NATIONAL CENTER FOR ELECTRON MICROSCOPY

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Aberration-corrected Electron Microscopy for Nanoelectronics Applications

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Supported by DoE’s Office of Science, Basic Energy Sciences

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Metrology 2002/3

Precision: < 1 Å ,Resolution: 0.8 Å

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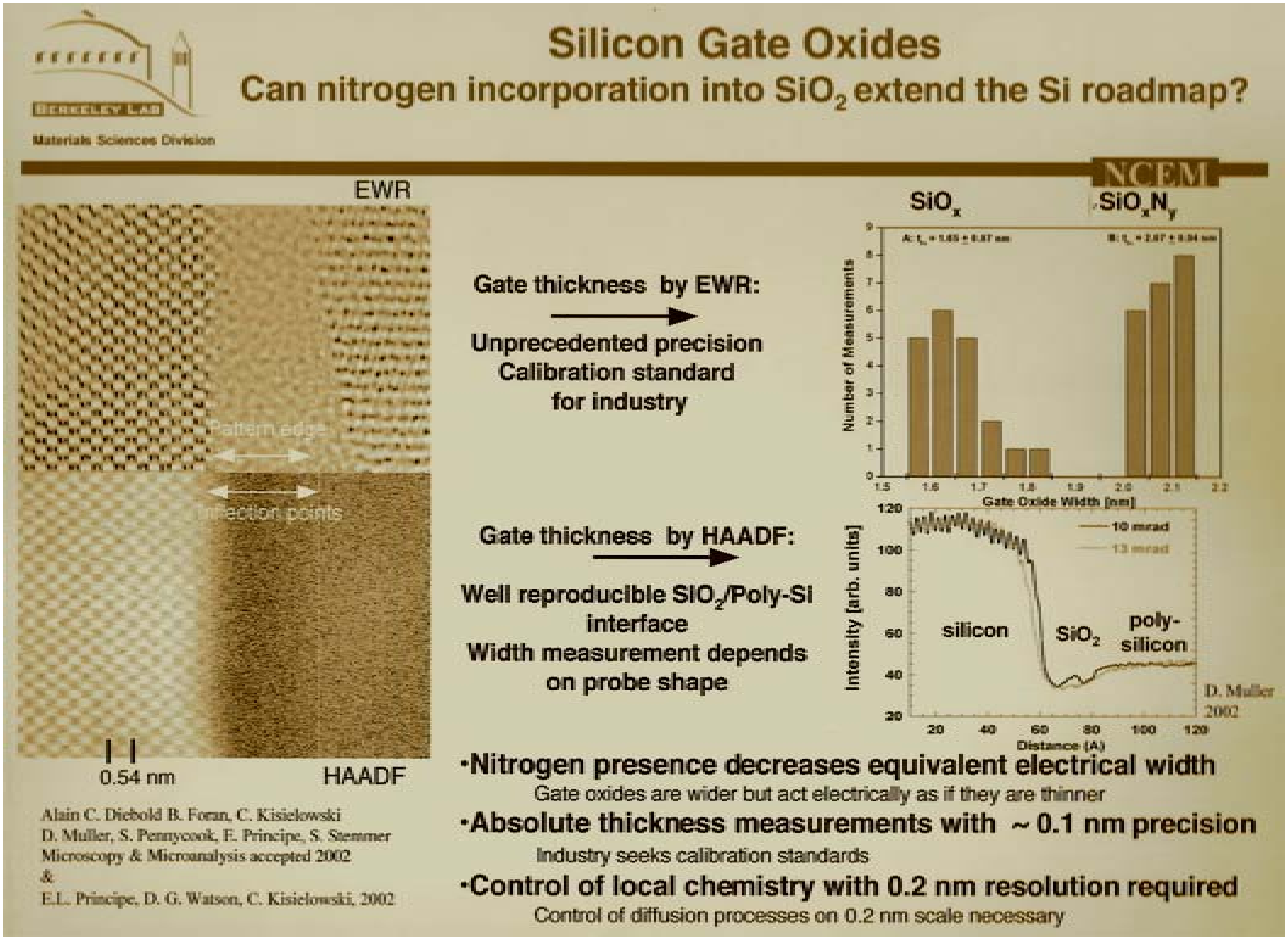

Why bother improving electron microscopy further?

LBNL in a Changing Environment

Changing needs

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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

Berkeley Lab Interim Director A. Paul Alivisatos

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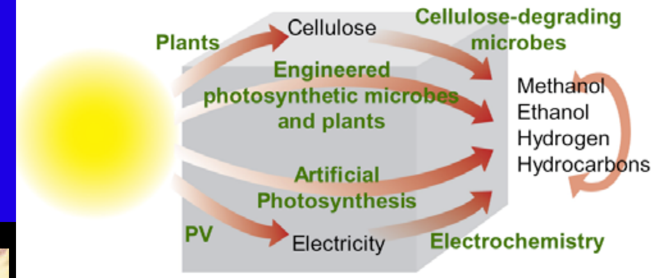
Helios

New Program



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SOLAR ENERGY RESEARCH CENTER

Overview | Goals & Challenges | Research Highlights | In the News | SERC Staff

Helios SERC Principle Staff

Management Team:

Paul Alivisatos, Director

Elaine Chandler, Deputy Director

Heinz Frei, Deputy Director

Melanie Sonsteng, Administrator

Components:

NanoPVs: Paul Alivisatos, Joel Ager, Jeff Neaton, Rachel Segalman, Wladek Walukiewicz

Catalysts: Don Tilley, Chris Chang, Cliff Kubiak (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, Vittal Yachandra, Don Tilley, Lin-Wang Wang, Peidong Yang, Nate Lewis (Caltech), Gabor Somorjai, Rachel Segalman, Paul Alivisatos, Berend Smit

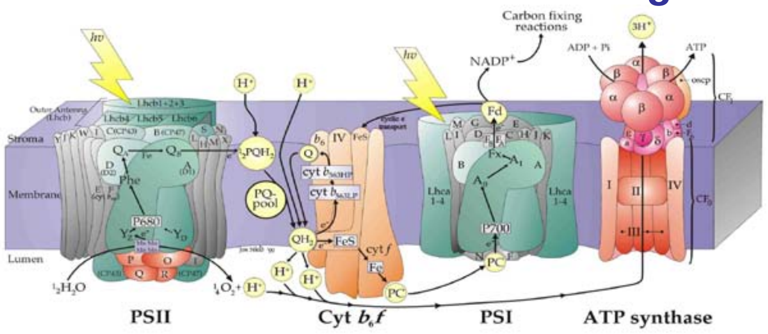
Cross-cutting Scientific Support:

Theory: David Chandler, Gavin Crooks, Jeff Neaton, Lin-Wang Wang, Phill Geissler, Steven G. Louie

Instrumentation: Mike Crommie, Christian Kisielowski

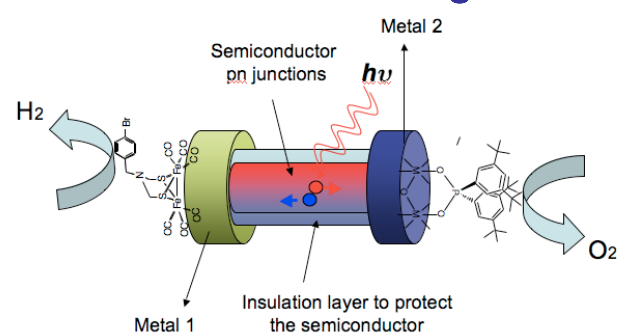
Natural photosynthesis

Efficient for sustaining life



Artificial photosynthesis

Efficient for making fuel?



http://www.lbl.gov/LBL-Programs/helios-serc/

Electron Microscopy : Status in 2002

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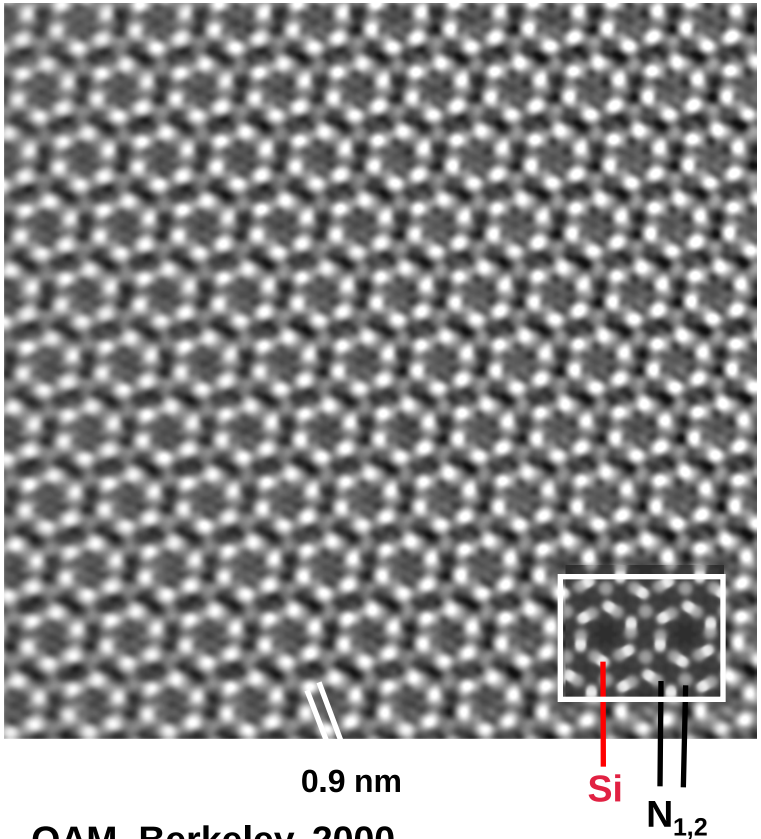
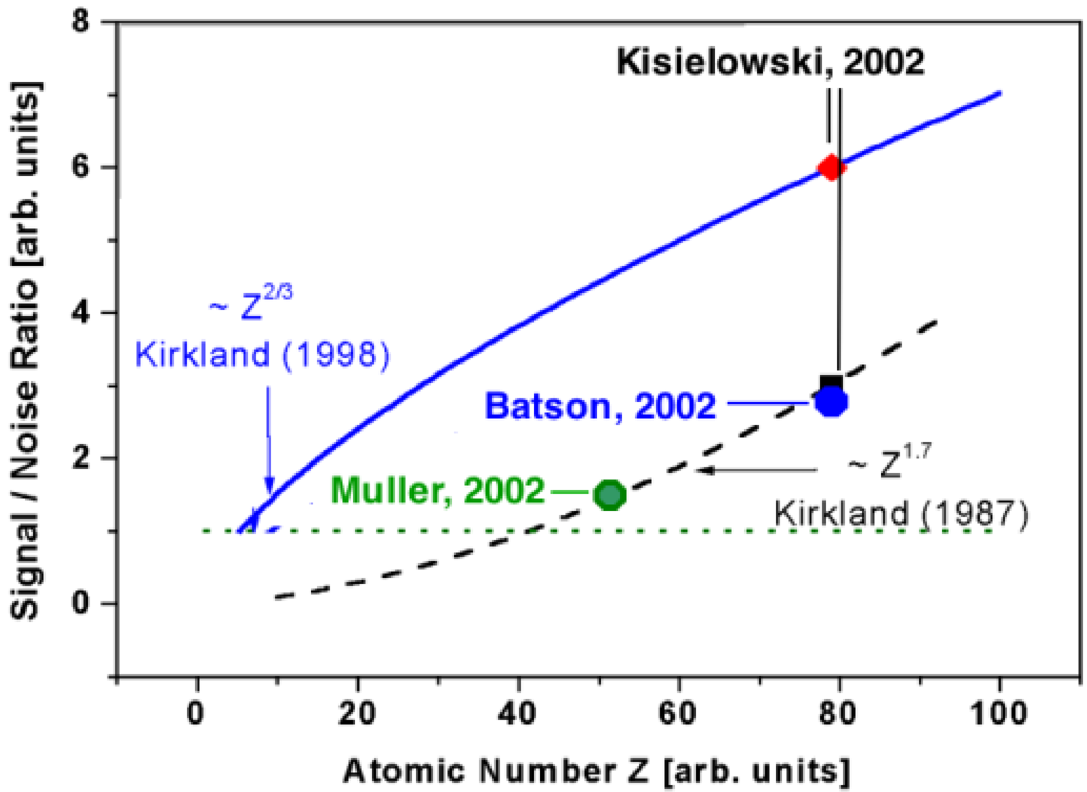


OAM, Berkeley, 2000



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

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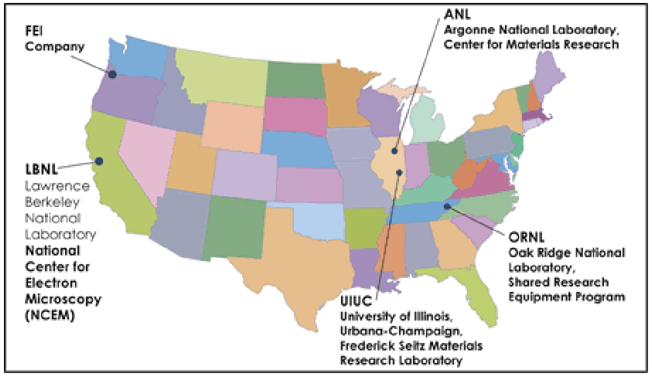

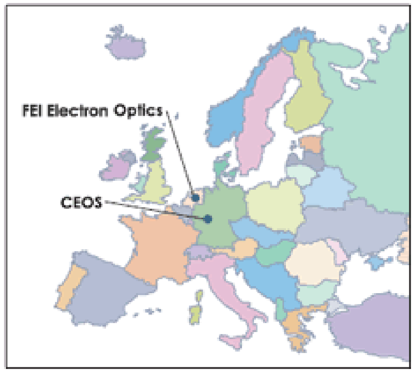

The TEAM team

- **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:** U. Dahmen, P. Denes, T. Duden, R. Erni C. Kisielowski, A.M. Minor, V. Radmilovic, Q.M. Ramasse, A.K. Schmid, M. Watanabe

TEAM Project begins

Begin stage development Lawrence Berkeley National Laboratory/ University of Illinois, Urbana-Champaign

Start probe corrector development Oak Ridge National Laboratory

Start C_c corrector development Argonne National Laboratory

Installation of TEAM 0.5 at NCEM, Lawrence Berkeley National Laboratory

TEAM 0.5 begins user Operations at NCEM

Installation of TEAM I at NCEM

TEAM I begins Facility operations at NCEM

04 | 05 | 06 | 07 | 08 | 09 | 10

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

Office of Science U.S. DEPARTMENT OF ENERGY

The Importance of Resolution
TEM Simulation: H adatom on graphene

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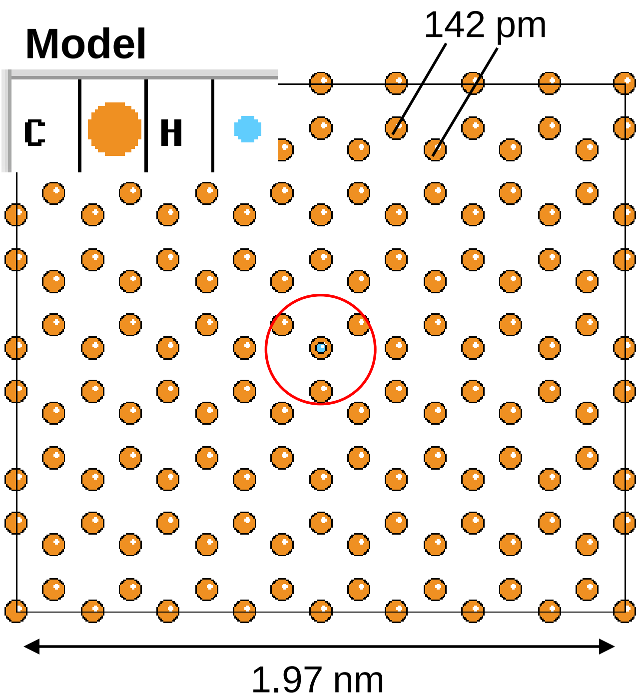
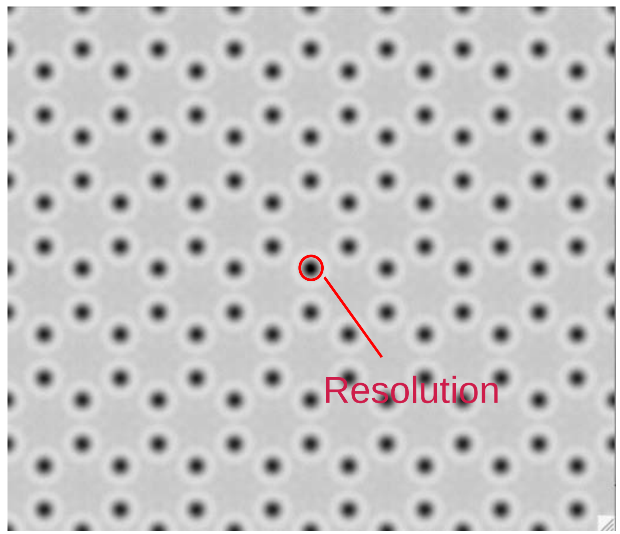
Model

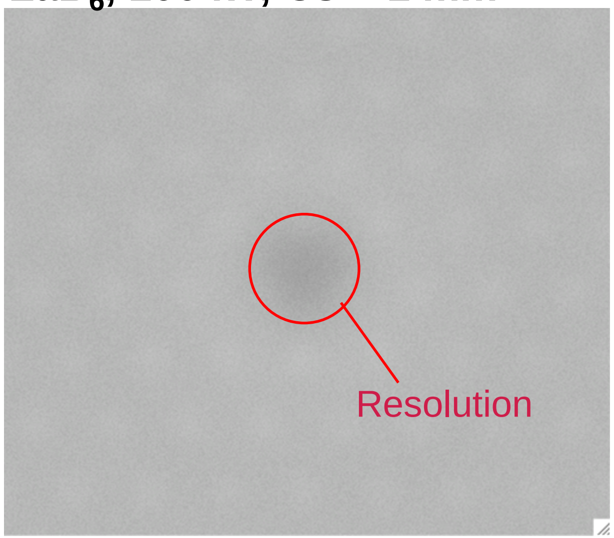


TEAM 0.5, 300 kV, Cs = 0 mm

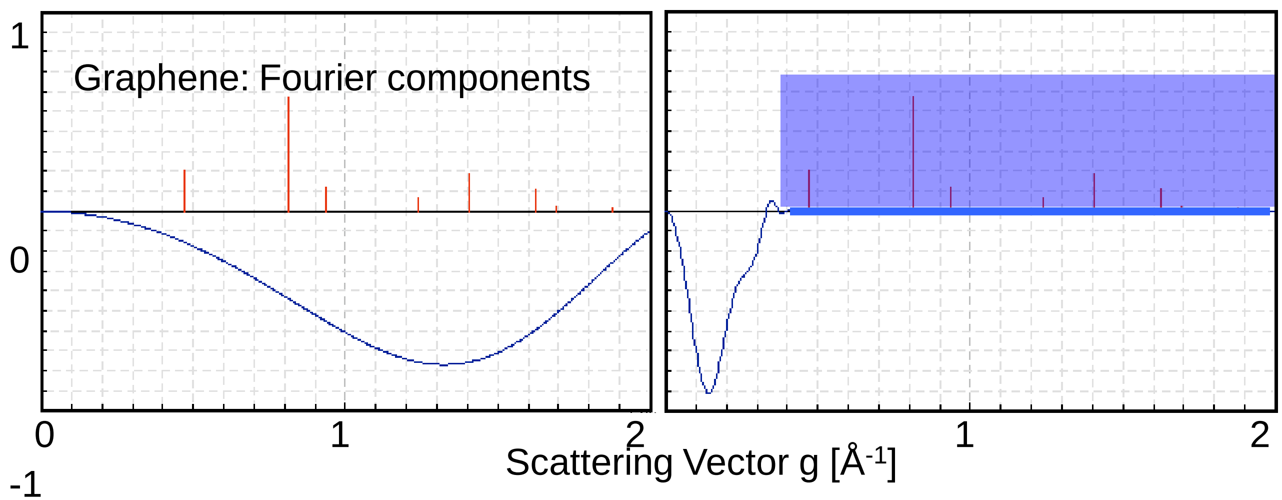


Scherzer image, Δf = -1.1 nm

LaB_6, 100 kV, Cs = 1 mm



Scherzer image, Δf = -75 nm



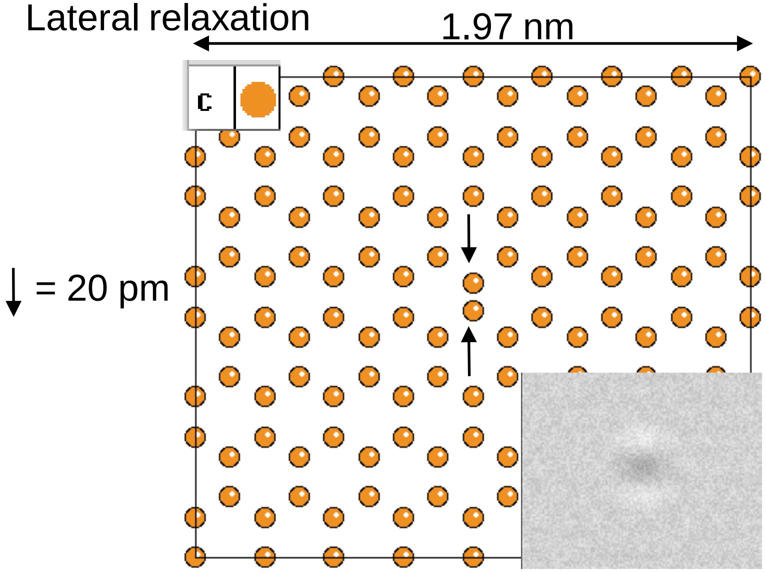
CTF describes frequency region of predictable phase shifts resulting in predictable atom positions

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

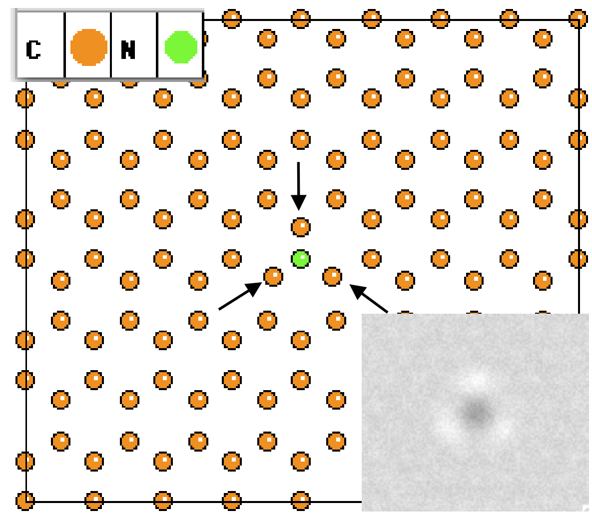
The Uniqueness Aspect

Defects in graphene

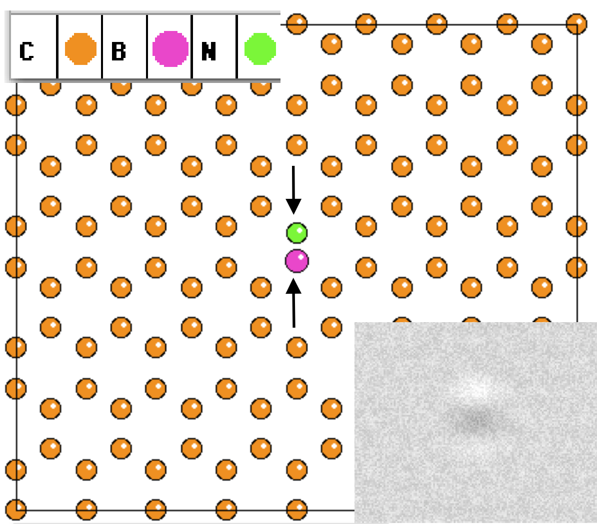
Lateral relaxation



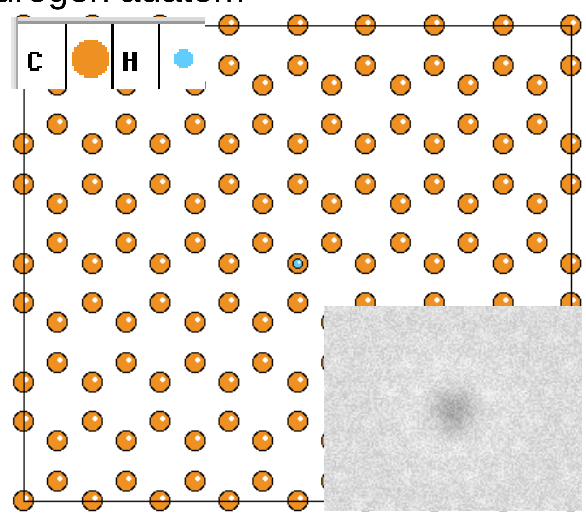
Nitrogen adatom (vacancy created)



Lateral relaxation & substitution



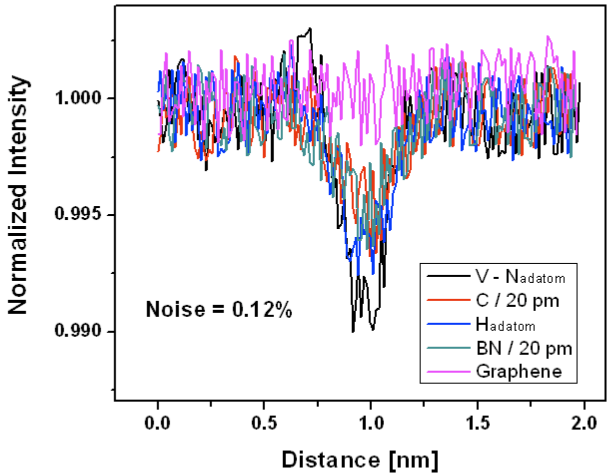
Hydrogen adatom



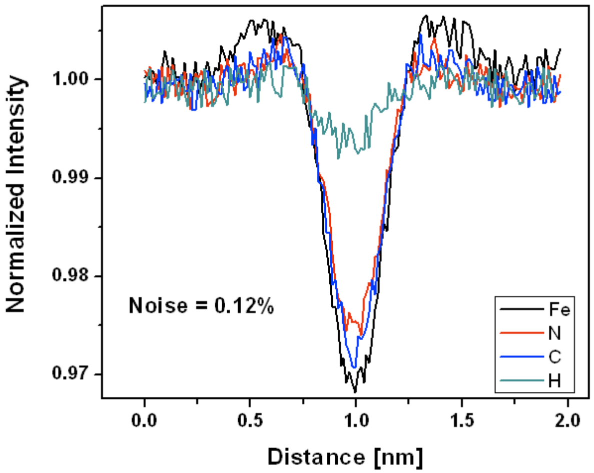
The Uniqueness Aspect

Reason: Limited resolution

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



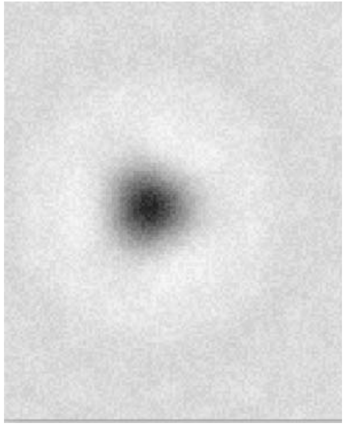
$V\text{-}N_{adatom}$, $C_{relaxed}$, BN, ….
can be confused with the "H contrast"



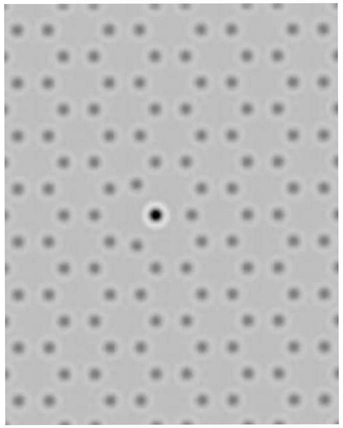
N_{adatom}, $Fe_{relaxed}$ (Z=26), …
can be confused with the "C contrast"

Resolution enhancement largely relaxes the uniqueness problem

LaB_6,
100 kV,
Cs = 1 mm
f = -75 nm



$Fe_{relaxed}$



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

Resolution and Noise

Simulation: S/N ratio for single light atoms

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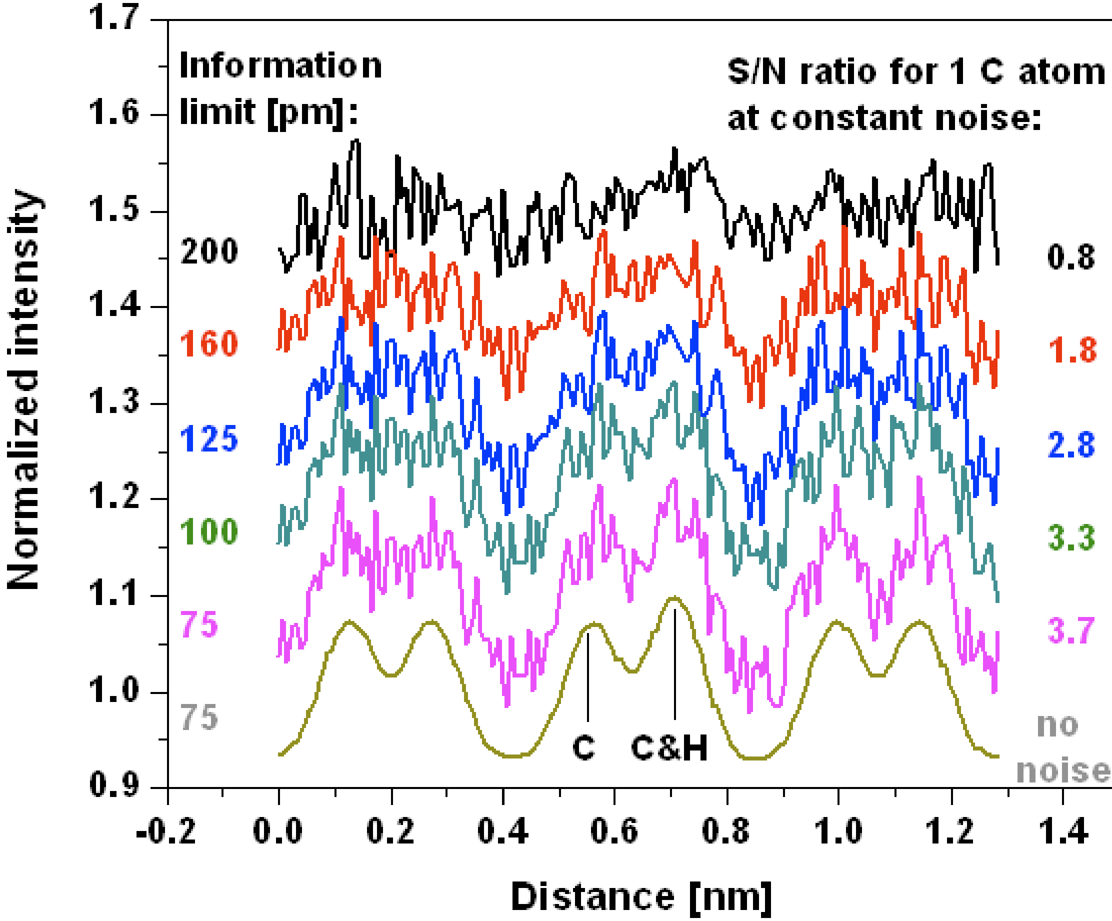
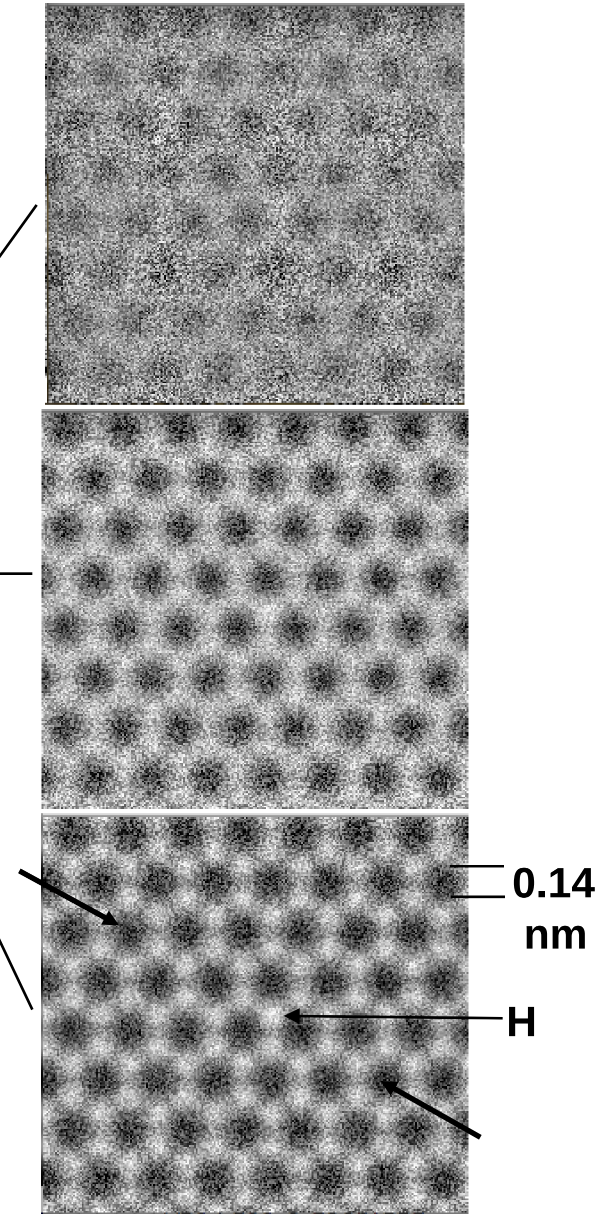
Simulation example: graphene (C) & hydrogen, 80 kV





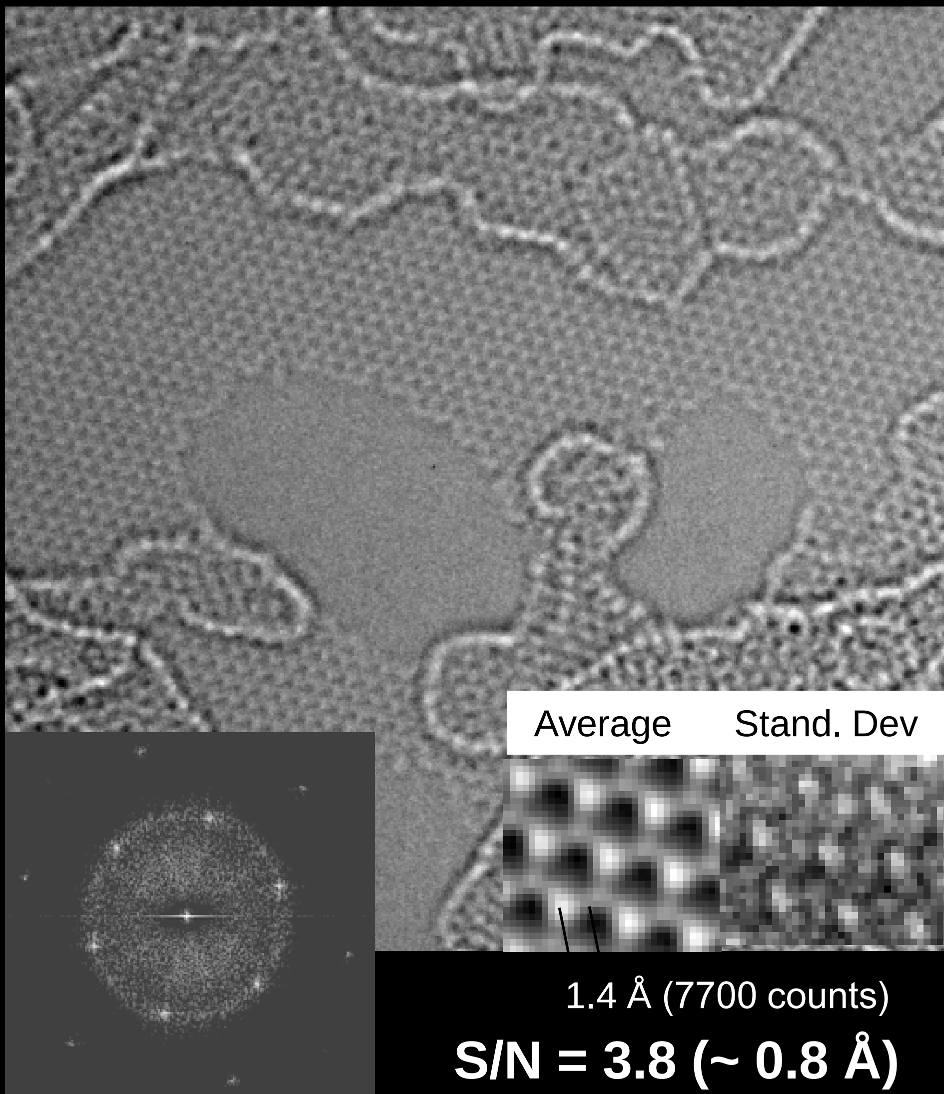
S/ N ratios are boosted by resolution improvement

It may be possible to detect H (radiation damage)

TEAM0.5 ---- Titan

Comparison / graphene

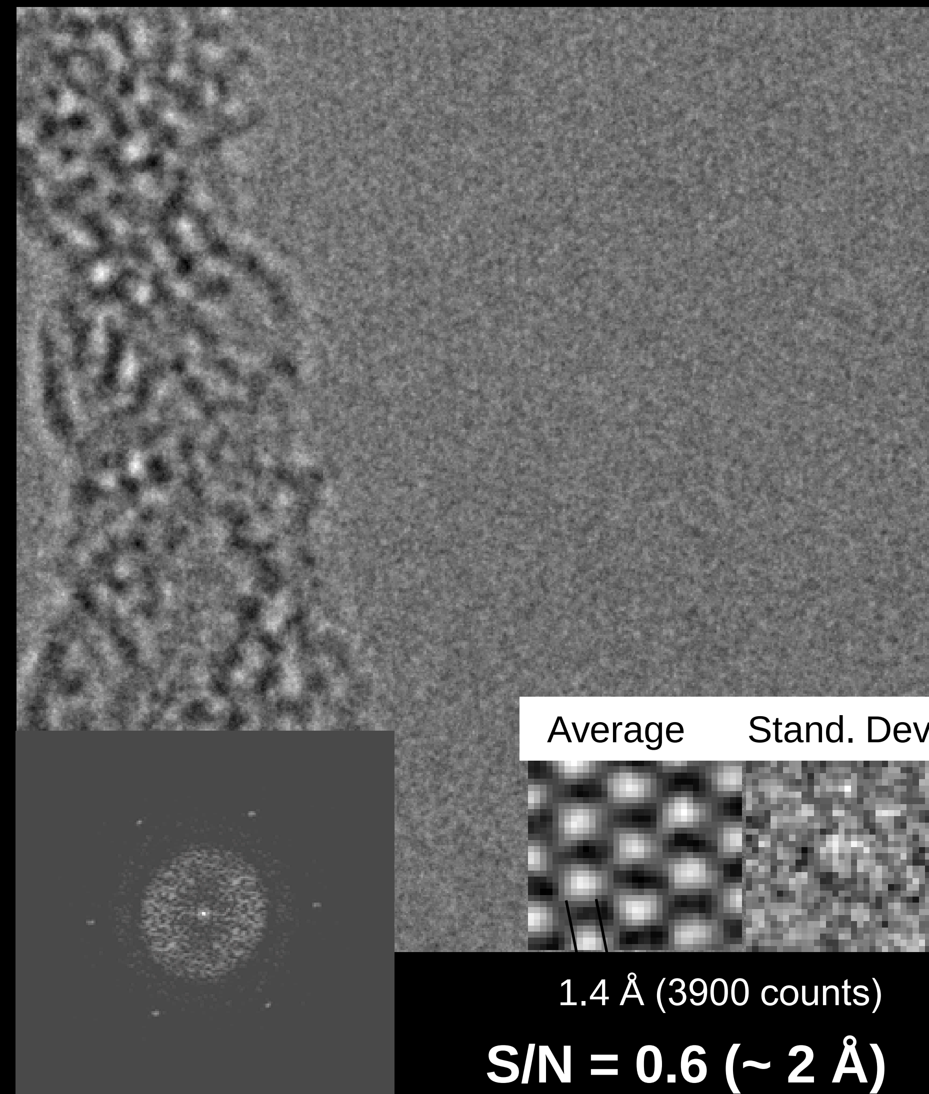
TEAM 0.5 (Cs-corrector / Monochromator)



1.4 Å (7700 counts)

S/N = 3.8 (~ 0.8 Å)

Titan (Cs-corrector)

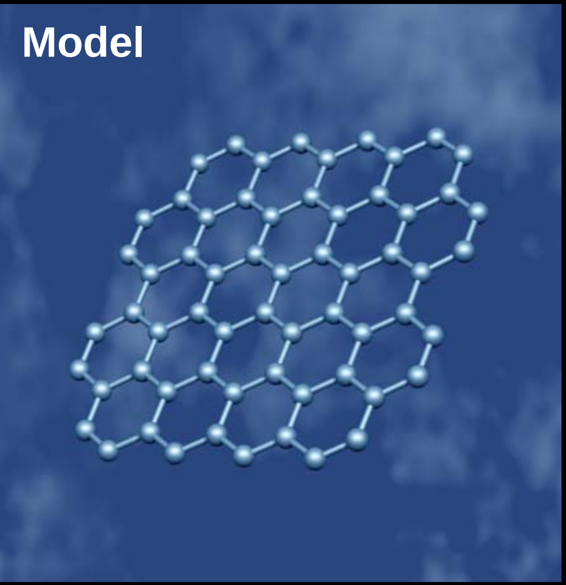


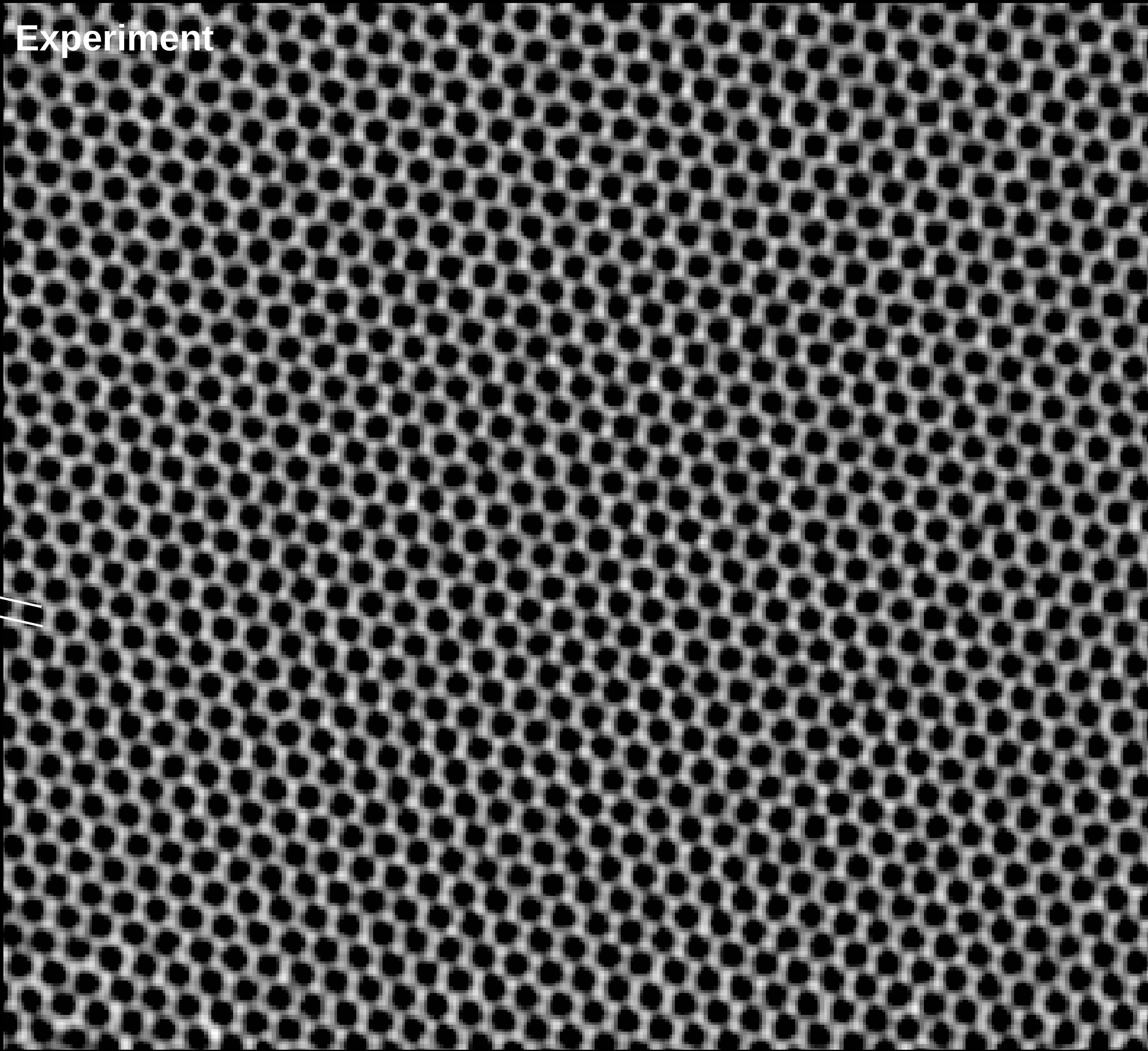
1.4 Å (3900 counts)

S/N = 0.6 (~ 2 Å)

TEAM 0.5 - 80 kV

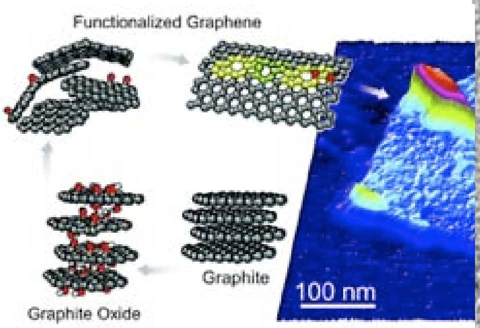
Graphene - ERW boosts sensitivity





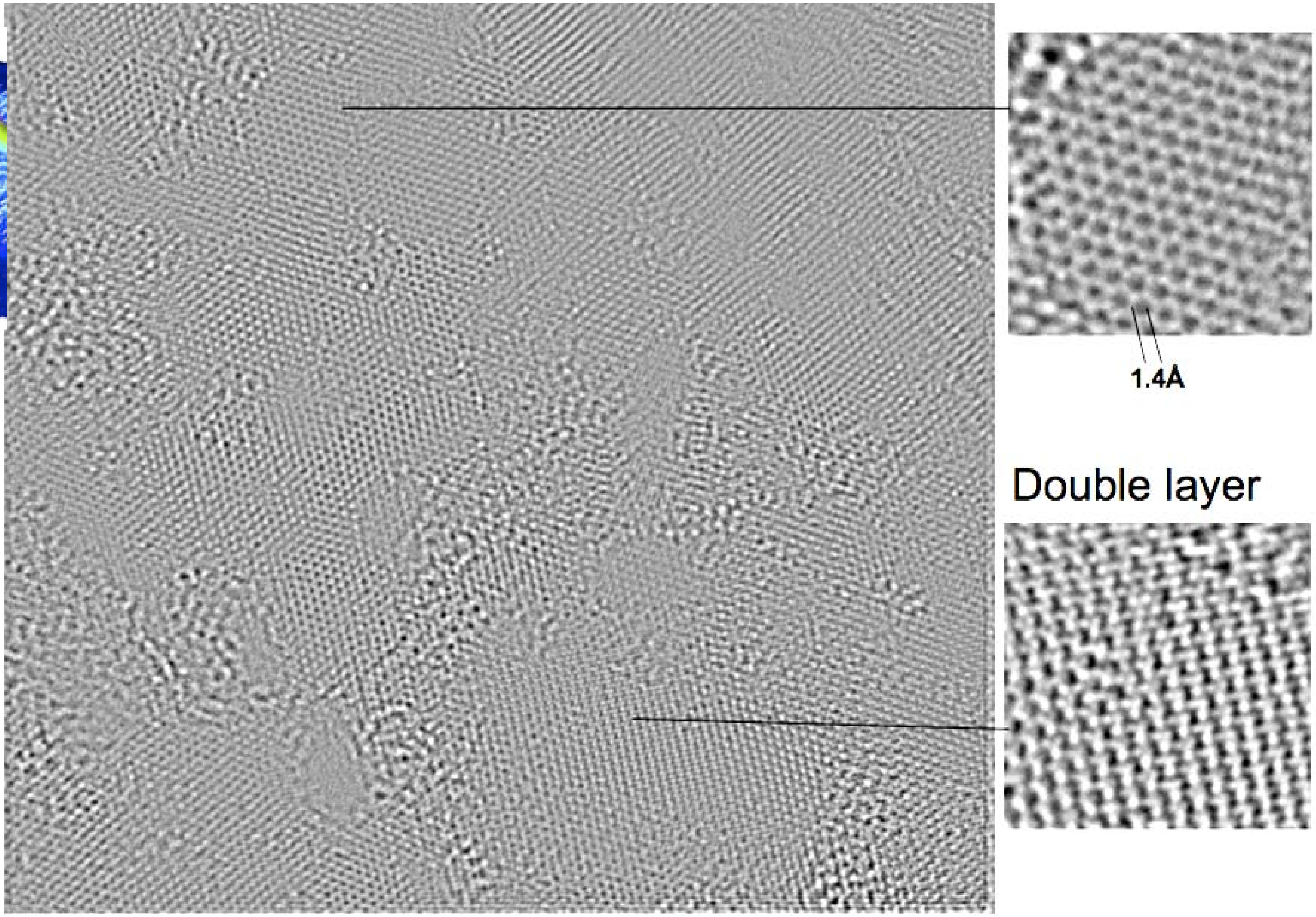
- 80 kV & Monochromator
- C_s = - 0.015 mm, C_5 = 5 mm
- Information Limit < 0.1 nm
- Reconstructed phase image

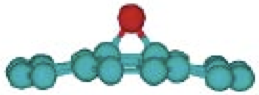
Functionalization or Doping?



Schniepp et al. 2006

Graphene oxide (O:30 %)





Defects in semiconductors
Or
Epoxy, hydroxyl, carboxyl groups

An Instrument Comparison

Reconstructed phase images of Au [110]

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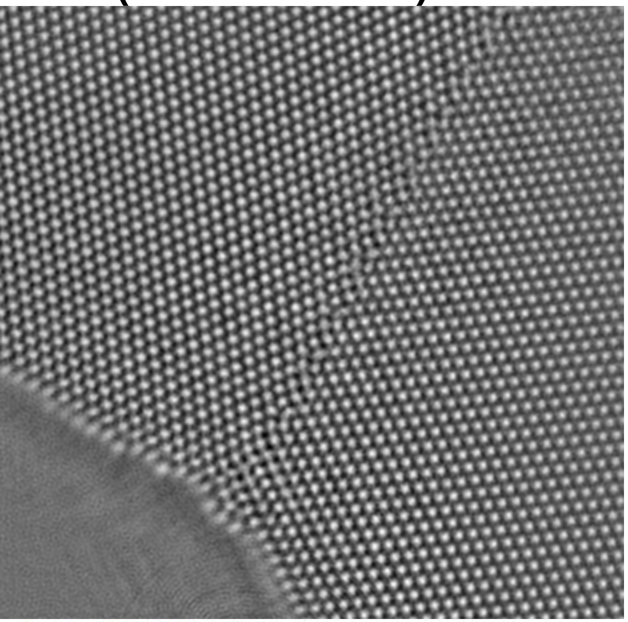
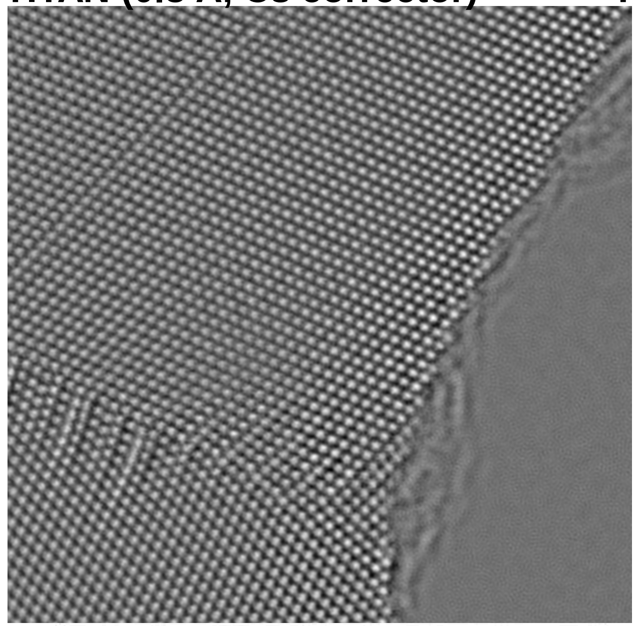
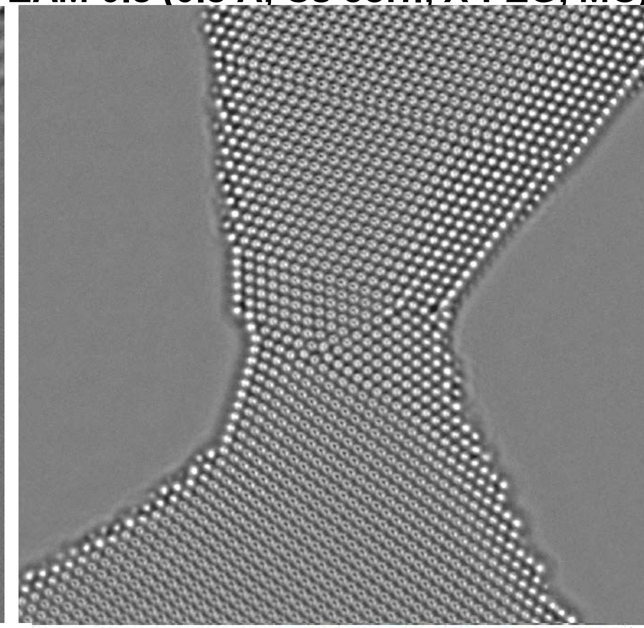
OAM (0.8 Å since 1999)

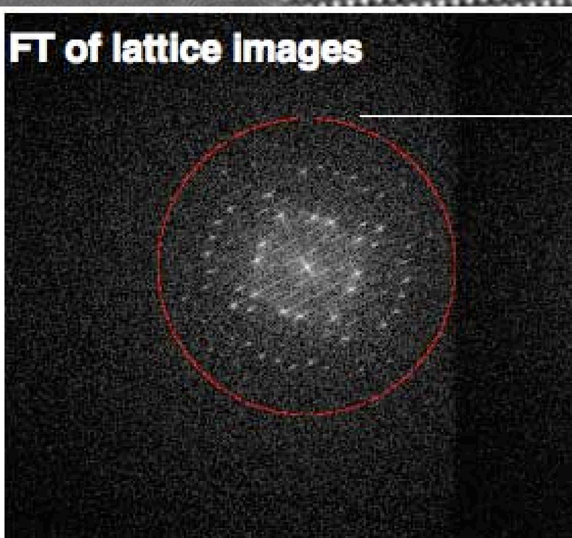


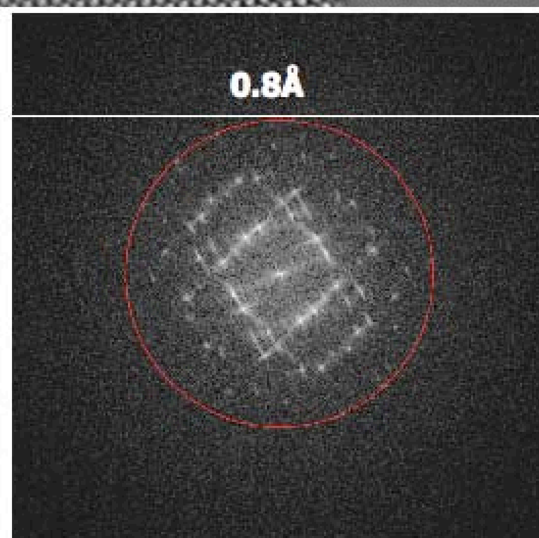
TITAN (0.8 Å, Cs corrector)

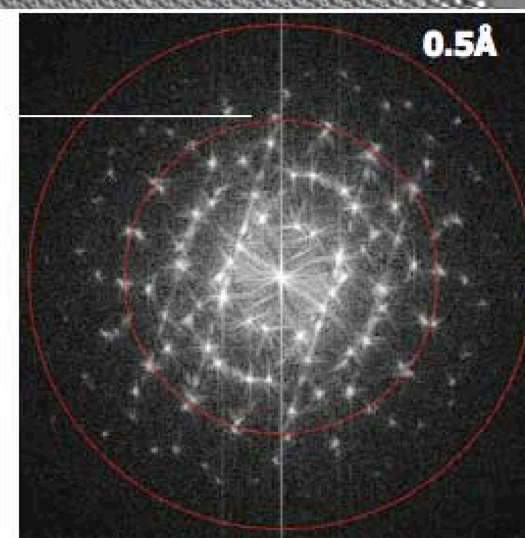


TEAM 0.5 (0.5 Å, Cs corr., X-FEG, MC)









Instrument Stability is Essential

OAM - TEAM 0.5

- **Focus stability reflects:**
  - Electrical stability
  - Mechanical stability
  - Temperature
  - Pressure
  - Noise
  - Sample stability
  - Measurement precision
  - Site

- **Next generation EM: Improvements are outstanding**

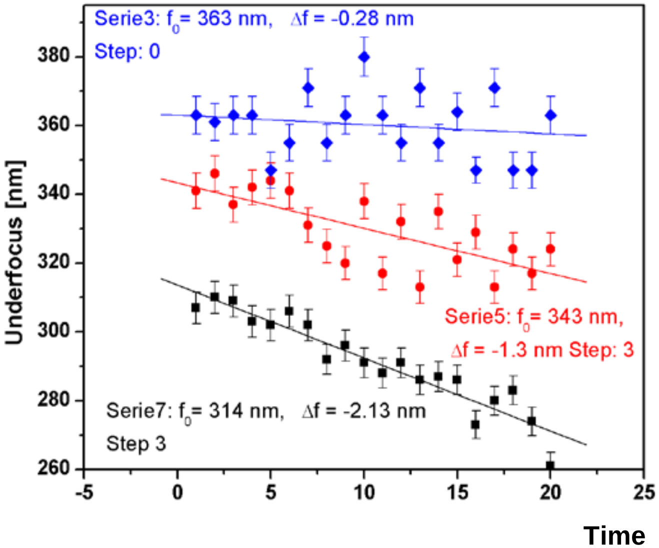
OAM

Resolution:

0.9 Å reliable

0.8 Å achievable

20 nm

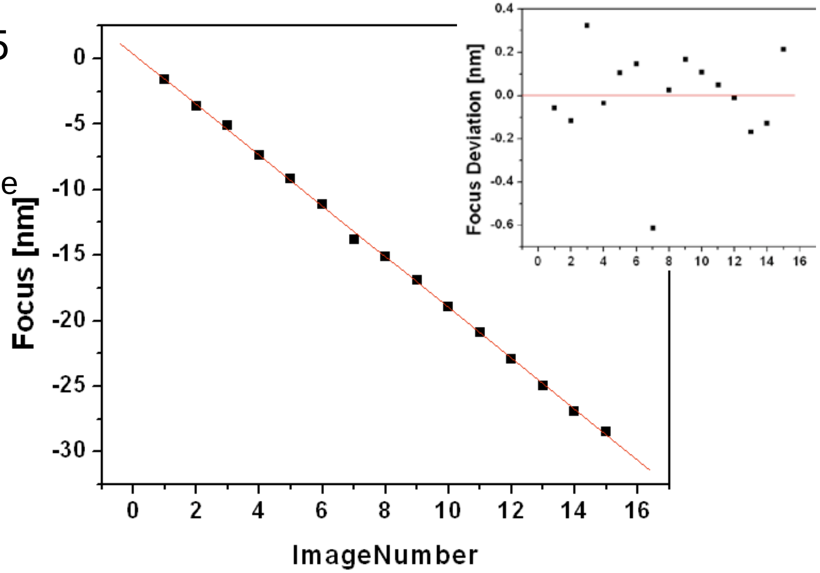


TEAM0.5

Resolution:

0.5 Å reliable

20 nm



TEAM0.5: Next Generation EM

Resolution tests @ 300kV

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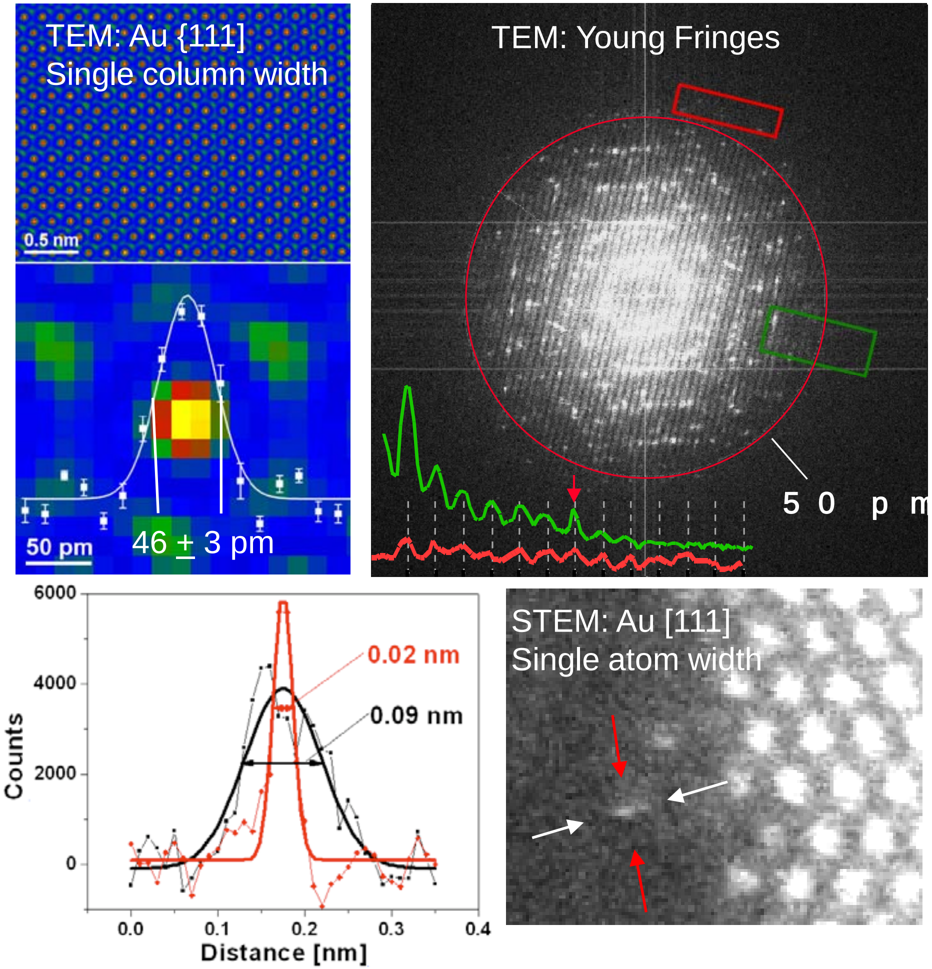
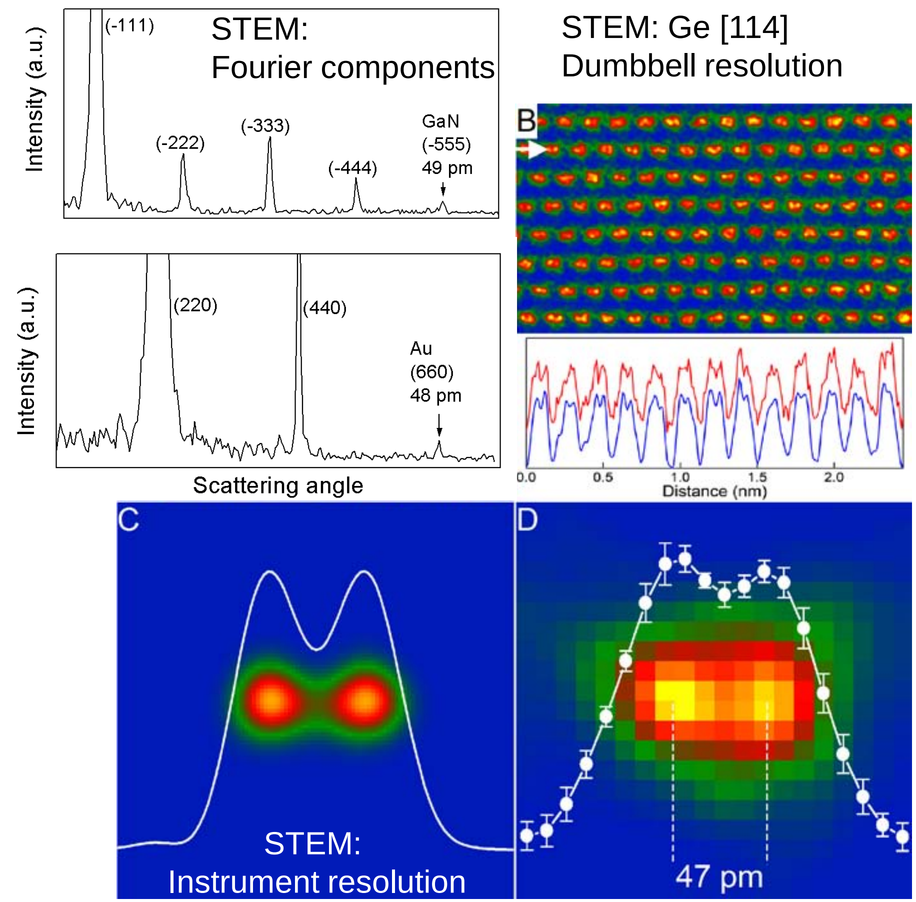


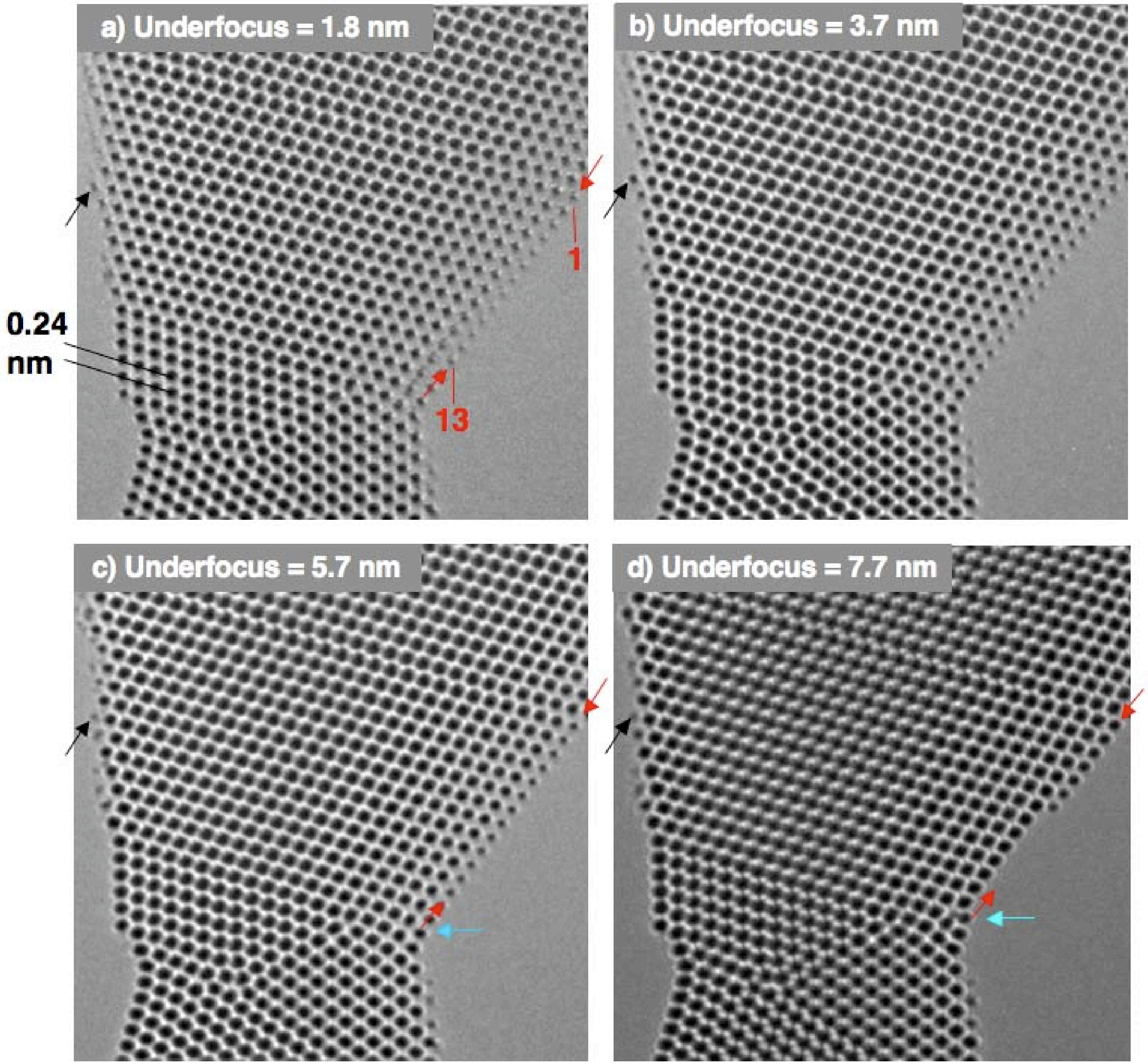


R. Erni, M.D. Rossell, C. Kisielowski, U. Dahmen, **Phys. Rev. Lett. 102,** 096101 (2009)

- **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



Contrast Interpretation

Focus spread TEAM: 0.7 Å

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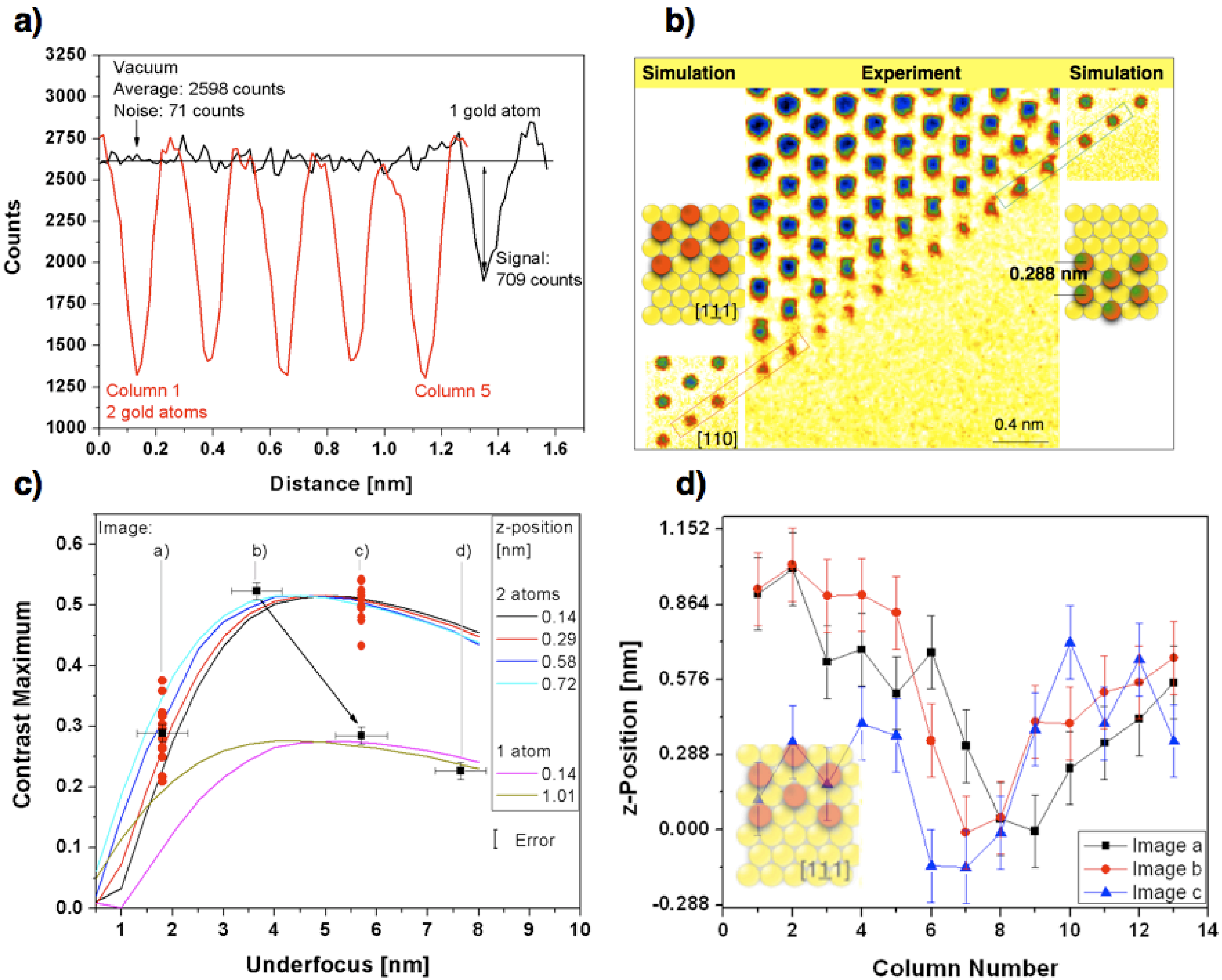

a)

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

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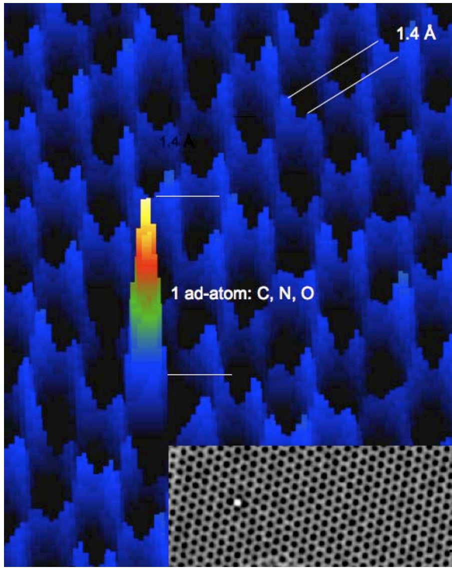
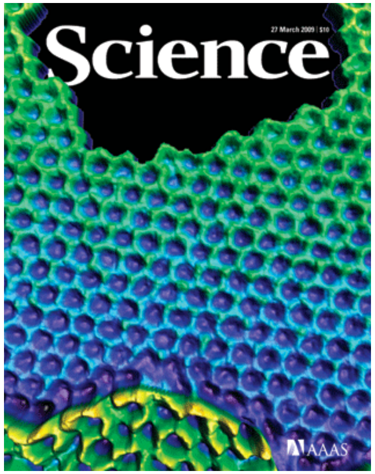
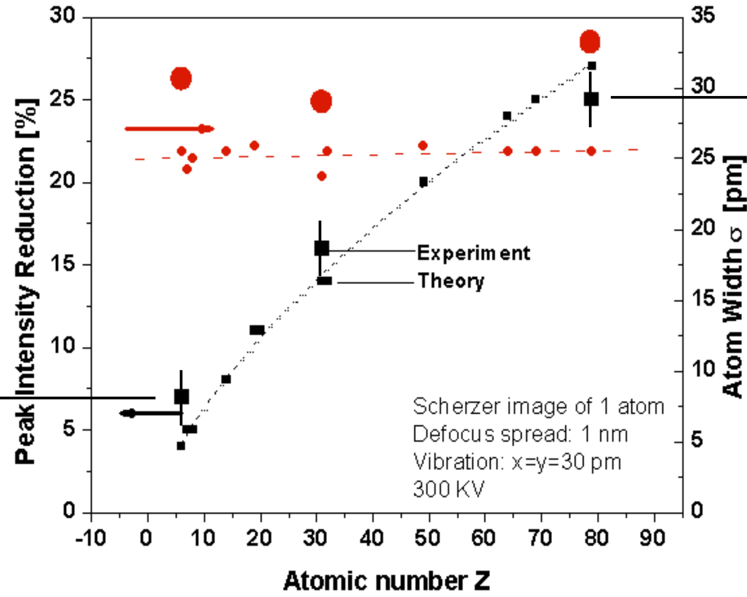
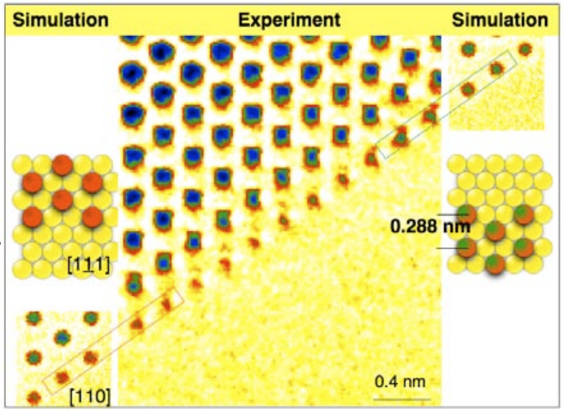
New: unprecedented S/N ratios (graphene)

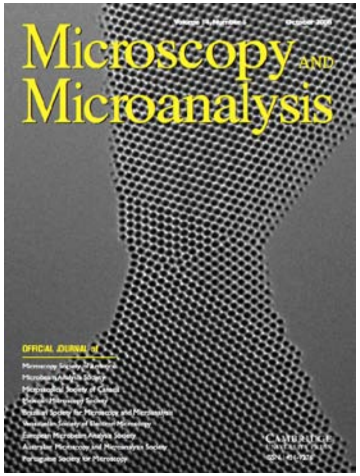
New: narrow focus spread < 10 Å (gold)







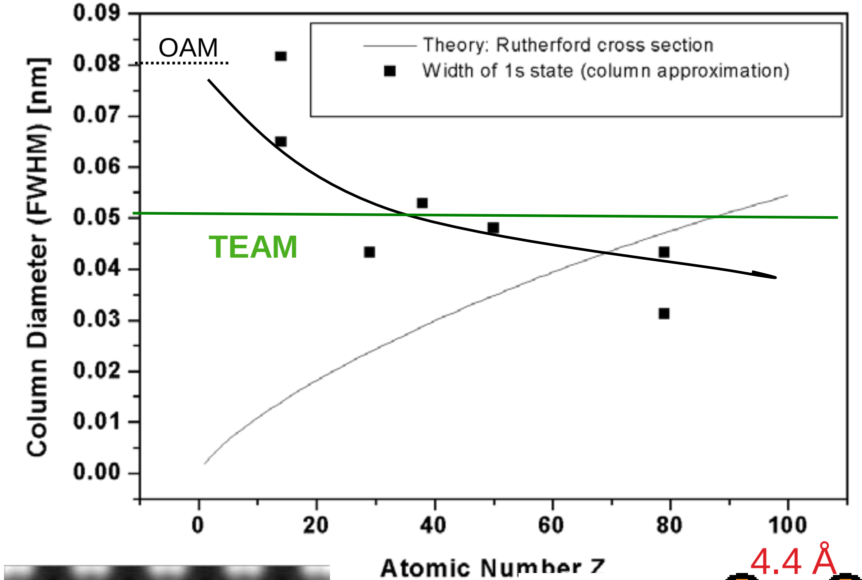


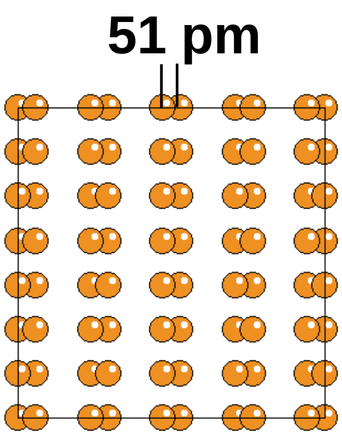


- **Single atom sensitivity across the Periodic Table**
- **Element identification by contrast interpretation**
- **Depth precision reaches interatomic distances**

Object-Limited Resolution

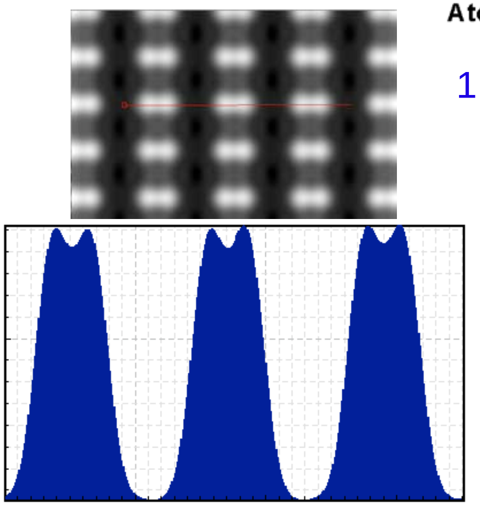
Light atoms: Simulation diamond [112]

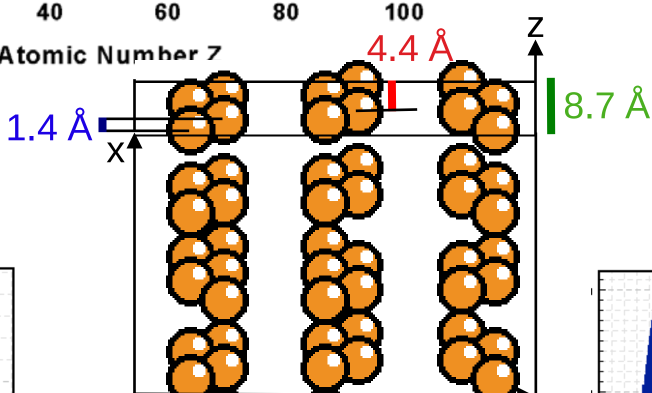




t = 4 Å

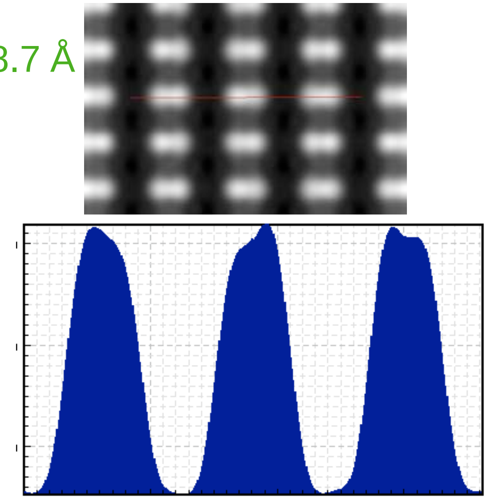
No depth-dependence included





t = 8.7 Å

Depth-dependence included

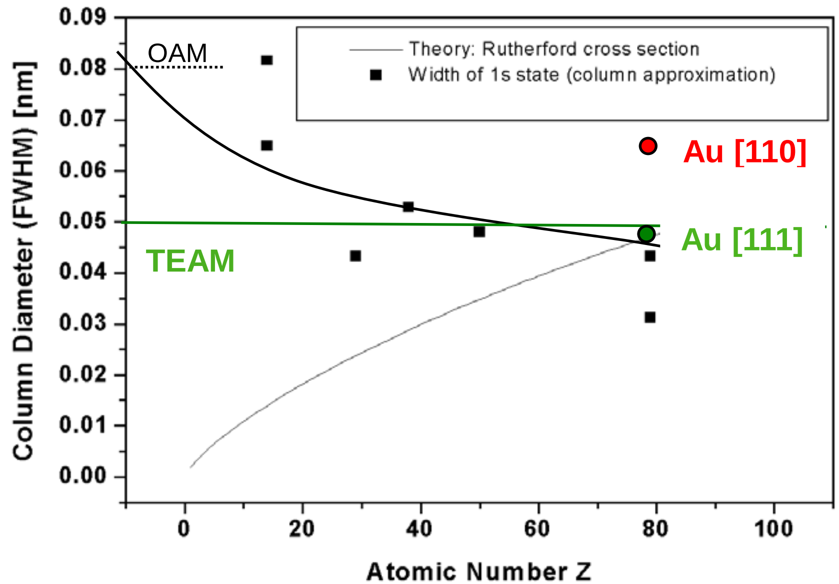


It is unreasonable to expect 0.51 Å "dumbbell" images from diamond [112]

Object-Limited Resolution
Heavy atoms: TEM experiments, gold

Experiment Au [110]

Experiment Au [111]



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

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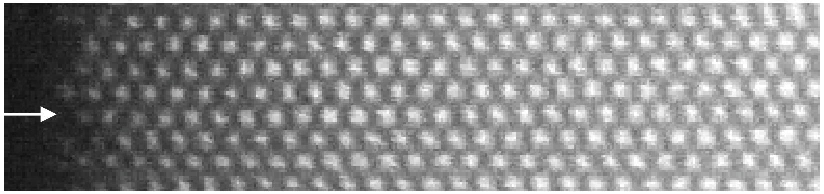
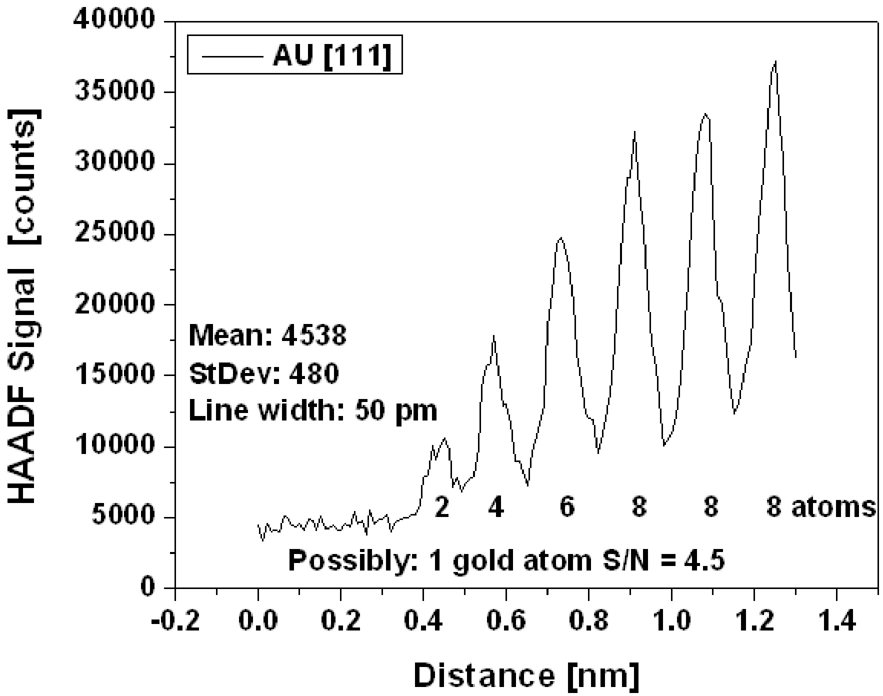
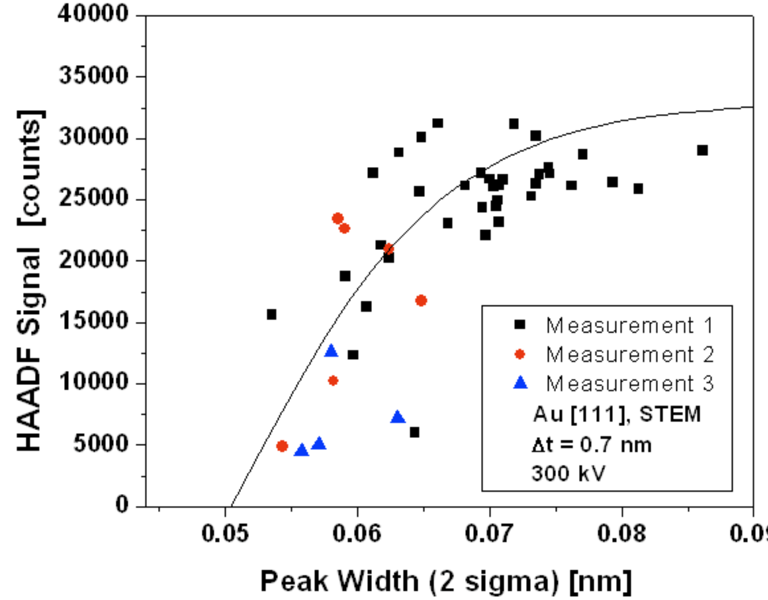
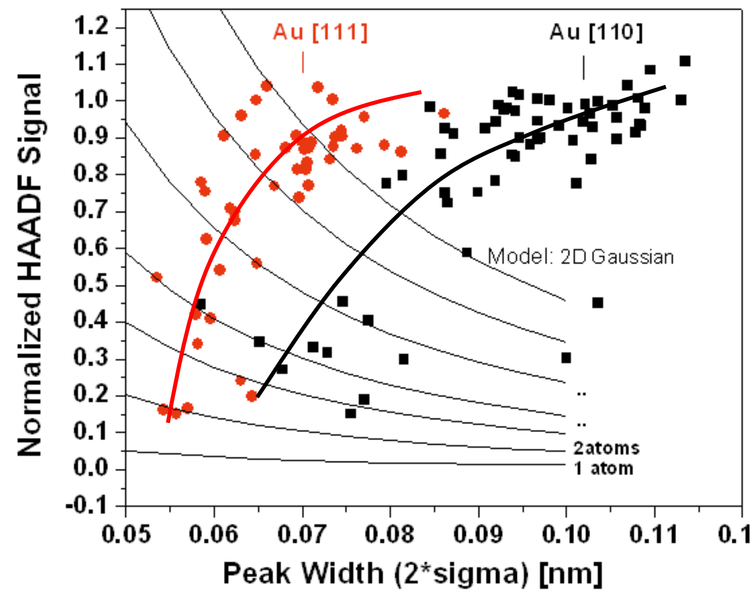




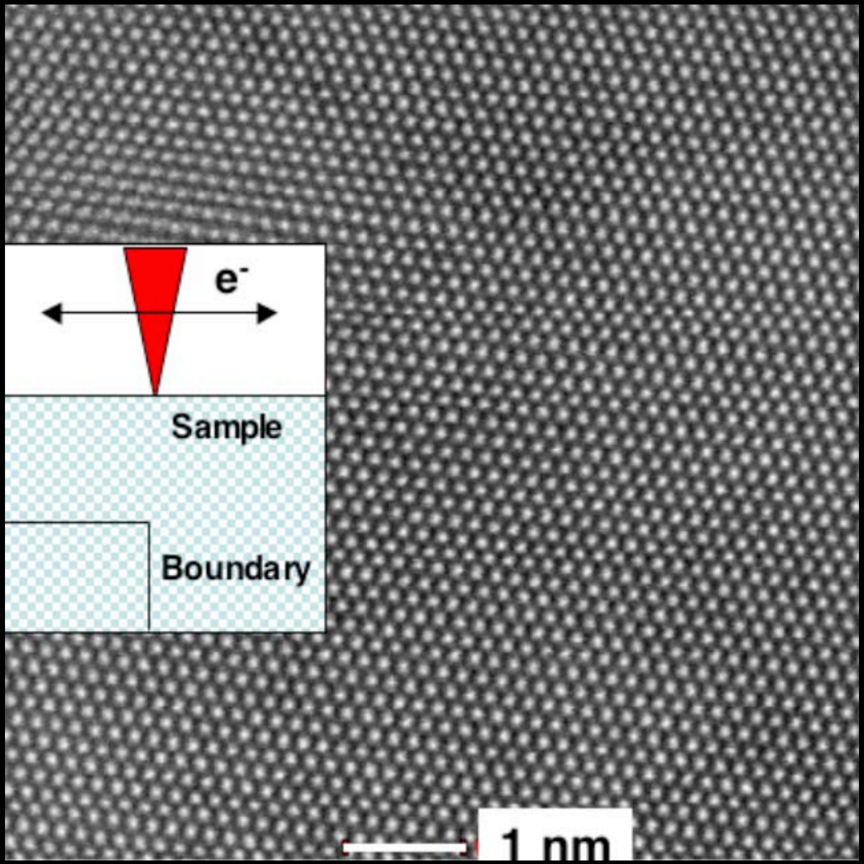


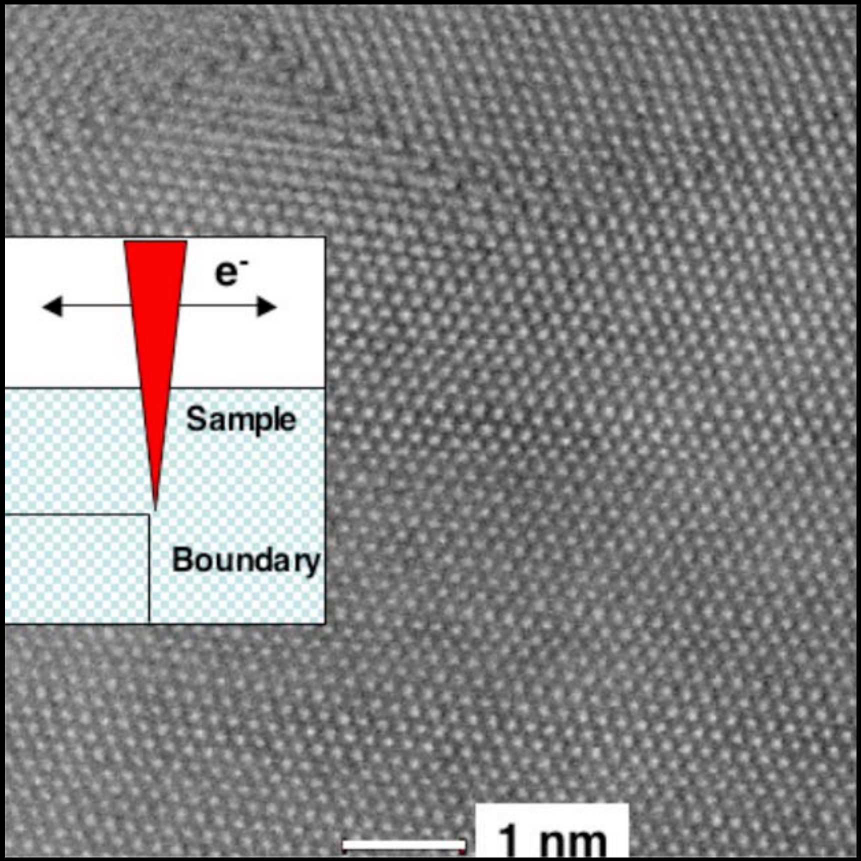


Electron channeling limits resolution in HAADF images, too

Towards 3D Electron Microscopy

STEM Depth sectioning (Au [110]): df = 6 nm - nm resolution





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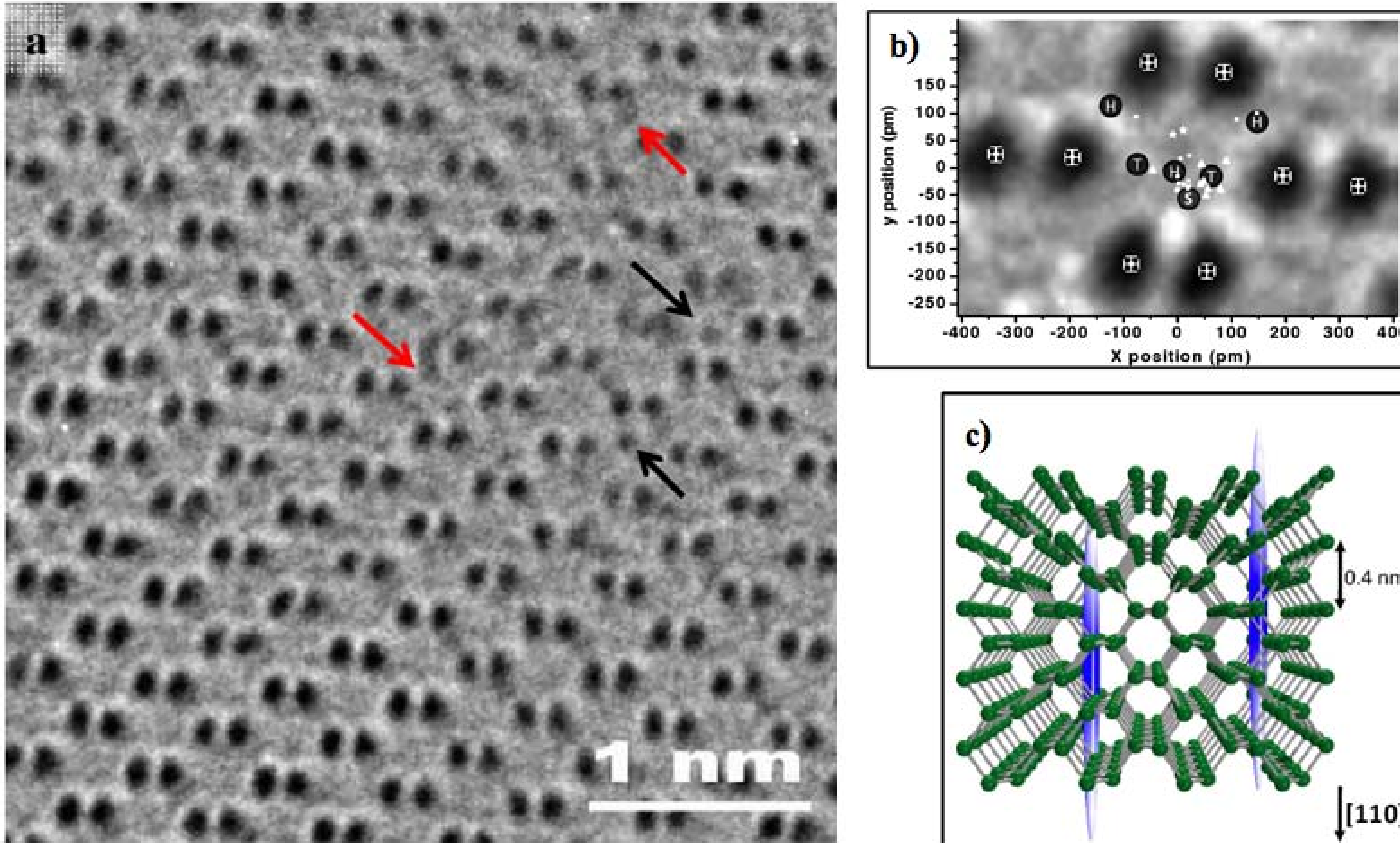
Self Interstitials in Ge [110]



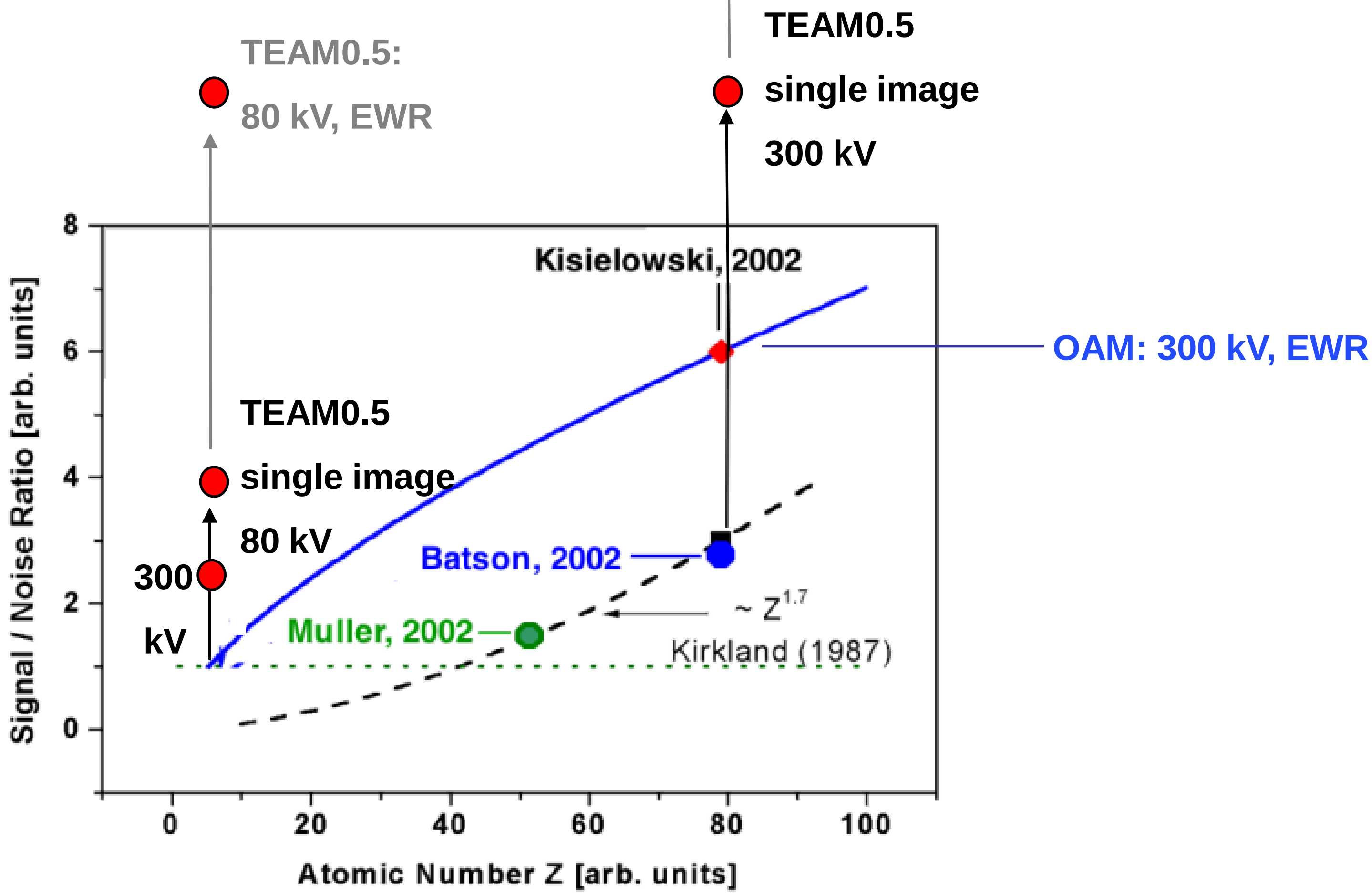
Ge[110], D. Alloyeau, B. Freitag, S. Dang. L.W. Wang, C.Kisielowski, Phys. Rev. B. 2009, in press

- **First detection of self interstitials & 3D reconstruction from single projection**
- **Dose limits now imaging of soft *and hard materials***

Progress

There is plenty of room for improvement (TEAM1)

S/N ratio of 20 for 1 gold atom is feasible



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Conclusions

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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

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