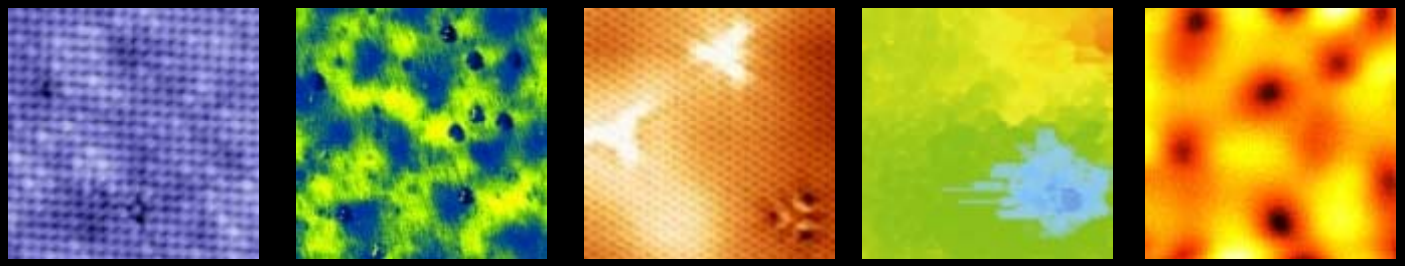
# Got Mott? Nanoscale Explorations of Electronic Transitions


Jenny Hoffman



Contributors:

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Thanks to:

NSEC

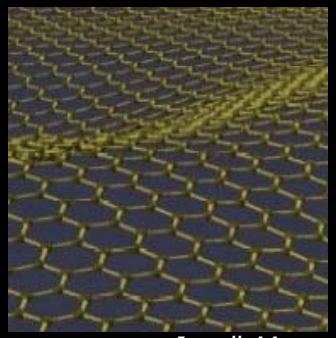
HARVARD

NSF

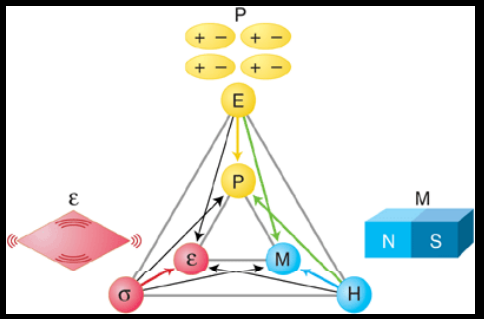
AIR FORCE OFFICE OF SCIENTIFIC RESEARCH
UNITED STATES AIR FORCE

# Correlated Electron Materials

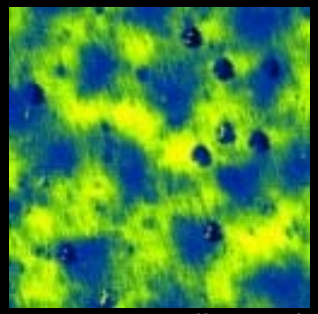
- High-Tc superconductors: $\rho = 0$ up to T = 135 Kelvin
- Colossal magnetoresistance: B-field changes electrical conductivity by 1000x
- Multi-ferroics: magneto-electric-elastic coupling
- Heavy fermion materials: $m^* = 1000\ m_e$
- Graphene: $m^* = 0$
- Topological insulators: dissipationless spin; fractional charges; magnetic monopoles



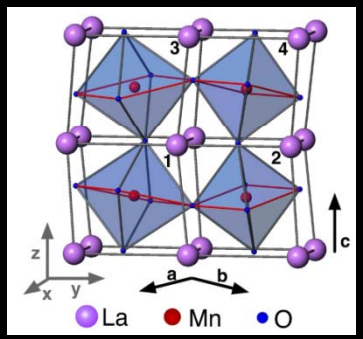
*Jannik Meyer*



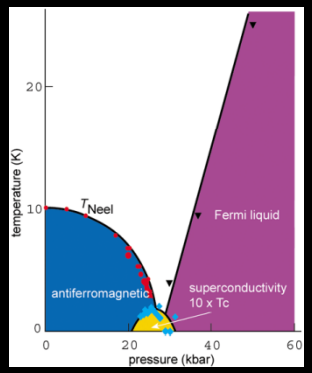

*Science 309, 391 (2005)*



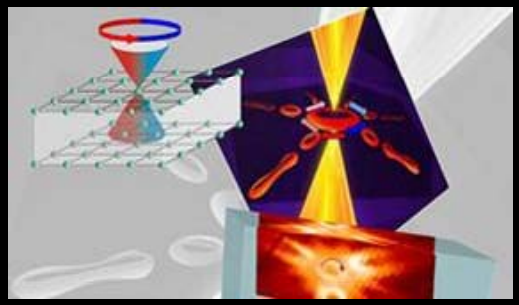
*Hoffman Lab*




*JARA, RWTH Aachen*




*Physics World*



*Zahid Hasan*

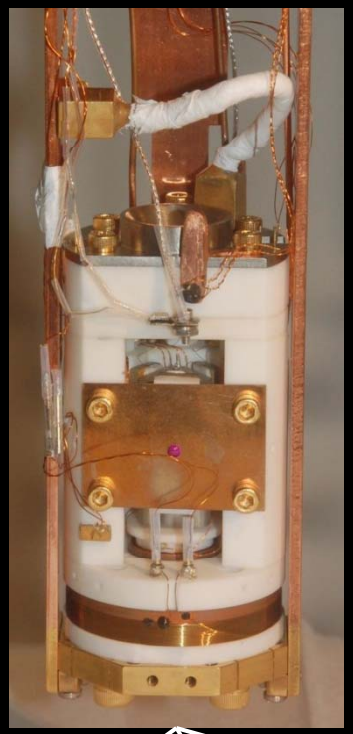
Metals: electronic Bloch states are homogeneous, no spatial variation

Correlated electron materials: weak screening, spatial inhomogeneity

→ We need local probes to study these materials.

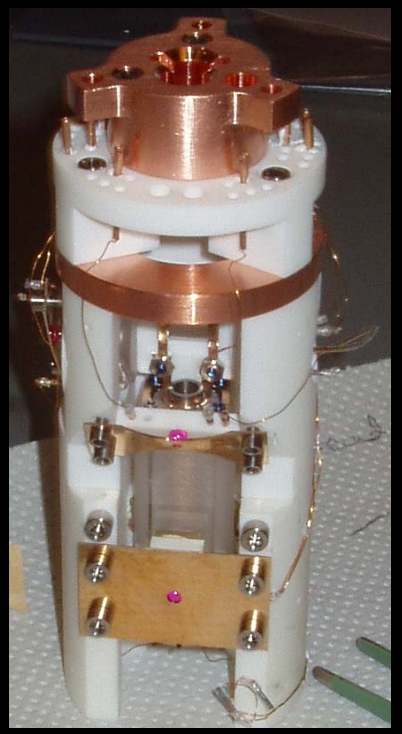
# Hoffman Lab Local Probes

Scanning Tunneling Microscope

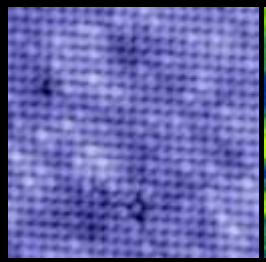


Force Microscope

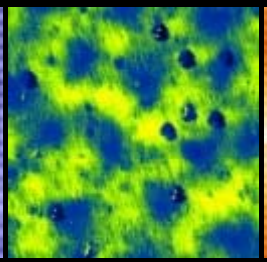
Ultra-high vacuum STM



coming on line soon!
- spin-polarized tunneling
- atom-moving

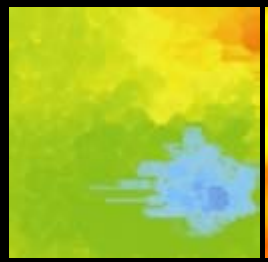


cuprate high-$T_c$ superconductor

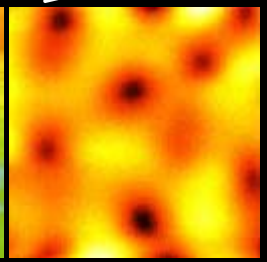


122 iron pnictide

topological insulator



metal-insulator transition



1111 iron pnictide

# Outline

High-Tc Superconductivity

- Can we understand it? → need to understand Mott
- Can we use it? → need to understand vortices
  - scanning tunneling microscope
    - → vortex imaging in $Ba(Co_xFe_{1-x})_2As_2$
  - force microscope
    - → vortex manipulation in $NdO_{1-x}F_xFeAs$

Metal-insulator transition in $VO_2$

- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

Outlook

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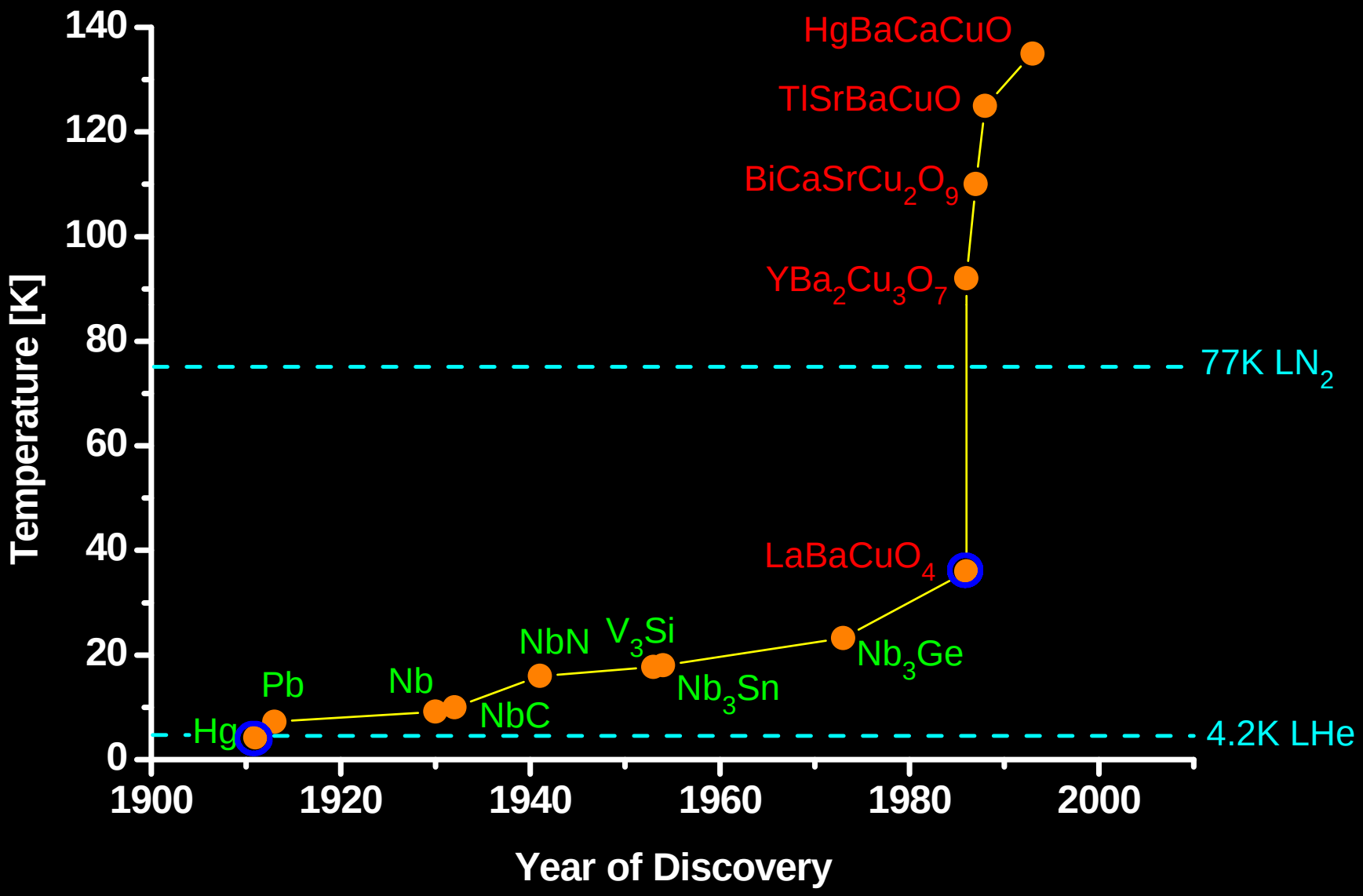
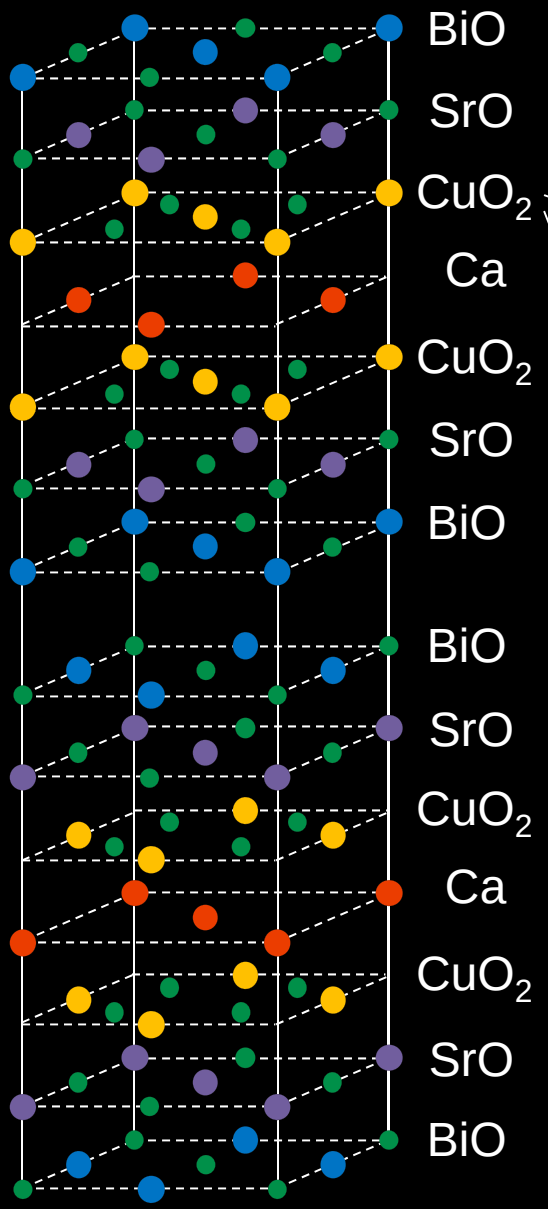
Outlook

# 100 Years of Superconductors



# Cuprate High-Tc Superconductor

$Bi_2Sr_2CaCu_2O_{8+d}$ ($T_c^{max}$ ~ 93 K)

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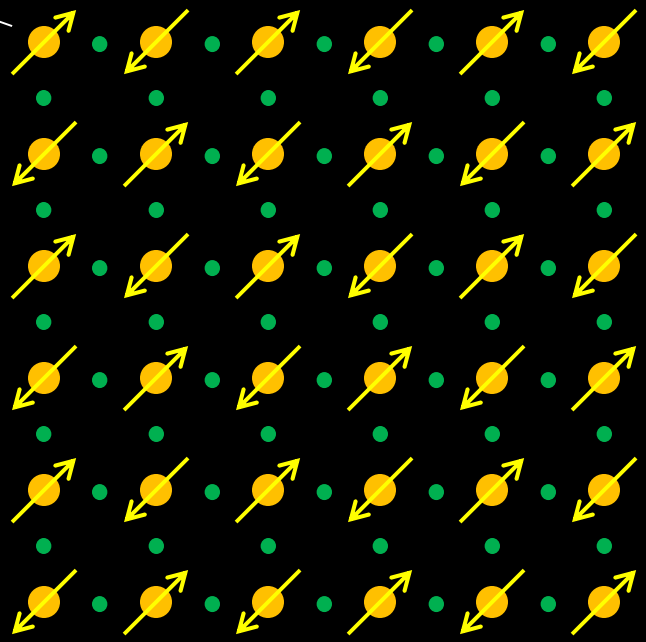
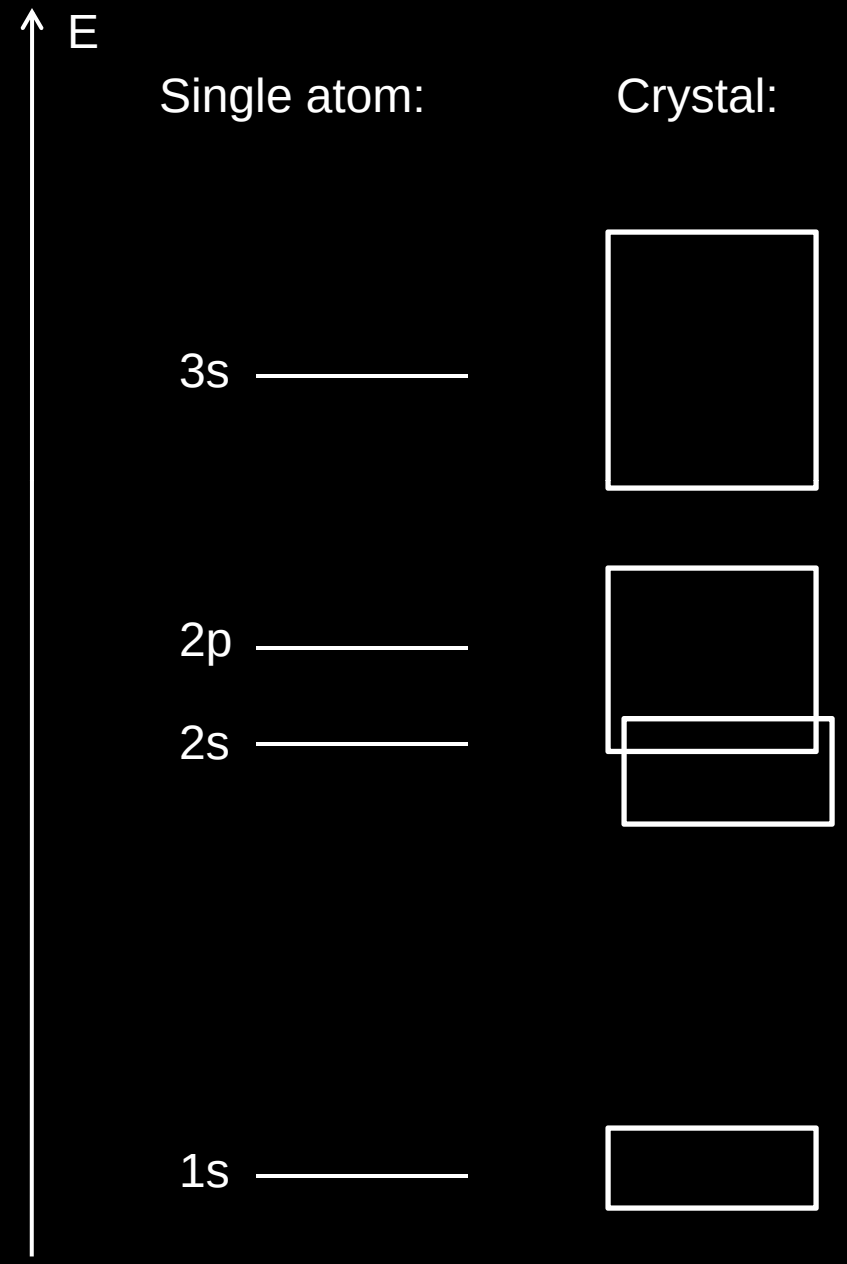
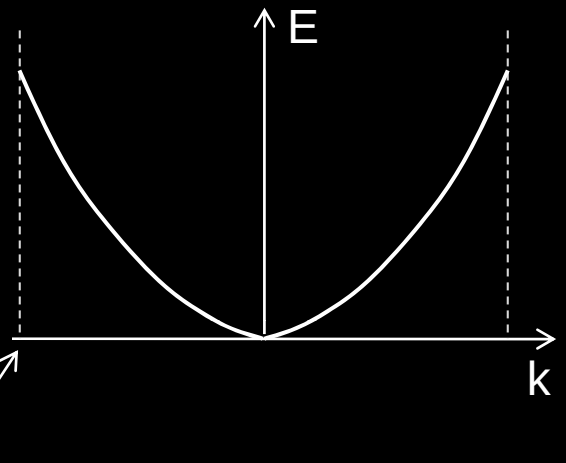


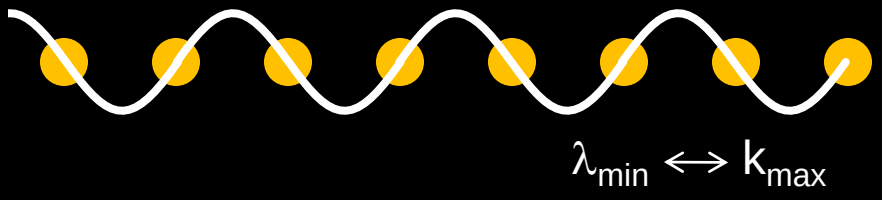
# Band Theory



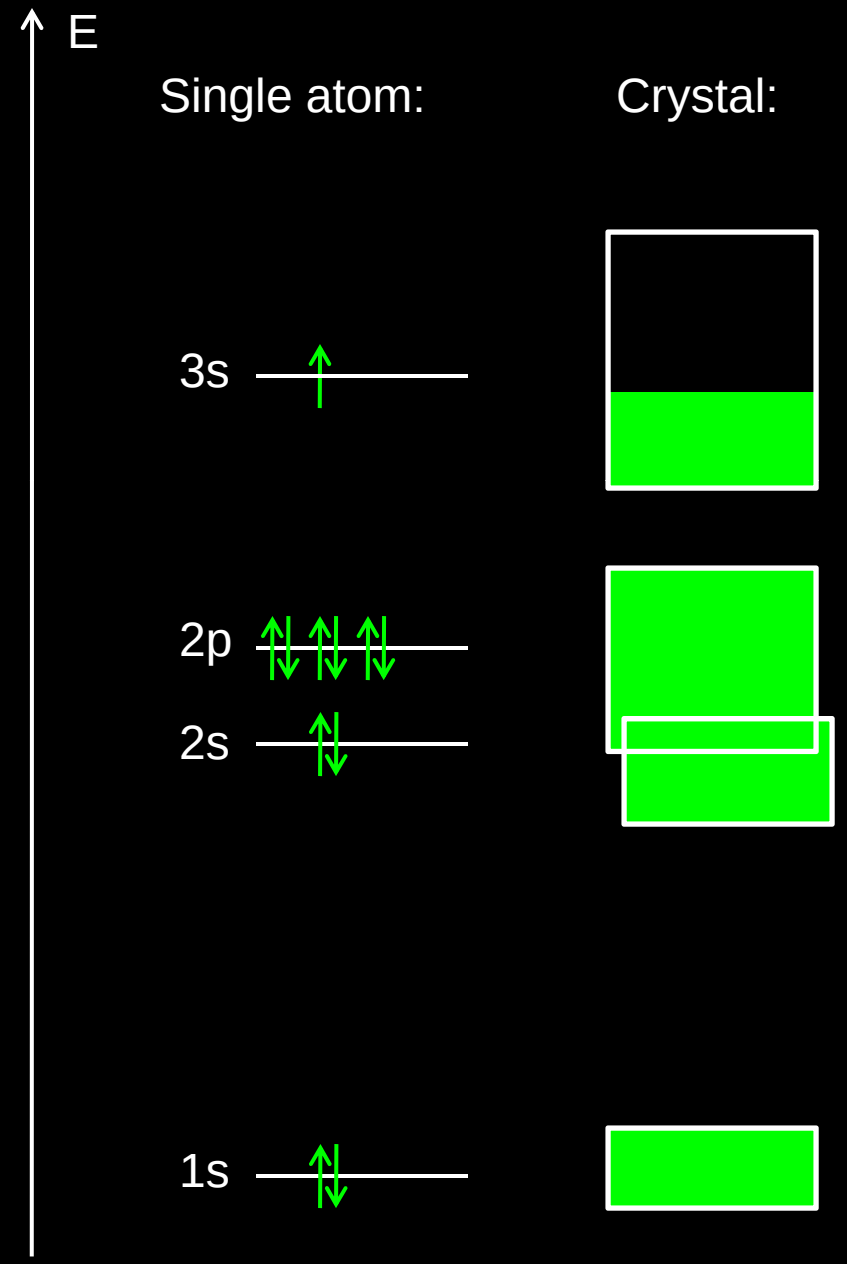

Add momentum information:




Brillouin zone edge:
highest unique wavevector k
(larger k's aren't sampled by the atoms so they are aliased back to smaller k's)



# Band Theory: Metal

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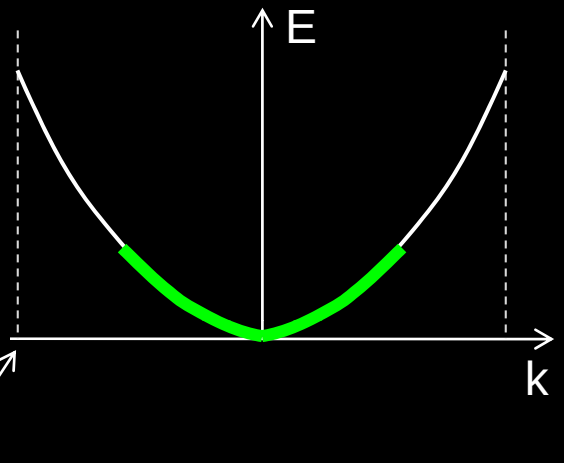


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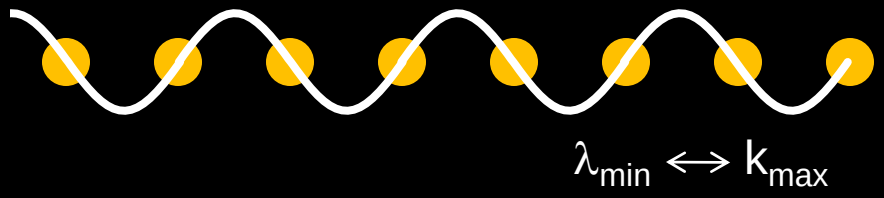
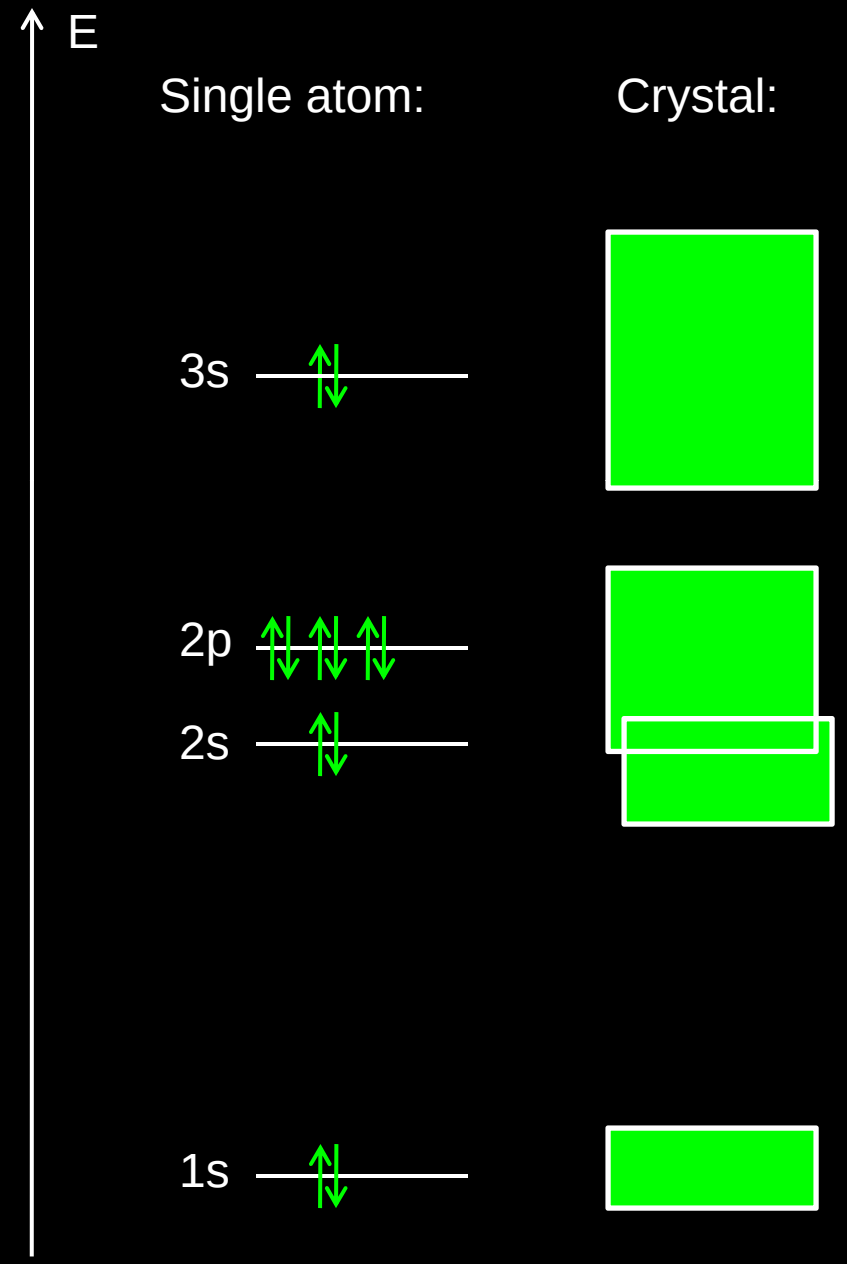


# Band Theory: Insulator

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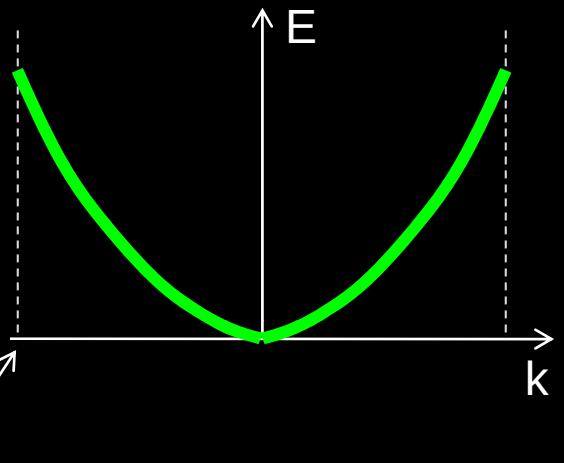


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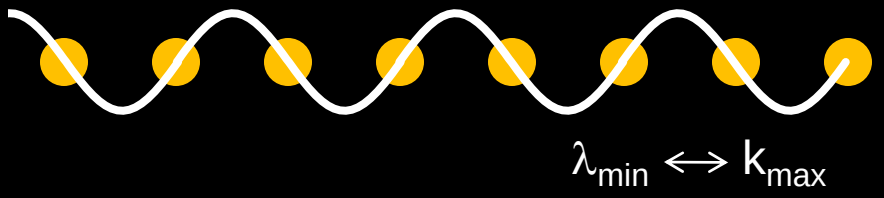

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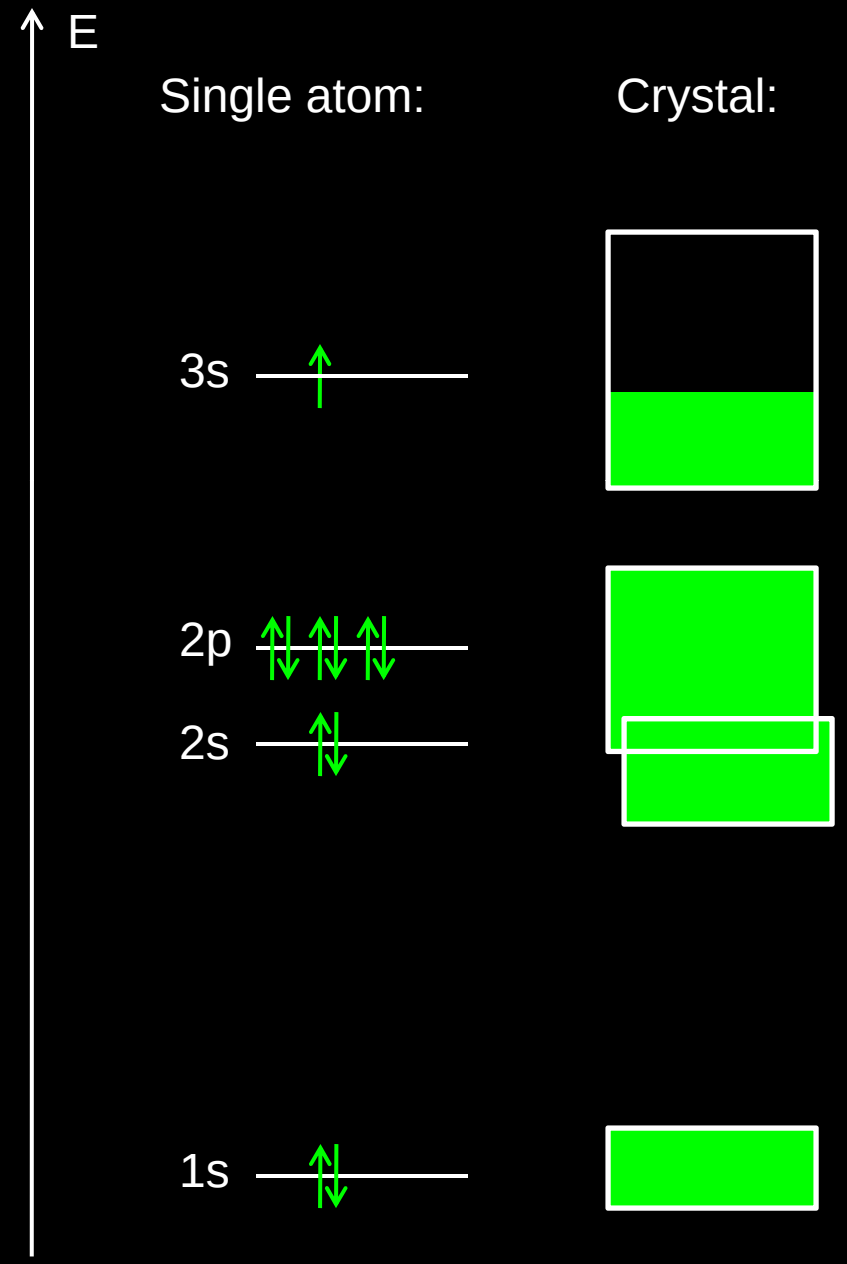


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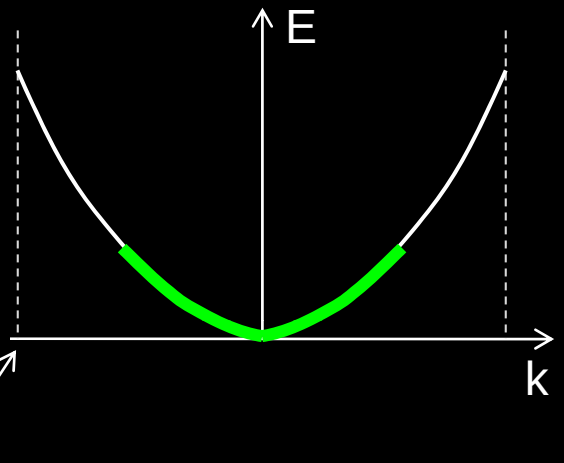


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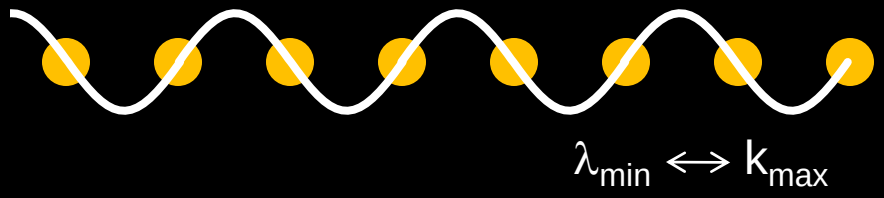
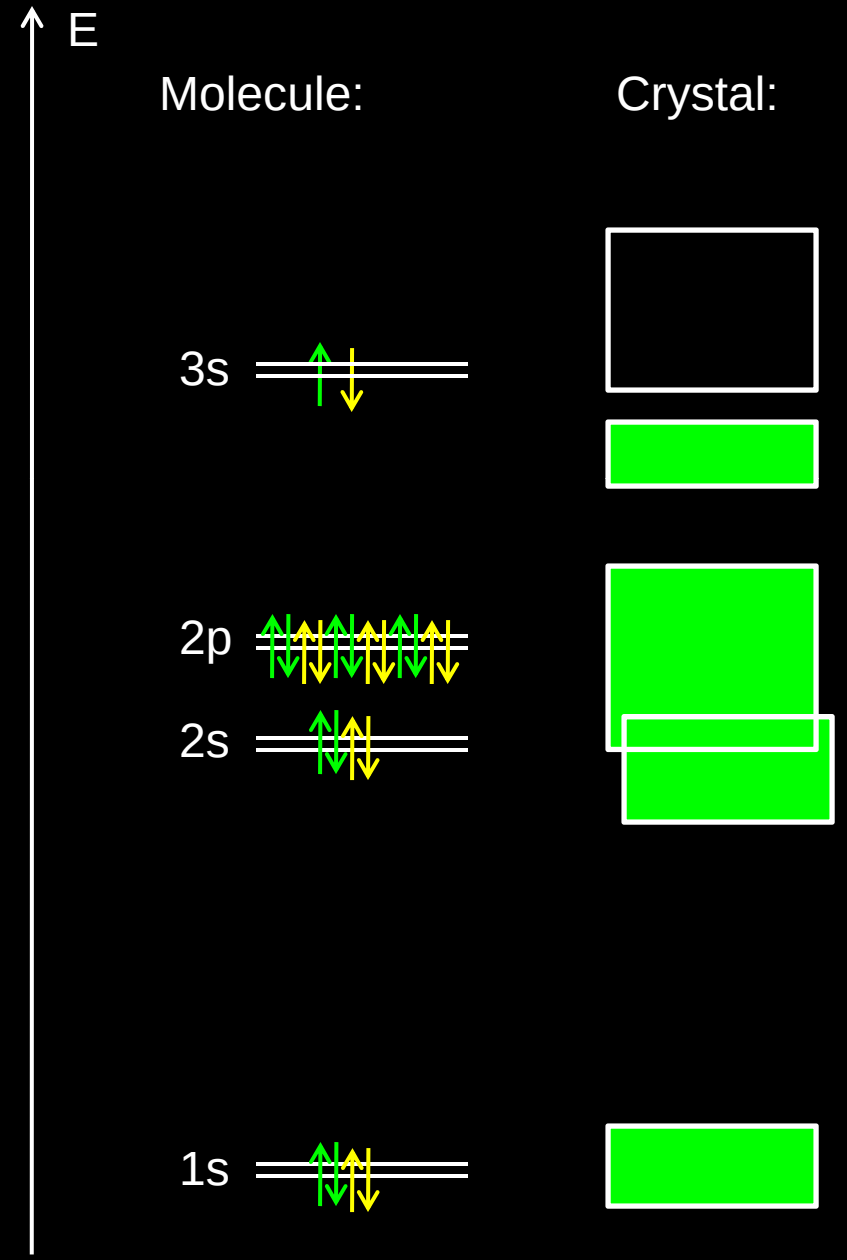


# Band Insulator: Peierls Transition

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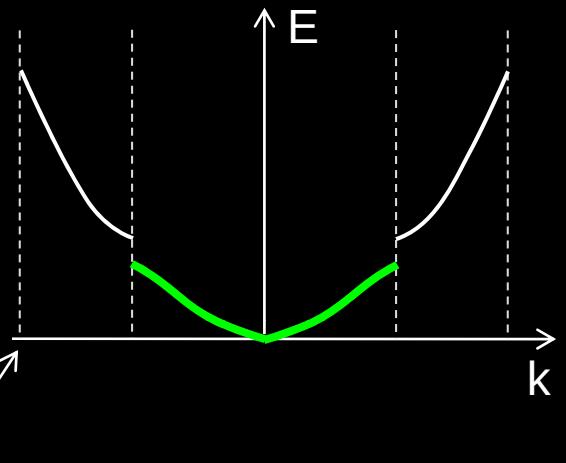


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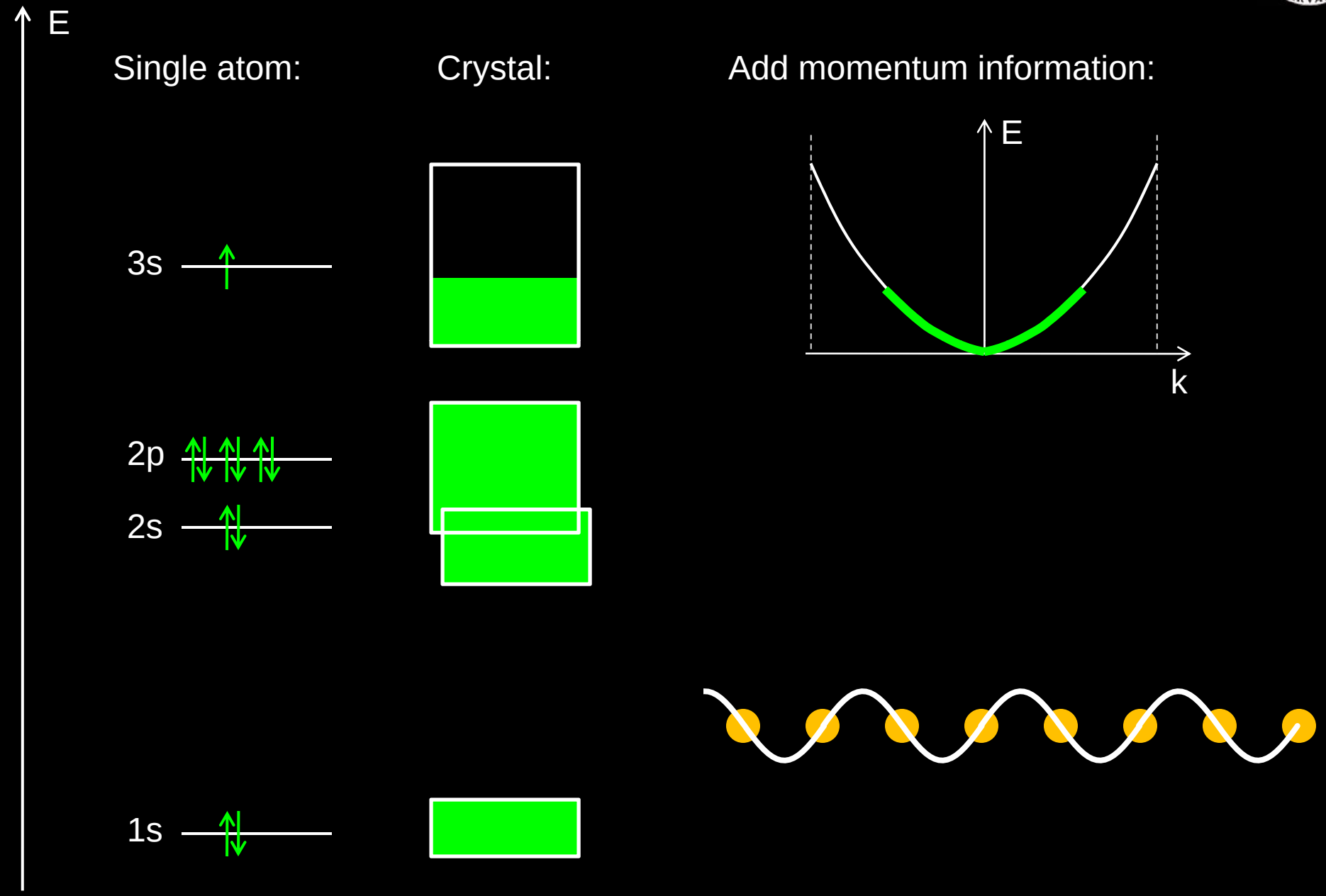
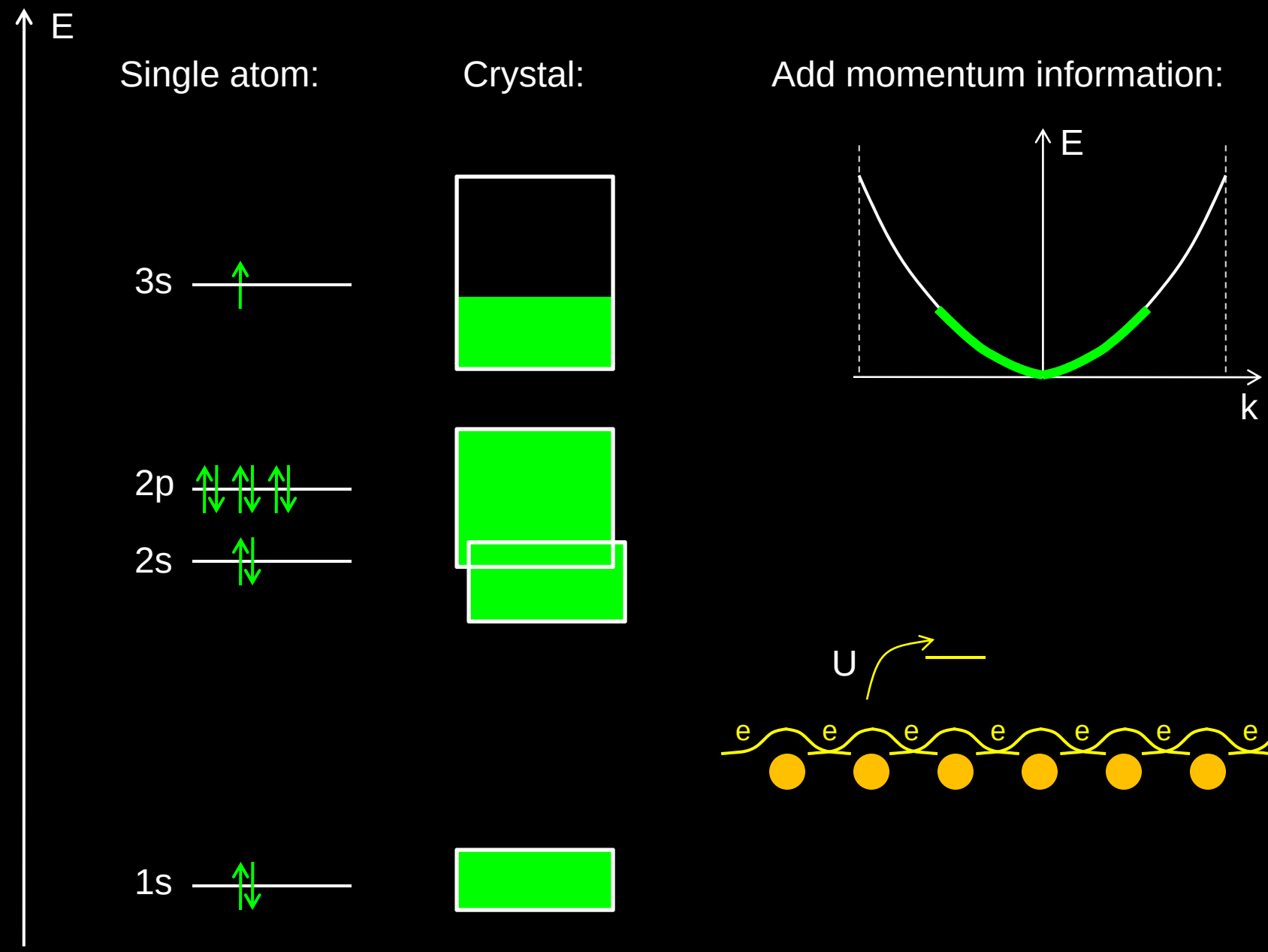
new $\lambda_{min}$ doubles $\leftrightarrow$ $k_{max}$ halves

# Band Theory: Metal



# Add e-e Correlations



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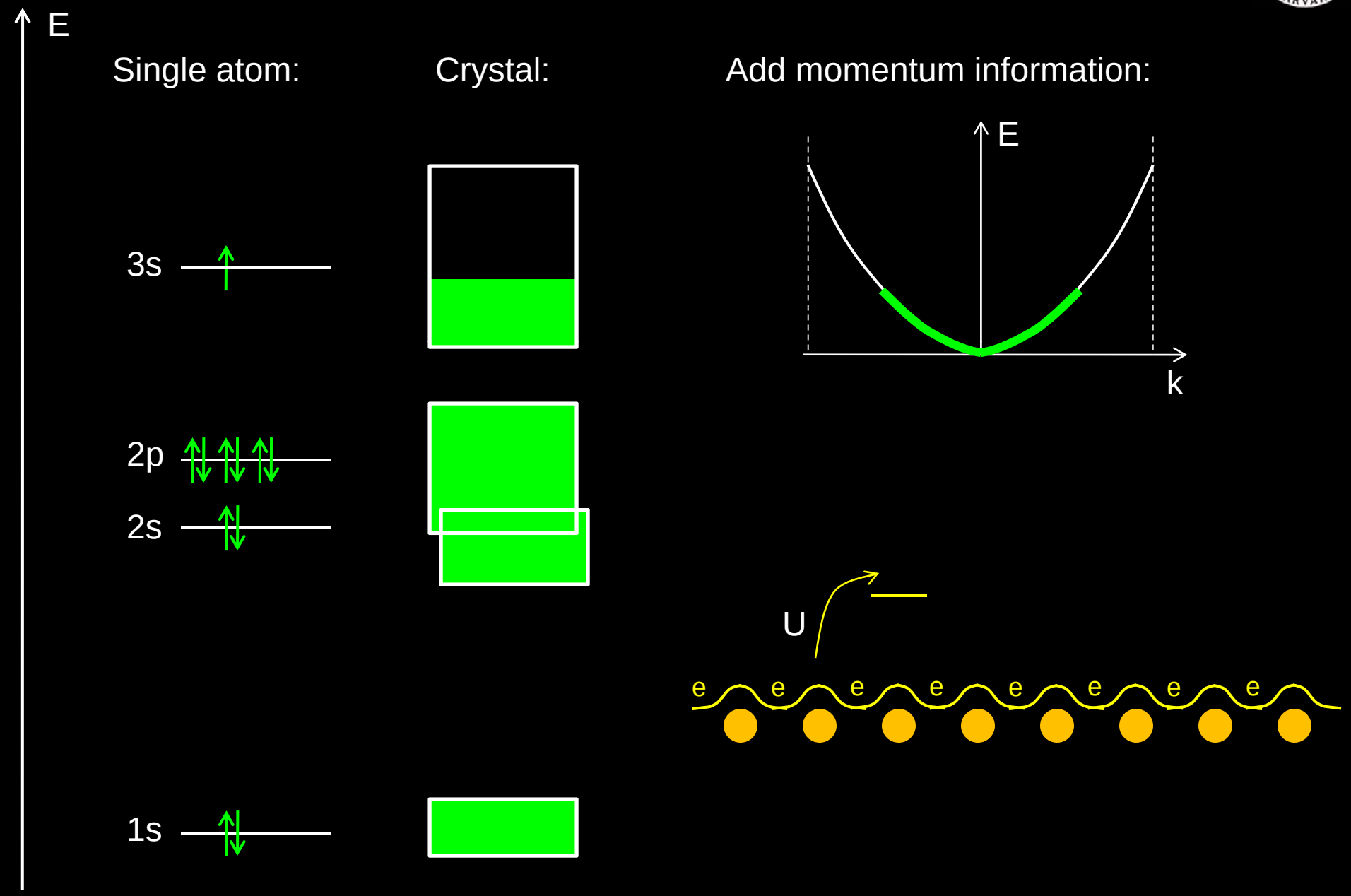

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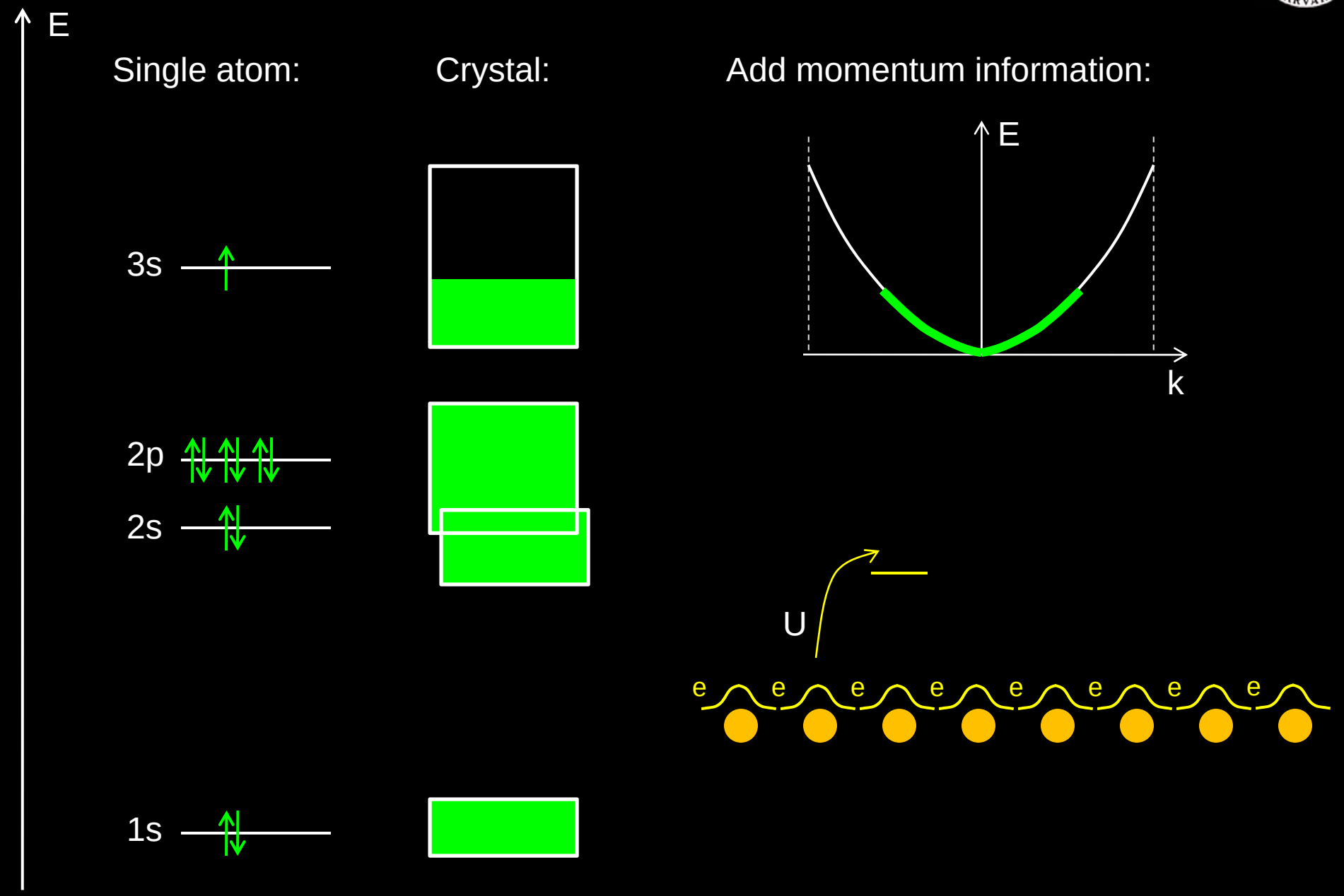
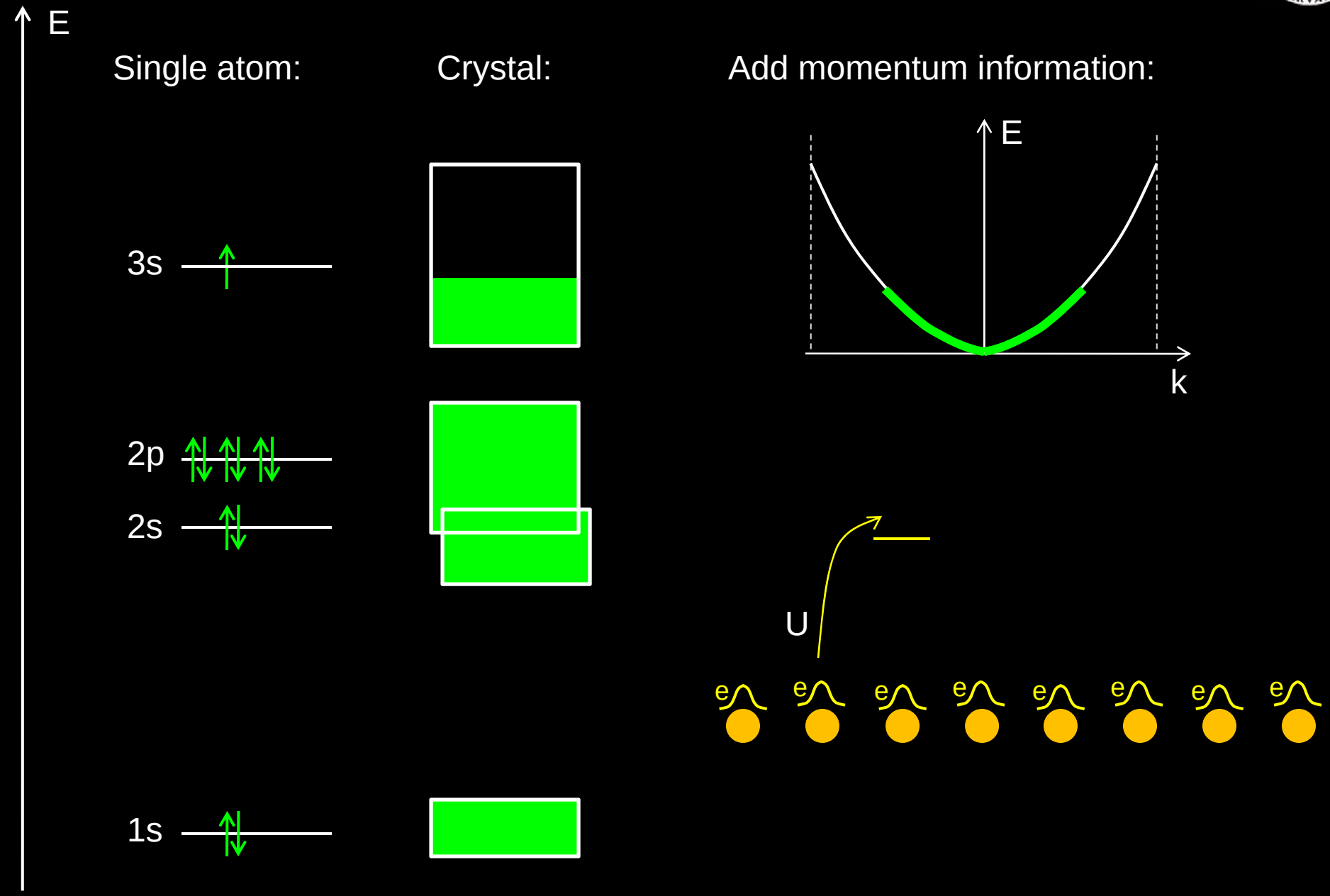


# Mott Insulator



# Mott Transition

localized

delocalized

further delocalized

# Cuprates: Doped Mott Insulators

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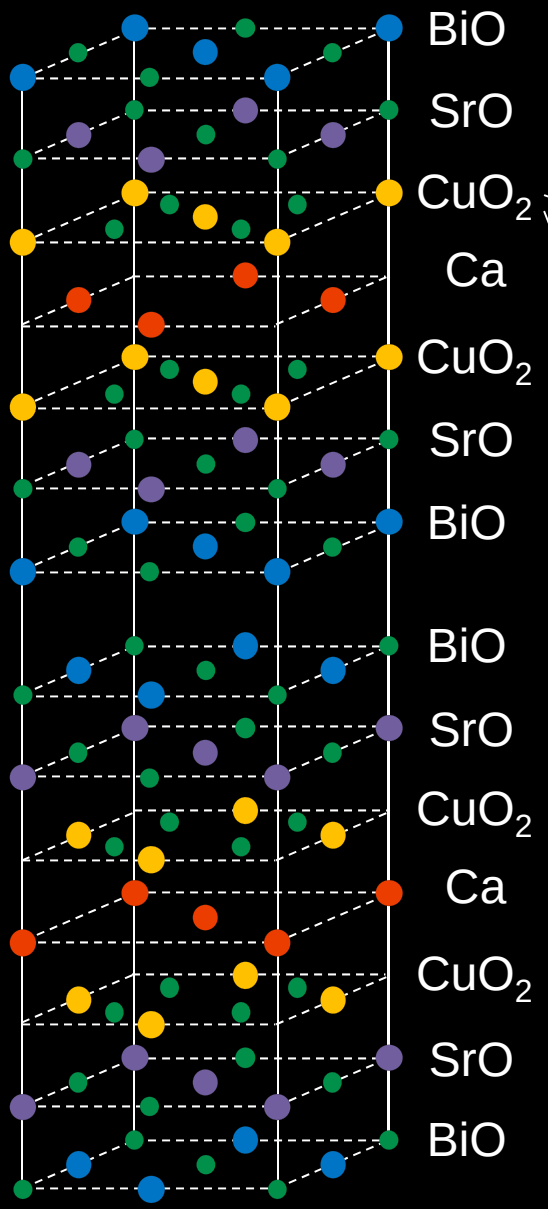


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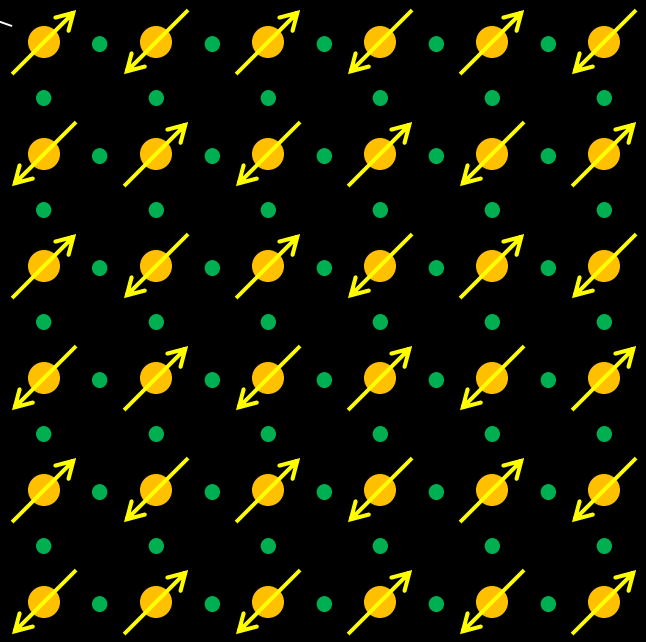
→ half-filled band, should be metal?

Strong Coulomb repulsion
→ Mott insulator



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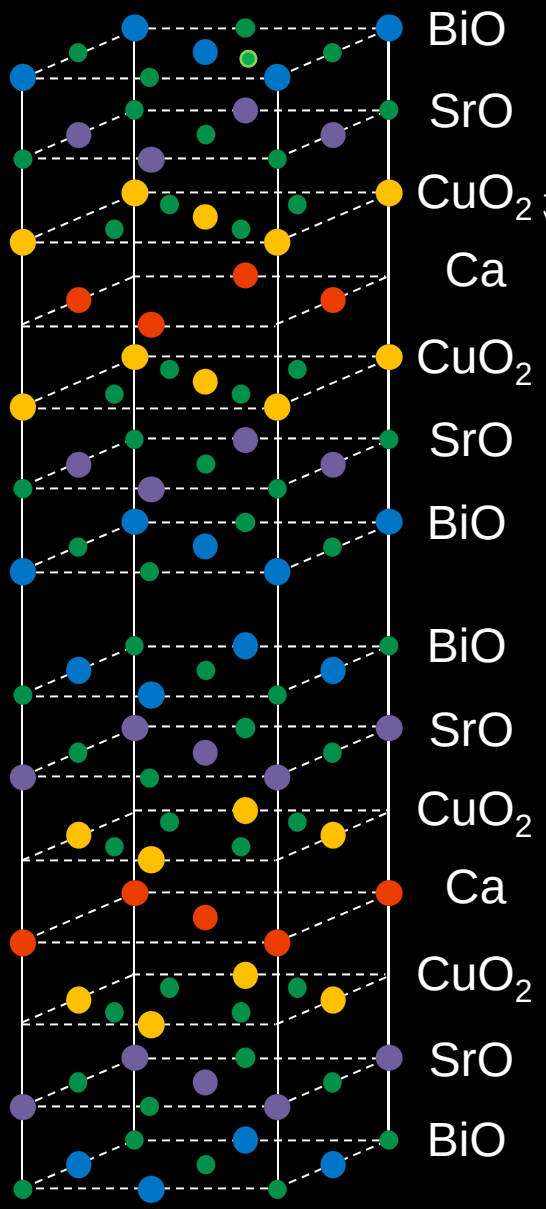


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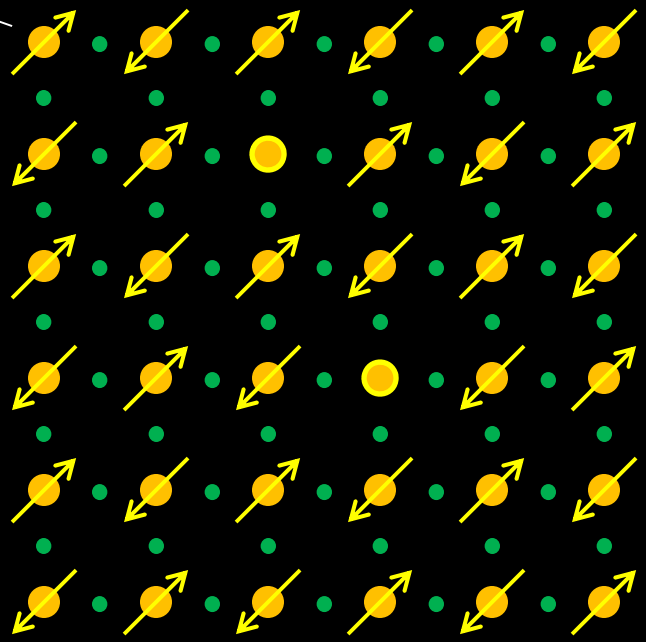

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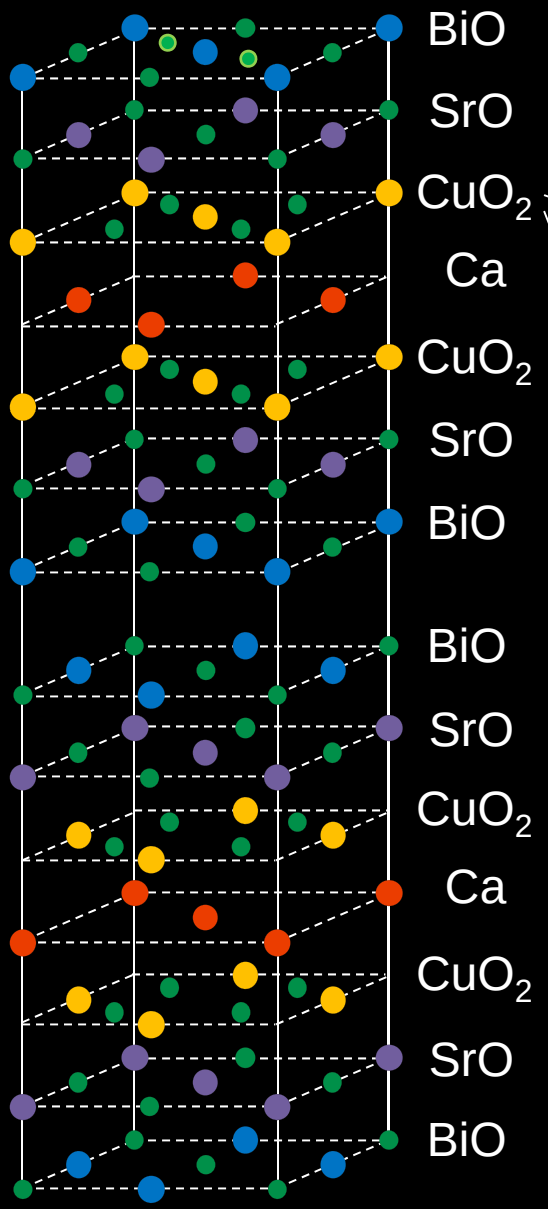


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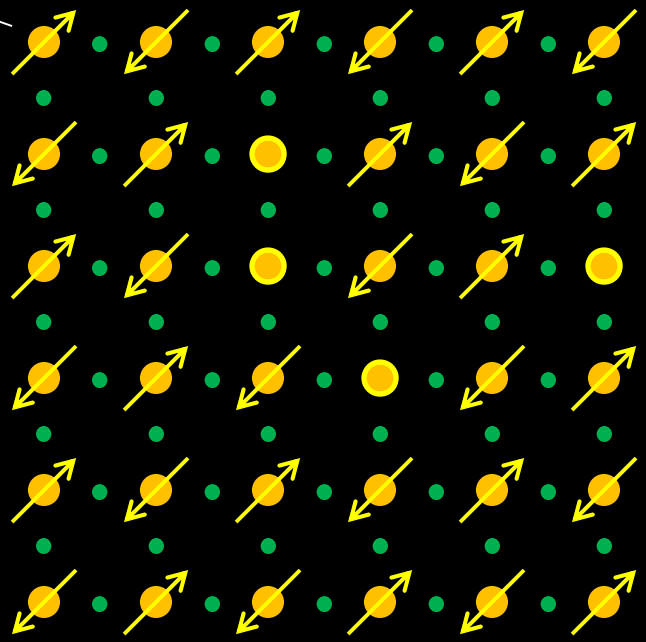
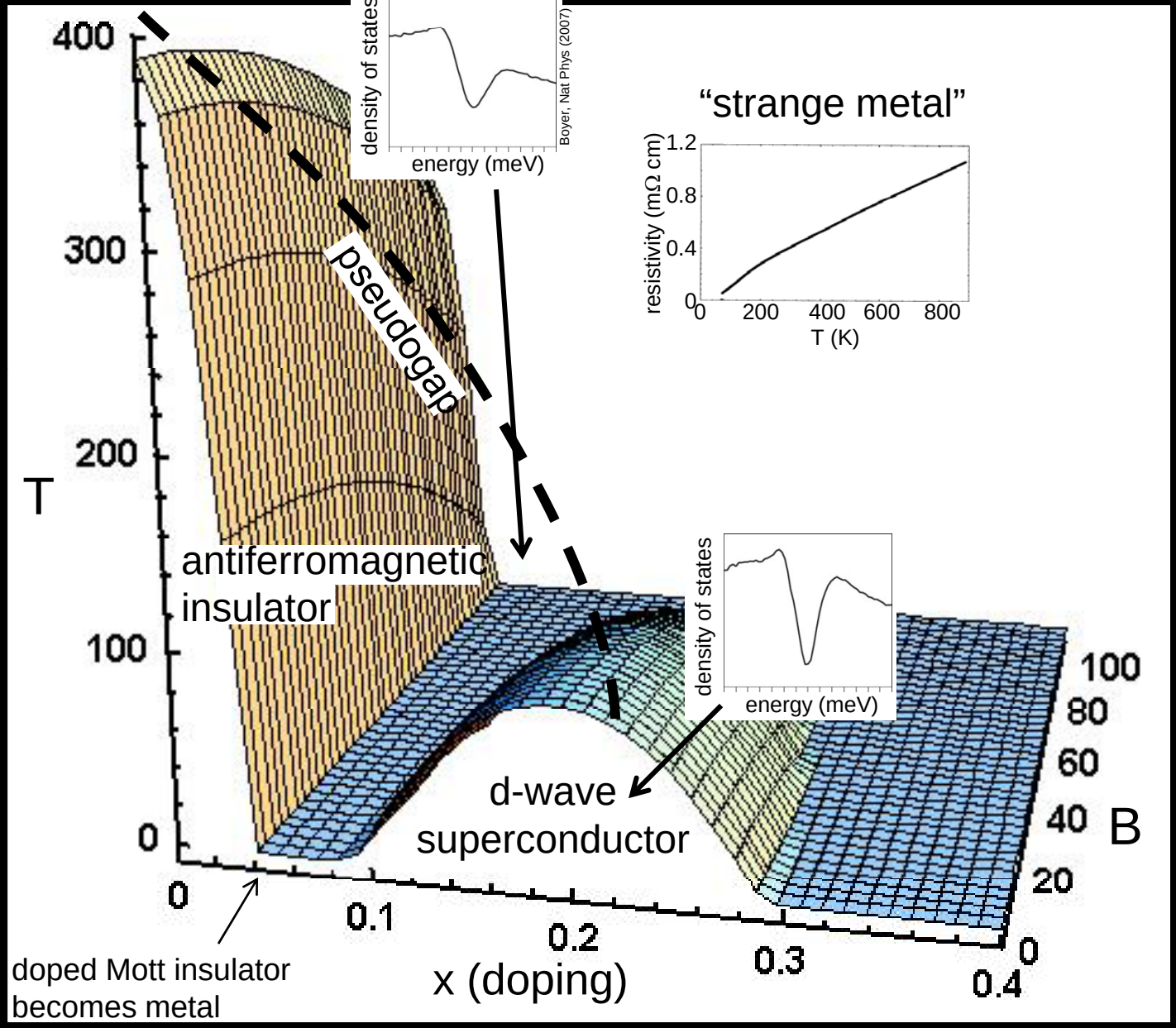


electronic interactions: drive holes apart

spin interactions: drive holes together

# Cuprate Phase Diagram



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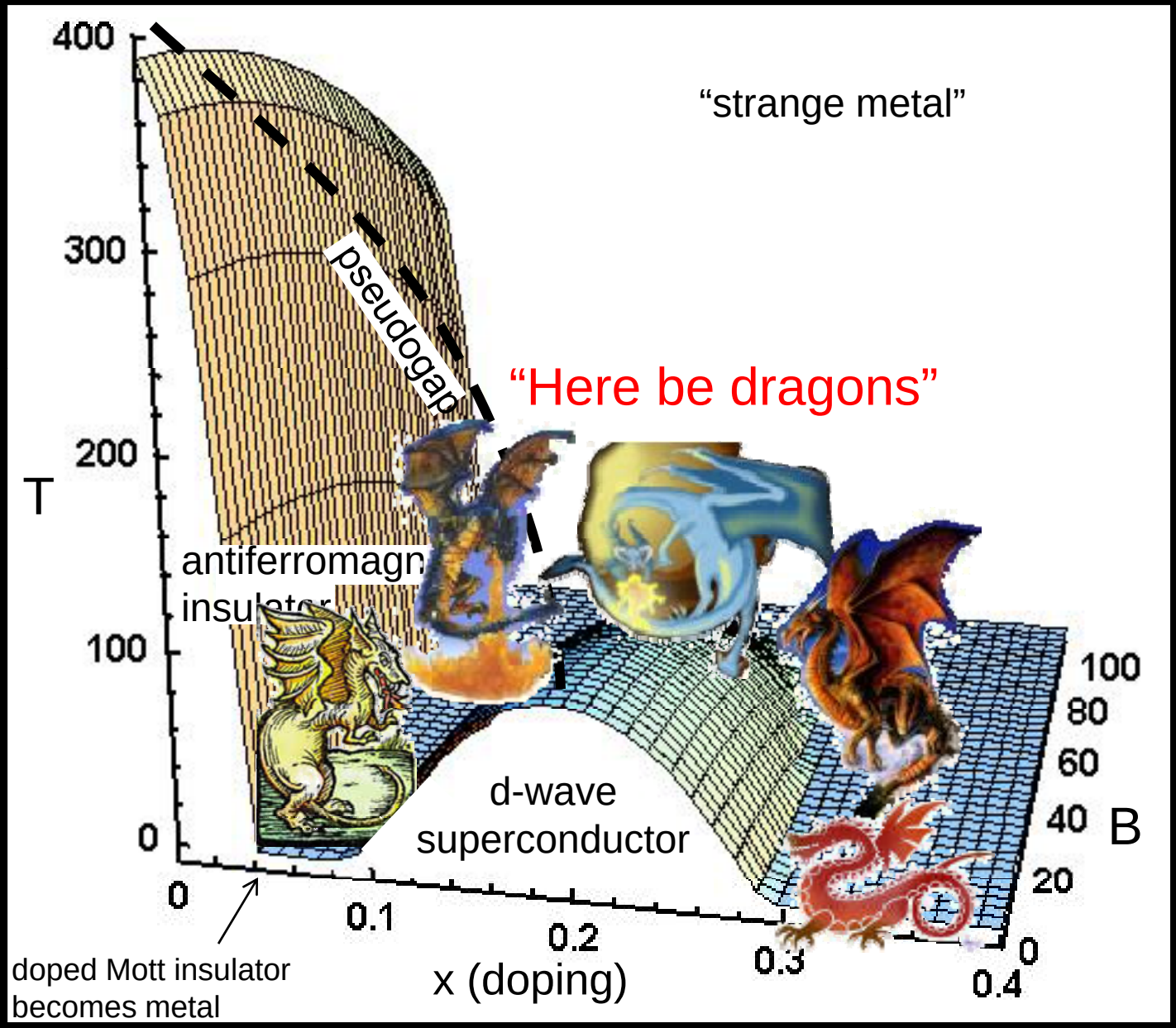

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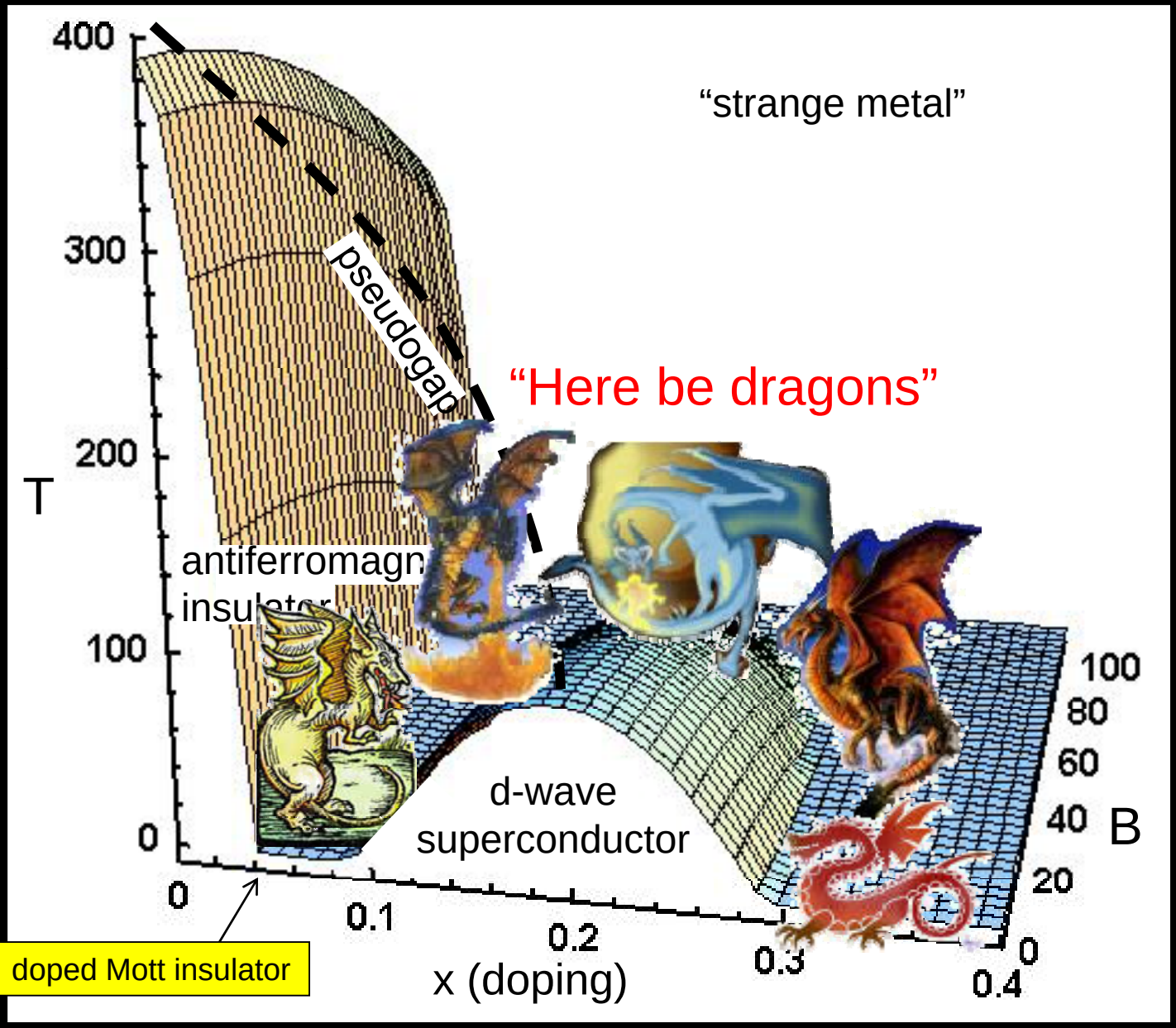
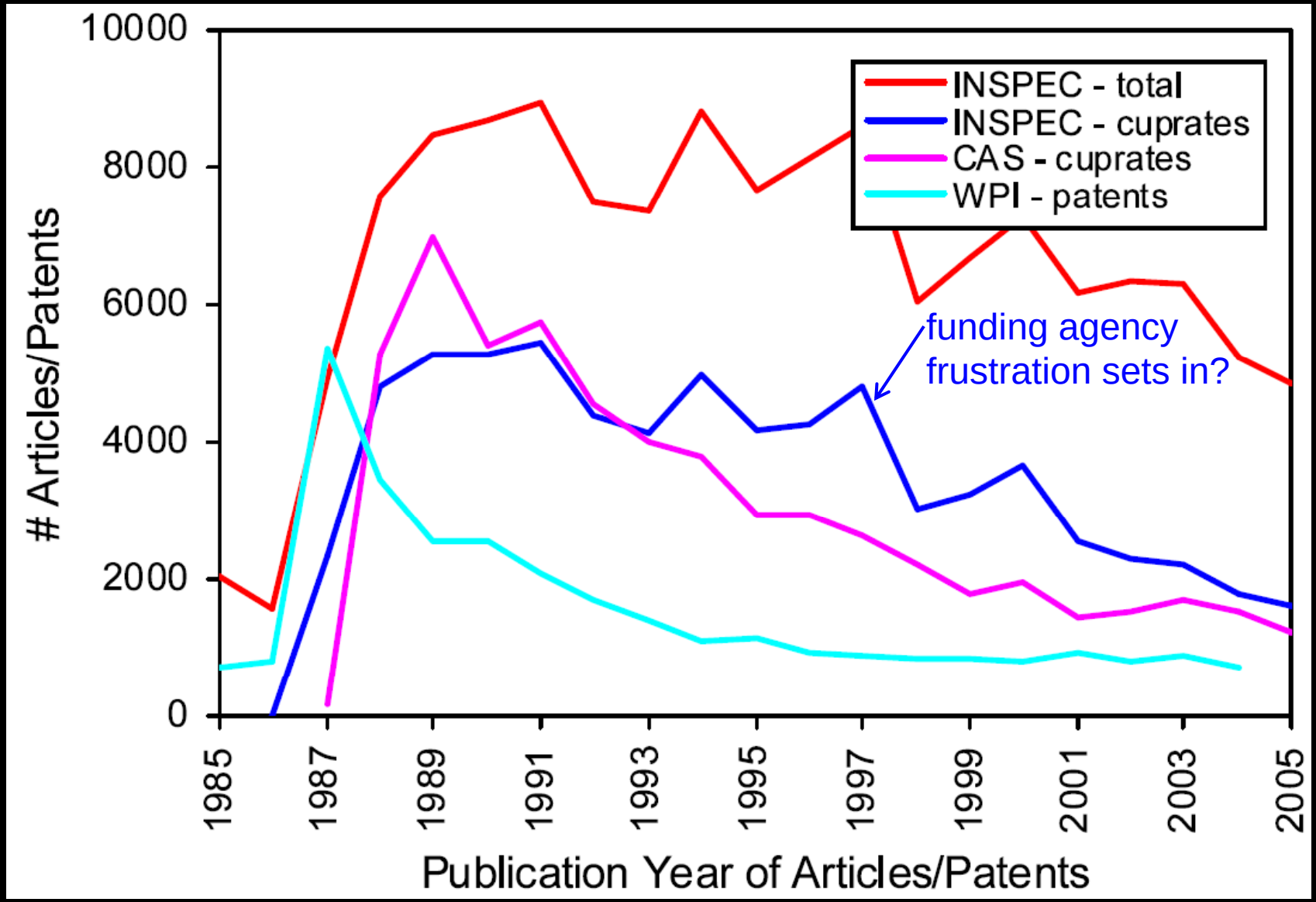


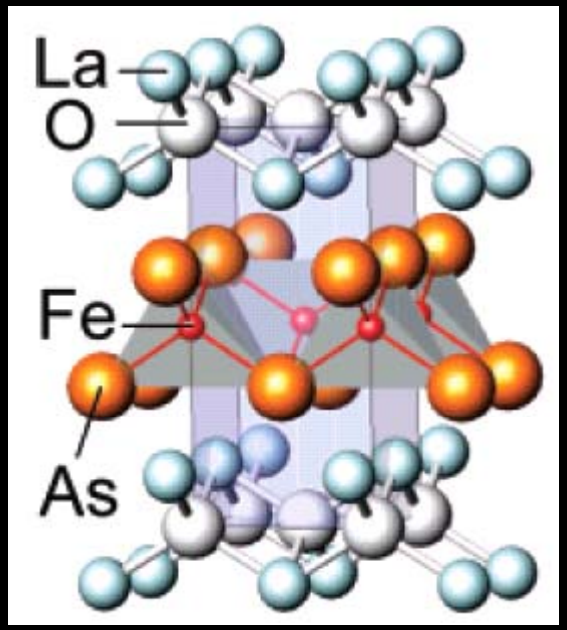
# Interest in High-$T_c$ Cuprates




*J Supercond Nov Magn 21, 113 (2008)*

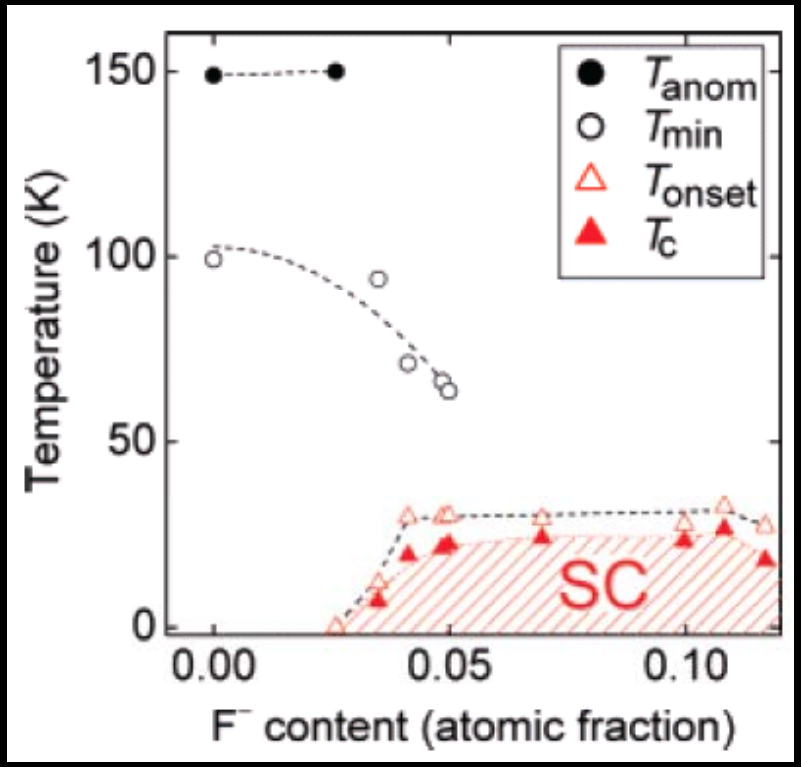
# A New Revolution in Superconductivity

Kamihara *et al*, J. Am. Chem. Soc. 130, 3296 (Feb 23, 2008)

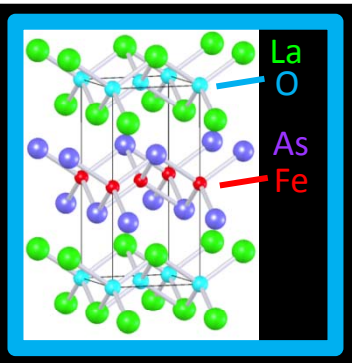



insulating layer

conducting layer

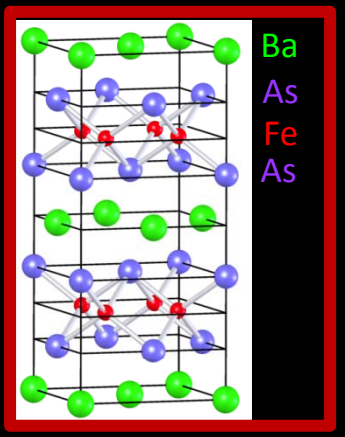


# History of Iron-Pnictide Superconductivity




1111

122




$T_c$ (K)

60
50
40
30
20
10
0

$SmFeAsO_{1-x}$
$Gd_{1-x}Th_xFeAsO$
$SmFeAsO_{1-x}F_x$
$PrFeAsO_{1-x}F_x$
$GdFeAsO_{1-x}$
$Tb_{1-x}Th_xFeAsO$
$NdFeAsO_{1-x}F_x$
$TbFeAsO_{1-x}F_x$
$DyFeAsO_{1-x}F_x$
$SmFeAsO_{1-x}F_x$
$CeFeAsO_{1-x}F_x$
$Sr_{1-x}K_xFe_2As_2$
$GdFeAsO_{1-x}F_x$
$Ba_{1-x}K_xFe_2As_2$
$Eu_{1-x}K_xFe_2As_2$
e-doped
$LaFeAsO_{1-x}F_x$
hole-doped
$La_{1-x}Sr_xFeAsO$
$Ca_{1-x}Na_xFe_2As_2$
$BaCo_xFe_{2-x}As_2$
$Eu_{1-x}La_xFe_2As_2$
$SrCo_xFe_{2-x}As_2$
$BaNi_xFe_{2-x}As_2$
$Li_{1-x}FeAs$
$FeSe_{0.5}Te_{0.5}$
$LaO_{1-x}F_xFeP$
$LaO_{1-x}NiBi$
$\alpha$-$FeSe_{1-x}$
$\alpha$-FeSe
LaOFeP
$BaNi_2P_2$
LaONiP
$SrNi_2As_2$

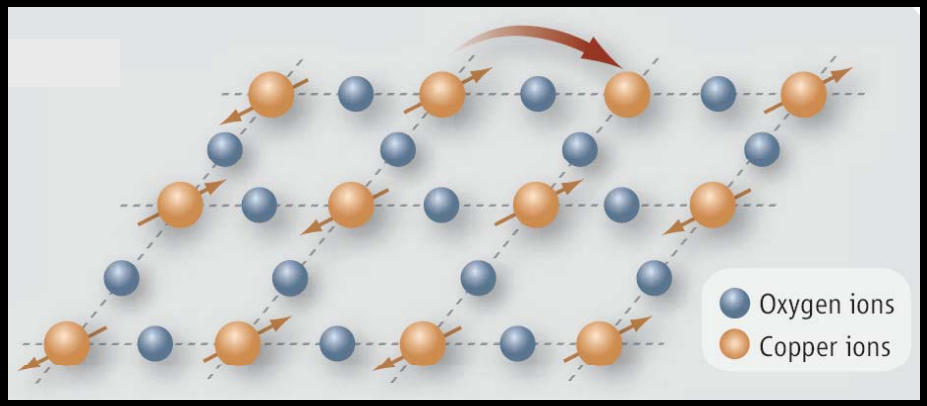
Feb Mar Apr May Jun Jul Aug Sep Oct Nov

2006-7

2008

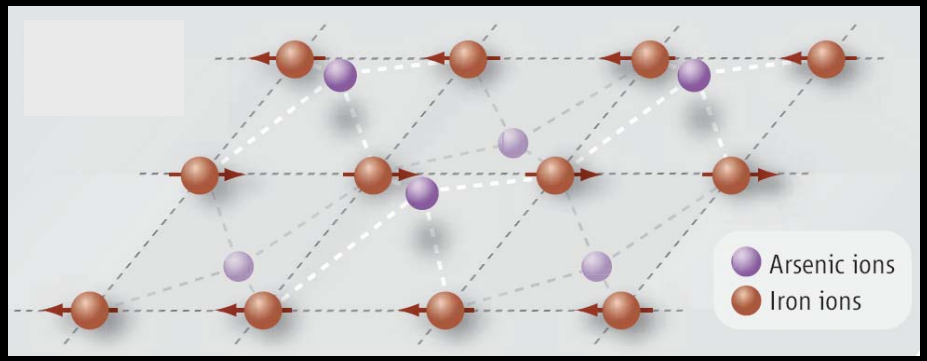
# Structure Comparison

## Cuprate Superconductors




antiferromagnetic
Mott insulator

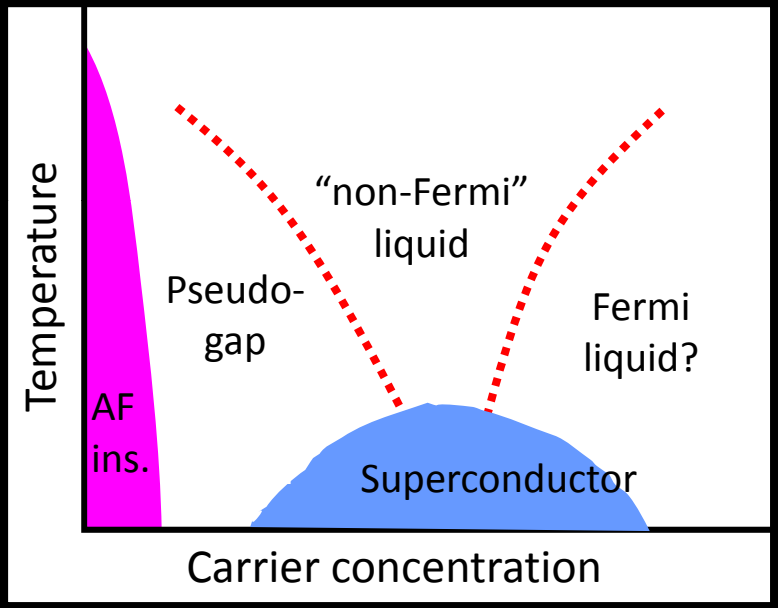
## Iron-Pnictide Superconductors




collinear antiferromagnet
semimetal

# Phase Diagram Comparison

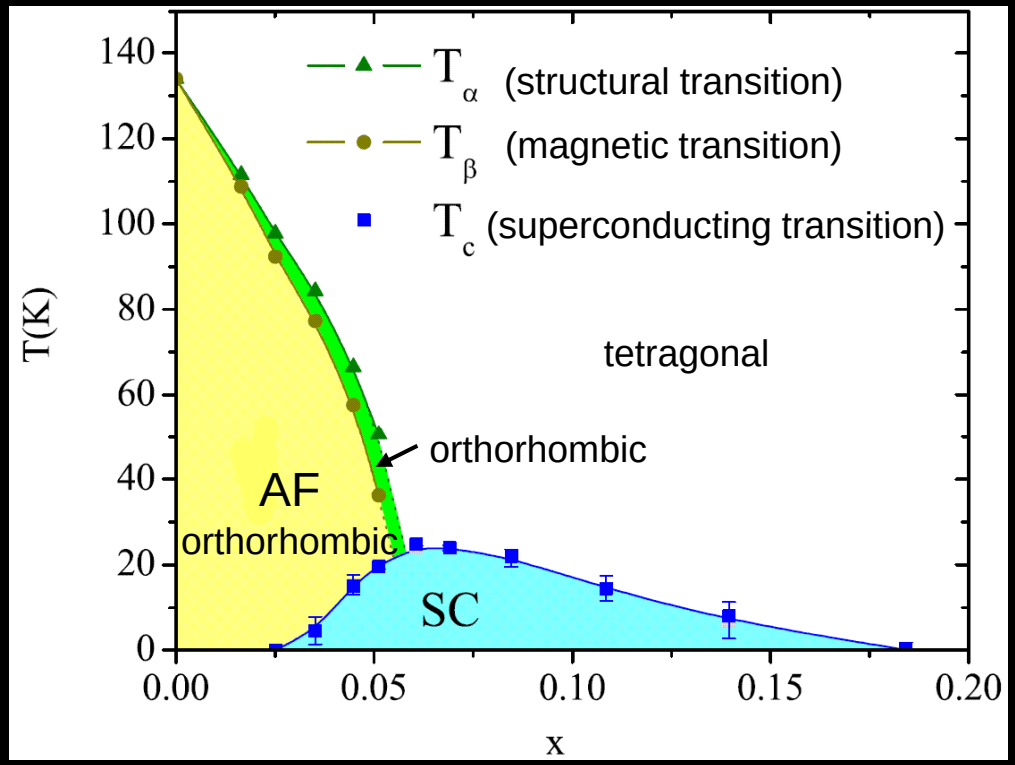
## Cuprate Superconductors




$T_c^{max} \sim 135$ K

## Iron-Pnictide Superconductors

$Ba(Co_xFe_{1-x})_2As_2$




$T_c^{max} \sim 57$ K

# Outline

High-Tc Superconductivity

- Can we understand it? → need to understand Mott
- Can we use it? → need to understand vortices
  - scanning tunneling microscope
    → vortex imaging in $Ba(Co_xFe_{1-x})_2As_2$
  - force microscope
    → vortex manipulation in $NdO_{1-x}F_xFeAs$

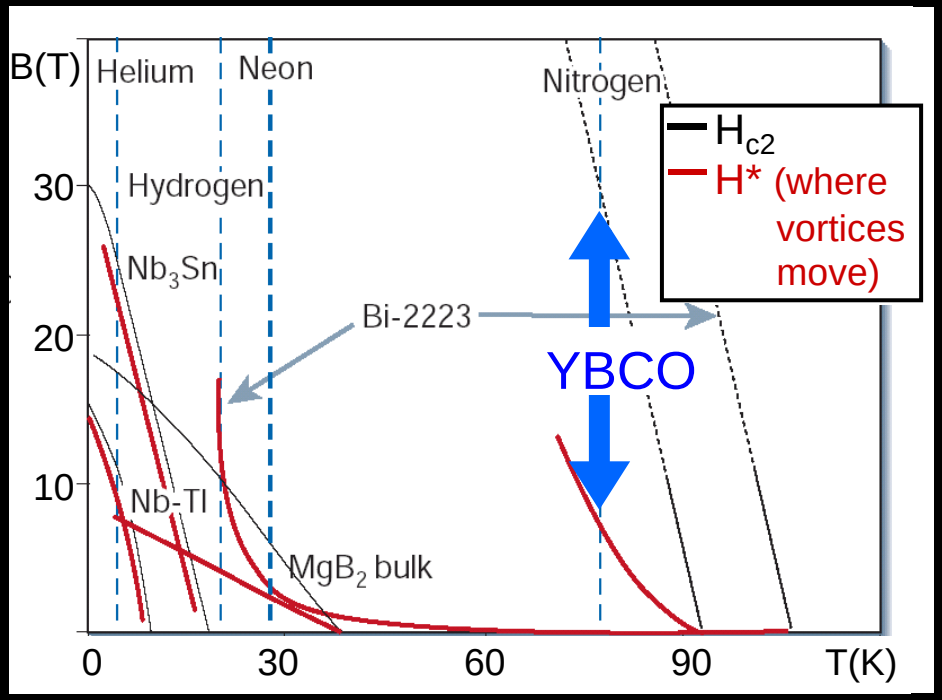
Metal-insulator transition in $VO_2$

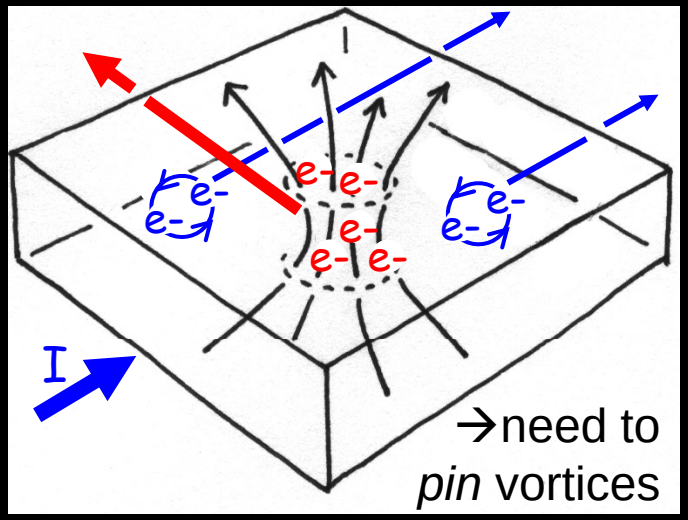
- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

Outlook

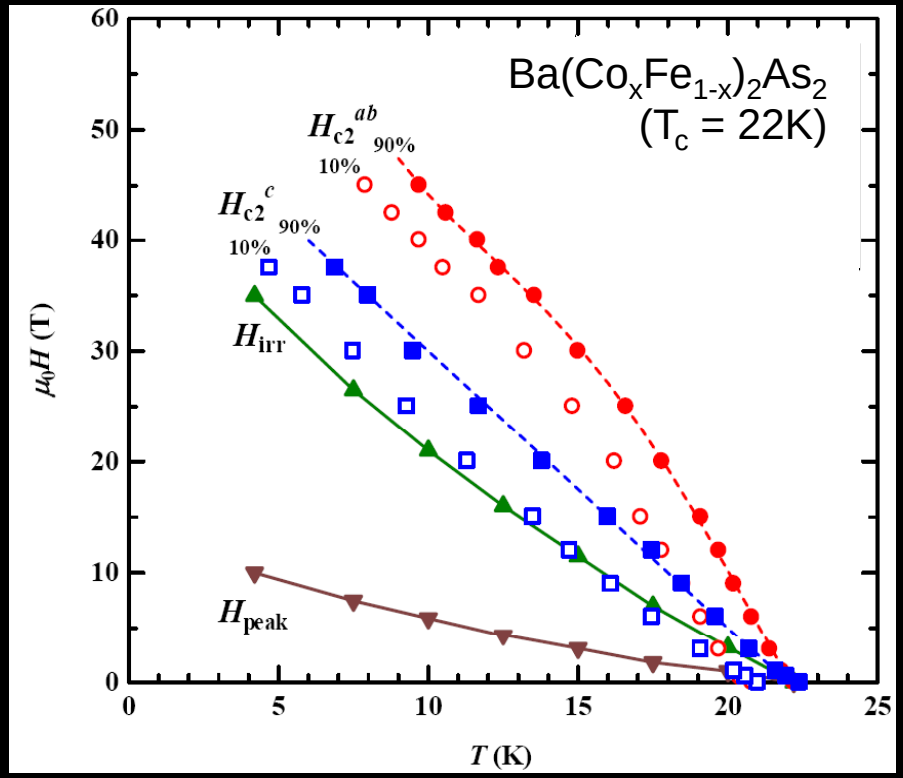
# Vortex Pinning Comparison

## Cuprate Superconductors




## Iron-Pnictide Superconductors




*Yamamoto,* APL 94, 062511 (2009)

Iron-Pnictides:

- strong pinning, speculation that it comes from nanoscale pinning sites, e.g. Co dopant inhomogeneities

→ need a local tool !

# Outline

High-Tc Superconductivity

- Can we understand it? → need to understand Mott
- Can we use it? → need to understand vortices
  - scanning tunneling microscope
    → vortex imaging in $Ba(Co_xFe_{1-x})_2As_2$
  - force microscope
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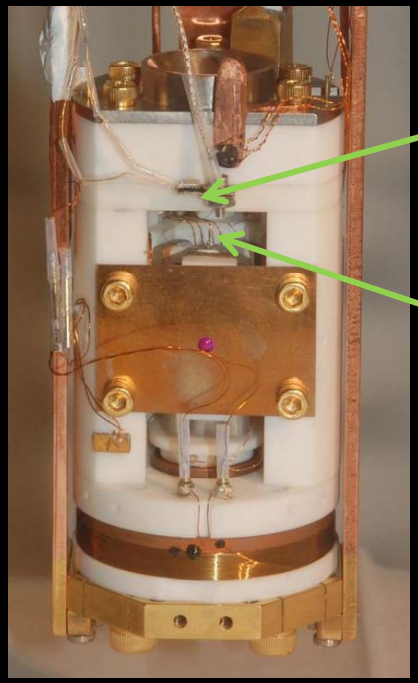
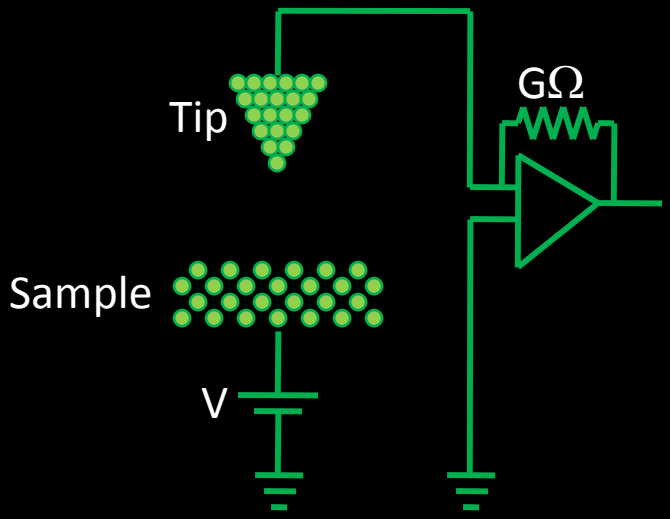
Metal-insulator transition in $VO_2$

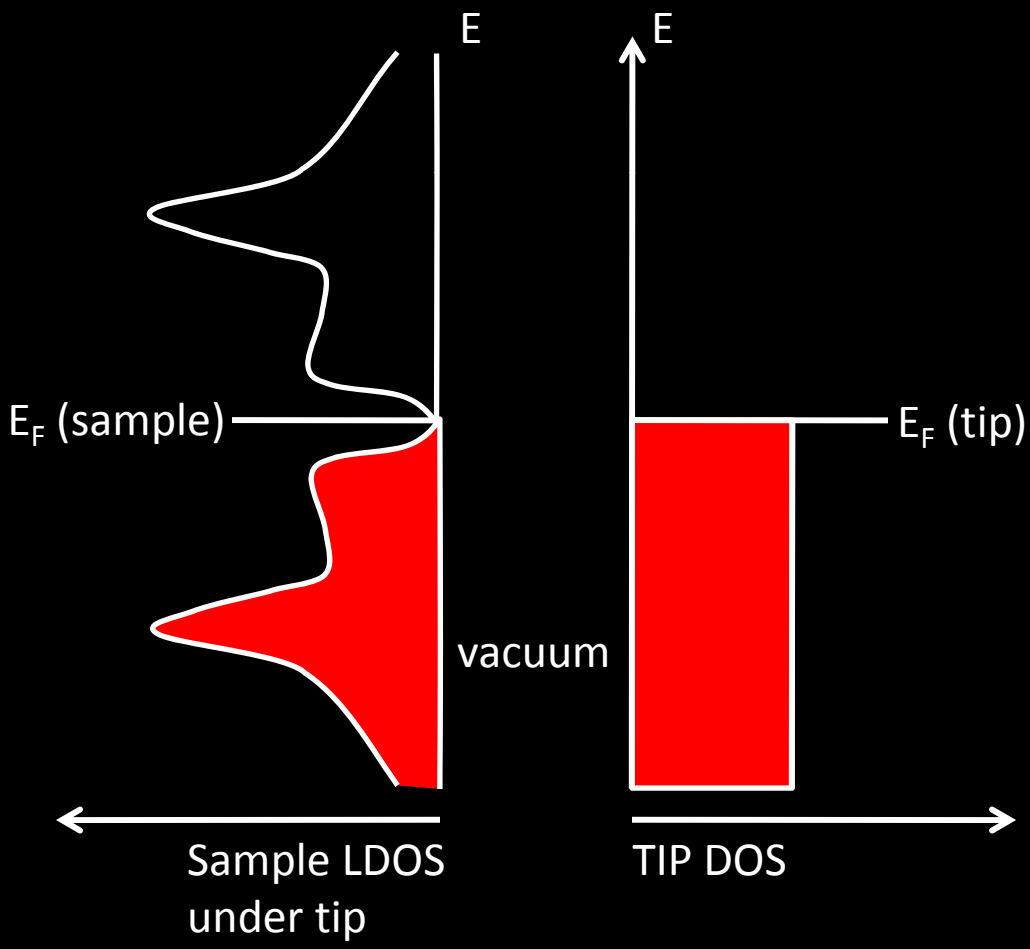
- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

Outlook

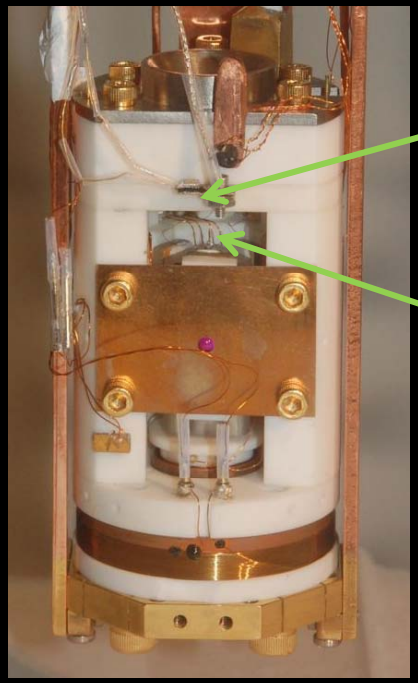
# Scanning Tunneling Microscopy



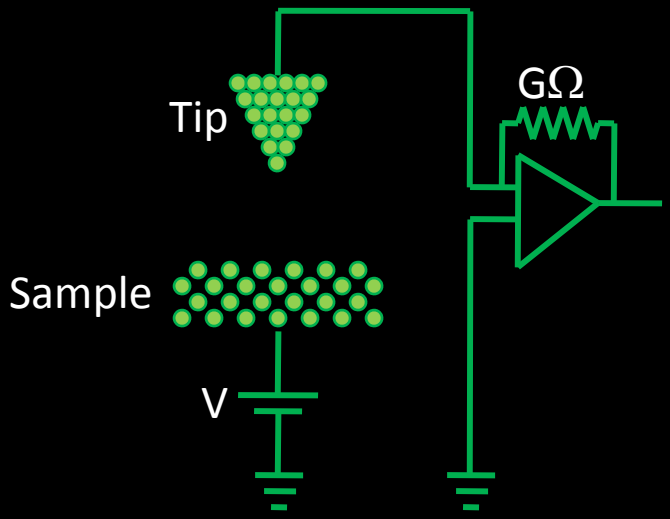



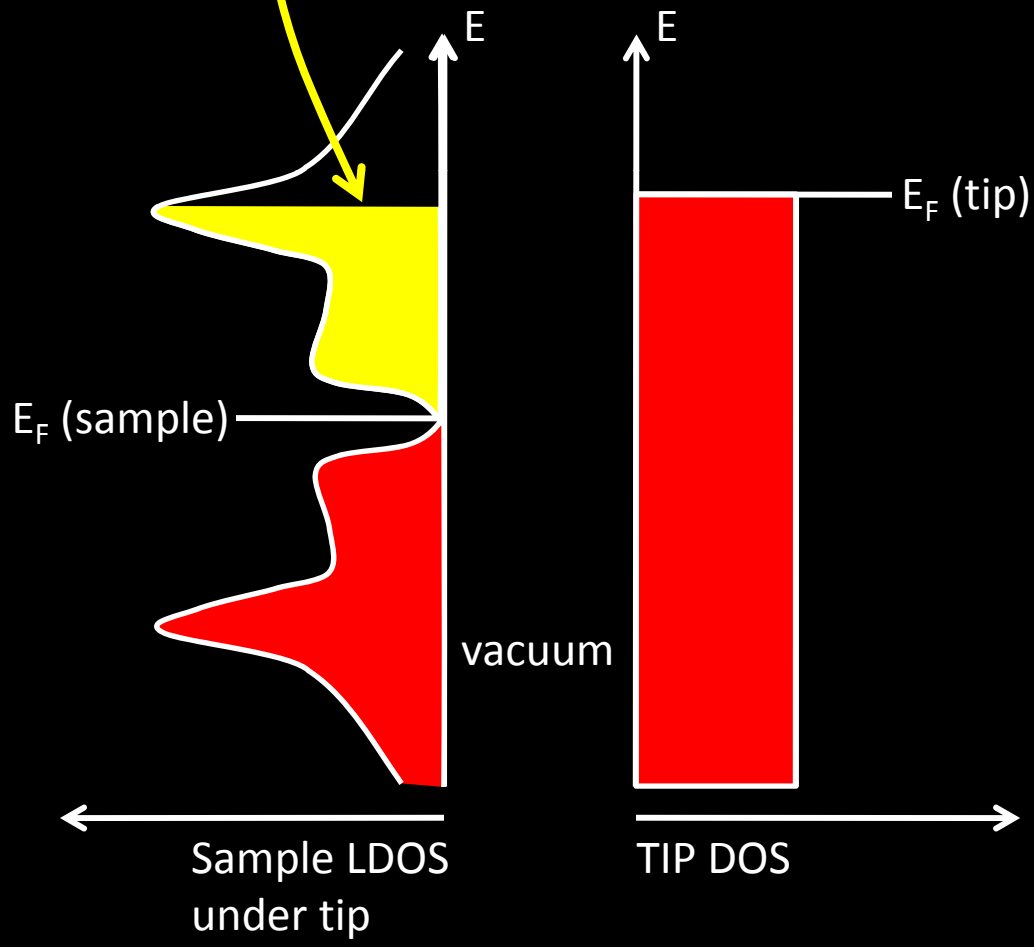



# Scanning Tunneling Microscopy

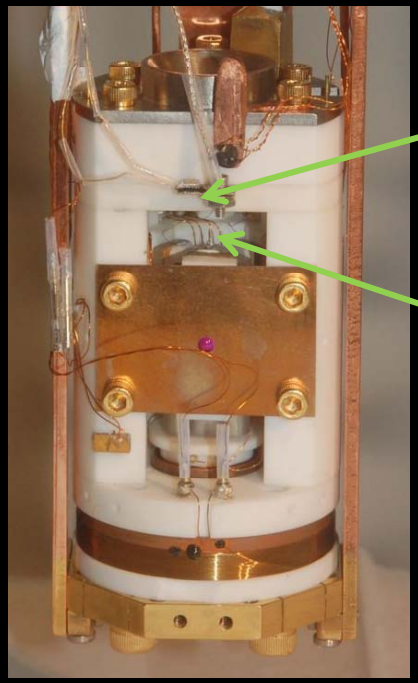



$$I(V) \propto \int_{E_F}^{eV} \mathrm{LDOS}(E)\,dE$$

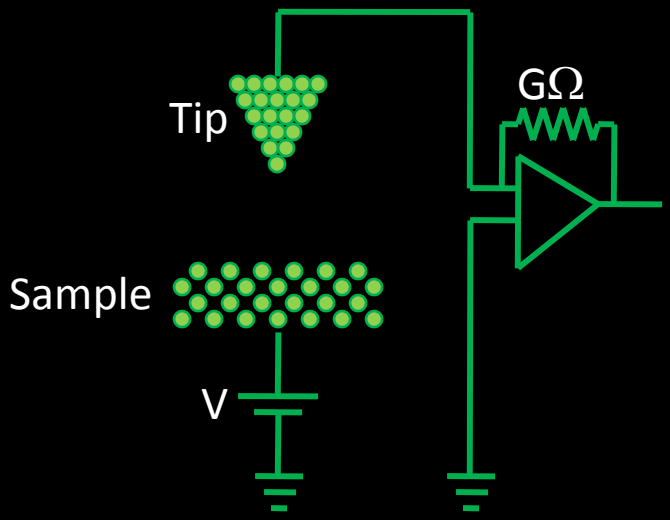


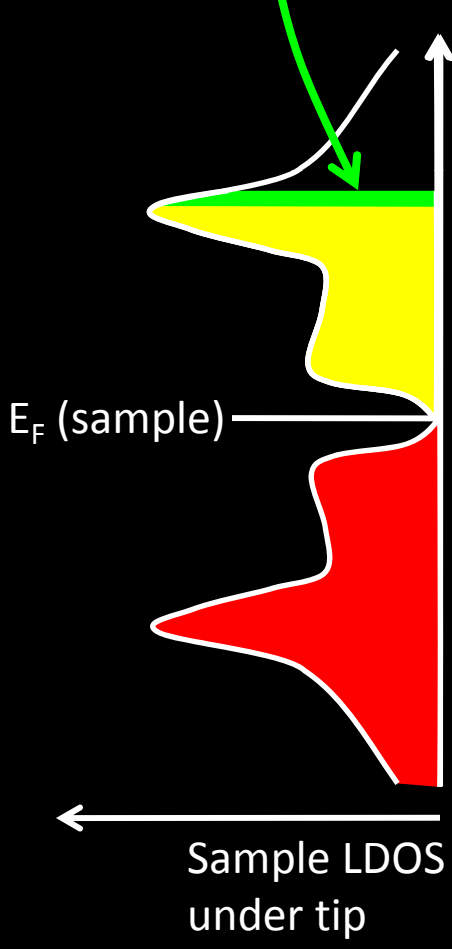



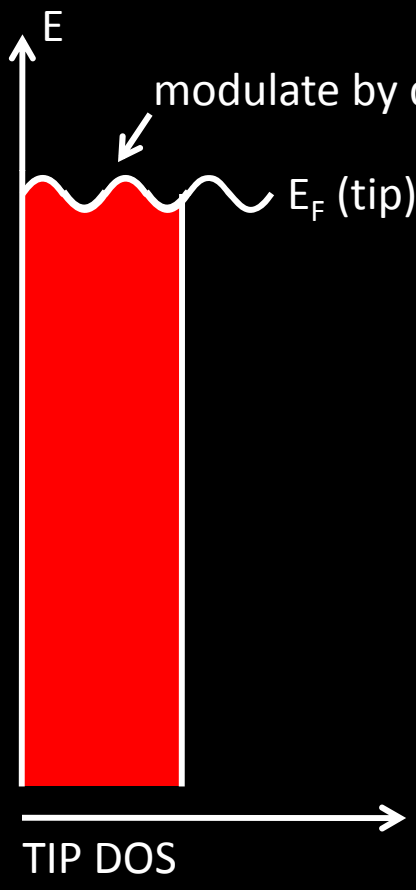
# Scanning Tunneling Microscopy



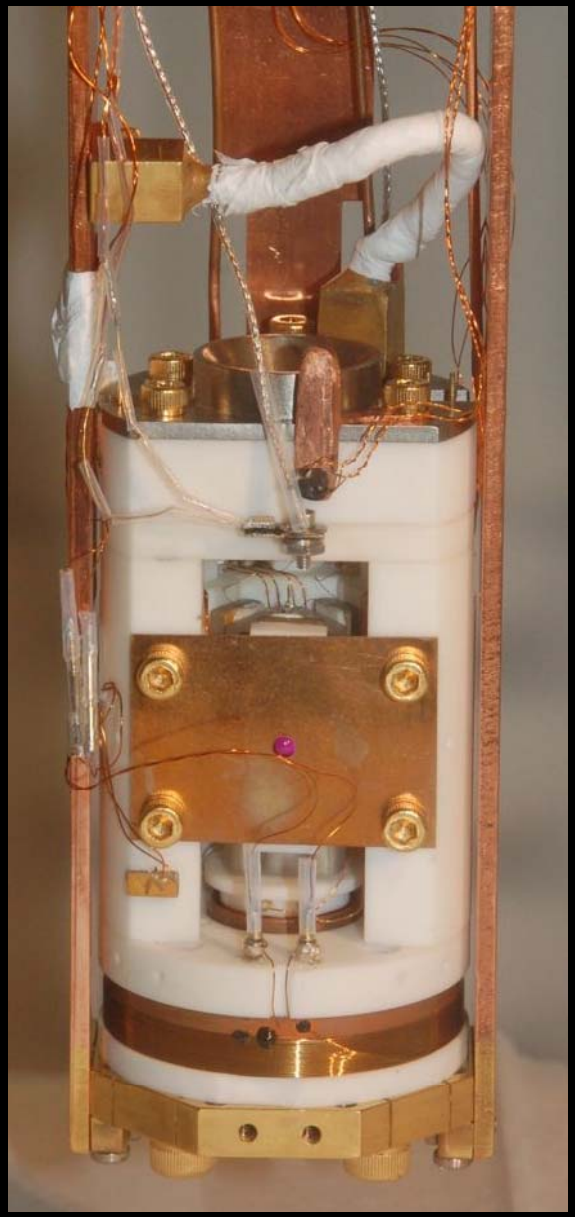

$$\frac{dI}{dV} \propto \mathrm{LDOS}(eV)\ e^{-z/z_0}$$



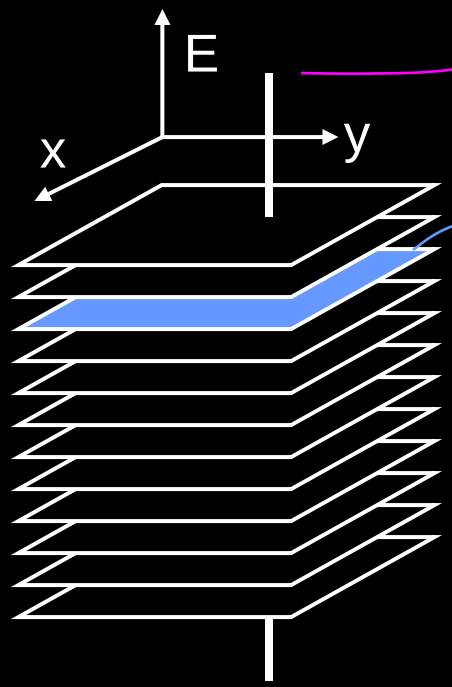



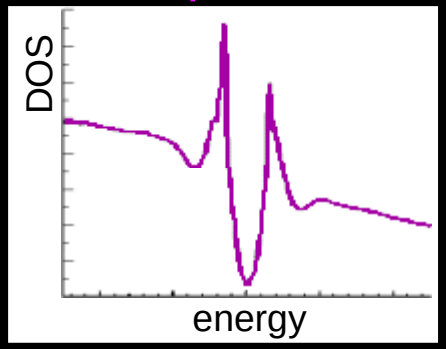
# Types of STM Measurements



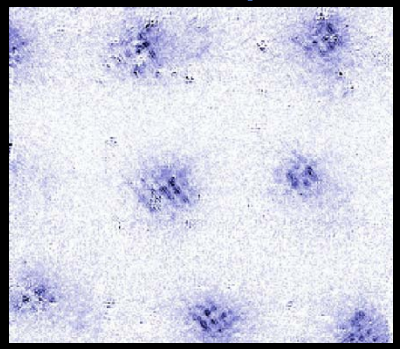
Local Density of States (x, y, E)



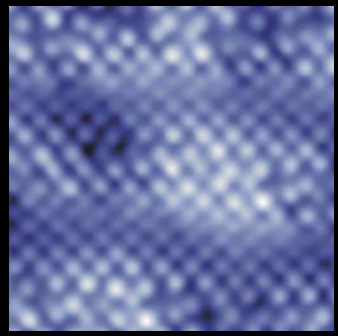

dI/dV Spectrum




dI/dV Map



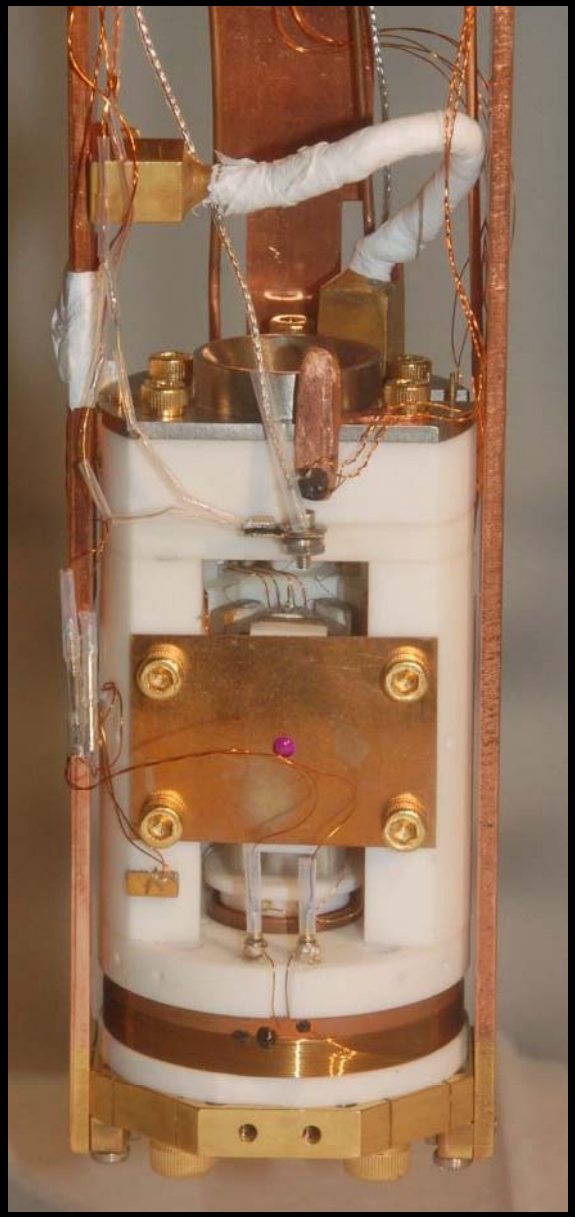
Topography




Constant current mode:

$$\int \frac{dI}{dV}$$

# Hoffman Lab Scanning Tunneling Microscope

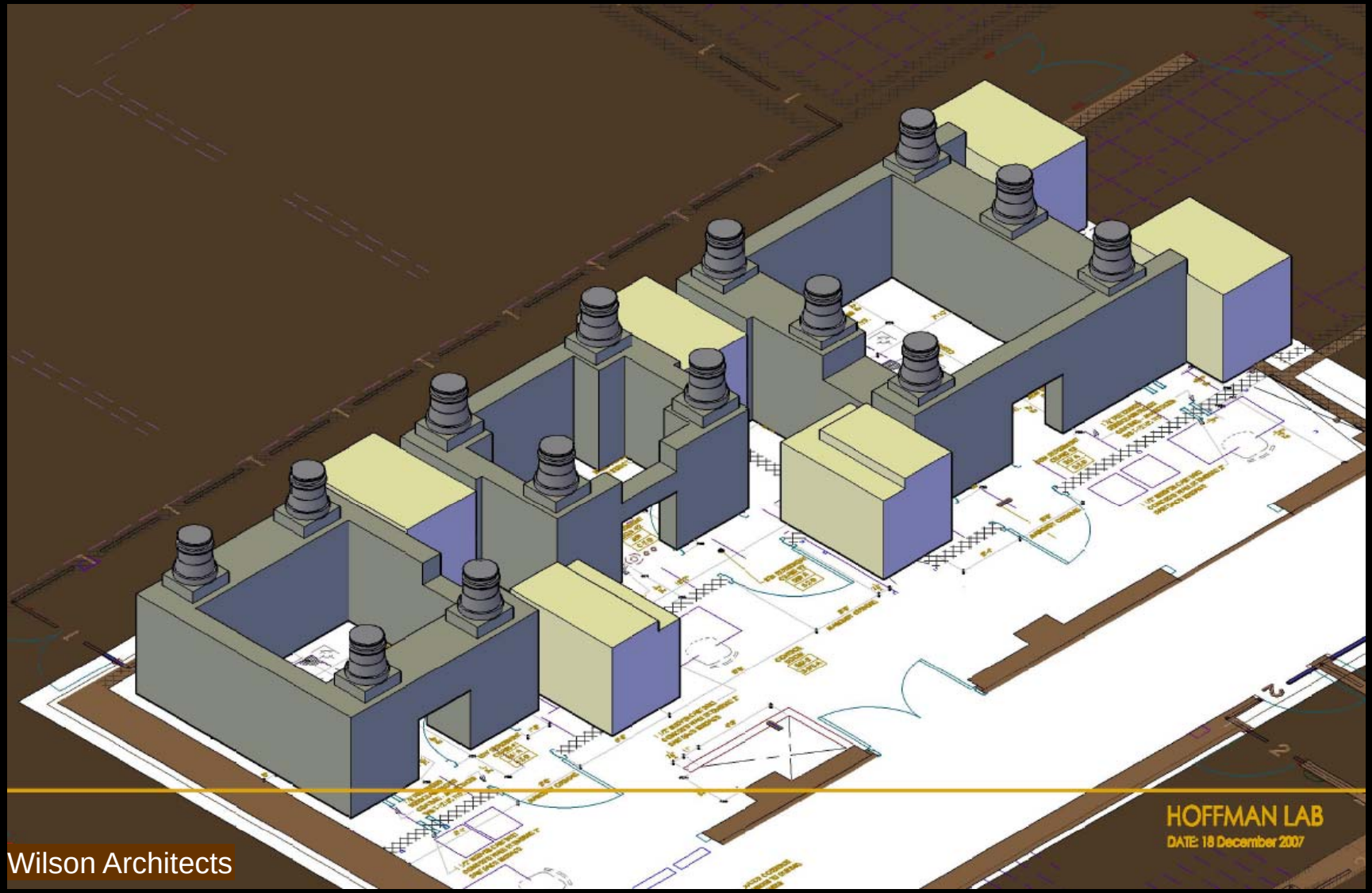


Optimized for the study of quasi-two-dimensional correlated electron materials:

- floating room, floating table for vibration isolation
- home-built continuous flow, low vibration $^3$He fridge
- in situ low temperature sample cleaver
- 3-axis magnetic field (9 T vertical, 3 T in plane)
- tracks the same atomically resolved locations over the full range of field

# Where are we?

# Laboratory Design

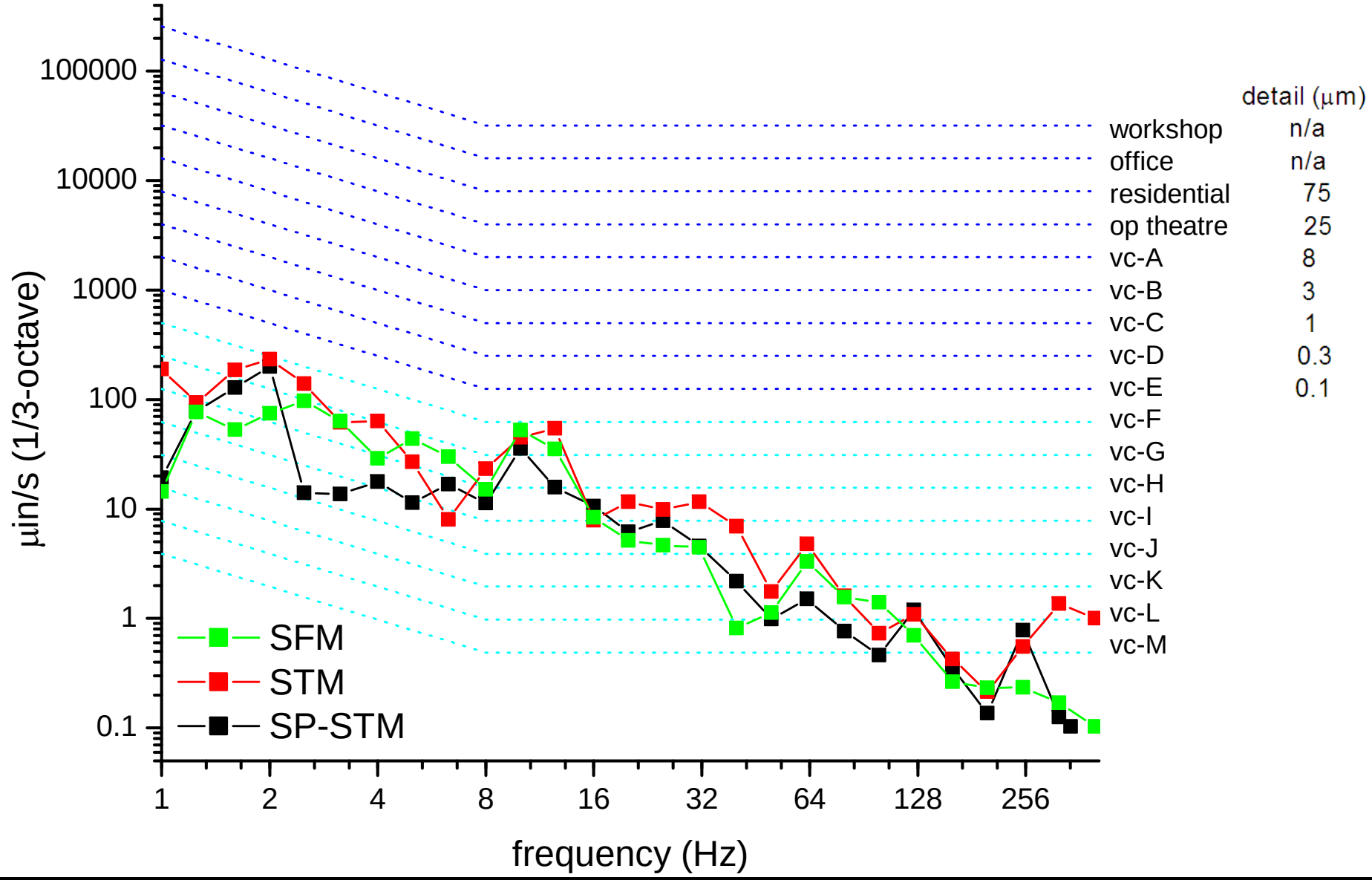



Wilson Architects

# Laboratory Design

Wilson Architects

# Vibration Results




This allows us to image samples with *atomic resolution* !!

# Outline

High-Tc Superconductivity

- Can we understand it? → need to understand Mott
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  - force microscope
    - → vortex manipulation in $NdO_{1-x}F_xFeAs$

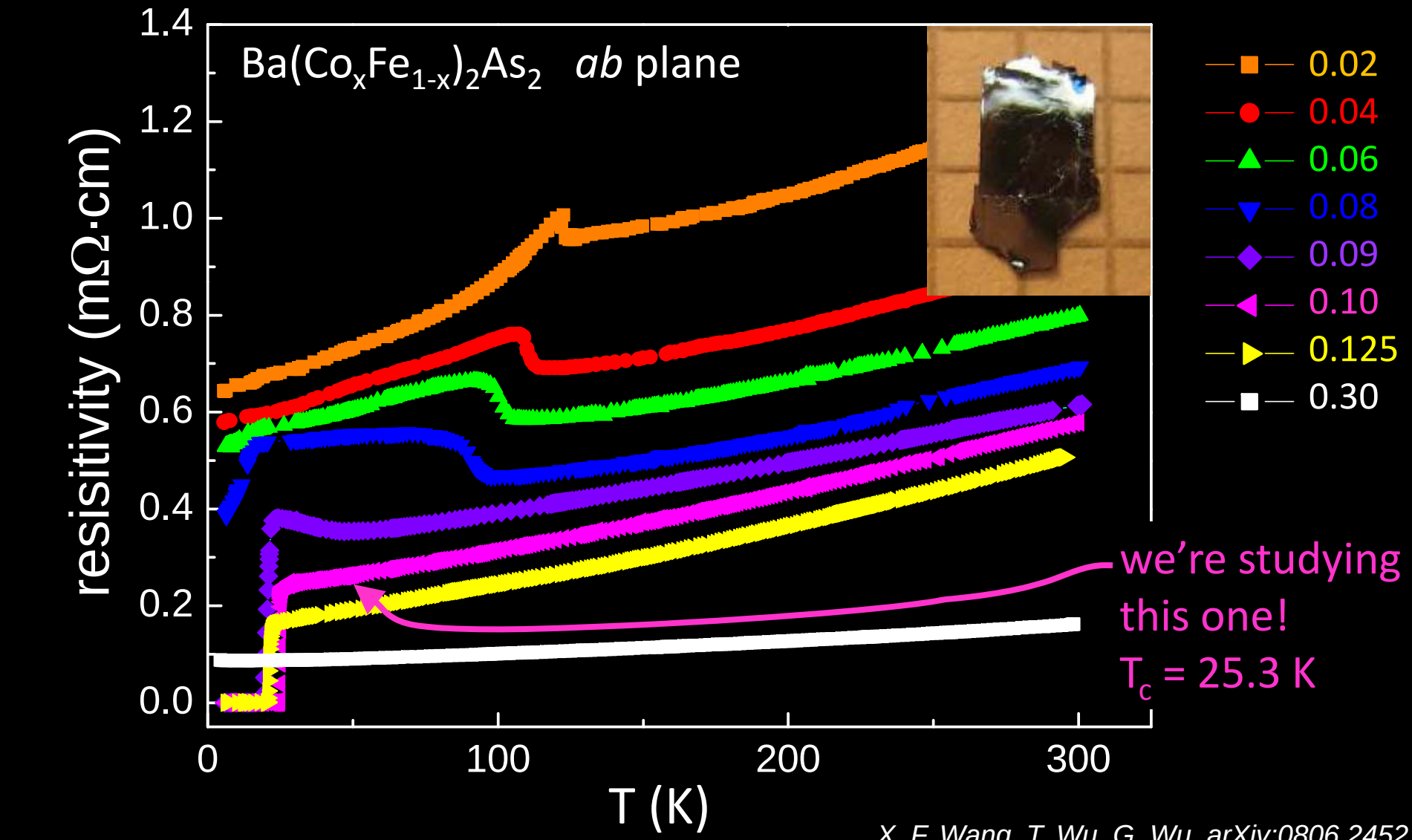
Metal-insulator transition in $VO_2$

- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

Outlook

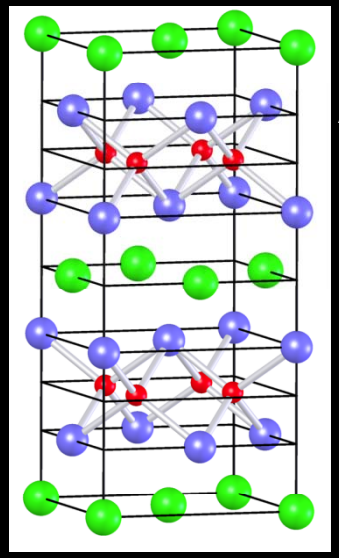
# $Ba(Co_xFe_{1-x})_2As_2$ Samples

single crystals grown by Prof. XianHui Chen

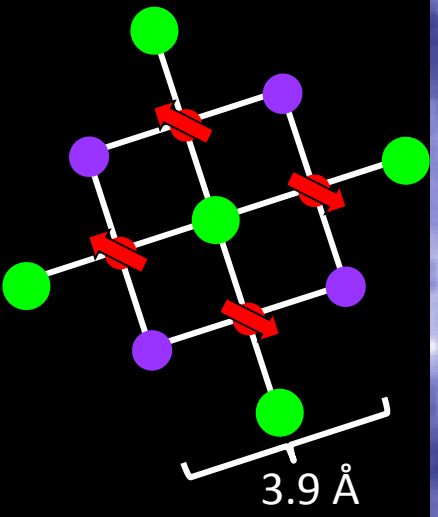


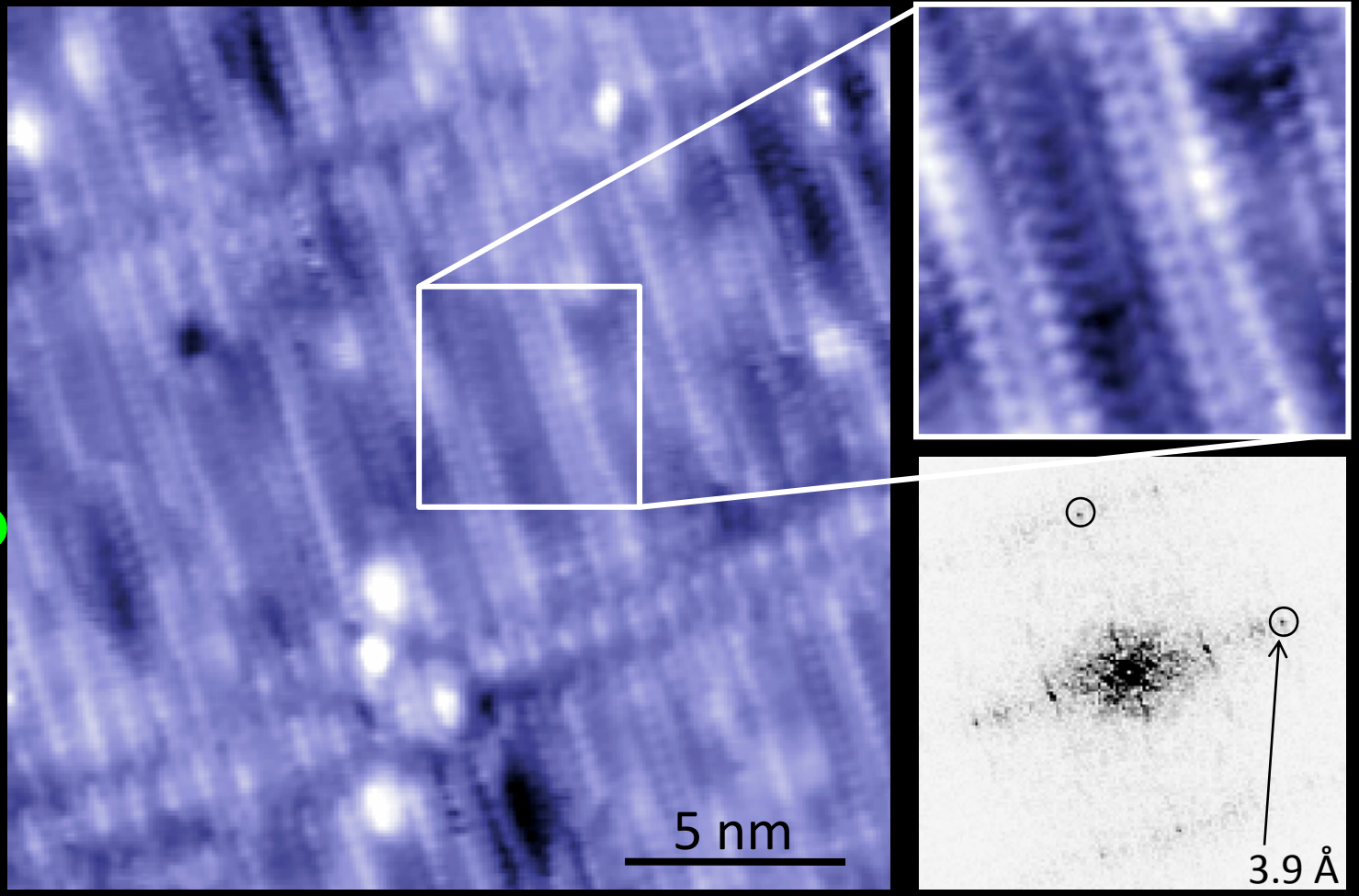

*X. F. Wang, T. Wu, G. Wu, arXiv:0806.2452 Phys. Rev. Lett.*

# Atomic Resolution Topography

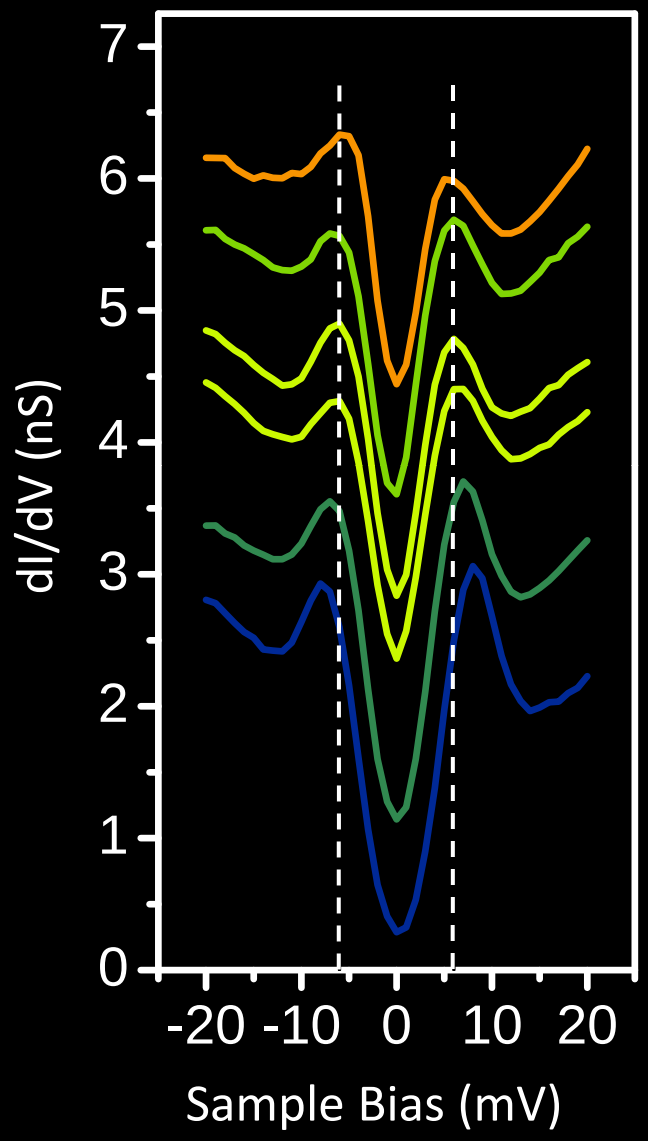


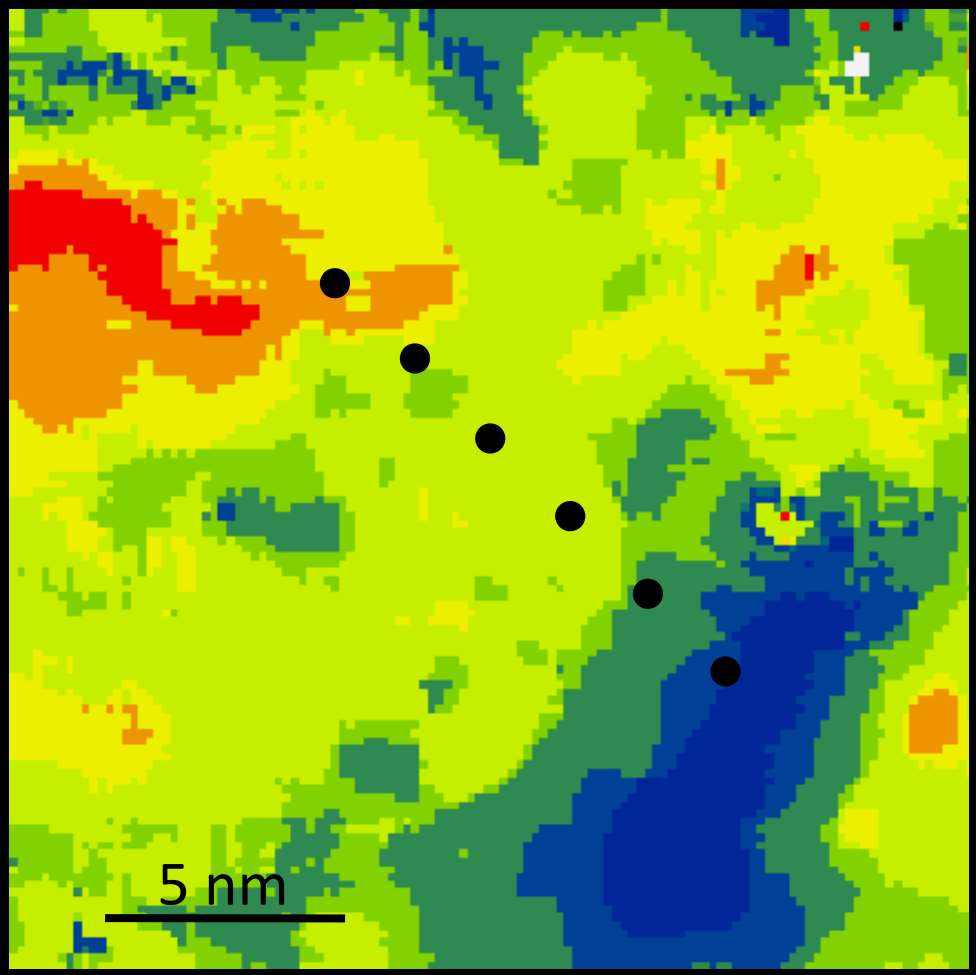

$Ba(Co_xFe_{1-x})_2As_2$
(x=0.1 nominal, $T_c$=25.3K)

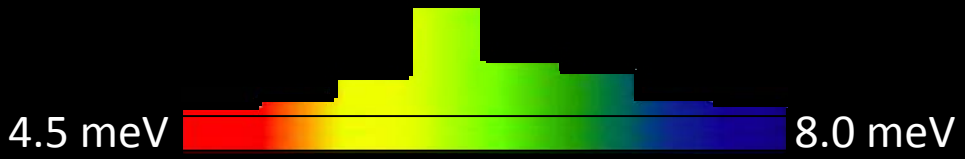


# Gap Mapping



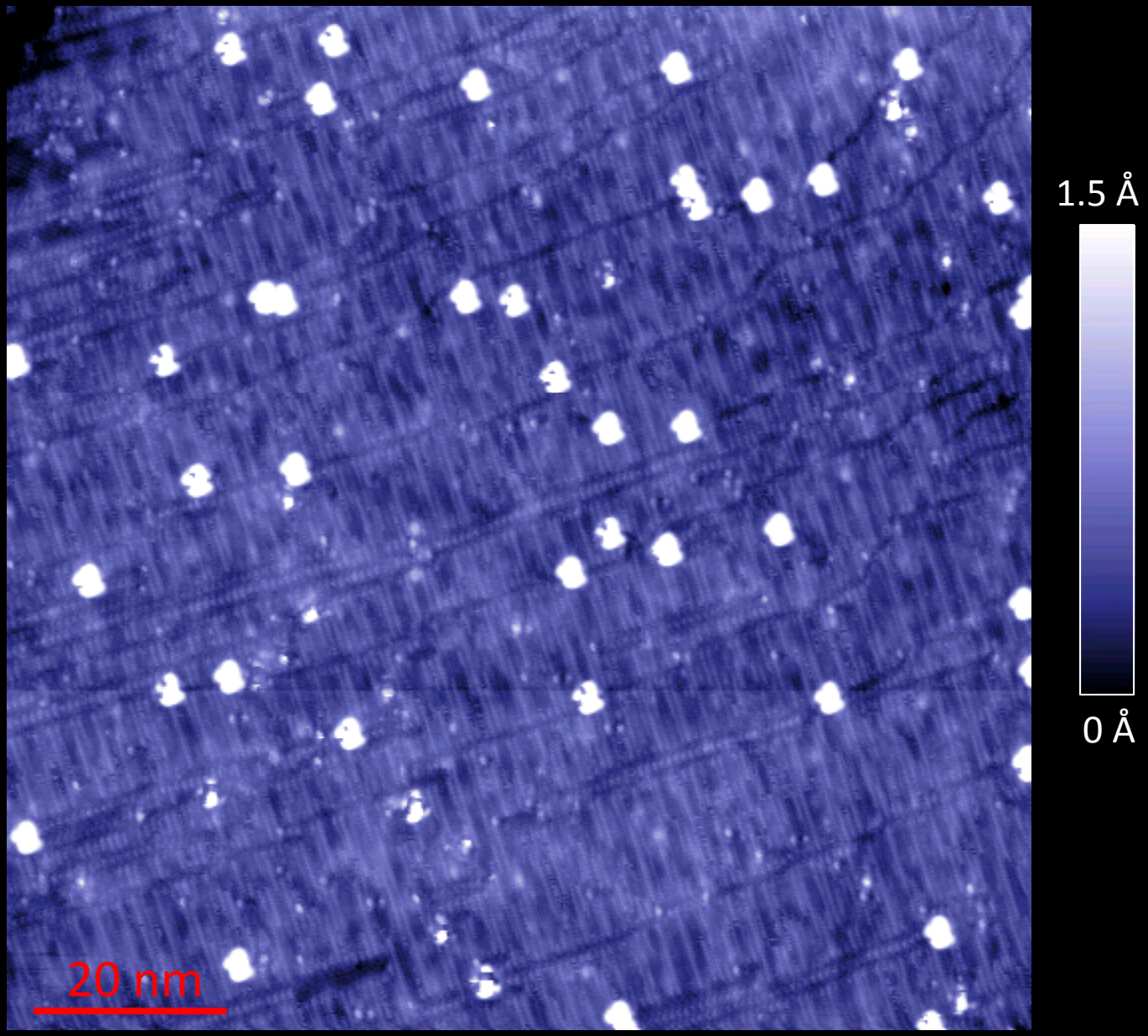




*measurements at T=6K; $k_BT$ = 0.5 meV*
*dV modulation = 1.5 meV*

$\overline{\Delta}$ = 6.25 ± 0.73 meV (12% variation)

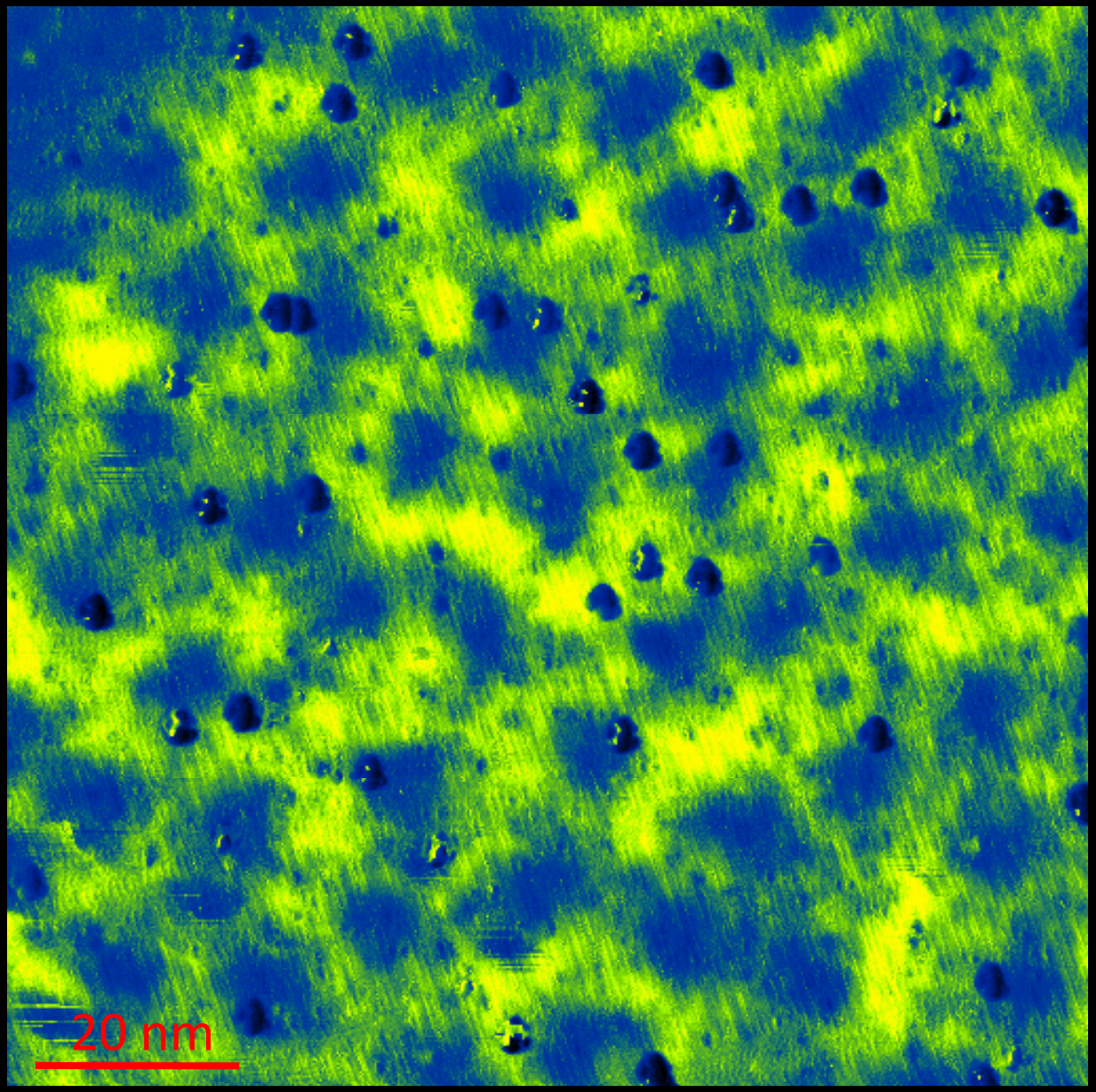
# Topography



# Vortices at 9T

dI/dV at 5 mV

(approximate coherence peak energy)



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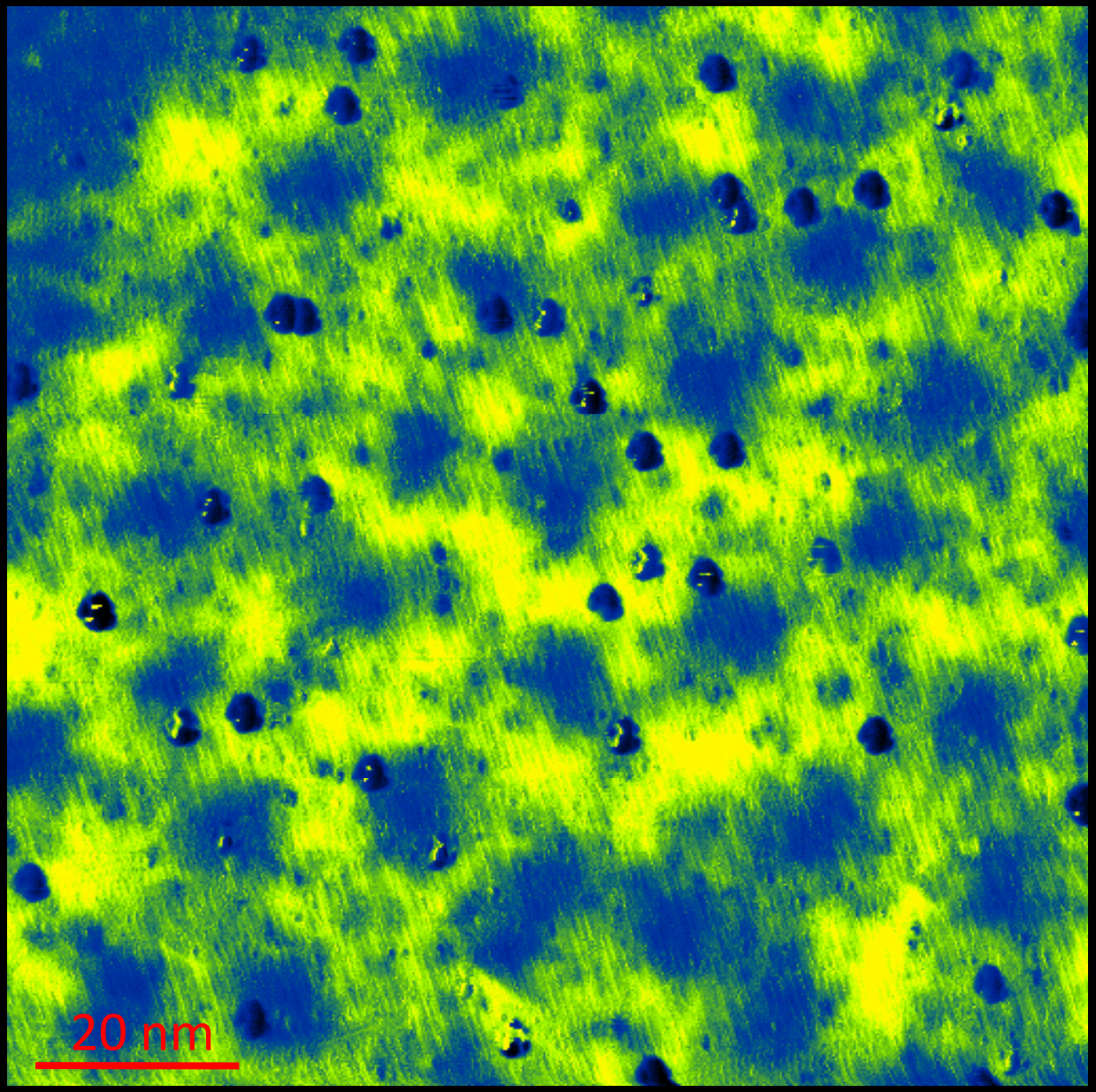
dI/dV at 5 mV

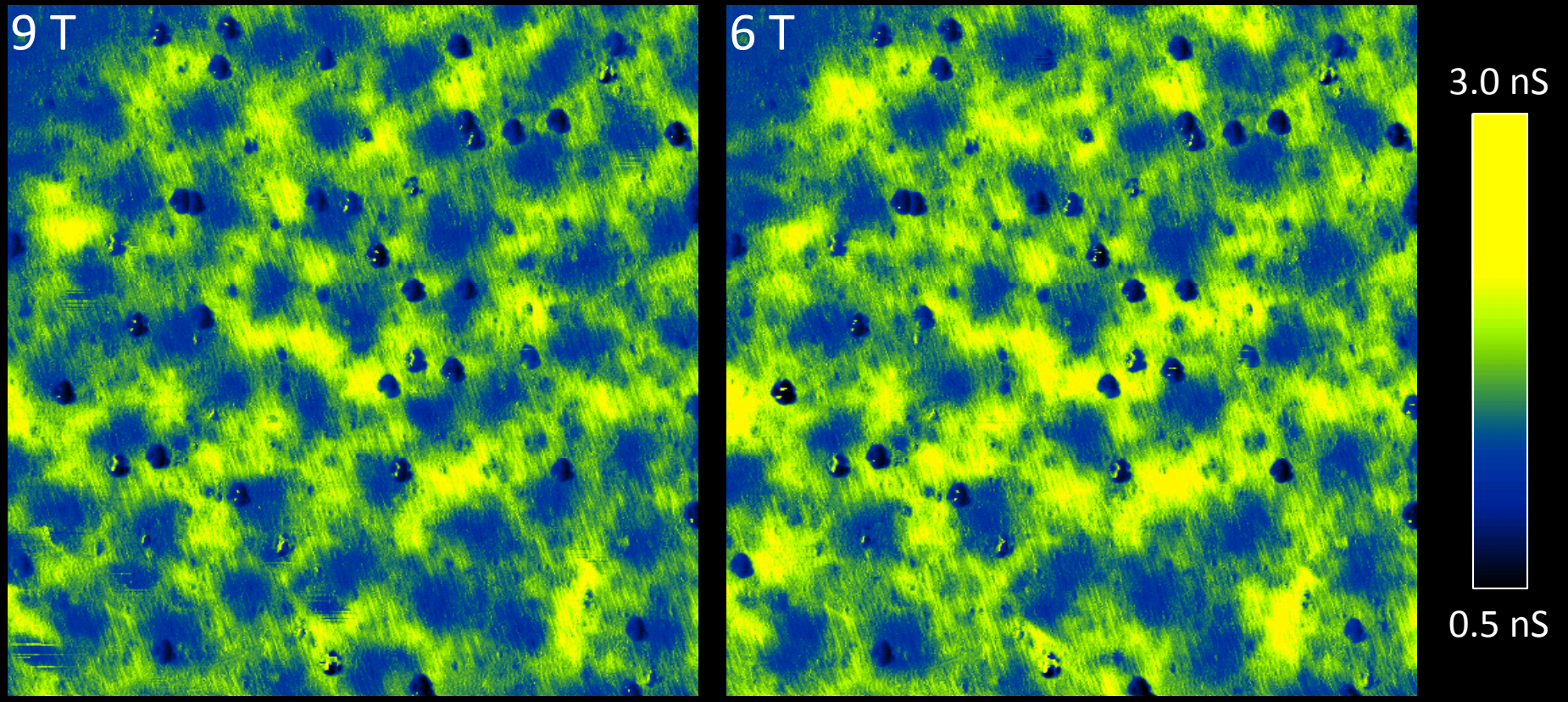
(approximate coherence peak energy)



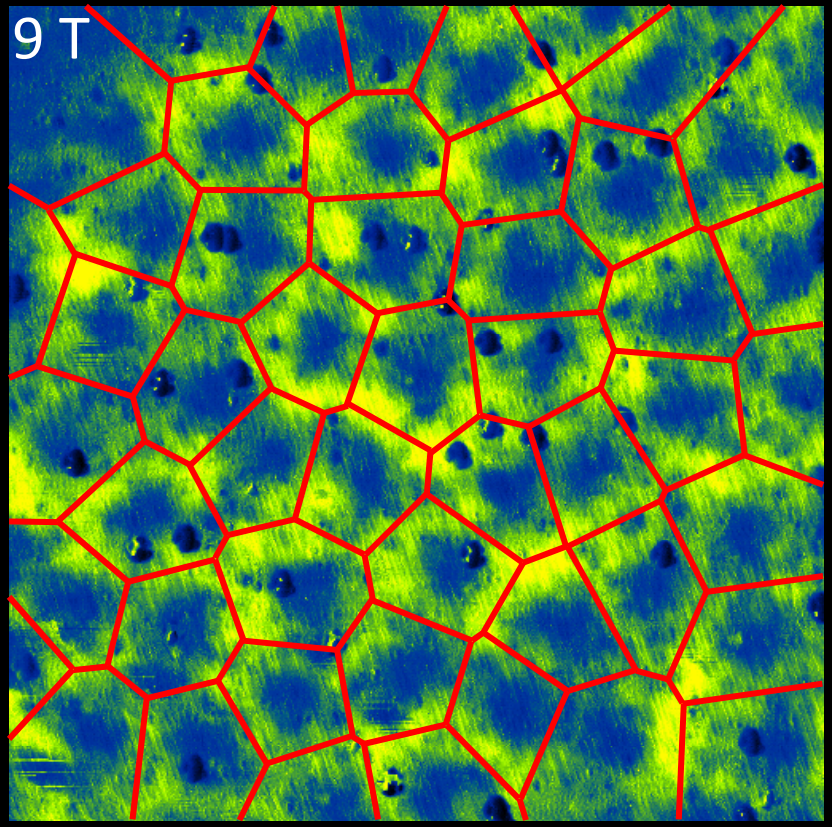

3.0 nS

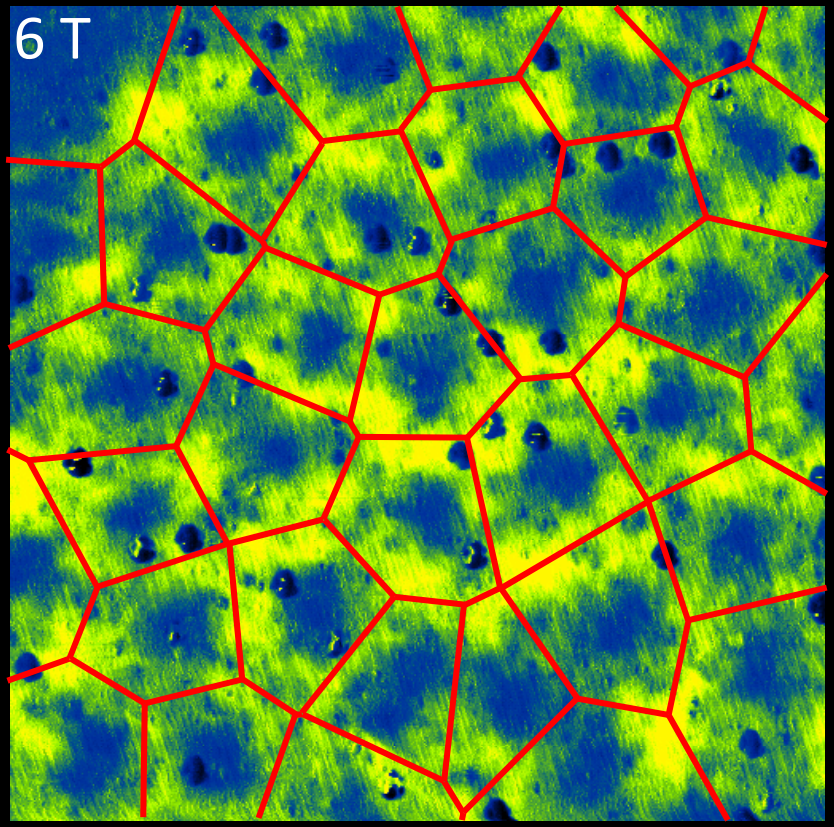
0.5 nS

# Flux Measurement



# Flux Measurement



average vortex area = 228 $nm^2$

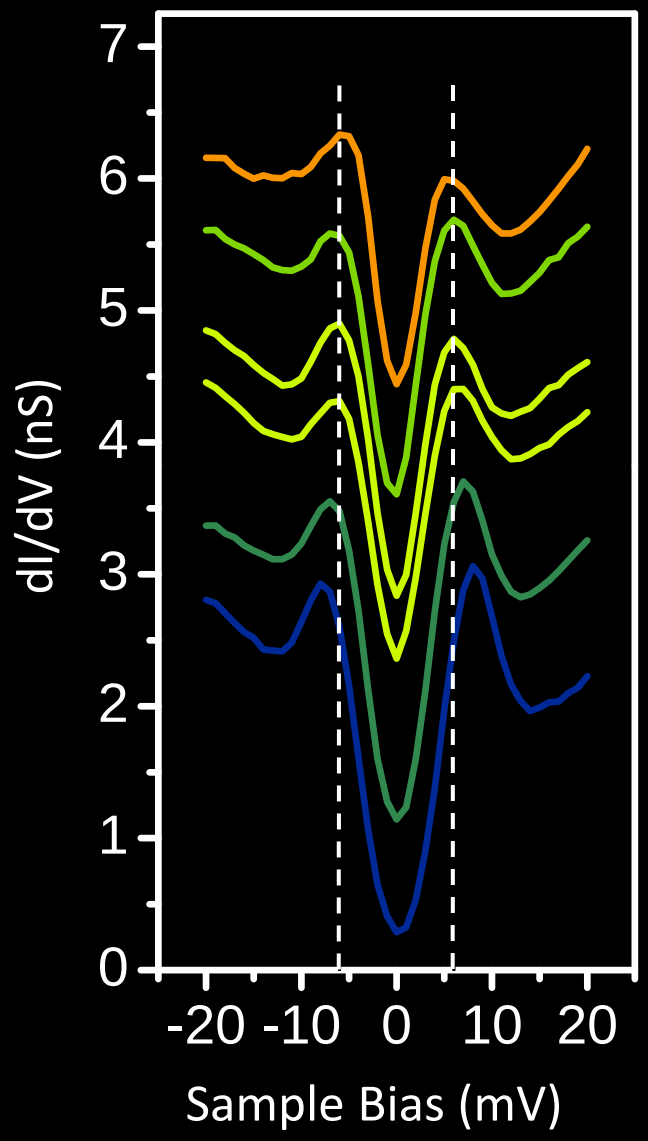
→ $\phi(9T) = 2.05 \times 10^{-15}$ T·$m^2$

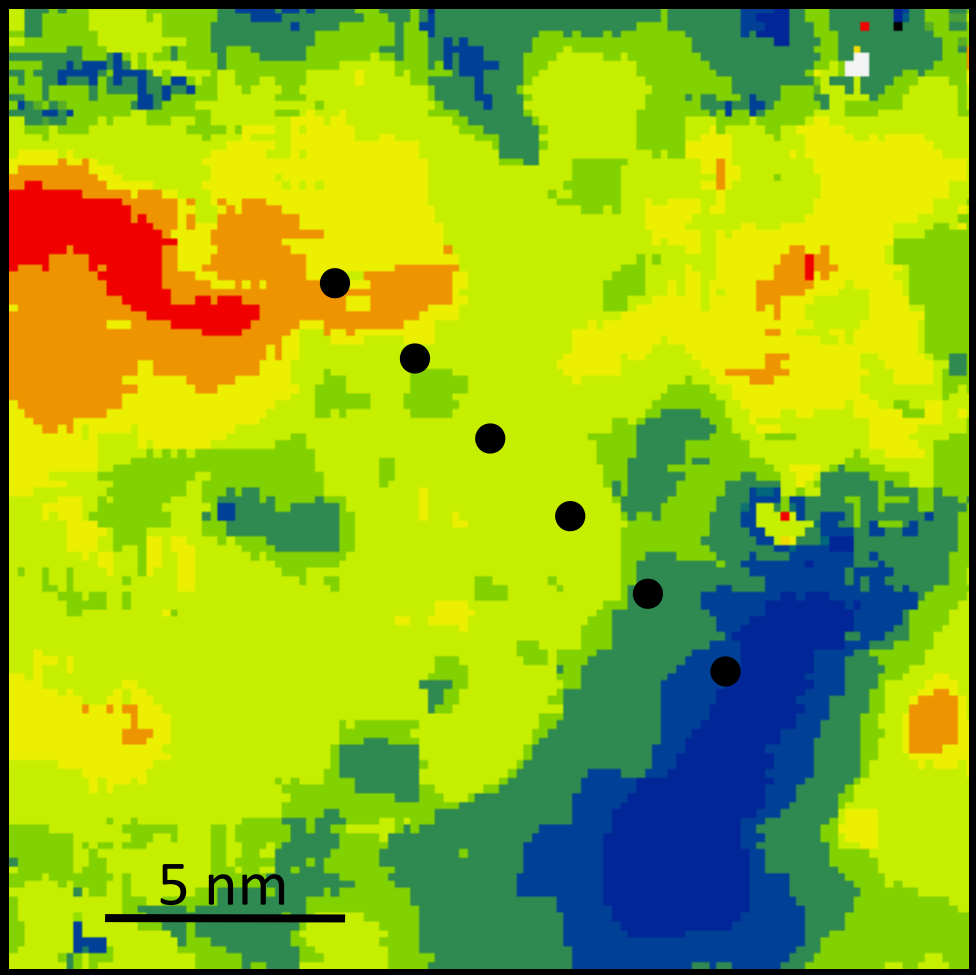
average vortex area = 362 $nm^2$

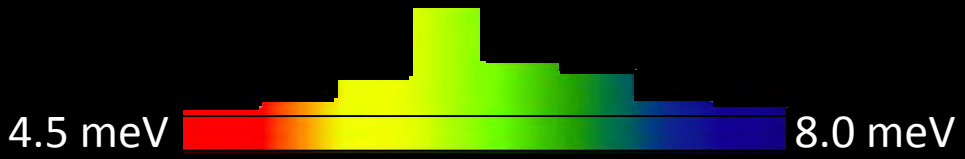
→ $\phi(6T) = 2.17 \times 10^{-15}$ T·$m^2$

Single magnetic flux quantum: $\Phi_0 = 2.07 \times 10^{-15}$ T·$m^2$

# Gap Mapping



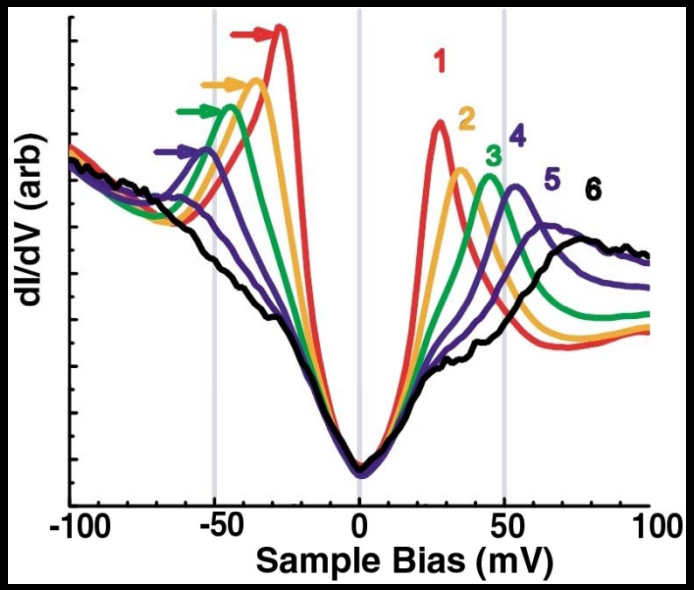

*measurements at T=6K; $k_BT$ = 0.5 meV*
*dV modulation = 1.5 meV*

$\overline{\Delta}$ = 6.25 ± 0.73 meV (12% variation)

# Correlation Comparison

## Cuprate Superconductors

$Bi_2Sr_2CaCu_2O_{8+d}$




*McElroy, PRL 94, 197005 (2005)*

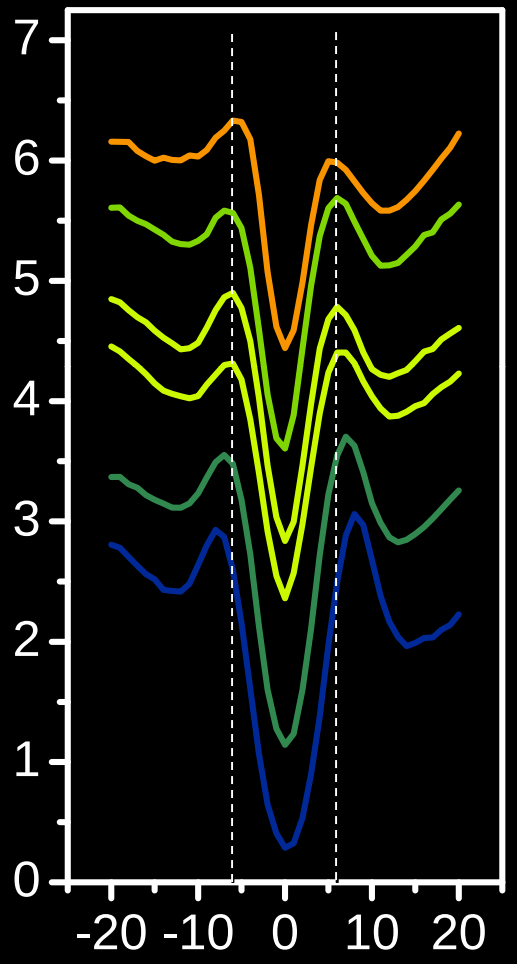
Measure of Correlation:

$\sigma = \delta\Delta / \overline{\Delta}$ = 21% variation

$2\Delta/k_BT_C$ = 6-10

## Iron-Pnictide Superconductors

$Ba(Co_xFe_{1-x})_2As_2$

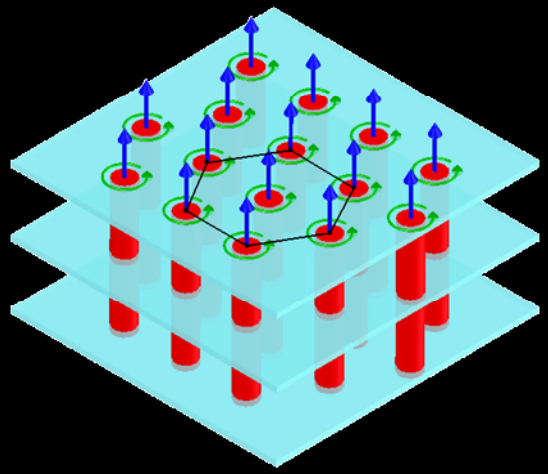



$\sigma = \delta\Delta / \overline{\Delta}$ = 12% variation

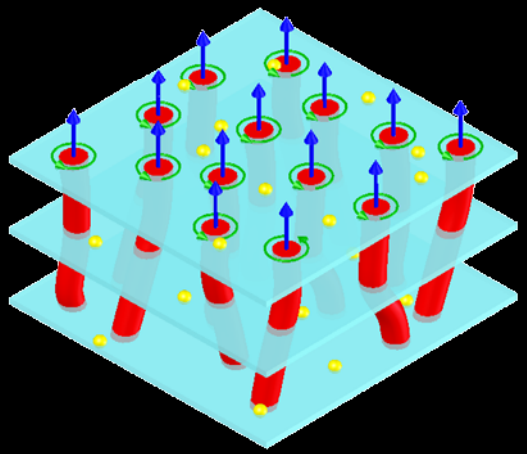
$2\Delta/k_BT_C$ = 5.3

# Vortex pinning possibilities

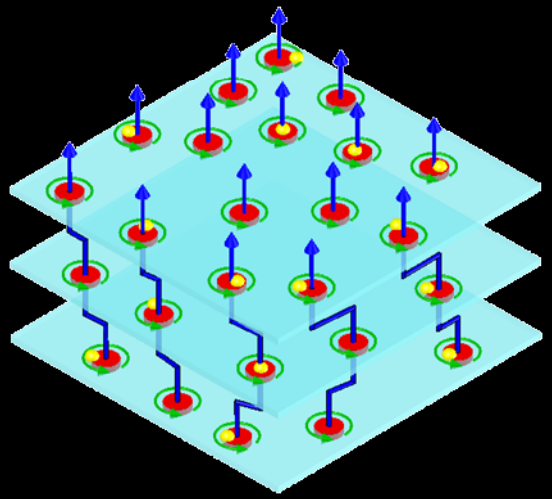
(1) no strong pinners
inter-vortex forces dominate
→ lattice formation



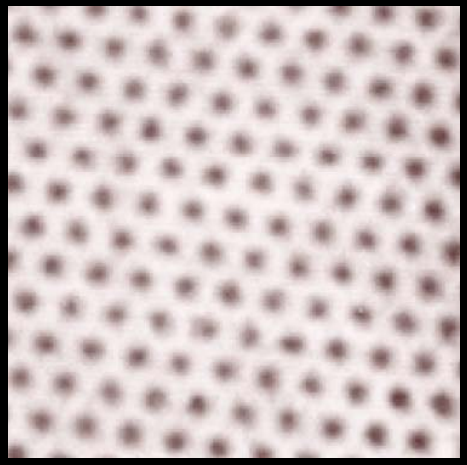
(2) strong pinners exist
low anisotropy
→ vortices bend slightly
to accommodate pinners



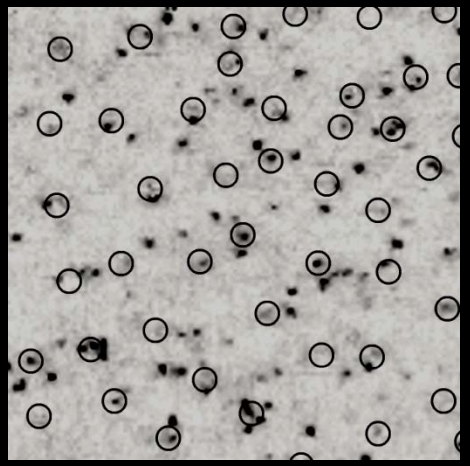
(3) strong pinners exist
high anisotropy
→ vortices pancake
each pancake pins independently



ideal case
for applications

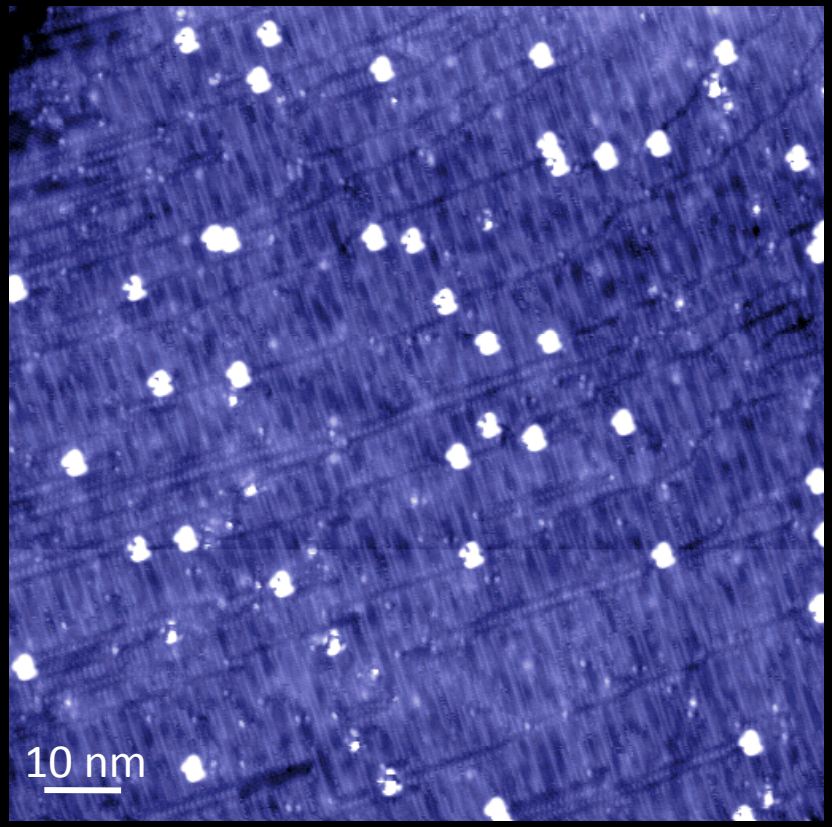


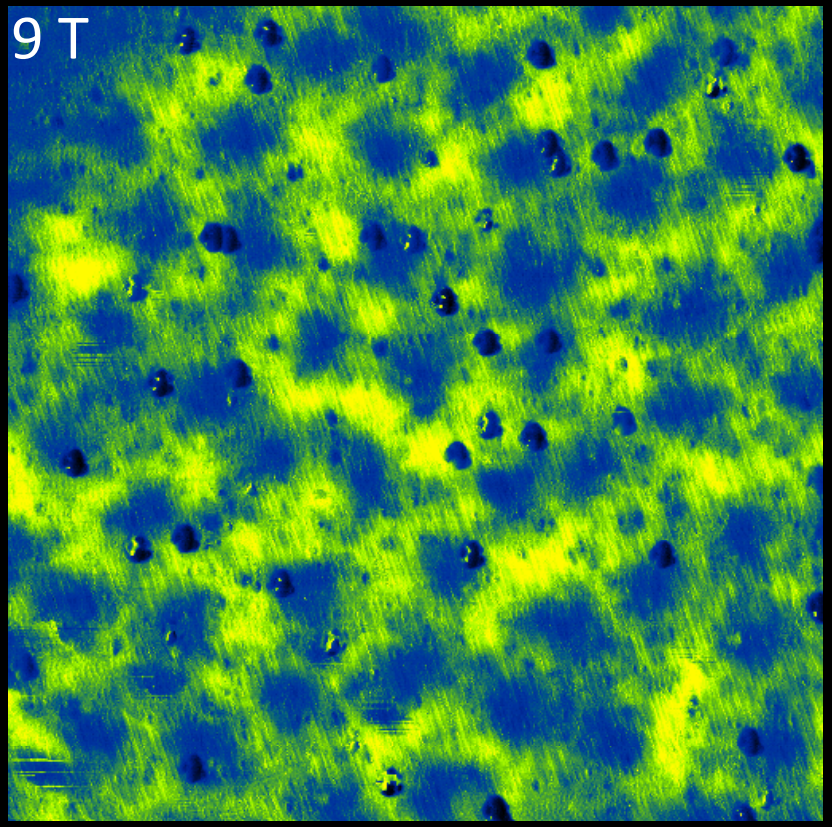
$NbSe_2$



$Bi_2Sr_2CaCu_2O_8$

# Are Vortices Pinned to Surface Impurities?

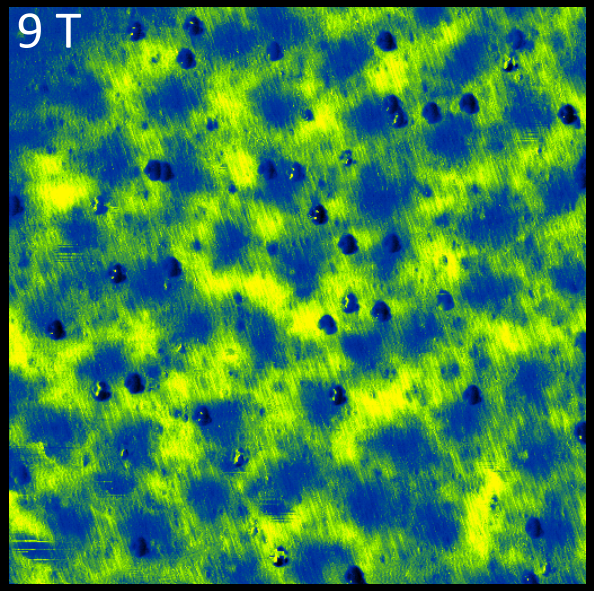


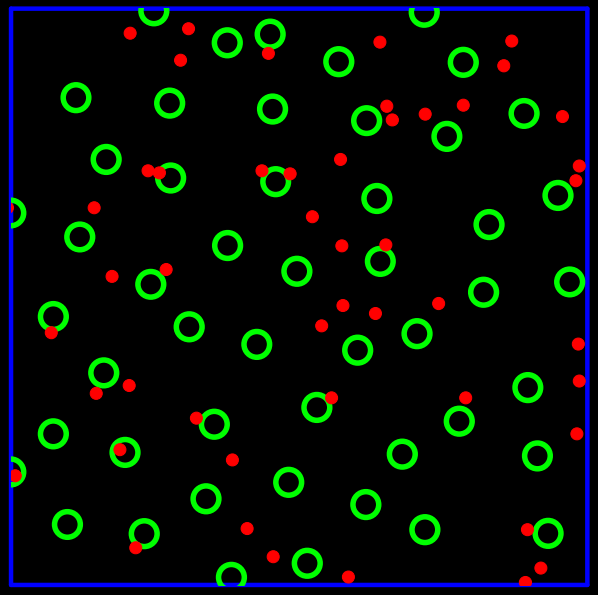



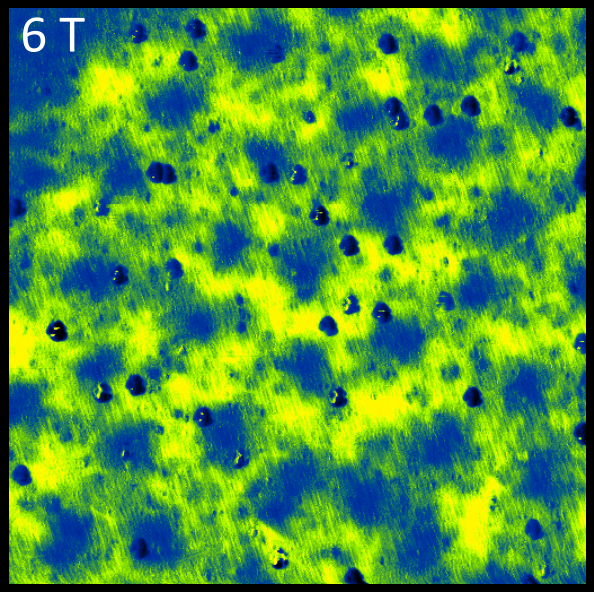
# Are Vortices Pinned to Surface Impurities?

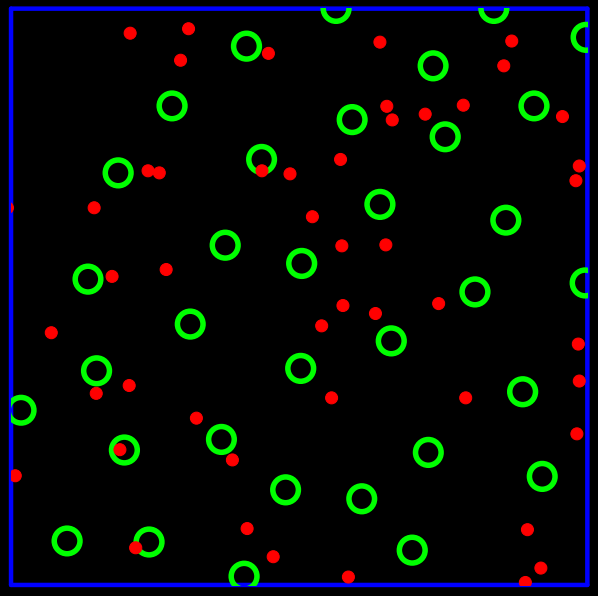
Raw Data

Idealized Data



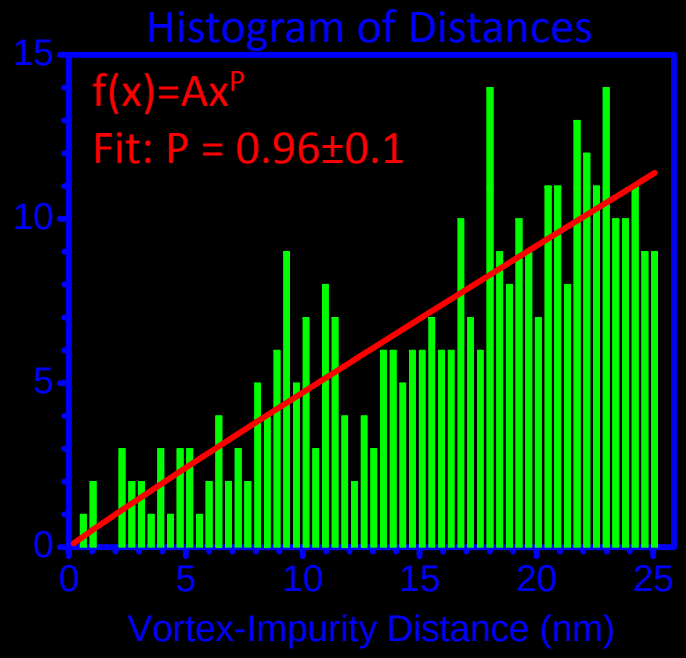





vortex, radius $\xi_0$ = 2.76 nm

impurity

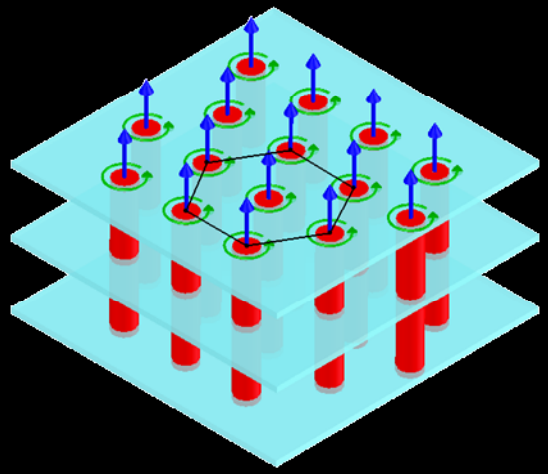


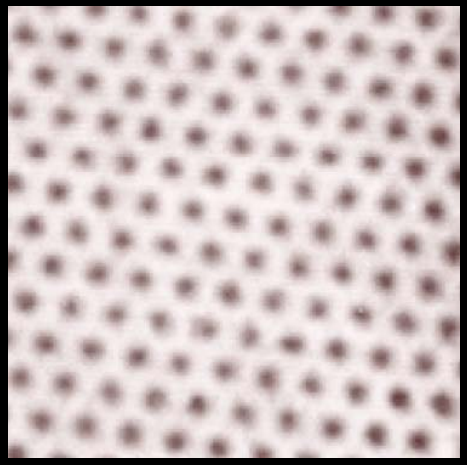

Linear fit!

→Vortices are <u>not</u> pinned to visible surface impurities

# Vortex pinning possibilities

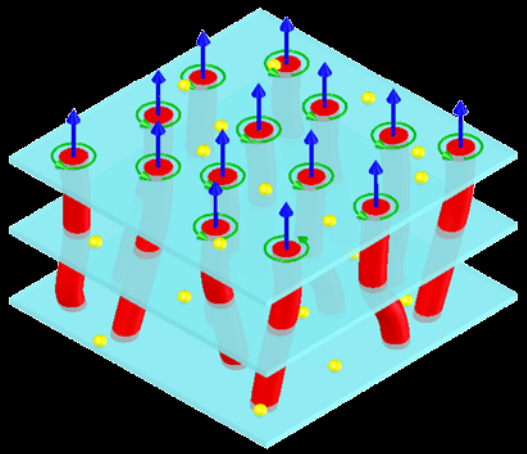
(1) no strong pinners
inter-vortex forces dominate
→ lattice formation

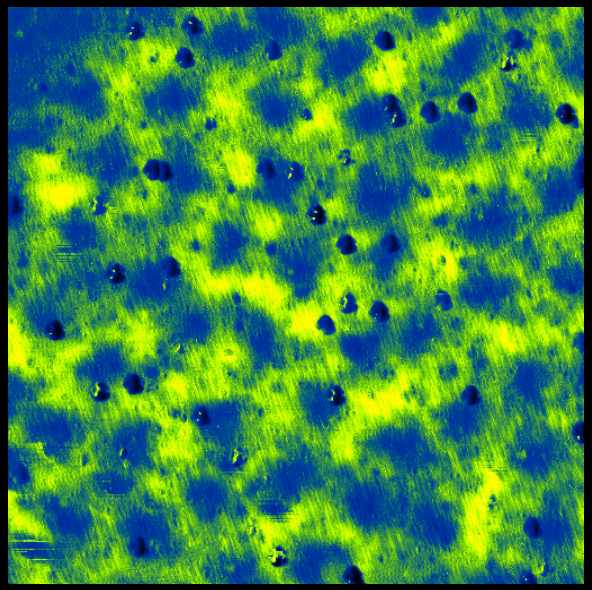




$NbSe_2$

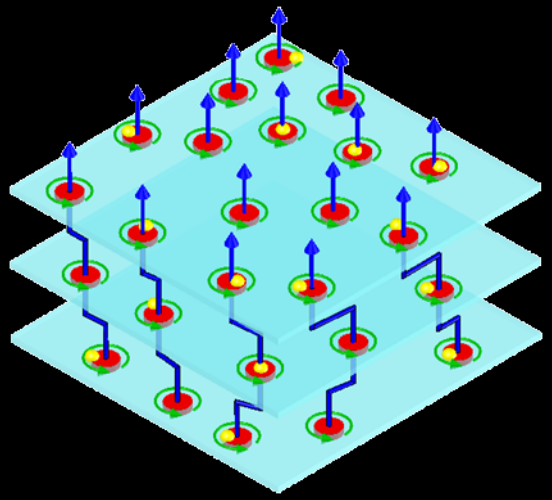
(2) strong pinners exist
low anisotropy
→ vortices bend slightly
to accommodate pinners

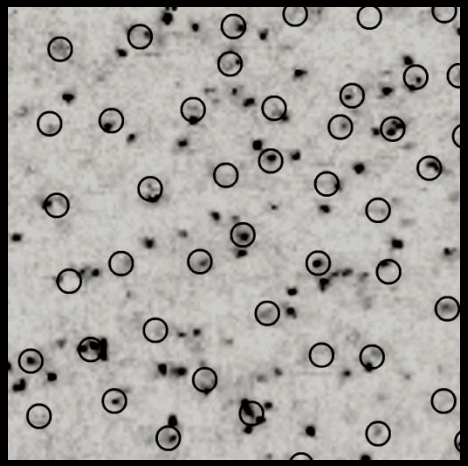




$Ba(Co_xFe_{1-x})_2As_2$

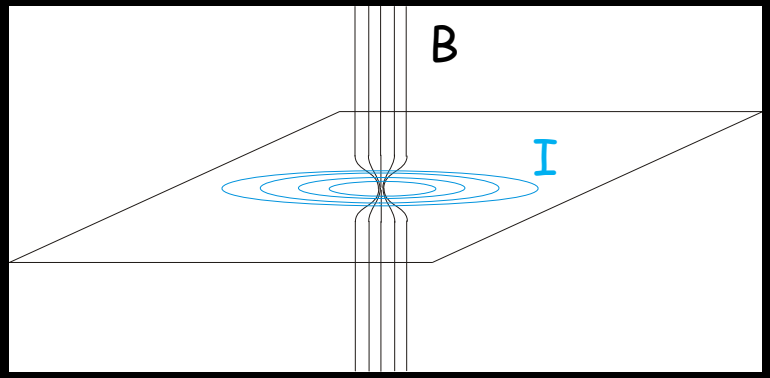
(3) strong pinners exist
high anisotropy
→ vortices pancake
each pancake pins independently





$Bi_2Sr_2CaCu_2O_8$

# Length Scales in Superconducting Vortices




$\Phi = 2.07 \times 10^{-15}\ T \cdot m^2$

B(r) = field penetration

$n_s(r)$ = density of Cooper pairs

vortex center

$\xi$

$\lambda$

r = distance from vortex center

"coherence length"
= distance over which Cooper pairs are broken (superconductivity is destroyed)

"penetration depth"
= distance over which magnetic field decays

# Outline

High-Tc Superconductivity

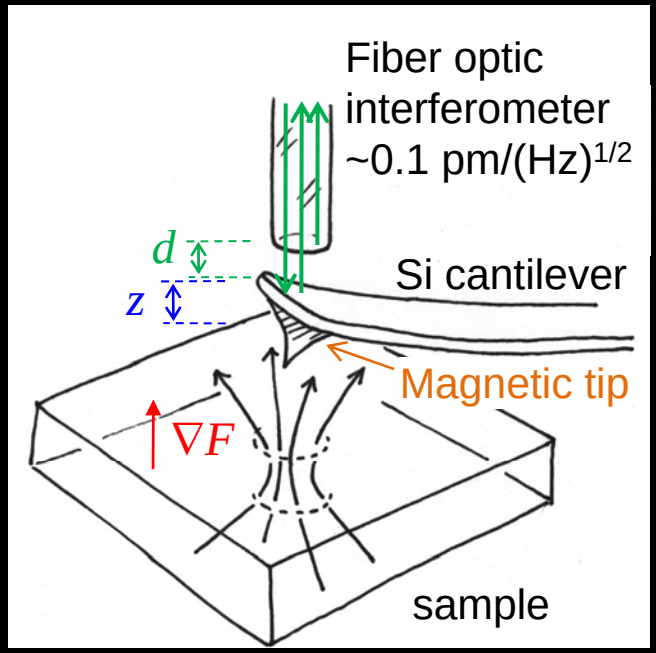
- Can we understand it? → need to understand Mott
- Can we use it? → need to understand vortices
  - scanning tunneling microscope
    - → vortex imaging in $Ba(Co_xFe_{1-x})_2As_2$
  - force microscope
    - → vortex manipulation in $NdO_{1-x}F_xFeAs$

Metal-insulator transition in $VO_2$

- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

Outlook

# Magnetic Force Microscope




Force between tip and sample:

$$\mathbf{F} = \nabla(\mathbf{m} \cdot \mathbf{B})$$

Frequency modulation imaging

(directly measures force gradient):

$$\frac{\Delta\omega}{\omega_0} = \frac{-1}{2k}\frac{dF_{ts}}{dz}$$

## Pros and Cons of MFM

*Tip Geometry*

*Con*: Imperfectly known

*Pro*: Up to 20 nm spatial resolution

*Other signals*

*Con*: See atomic forces too

*Pro*: Simultaneous topography

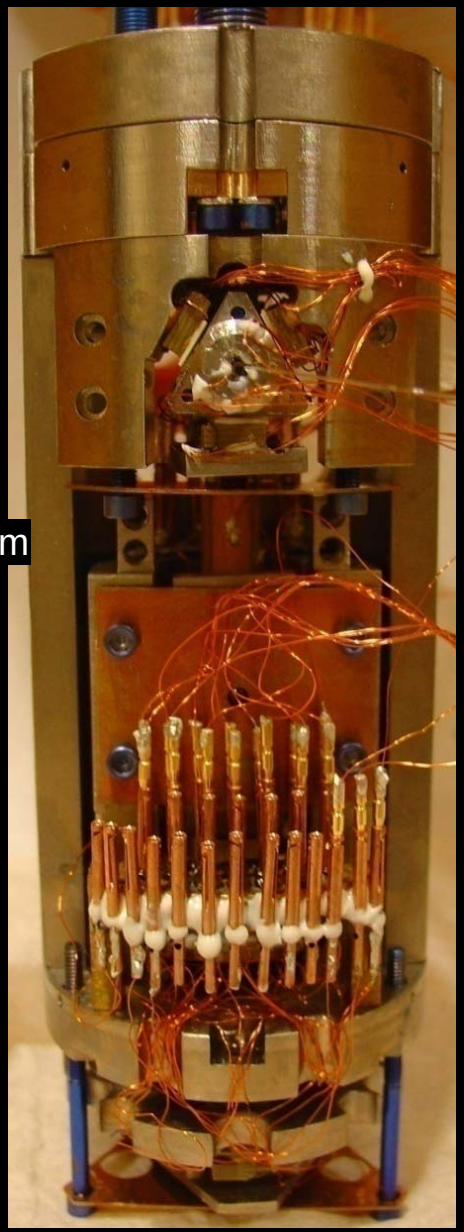
*Invasiveness*

*Con*: Tip exerts force on vortex

*Pro*: Tip exerts force on vortex

Vertical force gradient → imaging
Horizontal force → manipulation

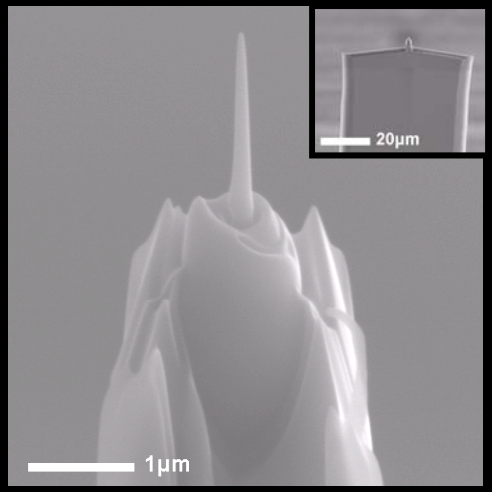
# Hoffman Lab Force Microscope




*built by Dr. Jeehoon Kim*

- 2 K to above 340 K
- 5 T vertical field
- lateral coarse motion (3 mm x 3 mm) allows imaging of isolated features in addition to bulk materials
- high-resolution, easily modelable tips fabricated in house via focused ion beam

Radius of curvature : 15-25 nm
Cone half-angles : 1-3°
Aspect ratios : 12-18




Versatility:

- vertical or lateral force measurement
- magnetic tips for magnetic imaging and manipulation
- conducting tips for local conductivity imaging and switching
- vertical cantilevers for friction imaging

# Outline

High-Tc Superconductivity

- Can we understand it? → need to understand Mott
- Can we use it? → need to understand vortices
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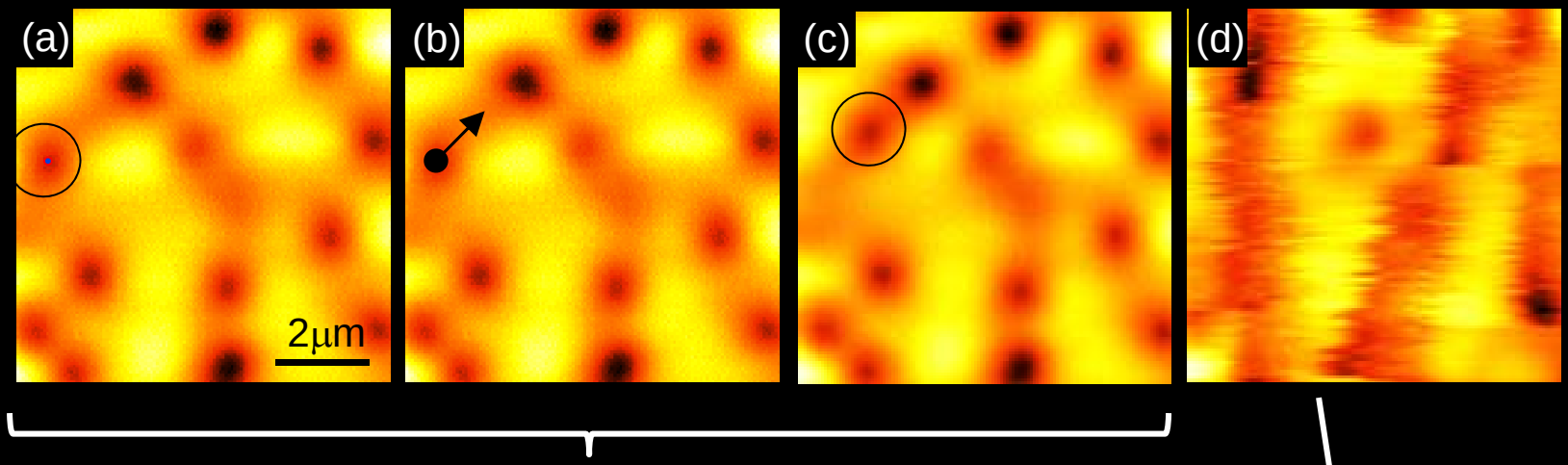
Metal-insulator transition in $VO_2$

- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

Outlook

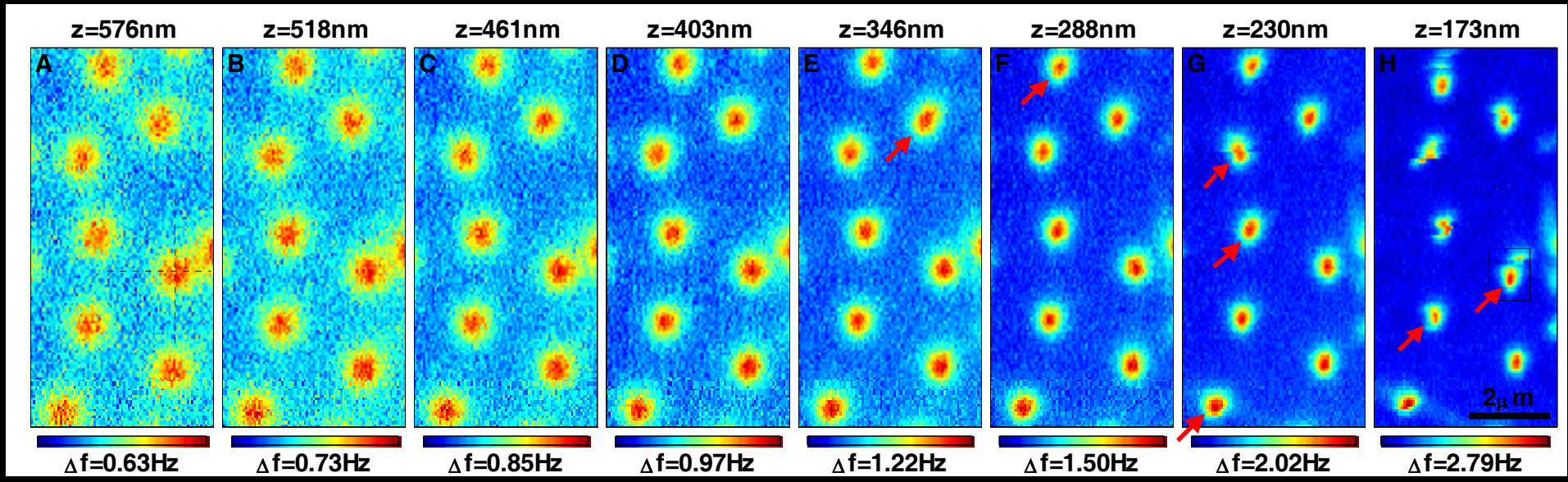
# Preliminary: Vortex Manipulation in $NdO_{1-x}F_xFeAs$

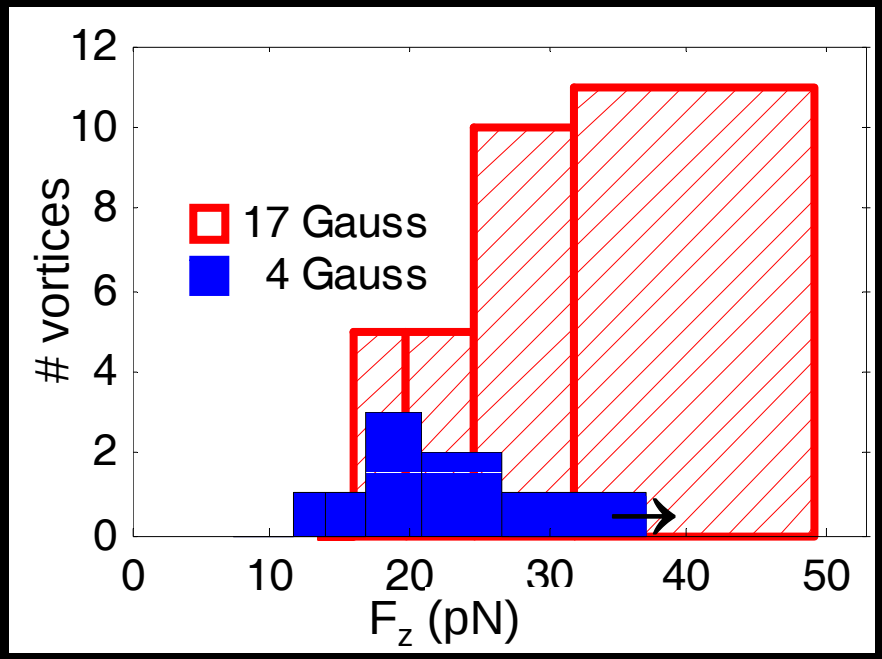



will histogram pinning forces

will measure pinning anisotropy

# Nb vortices: Pinning Force Histogram



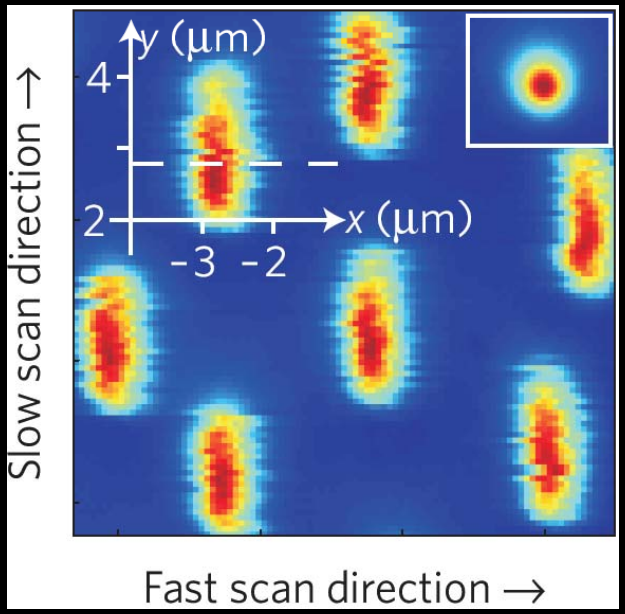




$F_{depin}$ ranges from
4 to 12 pN
at 5.5 K

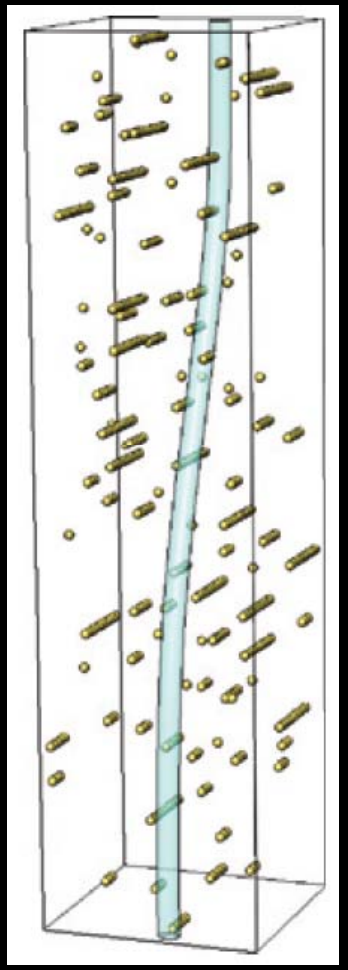
E. W. J. Straver, J. E. Hoffman,
O. M. Auslaender, D. Rugar, K. A. Moler,
***Appl. Phys. Lett.*** 93, 172514 (2008).

# $YBa_2Cu_3O_{7-d}$ vortices: Probe Bulk Pinning

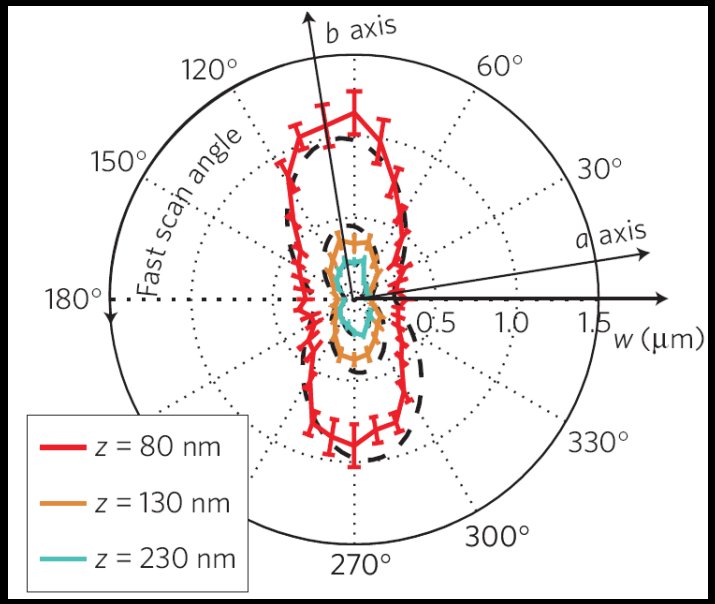
map anisotropy




deduce anisotropy of bulk pinning

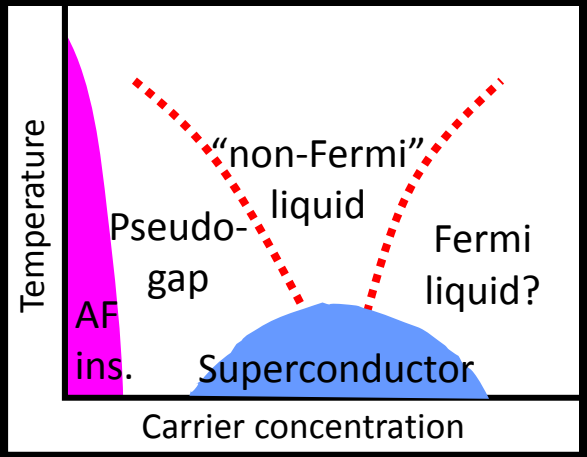
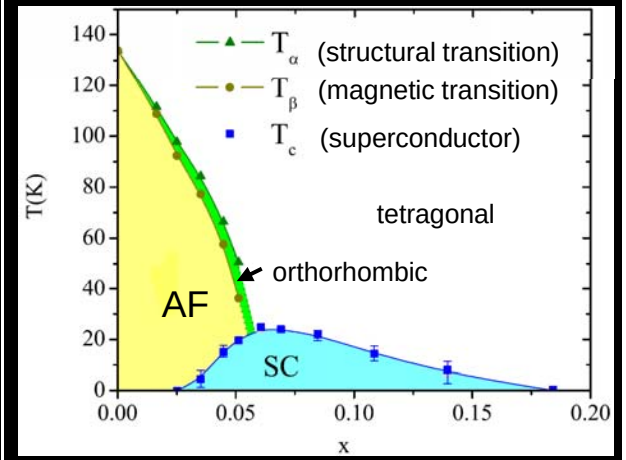


fit anisotropy

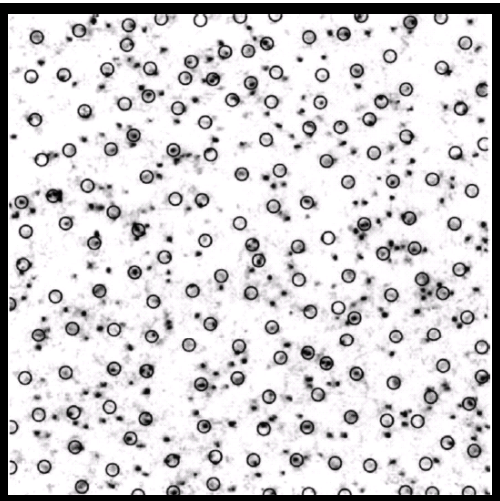
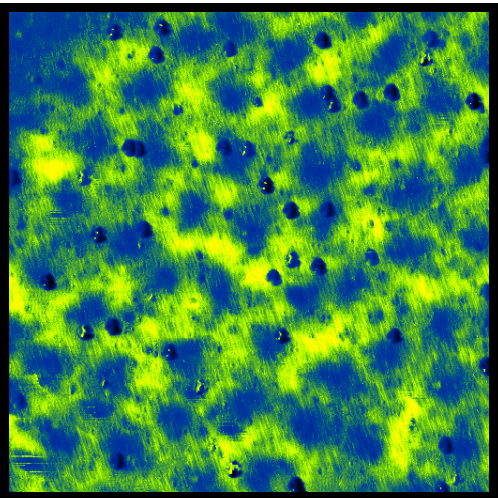



O. M. Auslaender, L. Luan, E. W. J. Straver, J. E. Hoffman, N. C. Koshnick, E. Zeldov, D. A. Bonn, R. Liang, W. N. Hardy, K. A. Moler, ***Nature Physics*** 5, 35 (2009).

# High-$T_c$ Cuprate-Pnictide Conclusions

| | Cuprate: $Bi_2Sr_2CaCu_2O_{8+d}$ | Pnictide: $BaCo_xFe_{2-x}As_2$ |
|---|---|---|
| phase diagram |  |  |
| ground state | antiferromagnetic Mott insulator | itinerant antiferromagnet semimetal |
| superconducting gap, $\Delta$ | $\Delta$ ~ 33 meV<br>$2\Delta/k_BT_c$ ~ 6-10 | $\Delta$ = 6.25 meV<br>$2\Delta/k_BT_c$ = 5.73 |
| gap inhomogeneity | $\sigma$ ~ 7 meV<br>$\sigma/\Delta$ ~ 21% | $\sigma$ = 0.73 meV<br>$\sigma/\Delta$ = 12% |
| gap symmetry | d-wave | s± |

# High-$T_c$ Cuprate-Pnictide Conclusions

| | Cuprate: $Bi_2Sr_2CaCu_2O_{8+d}$ | Pnictide: $BaCo_xFe_{2-x}As_2$ |
|---|---|---|
| coherence length, $\xi_0$ | 2.2 nm | 2.7 nm |
| Anisotropy | ~50 | 1-3 |
| vortex pinning |  vortices pinned to surface impurities |  vortices NOT pinned to surface impurities |

- Local probes access nanoscale inhomogeneity → "see" correlations
- Image and manipulate magnetic vortices
- Surface probes can access bulk properties

# Outline

High-Tc Superconductivity

- Can we understand it? → need to understand Mott
- Can we use it? → need to understand vortices
  - scanning tunneling microscope
    → vortex imaging in $Ba(Co_xFe_{1-x})_2As_2$
  - force microscope
    → vortex manipulation in $NdO_{1-x}F_xFeAs$

Metal-insulator transition in $VO_2$

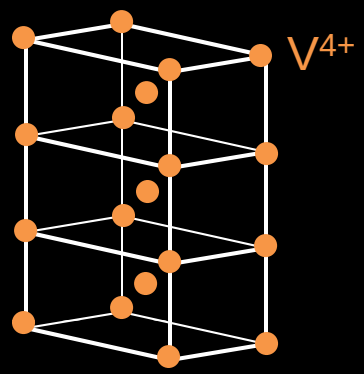
- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

Outlook

# $VO_2$ Metal-Insulator Transition

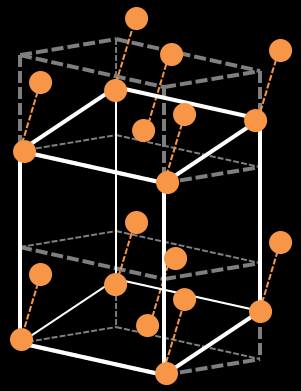
T (K)

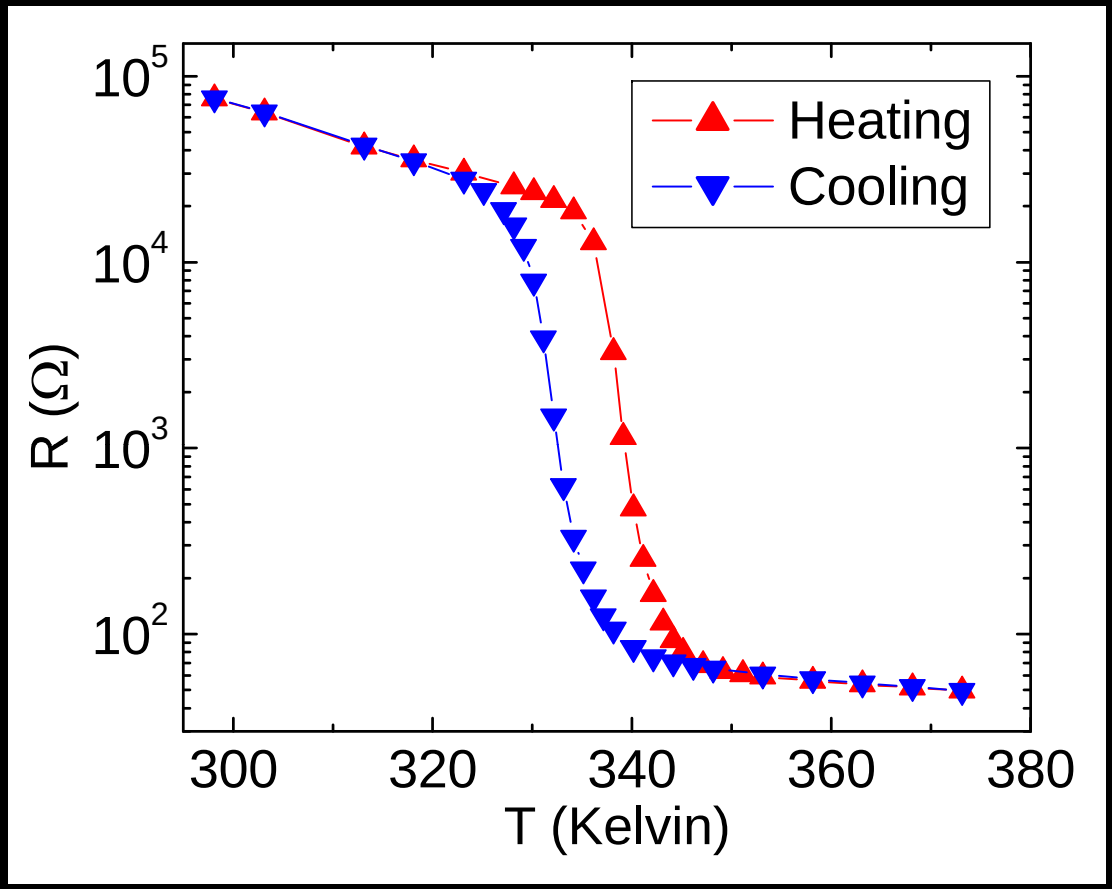
Tetragonal Metal




340 K

Monoclinic Insulator






Also transitions as a function of

- stress (~38 kbar)
- optical excitation
- applied voltage or current (~$10^7$ V/m ??)

→ up to 5 orders of magnitude change in conductivity

# Outline

High-Tc Superconductivity

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Metal-insulator transition in $VO_2$

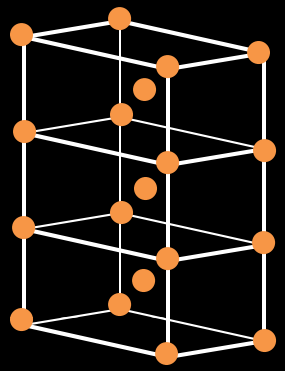
- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation

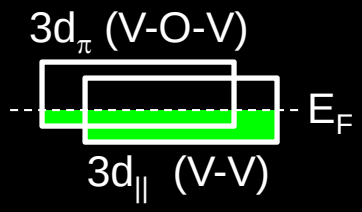
Outlook

# $VO_2$ Metal-Insulator Transition: Physics

T (K)

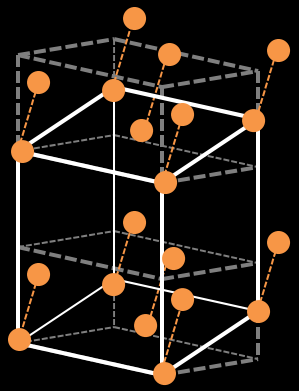
Tetragonal Metal

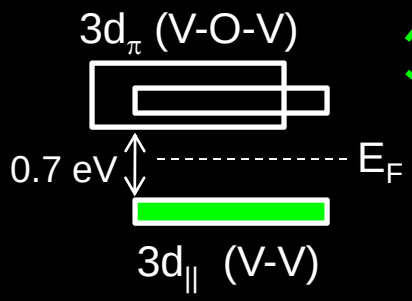





340 K

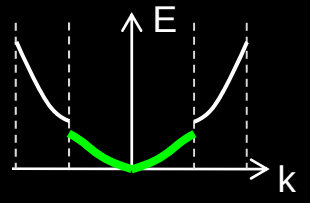
Monoclinic Insulator






?

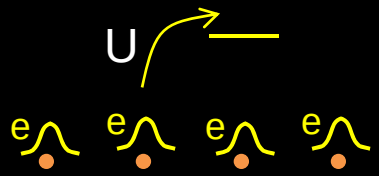
Band insulator?




- Goodenough, Phys. Rev. 117, 1442 (1960)
- Wentzcowitch, PRL 72, 3389 (1994)

?

Mott insulator?




- Zylbersztejn & Mott, PRB 11, 4383 (1975)

*Goodenough, J.Solid State Chem 3, 490 (1971)*

# $VO_2$ Questions

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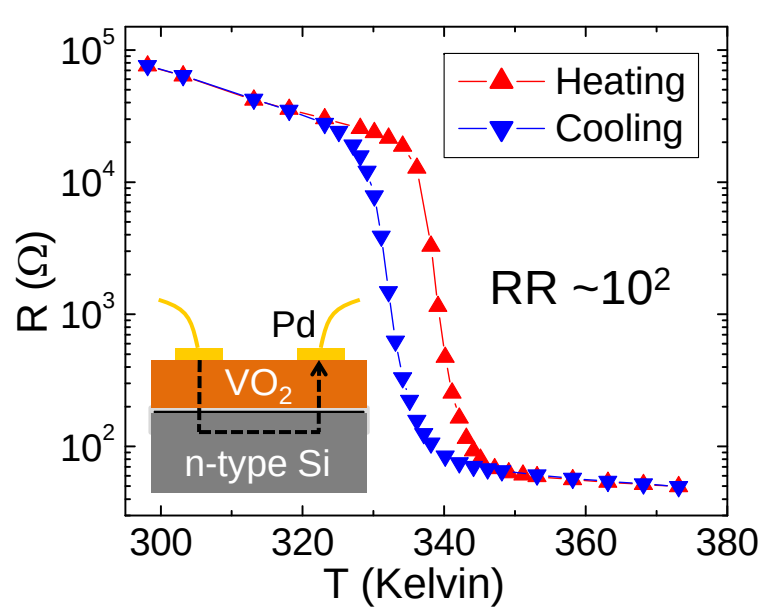
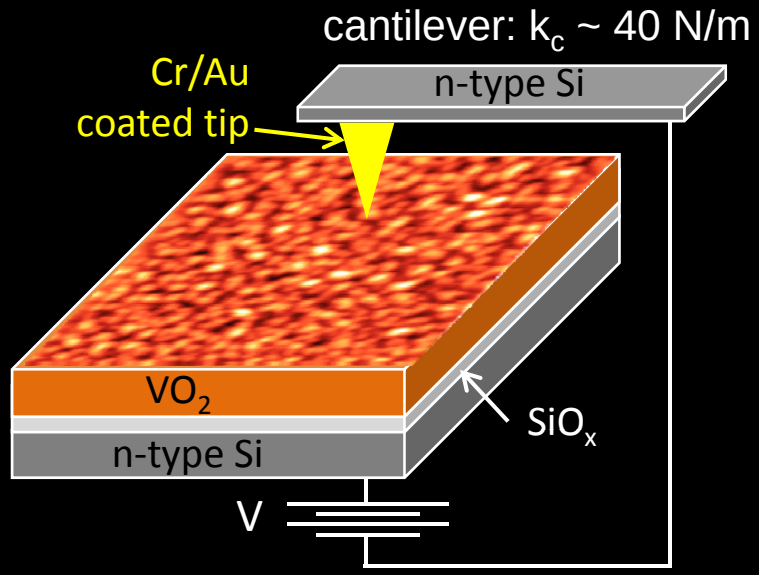
  *Gopalakrishnan, J. Mat. Sci. 44, 5345 (2009)*

# Bulk vs. Local

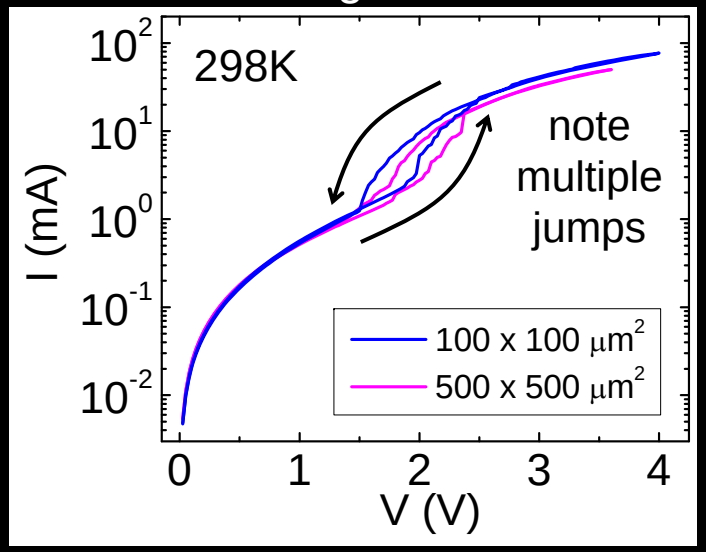
## Bulk thermal transition



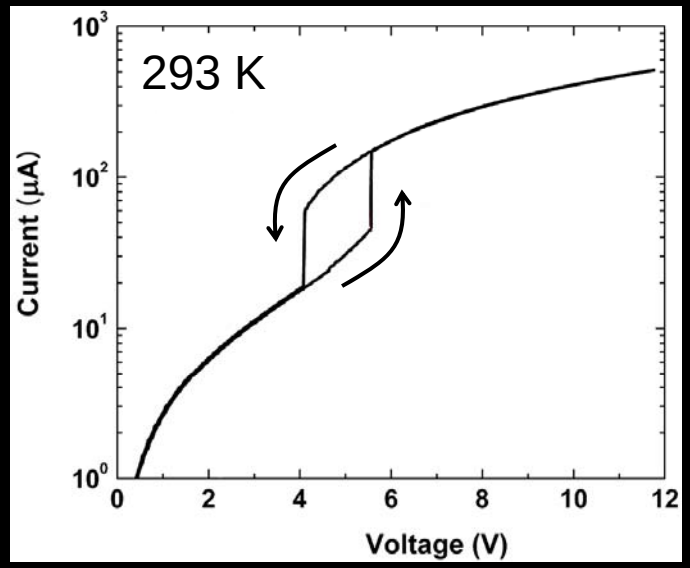

## Local geometry:



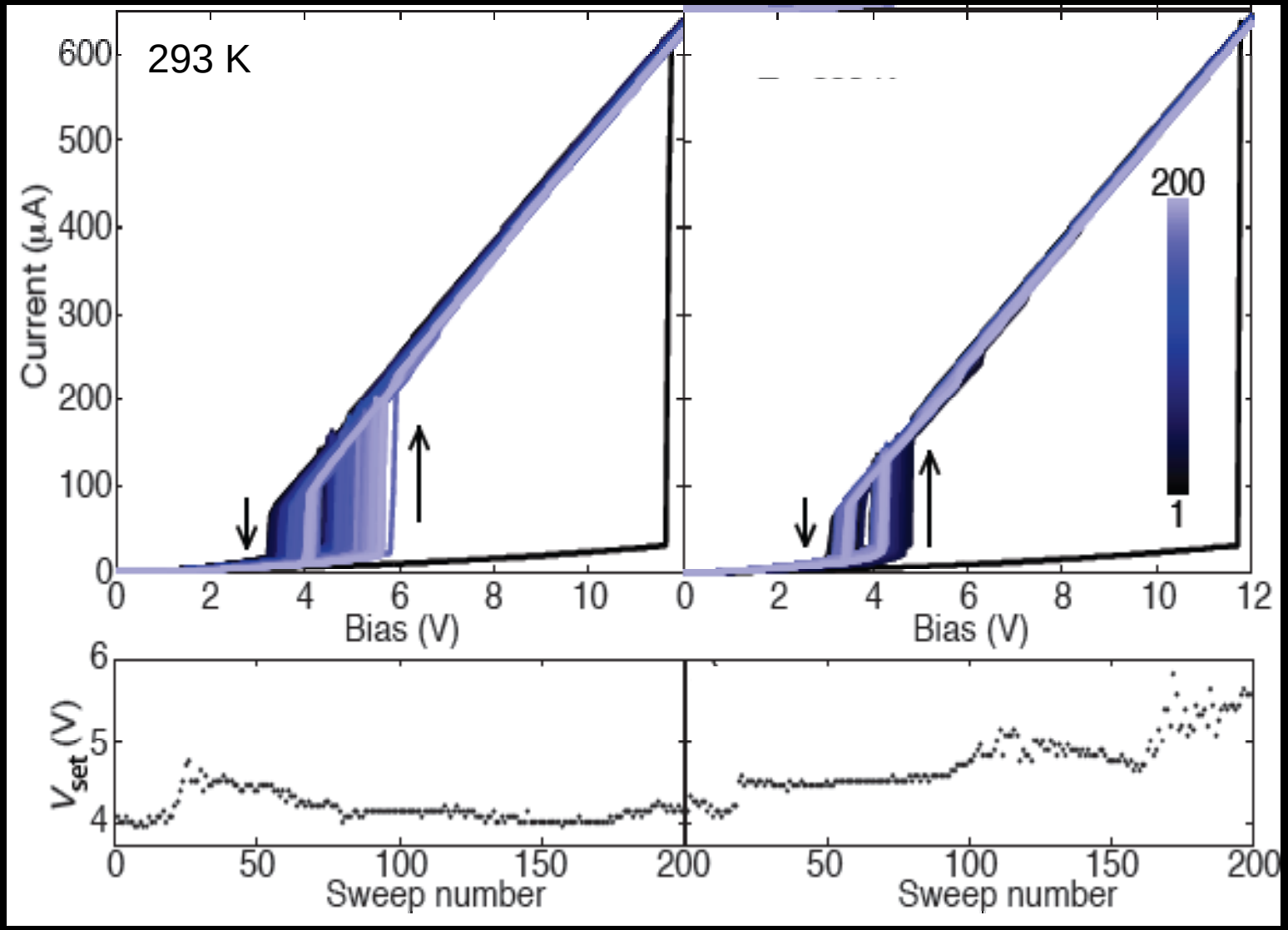

## Look for bulk voltage transition




## Single Point IV (conducting AFM):



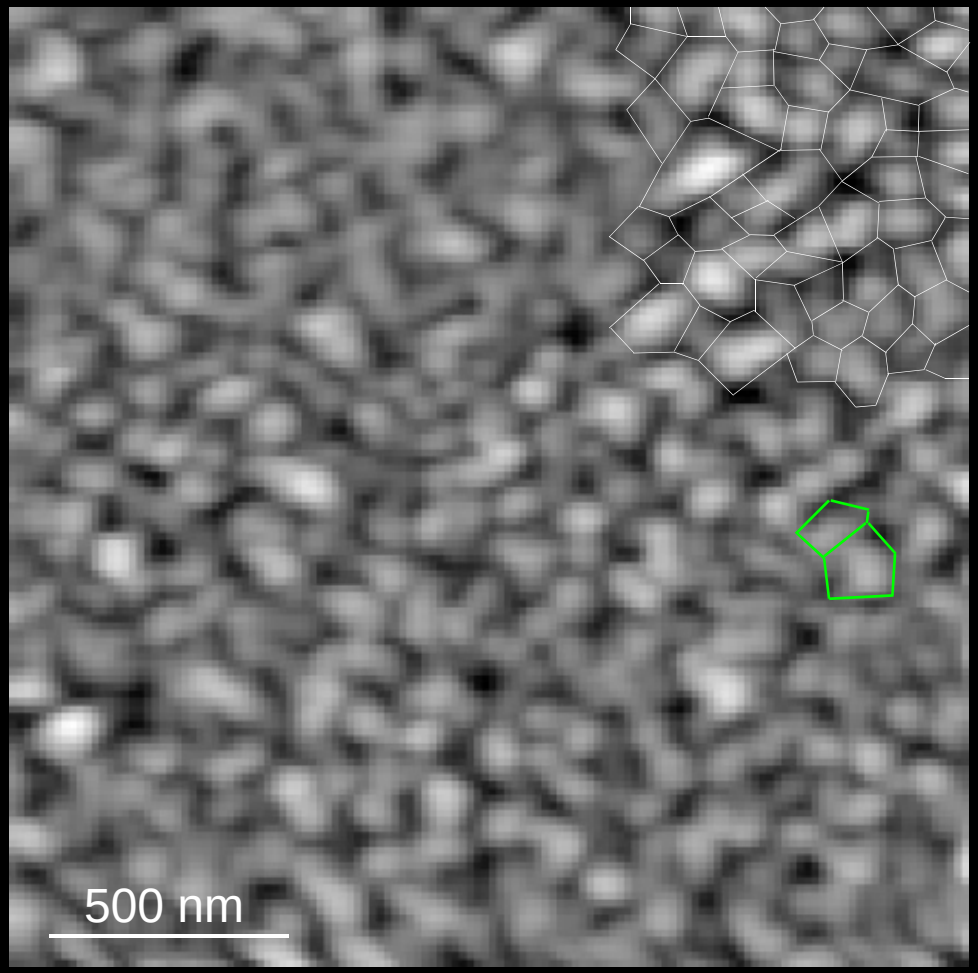
# Training & Repeatability




Training voltage: Typically one or several high voltage sweeps necessary before IV hysteresis loops roughly stabilize with ~5% jitter

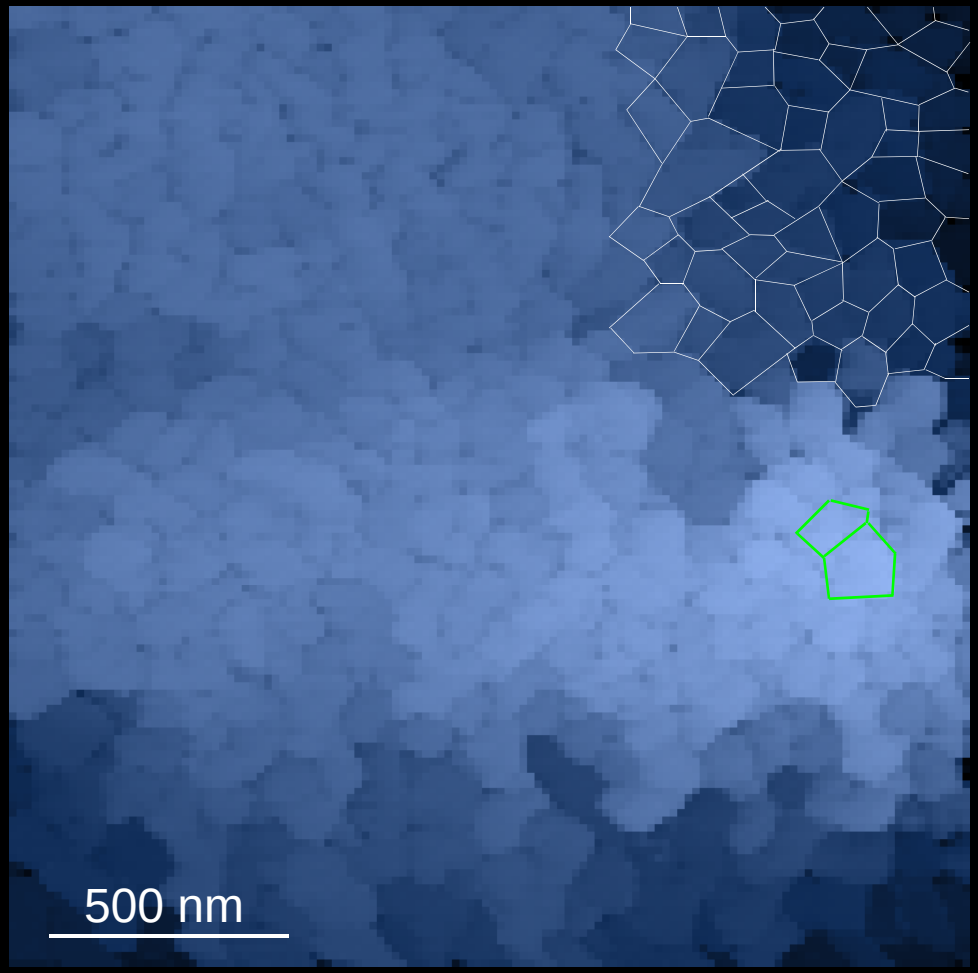
# Imaging the Metal-Insulator Transition

Topography



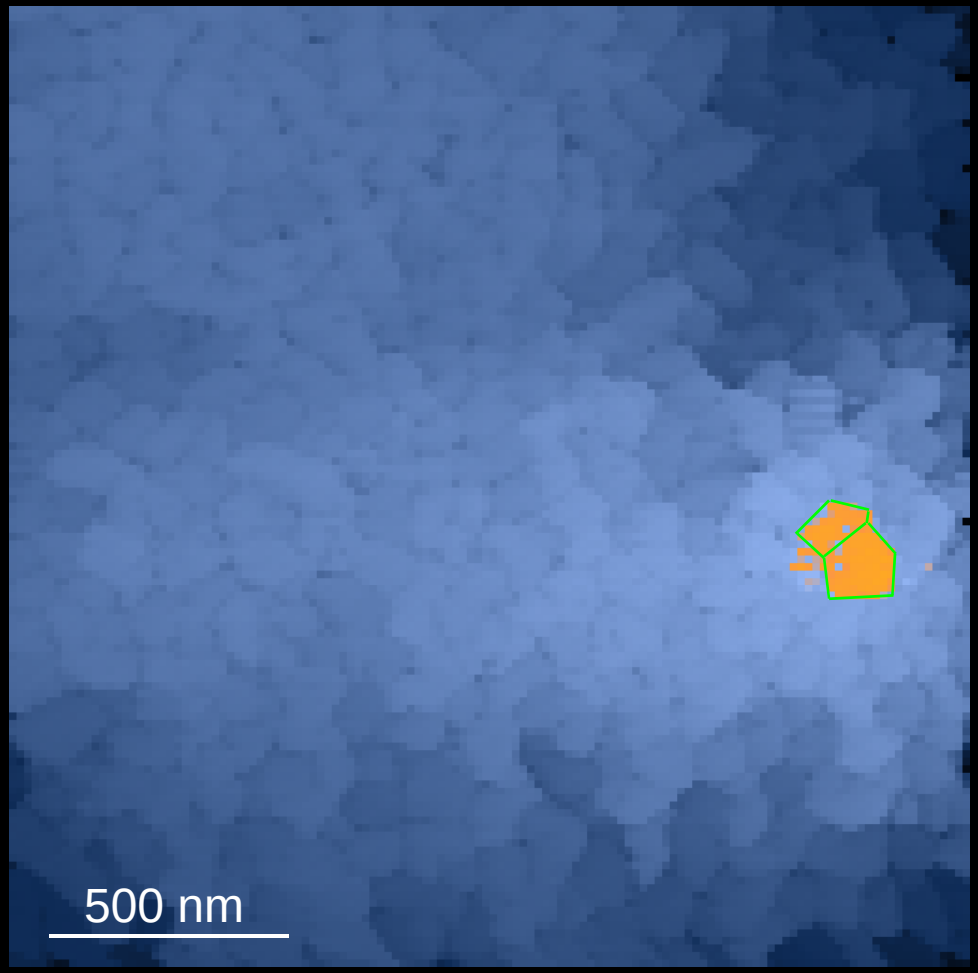
# Imaging the Metal-Insulator Transition

Current map
Tip bias: 3.45 V



# Imaging the Metal-Insulator Transition

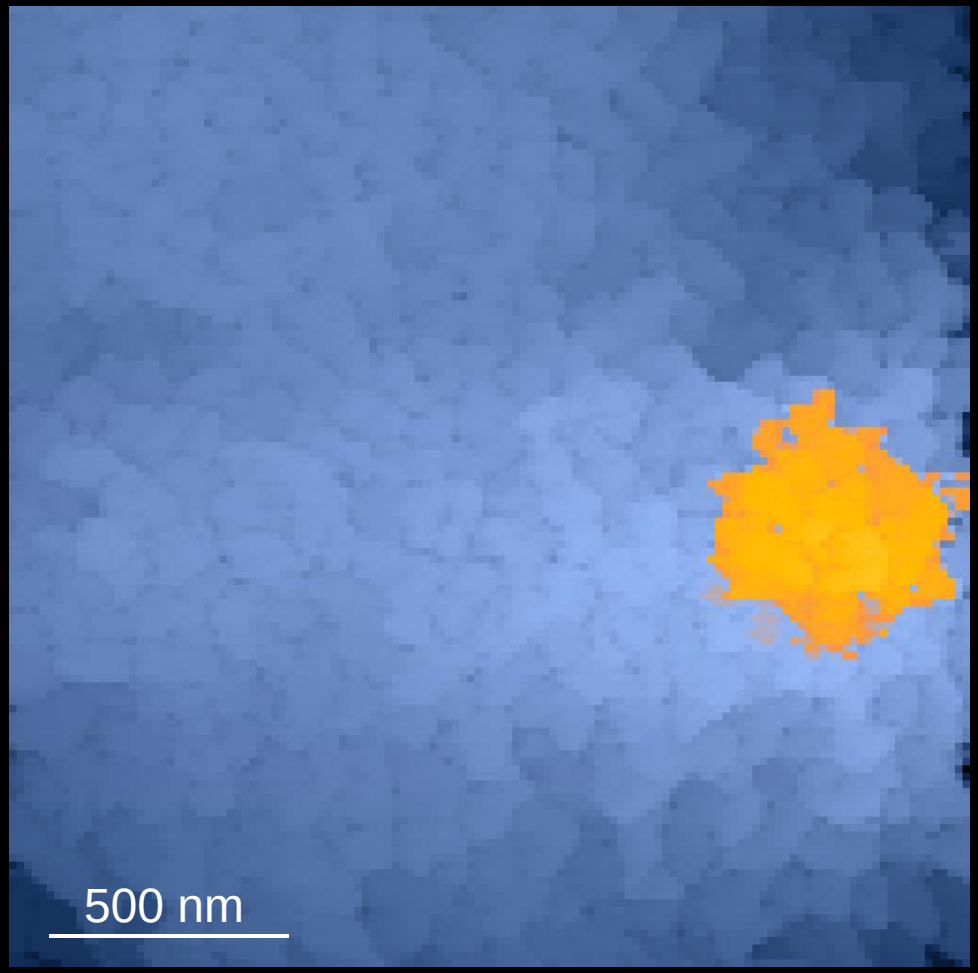
Current map
Tip bias: 3.95 V




→ seed grain is the one with highest conductivity in the insulating state

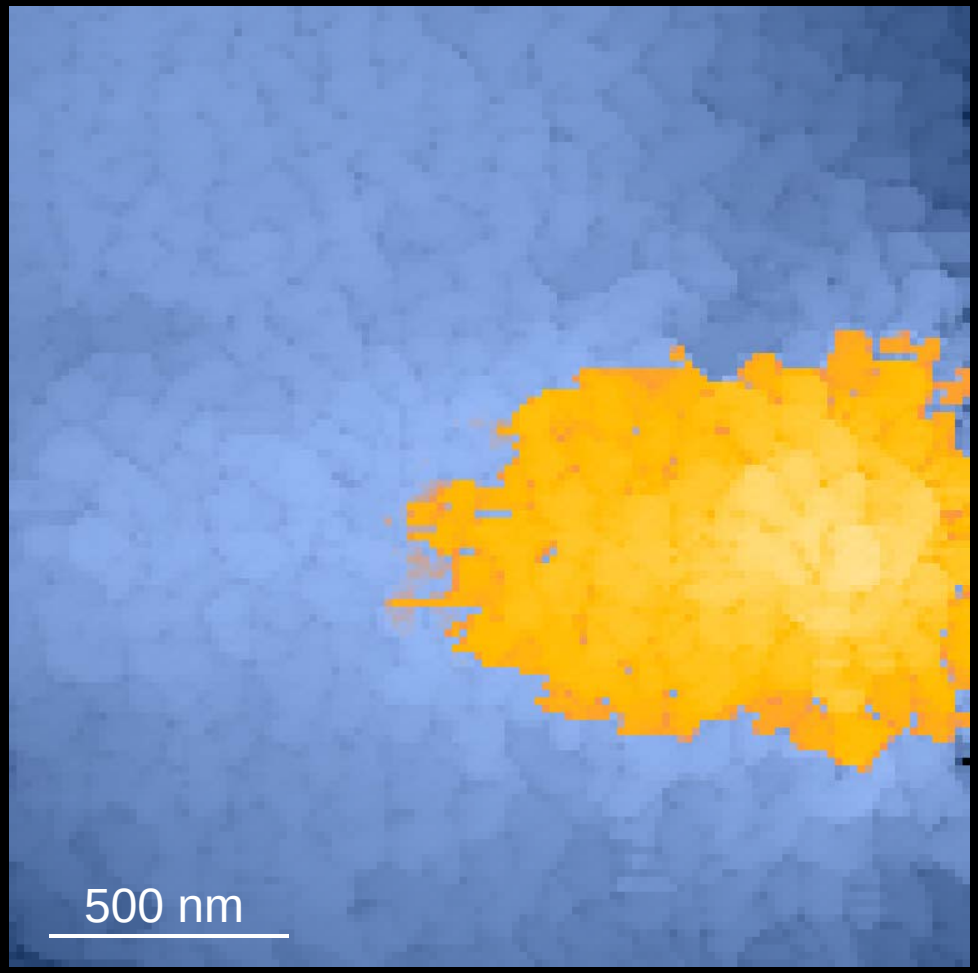
# Imaging the Metal-Insulator Transition

Current map
Tip bias: 4.44 V



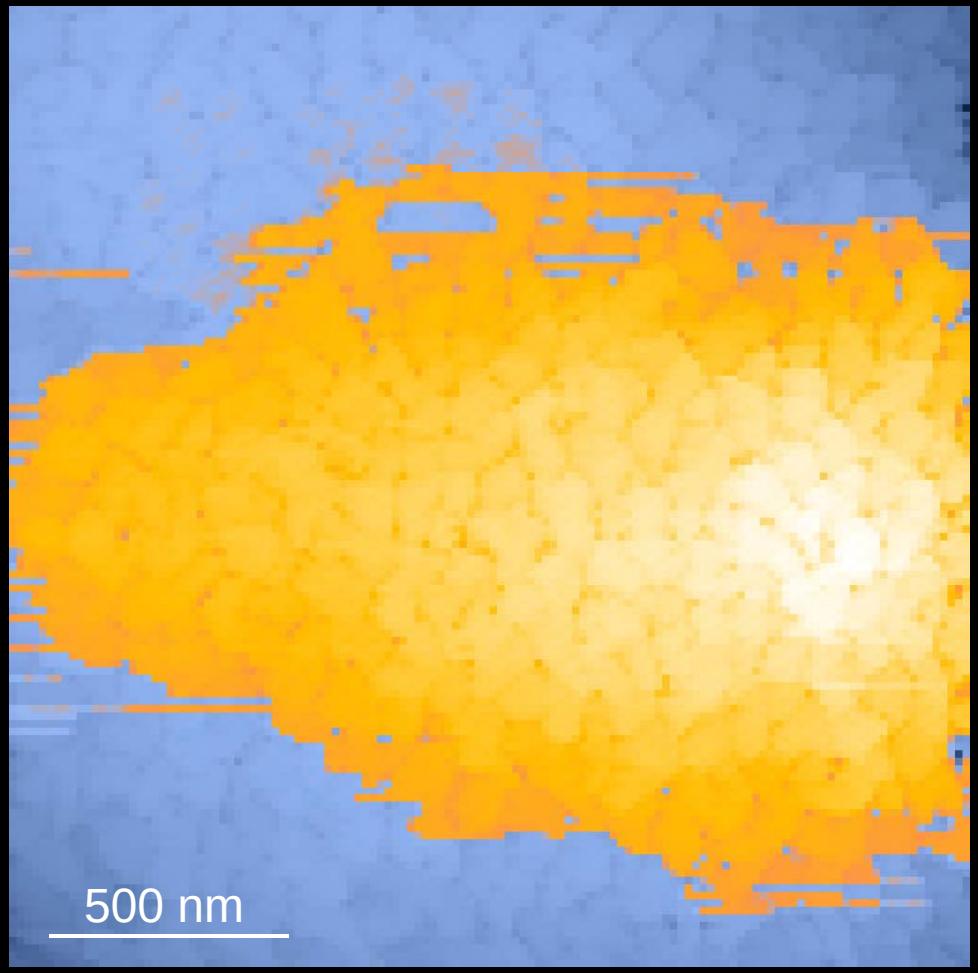
# Imaging the Metal-Insulator Transition

Current map
Tip bias: 4.93 V



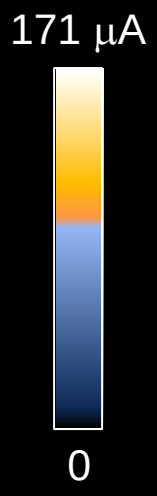
# Imaging the Metal-Insulator Transition

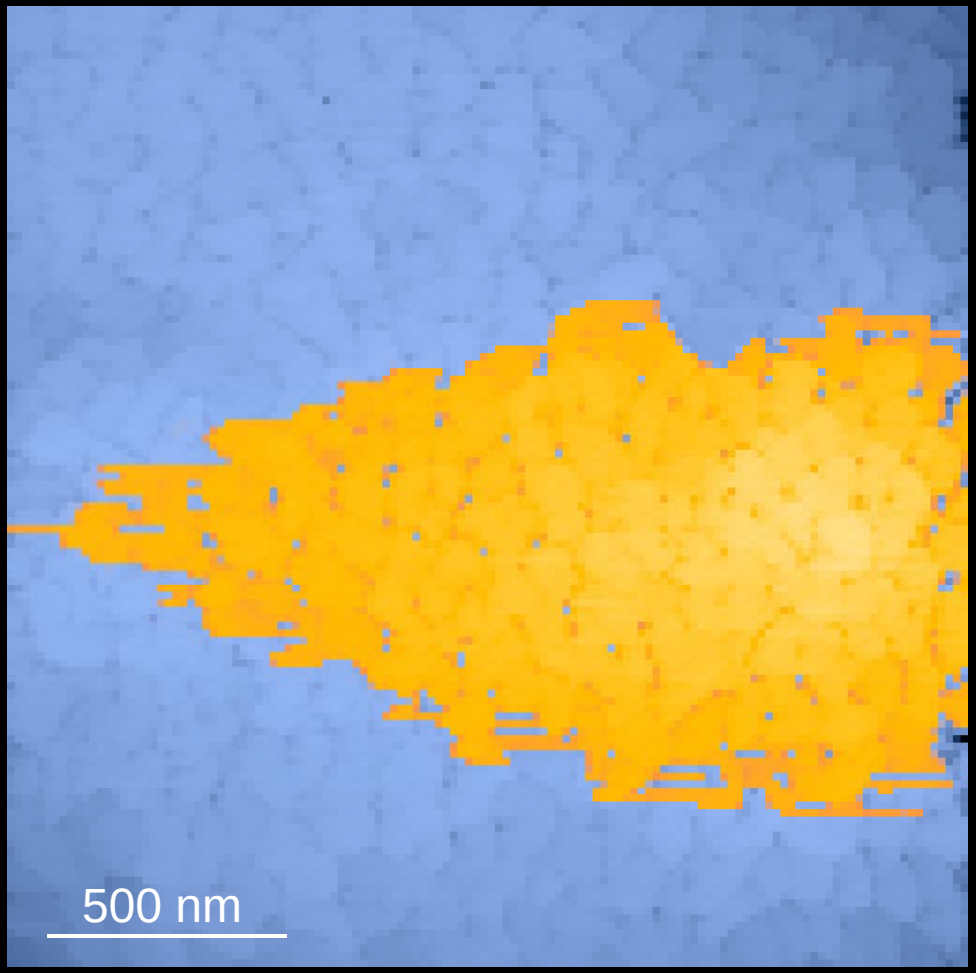
Current map
Tip bias: 5.43 V



# Imaging the Metal-Insulator Transition

Current map
Tip bias: 4.93 V



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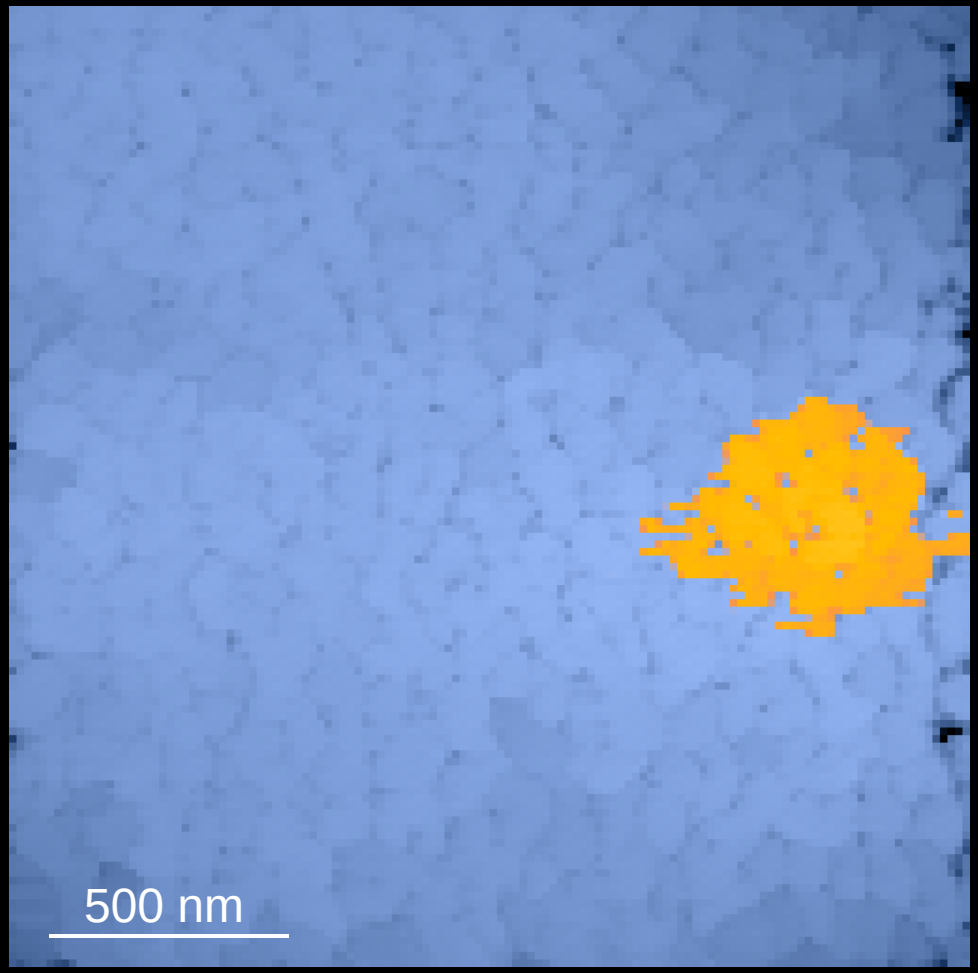
Tip bias: 4.44 V



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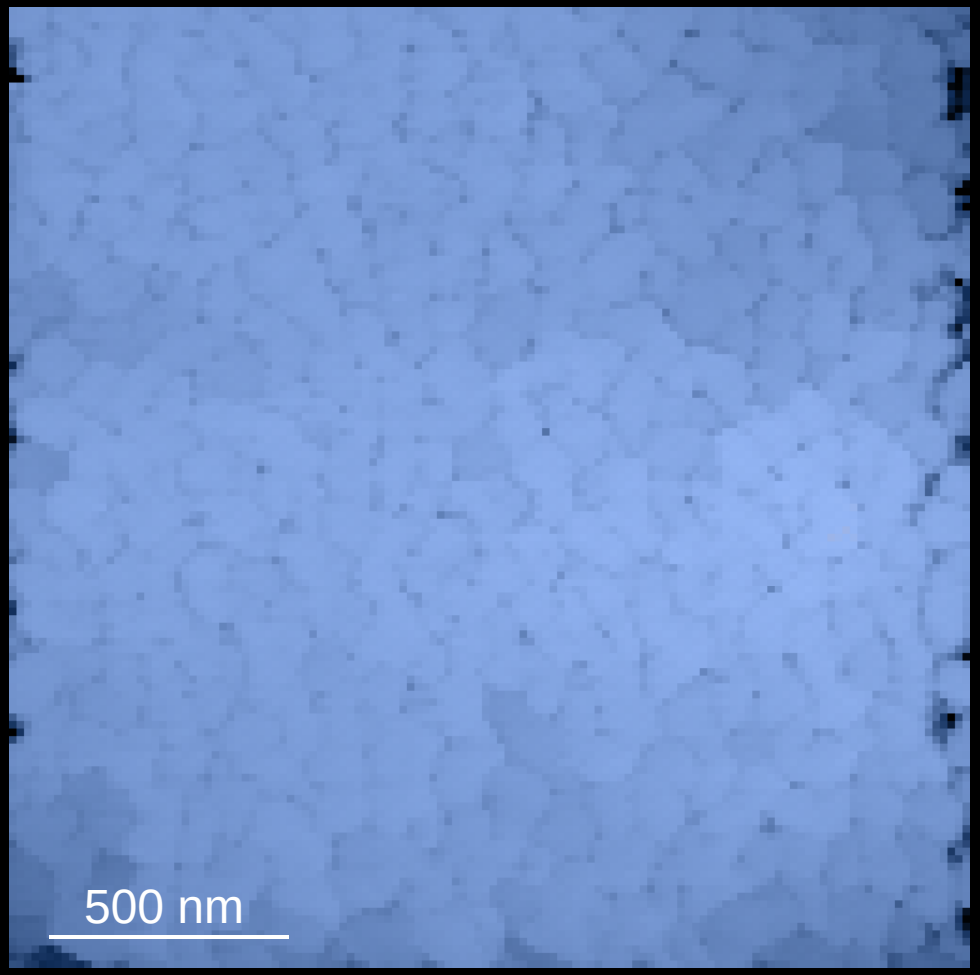

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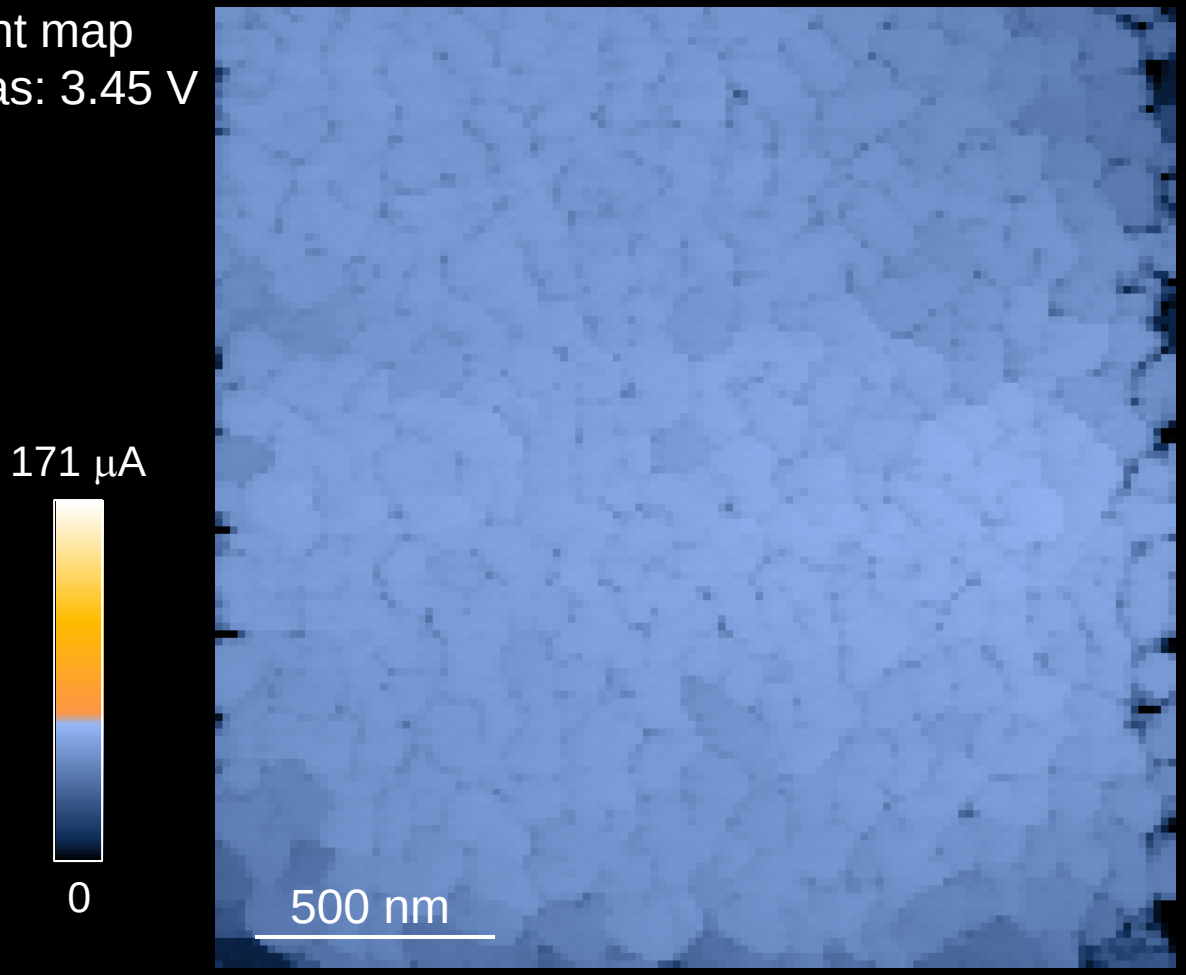
Tip bias: 3.45 V



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  *Qazilbash, unpublished APS March talk (2010)*

→ AFM detects structural; CAFM detect electronic; simultaneous independent info

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→ scanning gate microscopy

# Outline

High-Tc Superconductivity

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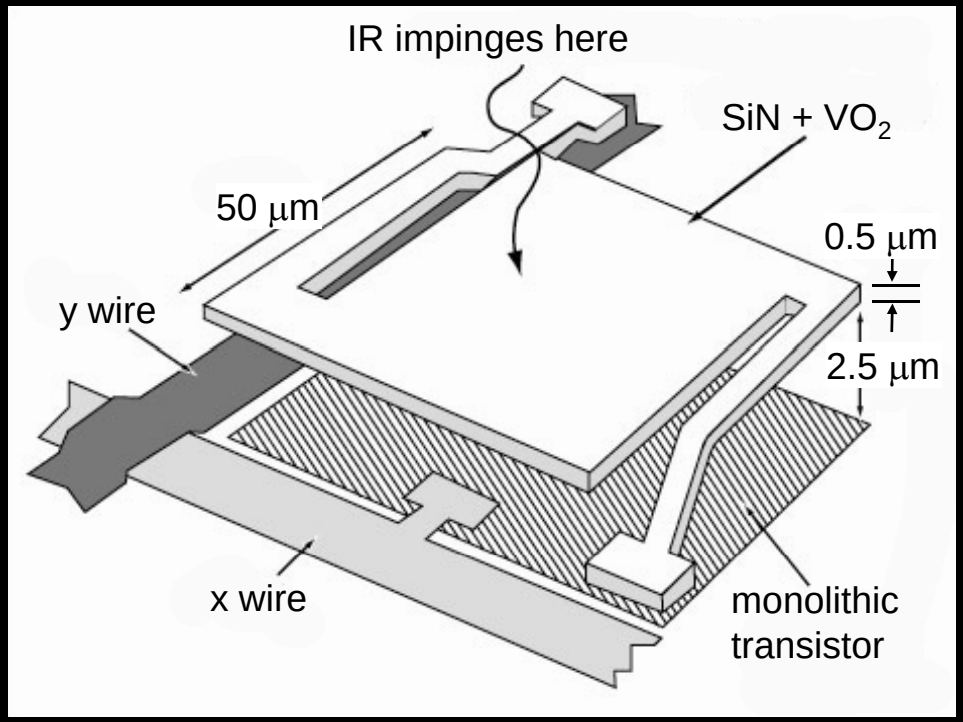
Metal-insulator transition in $VO_2$

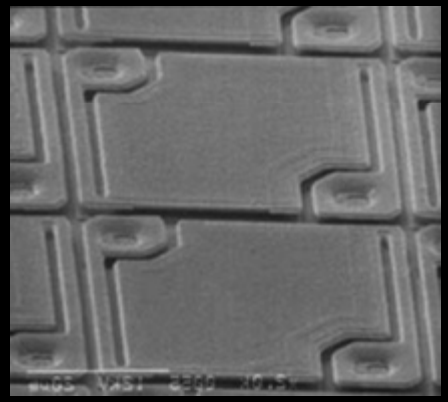
- Can we understand it? → isolate electronic & structural
- Can we use it? → Mott manipulation ?

Outlook

# $VO_2$ micro-bolometers for IR imaging




*http://www.thermodelta.hu*



*http://www.security-int.com*

- pixels size as small as ~ 25 μm
- pixel arrays up to 640 x 480

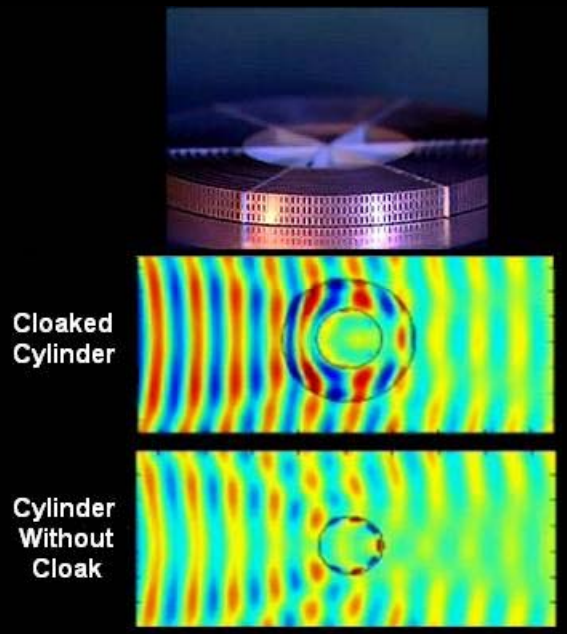
# $VO_2$ tunable metamaterials

"Invisibility cloak":
fabricate copper patterns on
standard circuit board
with fixed dielectric

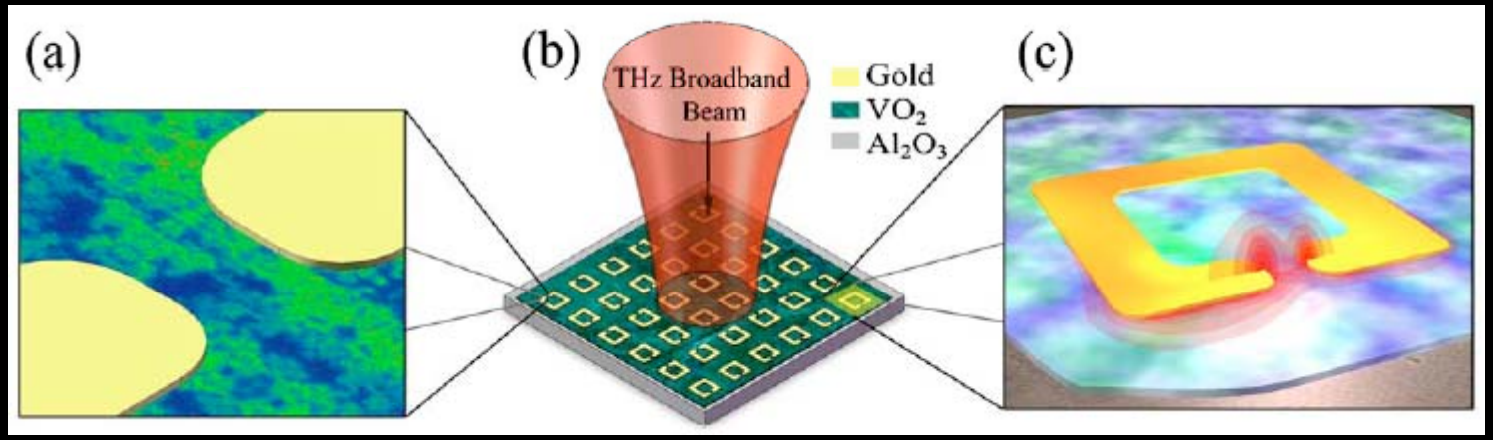
Problem: narrow, fixed bandwidth

Proposal: use $VO_2$ substrate
→ tunable dielectric
→ tunable metamaterial bandwidth

Prototype:

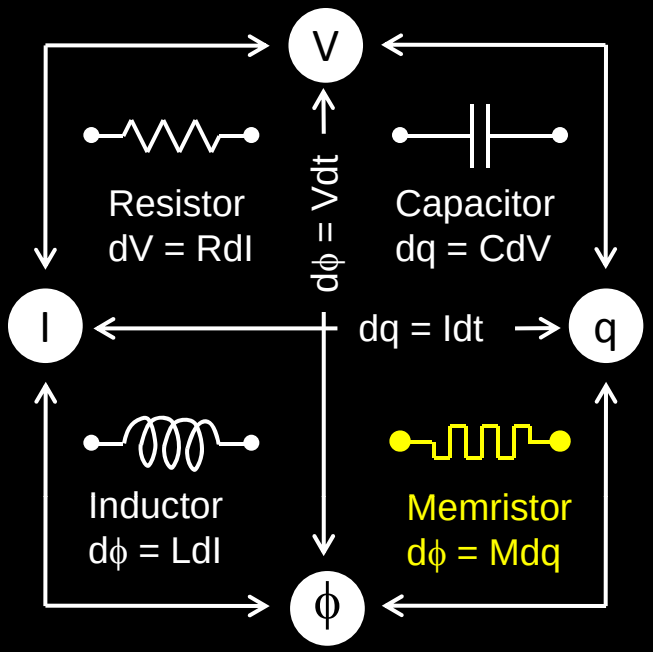



*David Smith, Duke University*




*Driscoll, APL 93, 024101 (2008)*

# Memristors




*L. Chua, IEEE Trans. Circuit Theory 18, 507 (1971)*
*D. Strukov, Nature 453, 80 (2008)*

## Applications

- Memory circuits
- Digital logic circuits
  (e.g. FPGA = Field Programmable Gate Array)
- Analog circuits
  (e.g. neural learning networks)

Integral form

$$\int d\varphi = \int M(q)dq$$

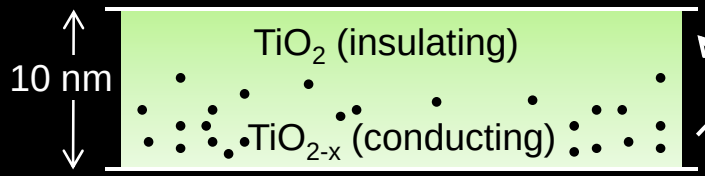
Differential form

$$M(q) = \frac{d\varphi}{dq} = \frac{d\varphi/dt}{dq/dt} = \frac{V}{I}$$

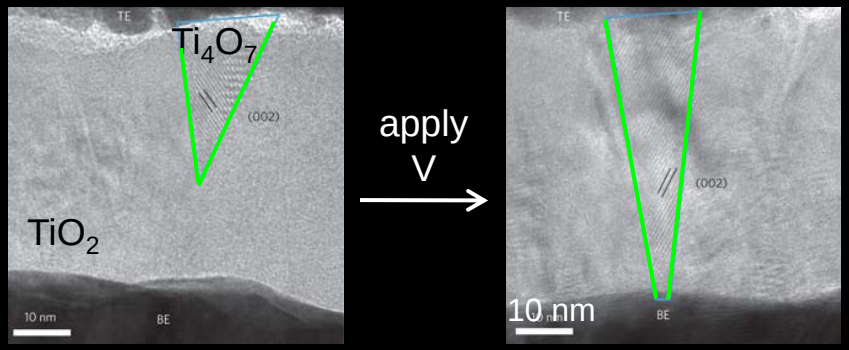
A memristor remembers its resistance, based on the history of charge flowing through it.

# $VO_2$ Memristors

Mechanism 1:
oxygen migration




Mechanism 2:
conductive channel formation

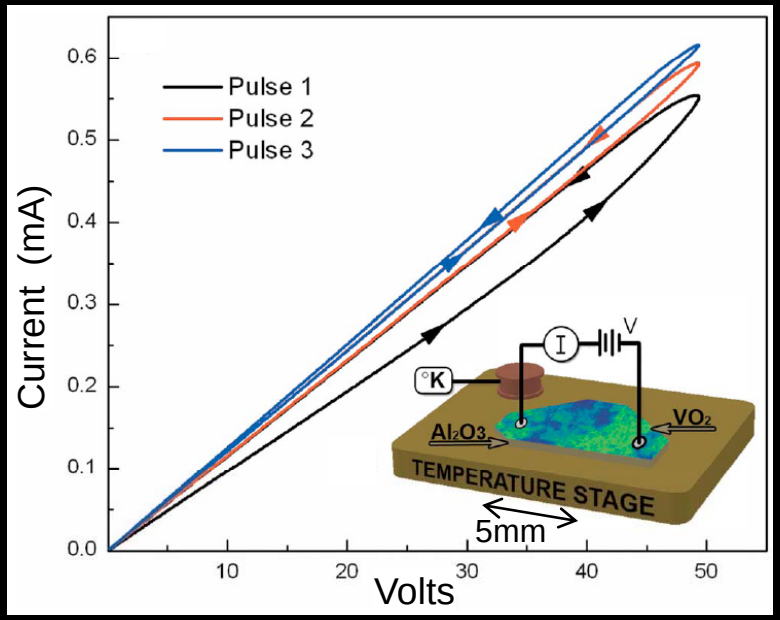



*Kwon, Nat Nano 5, 148 (2010)*

→ messy, disordered…
problems with reproducibility

Mechanism 3:
phase change

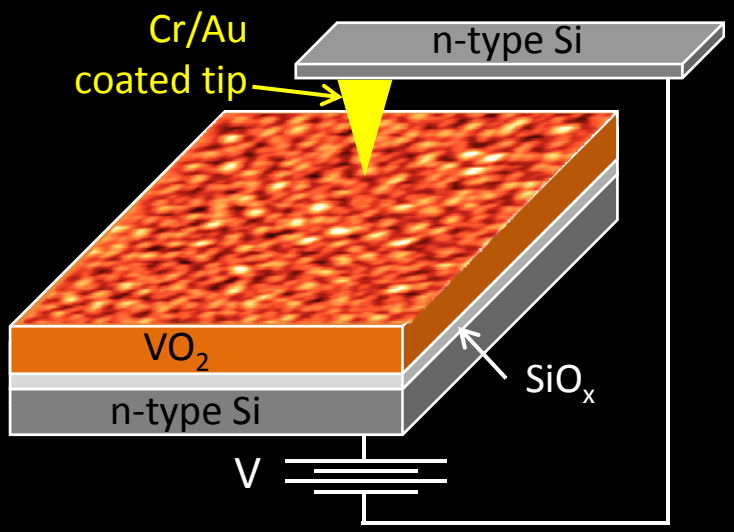
→ potentially cleaner, reproducible

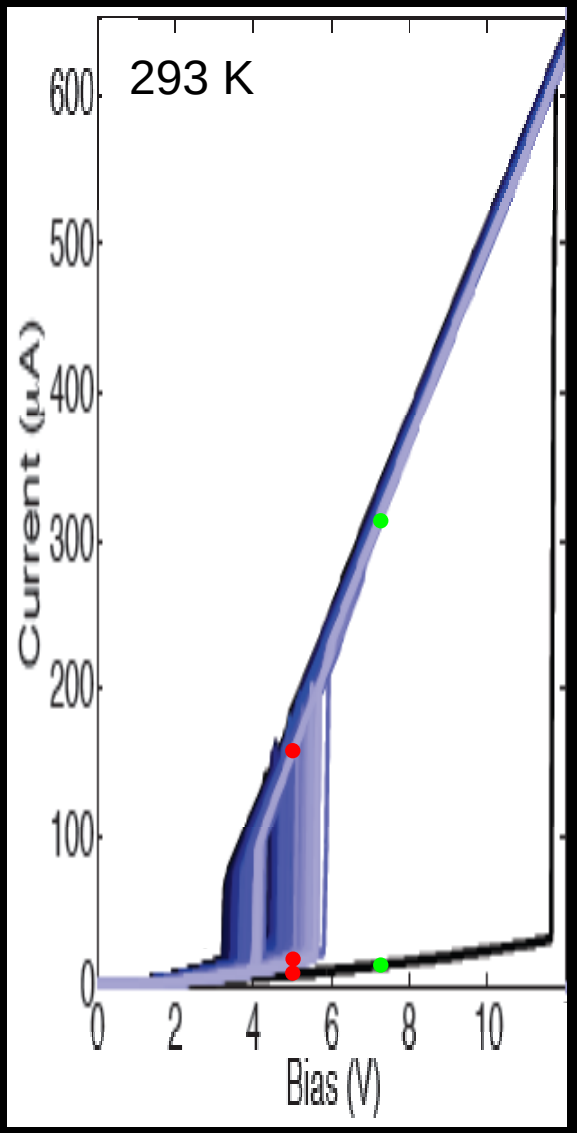



*Driscoll, APL 95, 043503 (2009)*

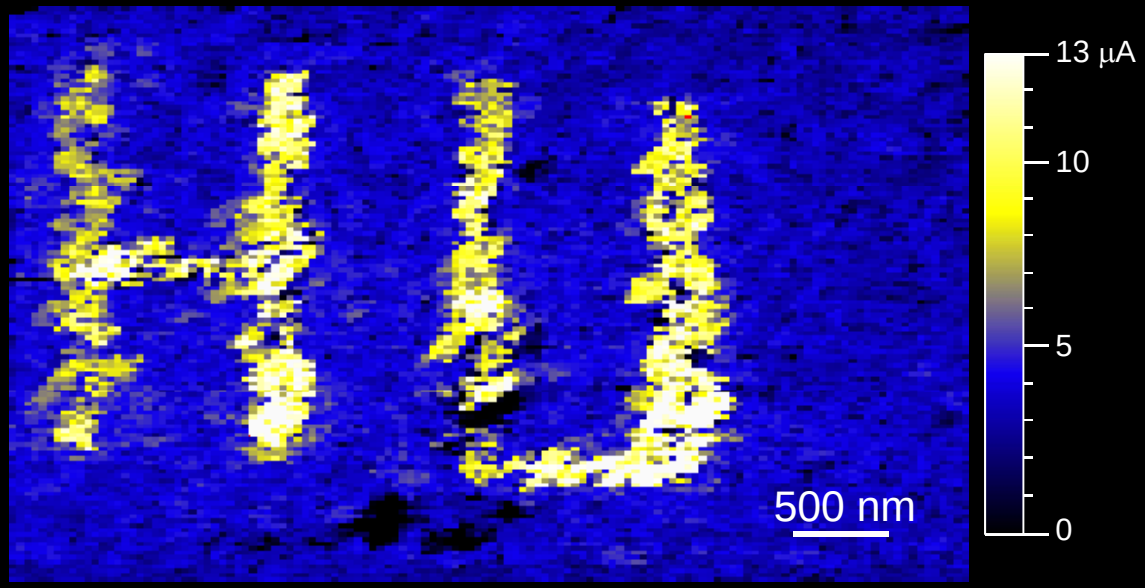
# Memristive Writing ?

Step 1: “write” with 13 V tip bias




Step 2: read current (local resistivity) with 3 V tip bias



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Metal-insulator transition in $VO_2$

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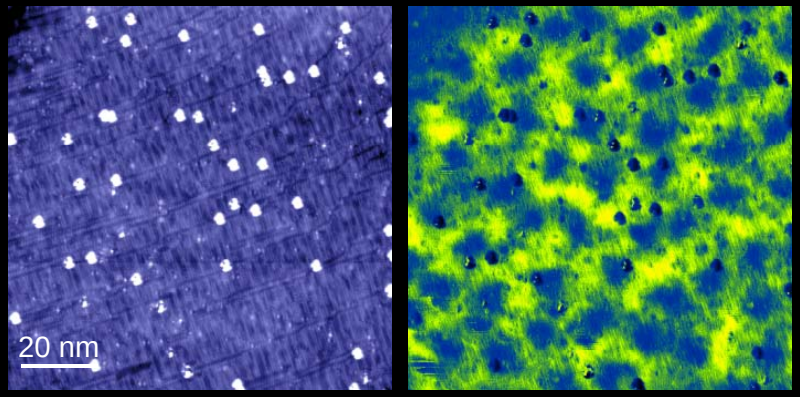
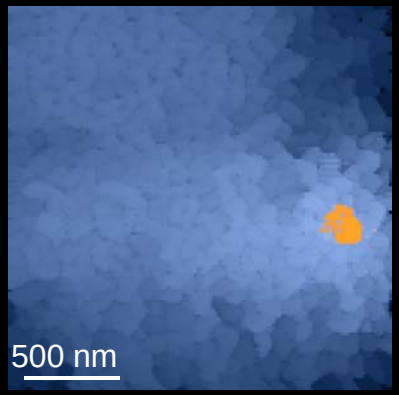
Outlook

# Conclusions

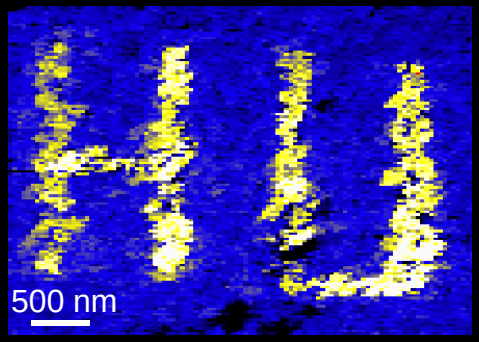
Iron-Pnictide Superconductors
- more homogeneous than cuprates → less highly correlated
- stronger vortex pinning




Vanadium Oxide
- controlled local transition (single grain, 10s of nanometers)
- after training, hysteresis is repeatable



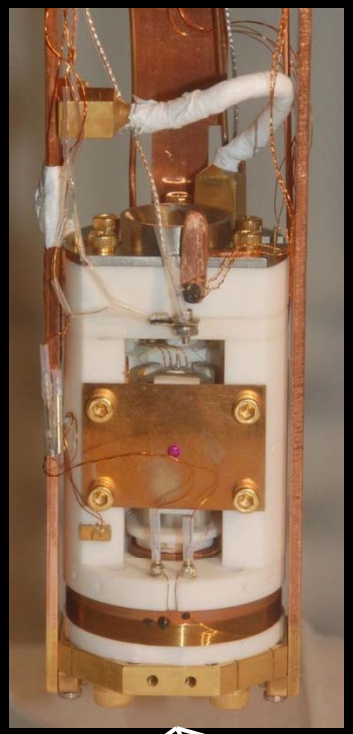

Outlook
- correlated electron systems → local inhomogeneity → need local probes
- surface probes can access bulk properties, e.g. vortex pinning
- passive imaging → active manipulation




To Do Next
- Quantify vortex pinning forces in iron-pnictide superconductors
- “Write” the Mott transition in vanadium oxide & other systems
- Image & manipulate other correlated systems, e.g. topological insulators

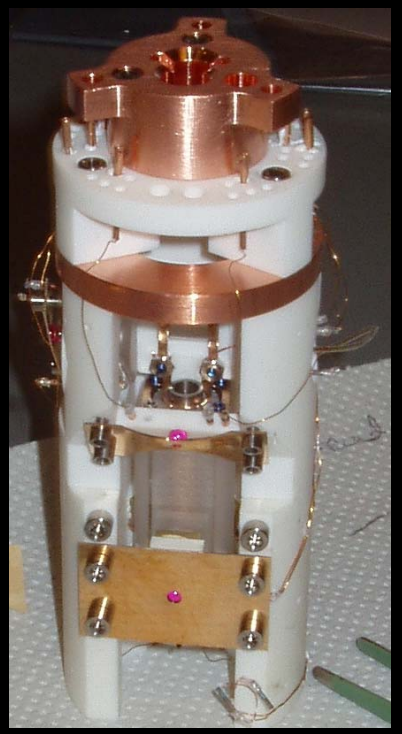
# Hoffman Lab Local Probes

Scanning Tunneling Microscope

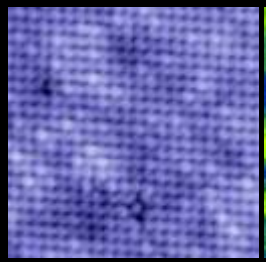


Force Microscope

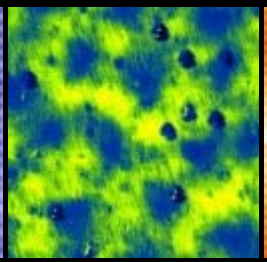
Ultra-high vacuum STM



coming on line soon!
- spin-polarized tunneling
- atom-moving

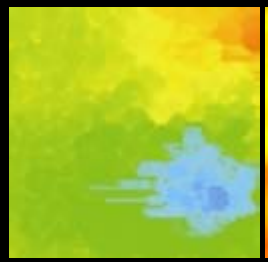


cuprate high-$T_c$ superconductor

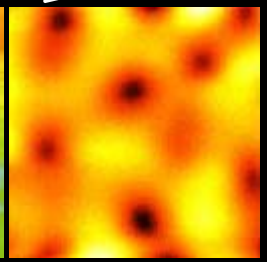


122 iron pnictide

topological insulator



metal-insulator transition



1111 iron pnictide