Aberration-corrected Electron Microscopy for Nanoelectronics Applications

C. Kisielowski
CFKisielowski@lbl.gov

Rolf Erni, Quentin Ramasse, P. Specht

National Center for Electron Microscopy
and Helios SERC
Ernest Orlando Lawrence Berkeley National Laboratory
Berkeley, CA 94720 / USA

Supported by DoE’s Office of Science, Basic Energy Sciences
Why bother improving electron microscopy further?
Berkeley Lab was founded in 1931 by Ernest Orlando Lawrence, a UC Berkeley physicist who won the 1939 Nobel Prize in physics for his invention of the cyclotron.

THE LAB AT A GLANCE

- 11 Nobel Laureates
- 13 National Medal of Science members
- 61 National Academy of Science members
- $700 Million Contributed to the local economy
- 800 University students trained each year
- 4,000 Employees
- 200 Site acreage
Helios
New Program

Efficient for sustaining life

Helios SERC Principle Staff

Management Team:
Paul Alivisatos, Director
Elaine Chanler, Deputy Director
Heinz Frei, Deputy Director
Melania Sonsteng, Administrator

Components:

NanoPVs: Paul Alivisatos, Joel Ager, Jeff Neaton, Rachel Segalman, Wladek Walukiewicz
Catalysts: Don Tilley, Chris Chang, Cliff Kubliak (UCSD)
Light Protection: Graham Fleming, Ana Moore, Tom Moore, Devens Gust (Arizona State)
Electrochemistry: Alex Bell, John Newman, Martin Head-Gordon, Rich Mathies

Integrated Systems:
Heinz Frei, Vital Yachandra, Don Tilley, Lin-Wang Wang, Peidong Yang, Nate Lewis (Caltech), Gabor Somorjai, Rachel Segalman, Paul Alivisatos, Barend Smit

Cross-cutting Scientific Support:
Theory: David Chandler, Gavin Crooks, Jeff Neaton, Lin-Wang Wang, Phil Geissler, Steven G. Louie
Instrumentation: Mike Crommie, Christian Kisielowski

Natural photosynthesis
Efficient for making fuel?

http://www.lbl.gov/LBL-Programs/helios-serc/
Electron Microscopy: Status in 2002

EM commonly images atom columns. Imaging of single atoms is an exception.

3D EM with atomic resolution requires single atom sensitivity.
The TEAM Project

New tools: Next generation electron microscopes

TEAM0.5:
- 2 Cs correctors
- High brightness gun
- Monochromator
- Improved electrical/mechanical stability

Spatial resolution: 0.5 Å
Energy resolution: 0.1 eV, 1 sec
User facility since 10/2008

Currently shipped to Berkeley
The Importance of Resolution

TEM Simulation: H adatom on graphene

CTF describes frequency region of predictable phase shifts resulting in predictable atom positions.

CTF = 0 describes unpredictable phase shifts resulting in unpredictable atom positions (resolution limit).

Graphene: Fourier components
The Uniqueness Aspect
Defects in graphene

Lateral relaxation

1.97 nm

\[ \downarrow = 20 \text{ pm} \]

Nitrogen adatom (vacancy created)

Hydrogen adatom

Lateral relaxation & substitution
The Uniqueness Aspect
Reason: Limited resolution

There is $4/3 \pi 150^3$ pm$^3$ of space available to place atoms (set phases) @ 3 Å resolution

Resolution enhancement largely relaxes the uniqueness problem

LaB$_6$, 100 kV, 
Cs = 1 mm 
f = -75 nm

Fe$_{\text{relaxed}}$

TEAM 0.5, 300 kV, 
Cs = 0 mm 
f = -1.1 nm

V-N$_{\text{adatom}}$, C$_{\text{relaxed}}$, BN, .... can be confused with the “H contrast”

N$_{\text{adatom}}$, Fe$_{\text{relaxed}}$ (Z=26), ...
can be confused with the “C contrast”
Resolution and Noise
Simulation: S/N ratio for single light atoms

Simulation example: graphene (C) & hydrogen, 80 kV

S/ N ratios are boosted by resolution improvement
It may be possible to detect H (radiation damage)
TEAM 0.5 (Cs-corrector / Monochromator)

Titan (Cs-corrector)

Comparison / graphene

S/N = 3.8 (~ 0.8 Å)

S/N = 0.6 (~ 2 Å)
TEAM 0.5 - 80 kV
Graphene - ERW boosts sensitivity

Model

Experiment

80 kV & Monochromator
$C_s = -0.015 \text{ mm}$, $C_5 = 5 \text{ mm}$

Information Limit $< 0.1 \text{ nm}$

Reconstructed phase image

$0.14 \text{ nm}$
Defects in semiconductors
Or
Epoxy, hydroxyl, carboxyl groups

Graphene oxide (O:30 %)

Double layer

1.4Å

Schniepp et al. 2006
An Instrument Comparison
Reconstructed phase images of Au [110]
Focus stability reflects:
- Electrical stability
- Mechanical stability
- Temperature
- Pressure
- Noise
- Sample stability
- Measurement precision
- Site

Next generation EM:
Improvements are outstanding

OAM
Resolution:
0.9 Å reliable
0.8 Å achievable

TEAM0.5
Resolution:
0.5 Å reliable
Results are consistent, information transfer & resolution < 50 pm
Resolution definition by column width & noise most useful
Natural column width (1s state) of ~ 0.5 Å is now a physical limit to resolution

Contrast Interpretation

Focus series

a) Underfocus = 1.8 nm

b) Underfocus = 3.7 nm

c) Underfocus = 5.7 nm

d) Underfocus = 7.7 nm
Contrast Interpretation

Focus spread TEAM: 0.7 Å

a) S/N ratio of one gold atom: 10
b) Depth precision reaches 2.9 Å
c) Atoms can be counted
d) 3D information from 1 projection
TEAM 0.5

Single atom detection and depth precision

New: unprecedented S/N ratios (graphene)
New: narrow focus spread < 10 Å (gold)

• Single atom sensitivity across the Periodic Table
• Element identification by contrast interpretation
• Depth precision reaches interatomic distances
It is unreasonable to expect 0.51 Å “dumbbell” images from diamond [112]
Object-Limited Resolution

Heavy atoms: TEM experiments, gold

Experiment

Au [110]

Electron channeling limits resolution to ~ 0.5 Å:

Au [110]: 0.5 Å not achievable

Au [111]: 0.5 Å achievable
Object-Limited Resolution

Heavy atoms: STEM experiments, gold

Electron channeling limits resolution in HAADF images, too
Towards 3D Electron Microscopy

STEM Depth sectioning (Au [110]): df = 6 nm - nm resolution
Towards 3D Electron Microscopy
Self Interstitials in Ge [110]


• First detection of self interstitials & 3D reconstruction from single projection
• Dose limits now imaging of soft and hard materials
There is plenty of room for improvement (TEAM1)
S/N ratio of 20 for 1 gold atom is feasible

TEAM0.5:
300 kV, EWR

TEAM0.5
single image
300 kV

OAM: 300 kV, EWR

Kisielowski, 2002

Batson, 2002

Muller, 2002

Kirkland (1987)

Signal / Noise Ratio [arb. units]

Atomic Number Z [arb. units]
Conclusions

Opportunities

• Single atom detection across the Periodic System is now possible
  Atomic resolution tomography, depth precision, catalysis,…

• Resolution debate has reached physically meaningful limits
  Instead, contrast (S/N ratios) becomes the important measure

• Electron tomography with atomic resolution becomes feasible

Challenges

• Radiation damage becomes a limiting factor even in hard materials
• Sample preparation is more demanding
• Image interpretation is increasingly demanding
  Seeing may be believing but understanding is still science
The TEAM team

- FEI Company
  B. Freitag, M. Bischoff, H. van Lin, S. Lazar, G. Knippels, P. Tiemeijer, M. van der Stam, S. von Harrach, M. Stekelenburg,
- CEOS
  M. Haider, S. Uhlemann, H. Müller, P. Hart,
- ANL
  B. Kabius, D. Miller,
- UCUI
  I. Petrov, E. A. Olson T. Donchev,
- ORNL
  E.A. Kenik, A.R. Lupini, J. Bentley, S.J. Pennycook
- NCEM

UC Berkeley: J. Meyer, A. Zettl,

CEN-DTU: R. Dunin-Borkowski, J. Jinschek