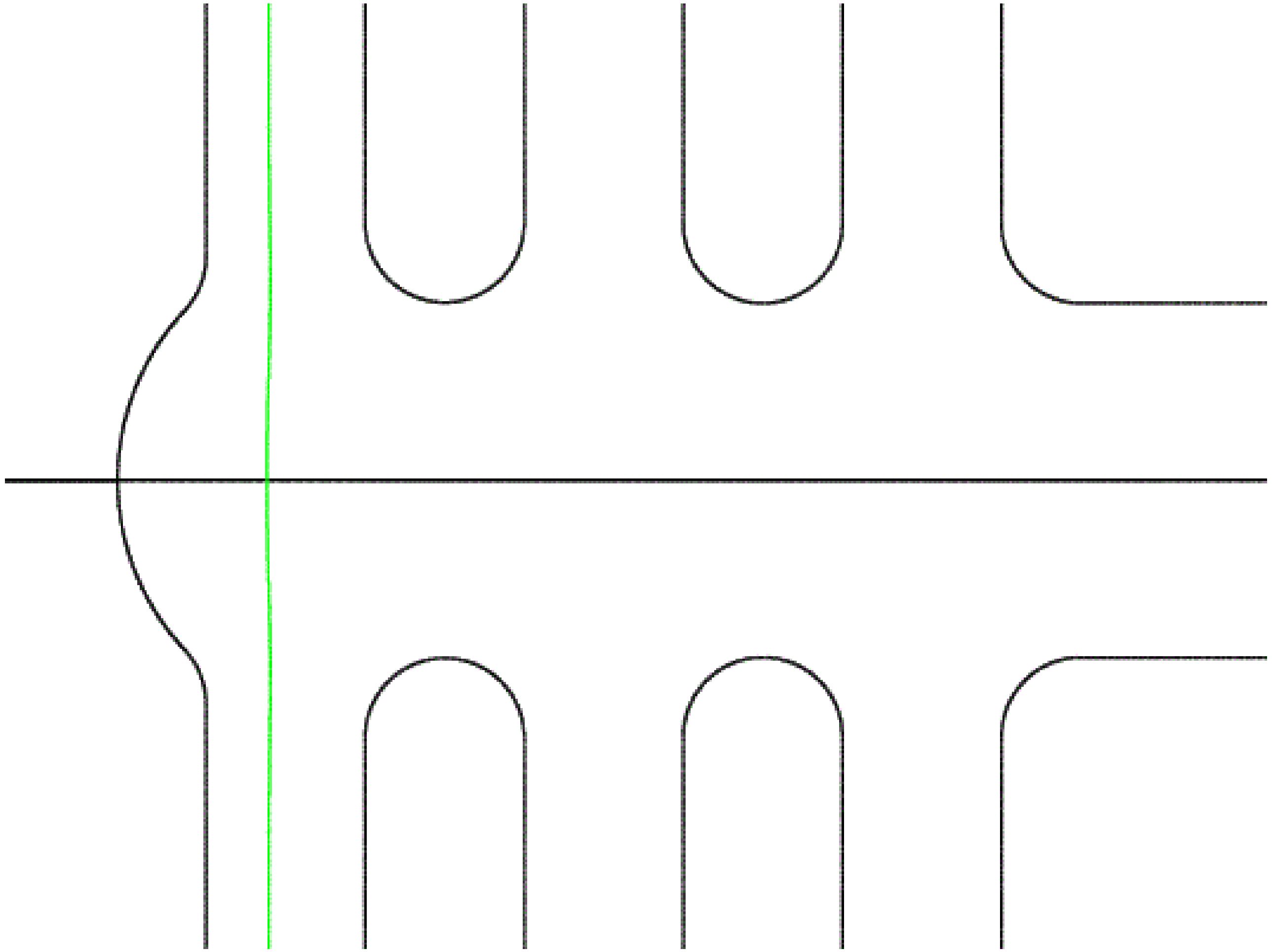
Does the shape of the mirror matter?





…or here….

The reflecting equipotential is far away, and can be either concave or convex...



How to design for desired properties? Can we flip the sign of C_{cc}?

Jim Hannon
Art Ellis

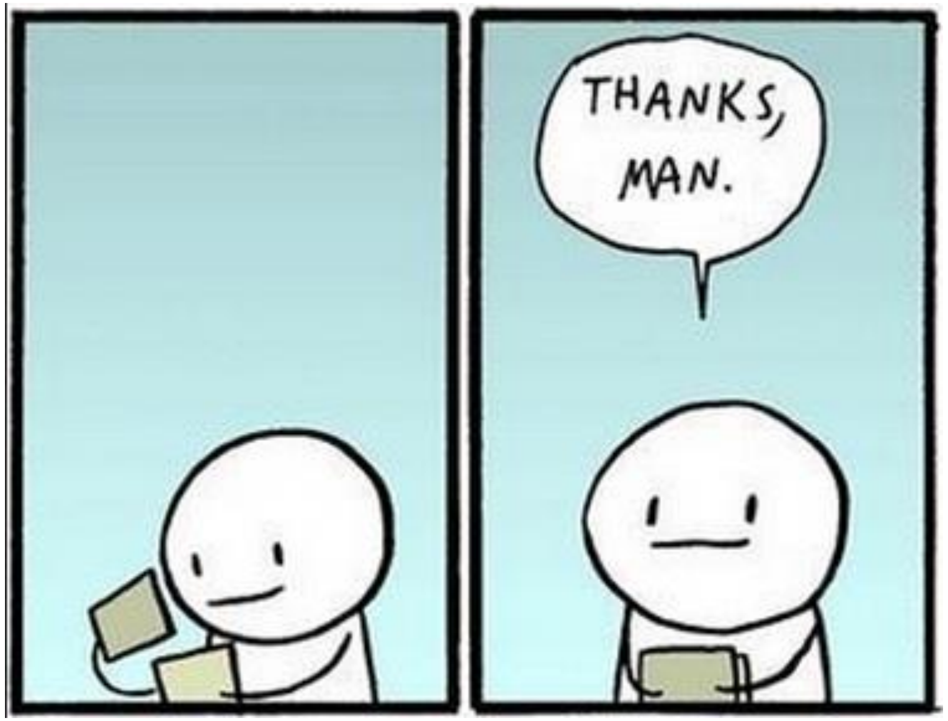
Weishi Wan

Johannes Jobst
Tobias de Jong
Daniel Geelen
Sense Jan van der Molen

Oliver Schaff
Alexander Kaiser
Andreas Berghaus

Eugene Krasovksii

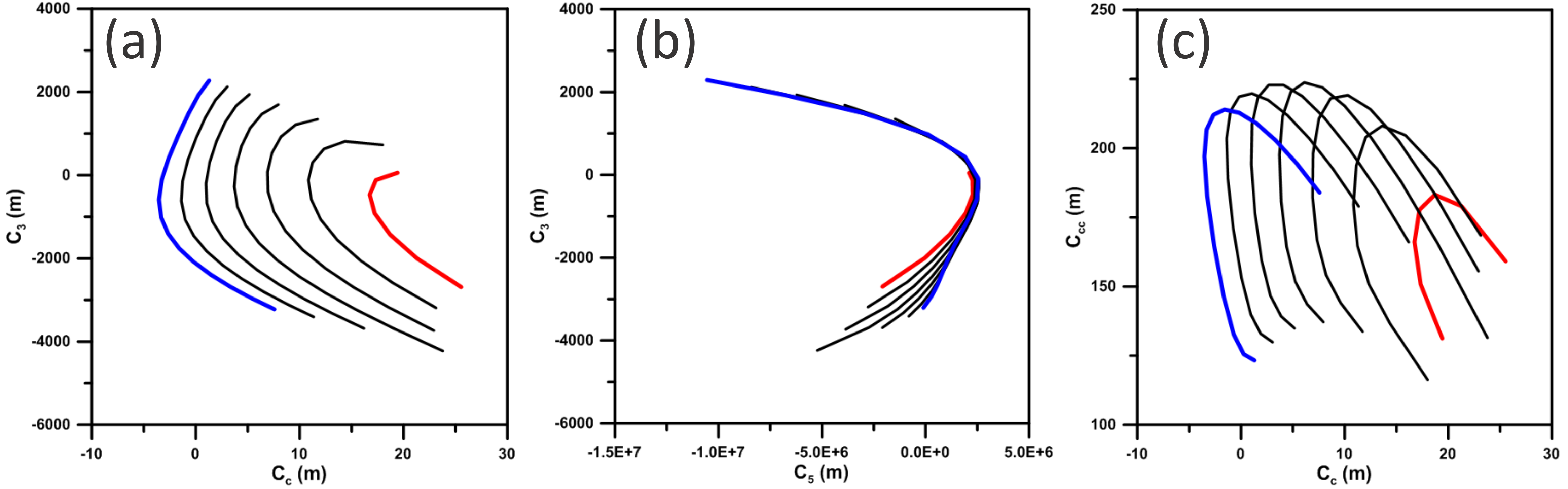
Frank Meyer zu Heringdorf



Mark Reuter
2017 AVS Hanyo Award

Arthur Ellis
2010 AVS Hanyo Award

Relevant geometric and chromatic aberrations



When, at the court of Chou, he first inspected the ancestral shrines and the arrangements for the great annual sacrifices to Heaven and Earth, he exclaimed: “As we use a glass to examine the forms of things, so must we study the past to understand the present.” (said about Confucius 551–479 BC)

IV scan in Convergent Beam

- Convergent Beam Diffraction
  - Graphene / Ir(111)
  - Incident electron beam is made convergent
  - LEED Spots fill entire screen (and overlap)
- Works With and Without Energy Filtering
  - This series is without energy filtering, as can be judged by the secondaries that disperse to the left of the picture at about STV=6 eV

Frank Meyer zu Heringdorf et al.

Data set: ZBVH

Overlap of beams obscures structure of interest