Computation and measurement of aberrations in low energy electron microscopy

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

R.M. Tromp, M. Mankos, M.C. Reuter, A.W. Ellis, M. Copel
## Lab-based LEEM/PEEM imaging modes

<table>
<thead>
<tr>
<th>PEEM imaging</th>
<th>LEED Atomic Structure</th>
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<tr>
<td>Hg light source</td>
<td>Selected Area LEED Local Atomic Structure 200 nm</td>
</tr>
<tr>
<td>Mirror Microscopy Topography Work Function</td>
<td>LEEM-EELS Local Electronic Structure Spectroscopy + Imaging</td>
</tr>
<tr>
<td>Bright field LEEM Phase contrast</td>
<td>eV-TEM Transmission imaging without damage</td>
</tr>
<tr>
<td>Bright field LEEM Reflectivity Structure factor</td>
<td>Transmission EELS Low energy loss on the nanoscale</td>
</tr>
<tr>
<td>Dark field LEEM Structure symmetry</td>
<td></td>
</tr>
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</table>
## Lab-based LEEM/PEEM imaging modes

<table>
<thead>
<tr>
<th>Method</th>
<th>Description</th>
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<tbody>
<tr>
<td>SPLEEM</td>
<td>Magnetic domain imaging</td>
</tr>
<tr>
<td>ARRES</td>
<td>Empty state band structure</td>
</tr>
<tr>
<td>PEEM-ARPES</td>
<td>Filled state band structure</td>
</tr>
<tr>
<td>LEEM-IV Imaging</td>
<td>Local Atomic Structure 2-5 nm</td>
</tr>
<tr>
<td>LEEM potentiometry</td>
<td>Contact-less nanoscale device measurements</td>
</tr>
<tr>
<td>IV-SPLEEM</td>
<td>Magnetic quantum Well asymmetry</td>
</tr>
<tr>
<td>SPA-LEED-PLD</td>
<td>Atomic Layer Oscillations during PLD growth</td>
</tr>
<tr>
<td>SPA-LEED</td>
<td>Local strain measurement</td>
</tr>
<tr>
<td>CBED</td>
<td>Local Atomic and Electronic structure</td>
</tr>
<tr>
<td>LEEM lithography</td>
<td>Structure fabrication with few eV electrons</td>
</tr>
</tbody>
</table>
## Synchrotron-based PEEM imaging modes

<table>
<thead>
<tr>
<th>PEEM-IV Imaging</th>
<th>Dynamic Imaging</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 nm resolution</td>
<td>In-situ processing</td>
</tr>
<tr>
<td>Local chemistry</td>
<td>Elemental/chemical</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Linear Magnetic</td>
<td>Picosecond</td>
</tr>
<tr>
<td>Dichroism</td>
<td>Resolution Imaging</td>
</tr>
<tr>
<td>Antiferromagnetism</td>
<td></td>
</tr>
<tr>
<td></td>
<td>100 ps</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Circular Magnetic</td>
<td>Localized</td>
</tr>
<tr>
<td>Dichroism</td>
<td>Spectroscopy</td>
</tr>
<tr>
<td>Ferromagnetism</td>
<td>Elemental, chemical</td>
</tr>
<tr>
<td></td>
<td>Magnetic, Valence</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Valence Band Imaging</td>
<td>Biological Imaging</td>
</tr>
<tr>
<td>Surface, bulk</td>
<td>Organic, Inorganic</td>
</tr>
<tr>
<td>Topological</td>
<td>Elemental, chemical</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Plasmonics</td>
<td>Cryo-PEEM</td>
</tr>
<tr>
<td>Dynamics, geometry</td>
<td>Solid State</td>
</tr>
<tr>
<td></td>
<td>Bio, soft matter</td>
</tr>
</tbody>
</table>
IBM/SPECS PEEM with an integrated imaging energy analyser: ARPES

Epitaxial Graphene/6H-SiC(0001)

Magnetic prism
Entrance slit
Sample
Objective
Selected area aperture
Contrast aperture
MCP screen

Real space
k-space

Dark-field imaging with bands

ARRES
Angle
Resolved
Reflected
Electron
Spectroscopy

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

H. Hibino et al.
PRB 77 (2008) 075413
Empty State Bands Bulk hBN

ARPES and ARRES for graphite and hBN


Eugene Krasovskii (Trieste, 2008)
‘Instead of angular scan of a parallel beam, why not use a convergent beam?’ asked Frank Meyer zu Heringdorf.
Ronchigrams in LEEM

CB-LEED, ronchigram, diffraction plane

Real space

Overexposed central spot
Spot diameter $< 100$ nm

Underexposed surroundings

SE

(0,0)

8 mrad $< \sim 100$ nm

(1,0)
Graphene/SiC, several tilts

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

CBED:
Valence-electron density

ARPES:
Valence-electron Band structure

CB-LEED/ARRES:
Conduction-electron Band structure
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.
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

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

Hyperbolic dipole

electrons

Light
# Boundary Element vs. Finite Difference

![Diagram](image.png)

<table>
<thead>
<tr>
<th>Author(s)</th>
<th>Voltage</th>
<th>image Z</th>
<th>$C_3$</th>
<th>$C_5$</th>
<th>$C_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>BEM</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CPO 'benchmark'</td>
<td>-3.198593</td>
<td>-0.119994(2)</td>
<td>-532.37(6)</td>
<td>-8.90(3)E5</td>
<td>-9.623(4)</td>
</tr>
<tr>
<td>D Preikszas</td>
<td>-3.198593</td>
<td>-0.120000</td>
<td>-532.9</td>
<td>-8.743E5</td>
<td>-9.62</td>
</tr>
<tr>
<td>R Tromp (Munro)</td>
<td>-3.198612</td>
<td>-0.120000</td>
<td>-532.89</td>
<td>-8.737E5</td>
<td>-9.624</td>
</tr>
<tr>
<td><strong>Finite Difference</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>D Preikszas</td>
<td>-3.1936</td>
<td>-0.11646</td>
<td>-531.8</td>
<td>-8.747E5</td>
<td>-9.62</td>
</tr>
<tr>
<td>B Lencova</td>
<td>-3.1936</td>
<td>-0.12008</td>
<td>-495</td>
<td>-1.18E6</td>
<td>-9.60</td>
</tr>
<tr>
<td>B Lencova/J Zlamal</td>
<td>-3.1936</td>
<td></td>
<td>-487</td>
<td>-1.29E6</td>
<td></td>
</tr>
</tbody>
</table>

Source: CPO user manual, xmpl2d17, mirror with negative aberrations, Frank Read
Correlation of first and higher order properties

V1 = -1200
V2 = 1800

First order properties, first order of business

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

http://dx.doi.org/10.1016/j.ultramic.2012.07.016
To correct we must measure

But what?
And how?

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

Measurement and Correction of $C_3$

\( C_3 = 0.3943 + 0.0001472C_{2,\text{mirror}} \)

\( C_3^m = -1173 - 3541/\sqrt{E_0} \)

An achromat for photons...... and electrons?

\[ C_c = -L \sqrt{\frac{E}{E_0}} \]
Chromatic aberration of the mirror is not constant.

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

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?

\[
\begin{align*}
\frac{E + dE}{E} &= 1 + \frac{dE}{E} \\
\frac{E + dE}{E} &\approx \frac{E}{E - dE} \\
\end{align*}
\]

\[dC_1 = C_c \frac{dE}{E}.\]
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 \textit{for the reference energy}.

\[
\frac{dV}{V} = \frac{dE}{E} \quad \text{(electrostatic lenses and electron mirror elements)}
\]

\[
\frac{dI}{I} = \frac{1}{2} \frac{dE}{E} \quad \text{(magnetic lenses)}
\]
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
- $= \circ$ to better than 1:10

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

\[
\Delta C_1 = -L \left( \frac{\sqrt{E_c} - \sqrt{E_0}}{\sqrt{E_cE_0}} \right)^2 - C_{cc}^{mir} \frac{(E_c - E_0)^2}{E^2}
\]
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**

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

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
IBM

Weishi Wan
Berkeley Lab

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

Oliver Schaff
Alexander Kaiser
Andreas Berghaus
SPECS

Eugene Krasovksii
Basque Foundation for Science

Frank Meyer zu Heringdorf
Universität Duisburg Essen

Mark Reuter
2017 AVS Hanyo Award

Arthur Ellis
2010 AVS Hanyo Award
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

Data set: ZBVH

Frank Meyer zu Heringdorf et al.
Overlap of beams obscures structure of interest