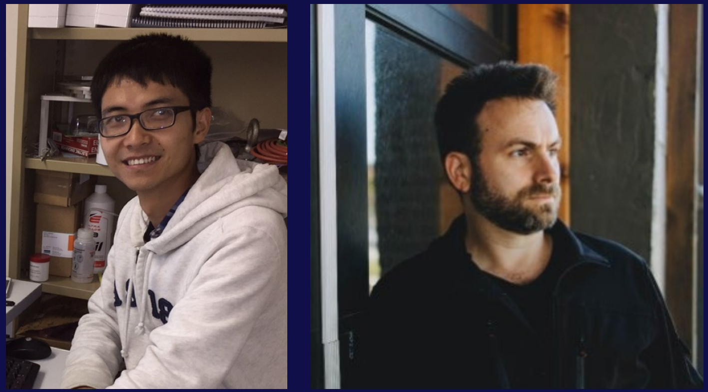
Recreating Cuprate Physics on a Silicon Platform

Hanno H. Weitering

University of Tennessee



F. Ming, X. Wu, C. Chen, K. D. Wang, P. Mai, T. A. Maier, J. Strockoz, J. W. F. Venderbos, C. Gonzalez, J. Ortega, S. Johnston, H. H. Weitering

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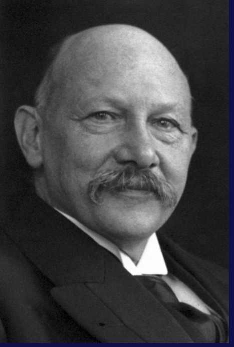
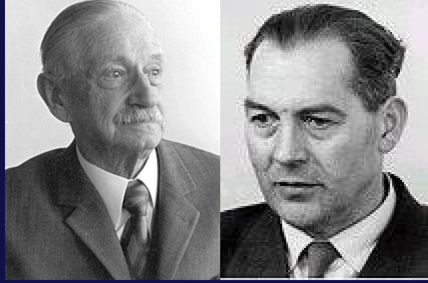
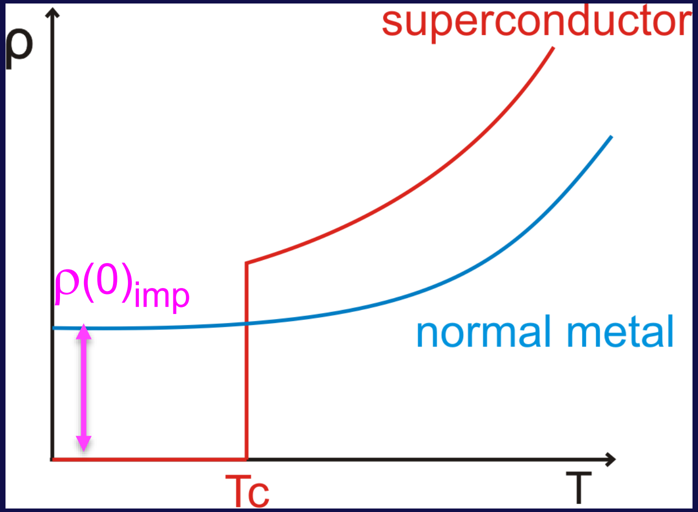
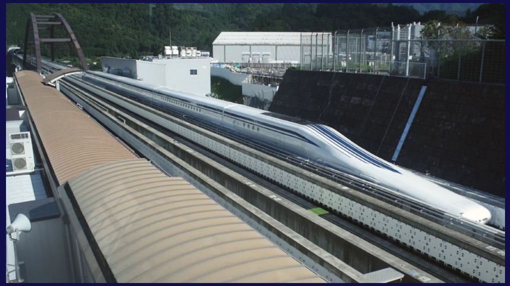


What it takes to be super…





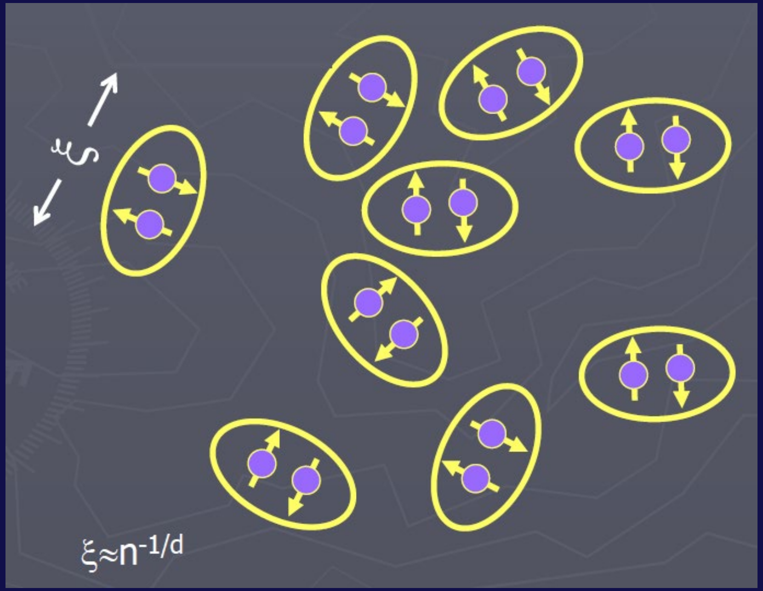


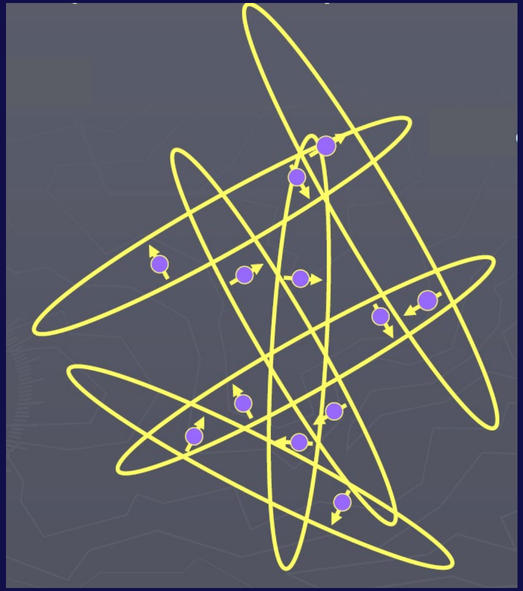
dissipationless electrical conductivity below T_c

perfect diamagnetism below T_c
(Anderson-Higgs mechanism
for ‘massive photons’)

Formation of a Cooper pair condensate

wrong picture

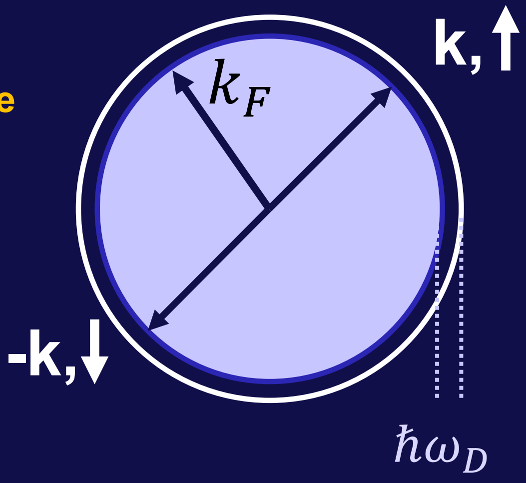


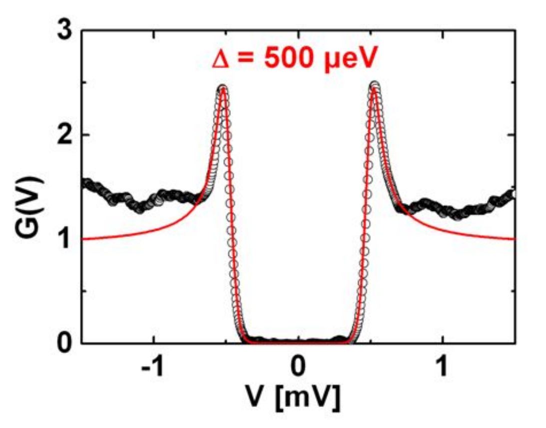


correct picture

credit P. Hirschfeld

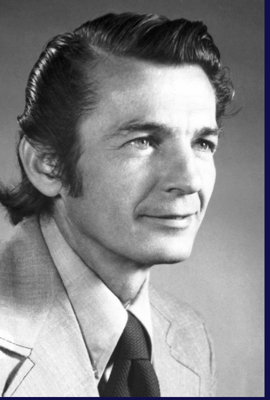
Fermi sphere



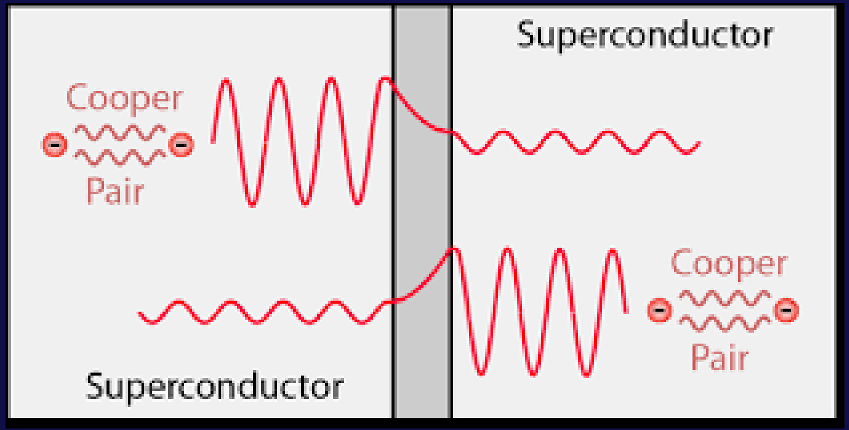


Superconducting gap

$$\Delta_0 = 2\hbar\omega_D e^{\frac{-1}{V_0 D(E_F)}}$$

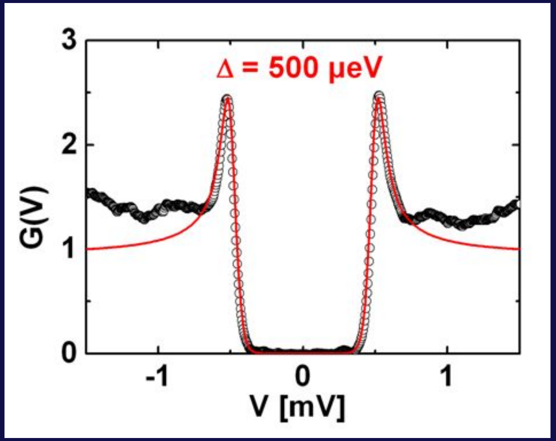


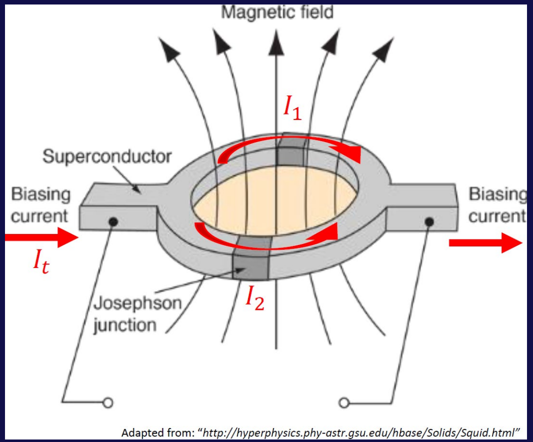
Superconducting Quantum Devices



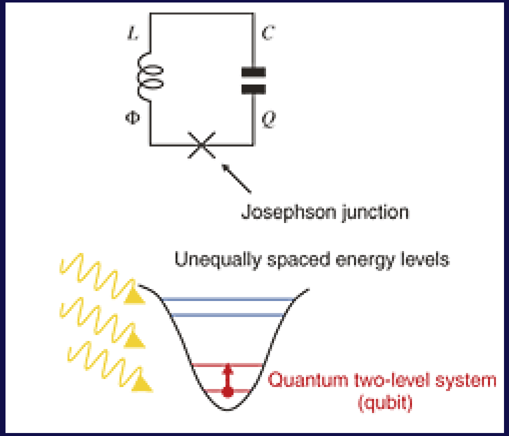
DC: $J = J_0 \sin(\theta_1 - \theta_2)$

AC: $\langle J(t) \rangle = J_c \sin[\theta(0) + \omega t]$ $\quad \omega = \frac{2e}{\hbar} V$





Adapted from: "http://hyperphysics.phy-astr.gsu.edu/hbase/Solids/Squid.html"

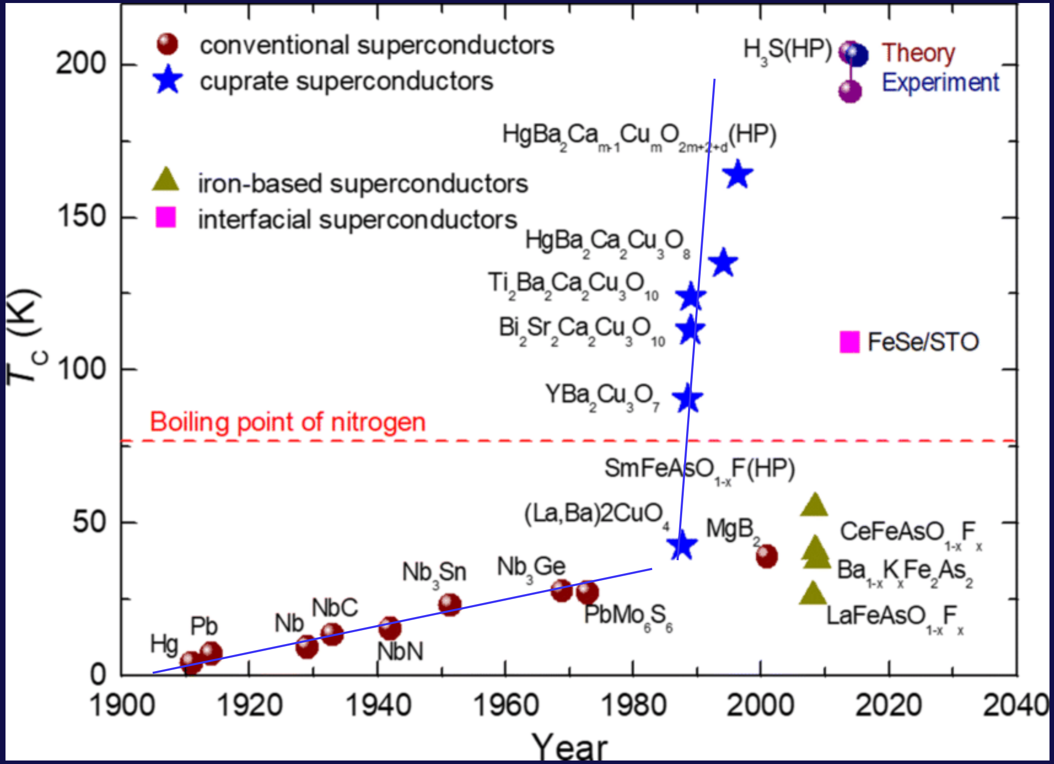


$\hbar\omega = 2eV$

1 mV ≅ 484 MHz

https://www.ntt-review.jp/archive/ntttechnical.php?contents=ntr200801sp6.html

‘The study of superconductivity is littered with disappointments, dead ends, and serendipitous discoveries’*



Article

Room-temperature superconductivity in a carbonaceous sulfur hydride

https://doi.org/10.1038/s41586-020-2801-z
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Accepted: 8 September 2020
Published online: 14 October 2020
Check for updates

Elliot Snider[1,6], Nathan Dasenbrock-Gammon[2,6], Raymond McBride[1,6], Mathew Debessai[3], Hiranya Vindana[2], Kevin Vencatasamy[2], Keith V. Lawler[4], Ashkan Salamat[5] & Ranga P. Dias[1,2]✉

One of the long-standing challenges in experimental physics is the observation of room-temperature superconductivity[1,2]. Recently, high-temperature conventional superconductivity in hydrogen-rich materials has been reported in several systems under high pressure[3–5]. An important discovery leading to room-temperature superconductivity is the pressure-driven disproportionation of hydrogen sulfide (H_2S) to H_3S, with a confirmed transition temperature of 203 kelvin at 155 gigapascals[3,6]. Both H_2S and CH_4 readily mix with hydrogen to form guest–host structures at lower pressures[7], and are of comparable size at 4 gigapascals. By introducing methane at low pressures into the $H_2S + H_2$ precursor mixture for H_3S, molecular exchange is allowed within a large assemblage of van der Waals solids that are hydrogen-rich with H_2 inclusions; these guest–host structures become the building blocks of superconducting compounds at extreme conditions. Here we report superconductivity in a photochemically transformed carbonaceous sulfur hydride system, starting from elemental precursors, with a maximum superconducting transition temperature of 287.7 ± 1.2 kelvin (about 15 degrees Celsius) achieved at 267 ± 10 gigapascals. The superconducting state is observed over a broad pressure range in the diamond anvil cell, from 140 to 275 gigapascals, with a sharp upturn in transition temperature above 220 gigapascals. Superconductivity is established by the observation of zero resistance, a magnetic susceptibility of up to 190 gigapascals, and reduction of the transition temperature under an external magnetic field of up to 9 tesla, with an upper critical magnetic field of about 62 tesla according to the Ginzburg–Landau model at zero temperature. The light, quantum nature of hydrogen limits the structural and stoichiometric determination of the system by X-ray scattering techniques, but Raman spectroscopy is used to probe the chemical and structural transformations before metallization. The introduction of chemical tuning within our ternary system could enable the preservation of the properties of room-temperature superconductivity at lower pressures.

In the past decade there has been an emergence of interest in the discovery of materials relevant to room-temperature superconductivity. Extreme pressure has already been proven to be the most versatile order parameter because it facilitates the production of new quantum materials with unique stoichiometries and a mechanism for pressure-induced metallization[8–10]. This has been most essential for non-metallic starting materials[11–13]. All systems with high superconducting critical temperature ($T_c > 200$ K) that have been accessed under pressure so far are hydrogen-rich materials, in which the superconductivity is driven by strong electron–phonon coupling to high-frequency hydrogen phonon modes[14,15]. However, the specific stoichiometry (that is, XH_n) does not seem to be as critical as having a hydrogen-rich chemical environment that mimics the properties (electron density near/at the Fermi surface and high-frequency phonon modes) of idealized pure metallic hydrogen[16–19]. This is highlighted by the difference between purely covalent systems such as H_3S compared to metal hydride systems. The most recent example of a metal hydride is lanthanum hydride ($LaH_{10\pm x}$), which has $T_c = 250$–260 K at 180–200 GPa (refs. [4,5]). A lanthanum ‘superhydride’ has been experimentally realized, although a precise determination of its stoichiometry is lacking, as are the tools to determine such parameters. Investigation of the predicted band structures of rare-earth (La and Y) superhydrides implies an ionic heavy atom that donates its valence electrons to the hydrogen network, stabilizing a clathrate-like hydrogen cage structure[20–22]. Despite the large number of theoretical predictions for possible hydrogen-rich materials at high pressure, very few demonstrate superconducting behaviour at

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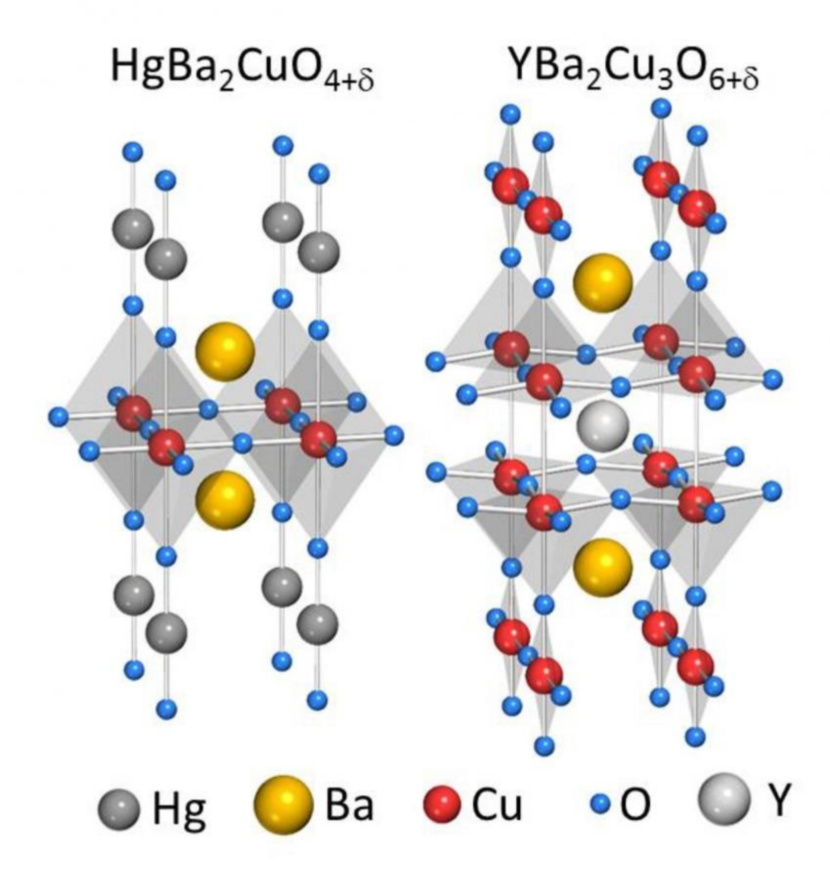
*Prof. Antia Botana, ASU

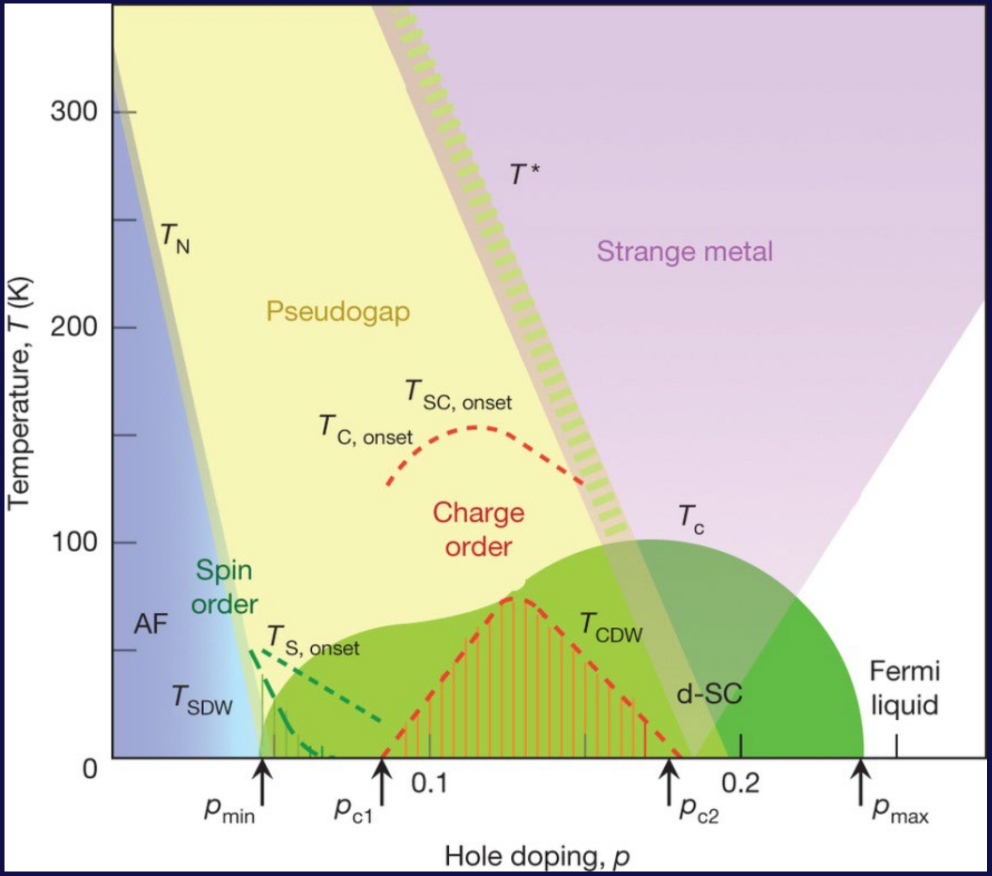
Outline

- Background & overview
- **Recreating cuprate physics**: Triangular tin lattice on a silicon template
- Modulation doping and evidence for Mott physics
- Superconducting properties
- Time reversal symmetry breaking and d-wave pairing
- Conclusions and outlook

- Electron doping and charge ordering

High temperature superconductors

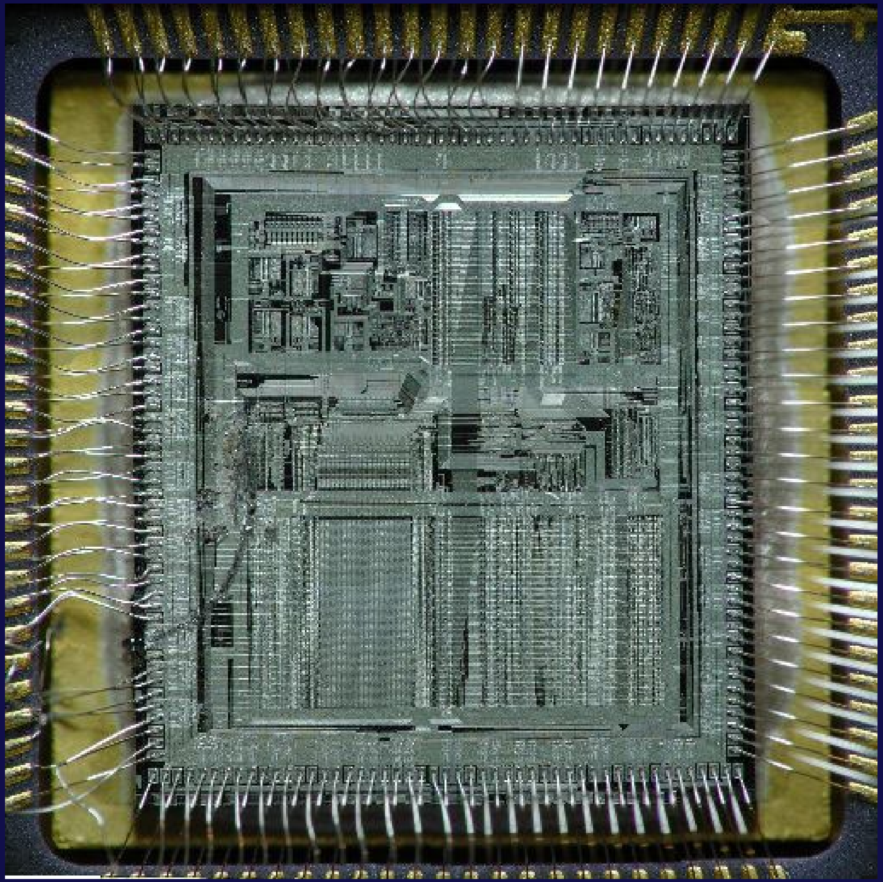


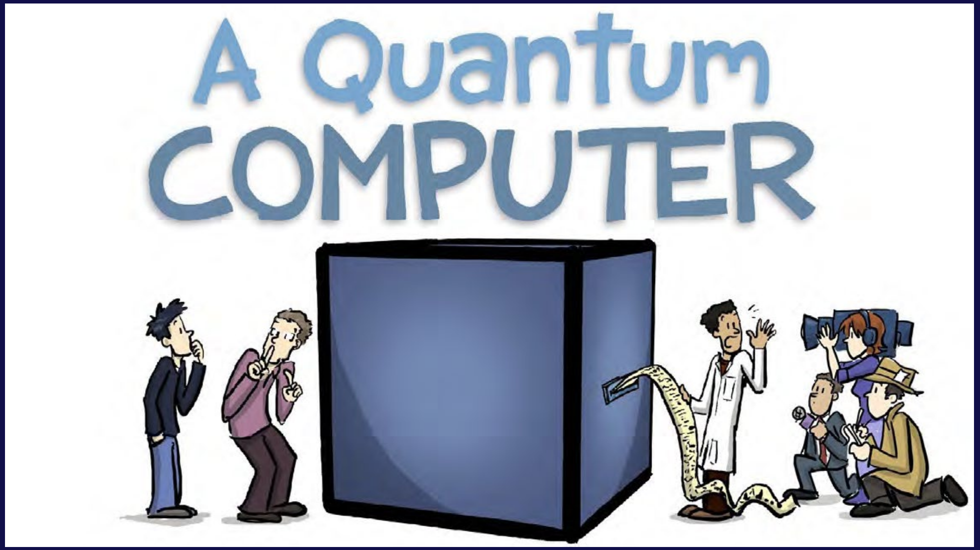


- **Doped Mott insulators**
- **Quasi 2D phenomenon**

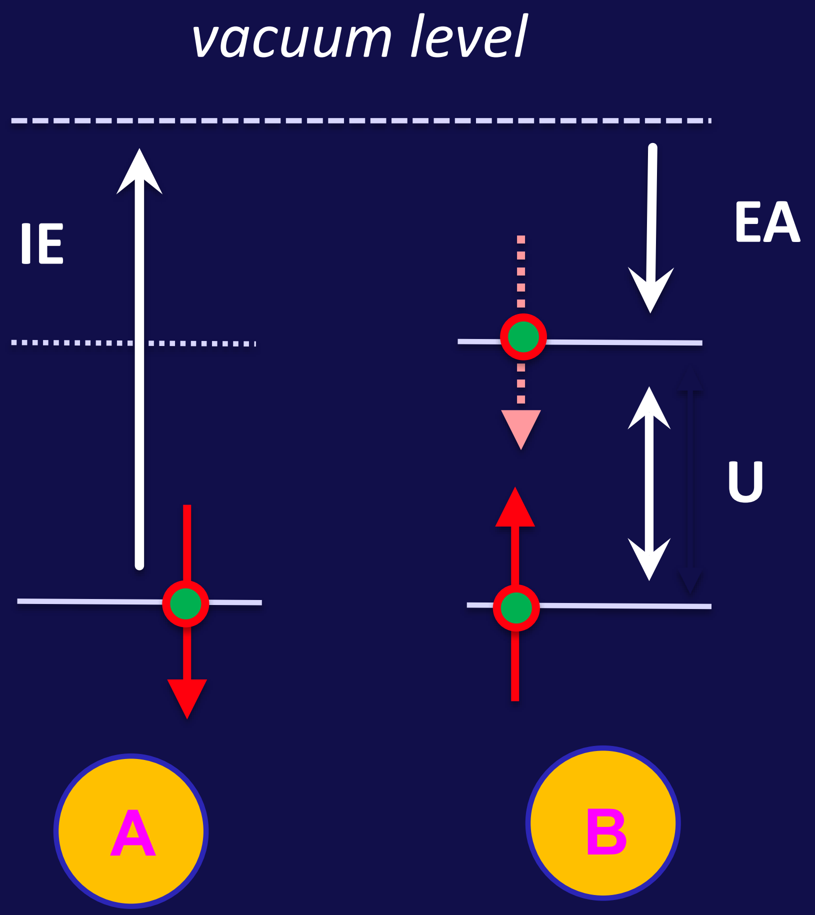
Keimer et al. Nature '15

a dream…..

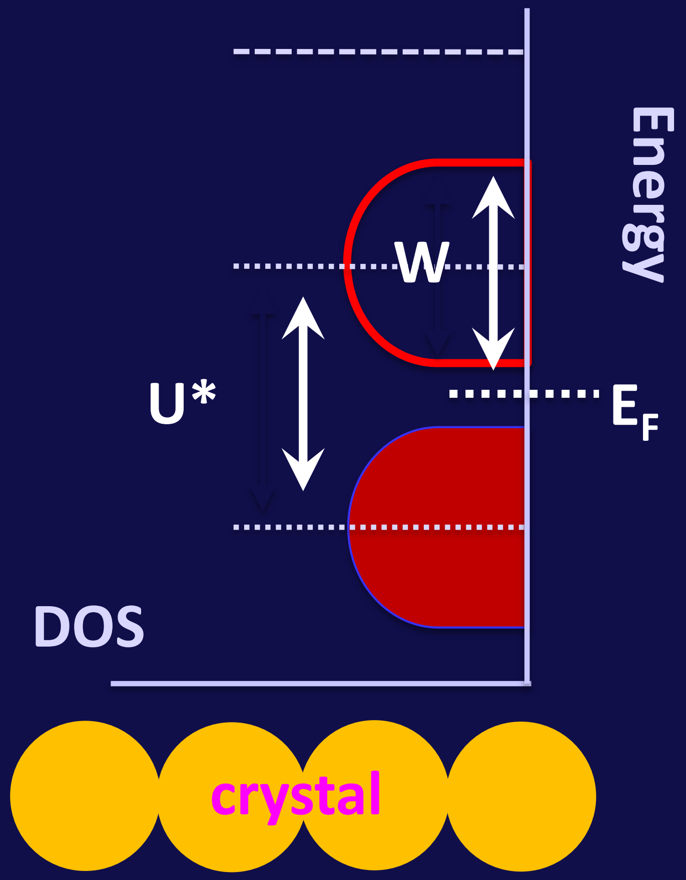




Mott Physics 101



atomic limit

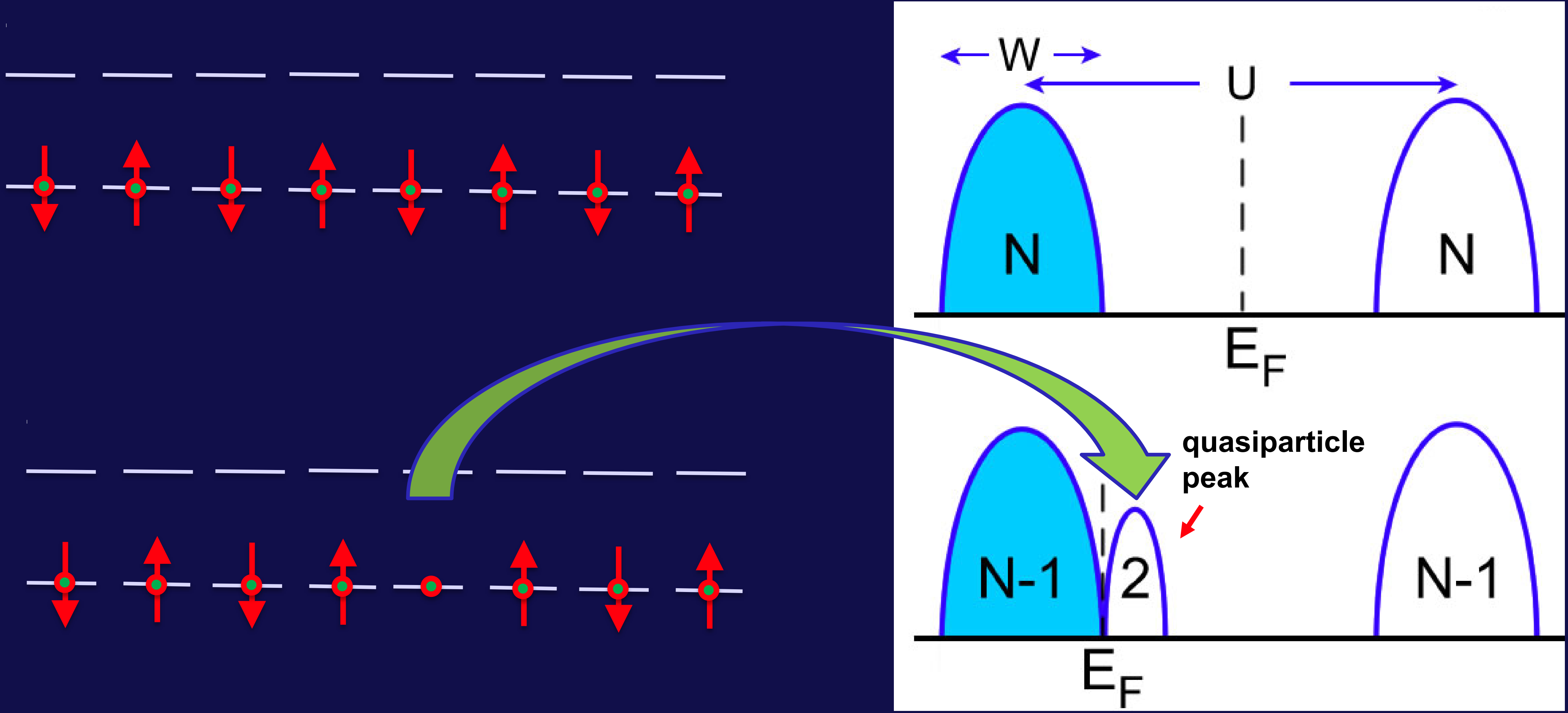


condensed matter

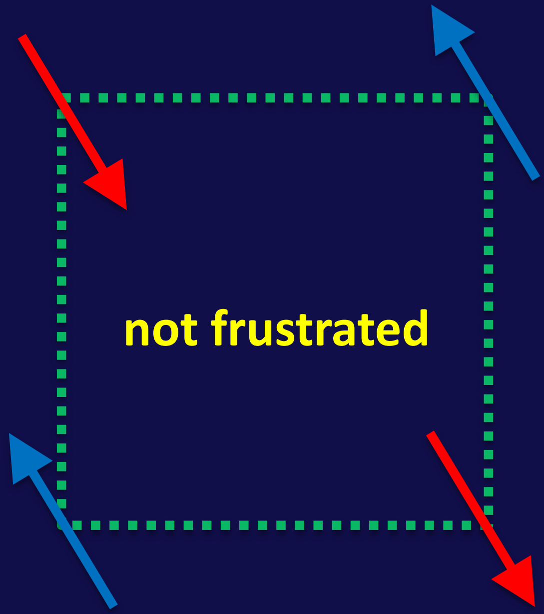
- Insulator – metal (Mott) transition when $U^* \cong W$

Doping a Mott Insulator

M. B. J. Meinders, H. Eskes, and G. A. Sawatzky,
Phys. Rev. B 48, 3916 (1993).

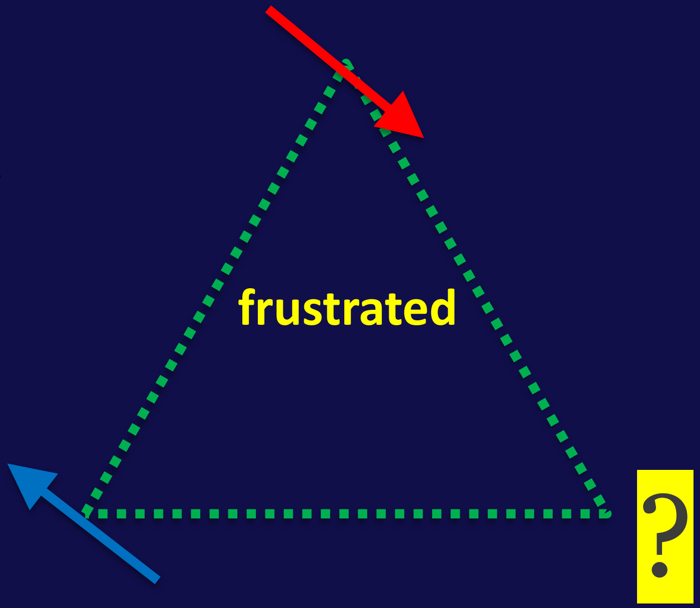


2D Spin 1/2 Antiferromagnet



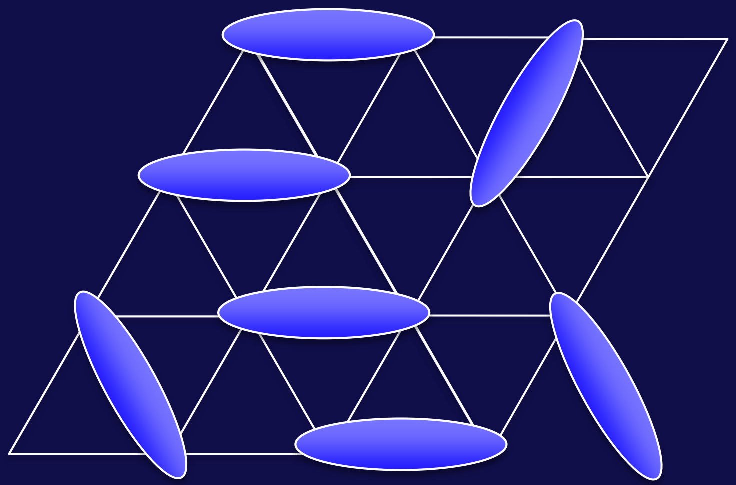
$$\hat{H} = 1/2 \sum_{i,j} J_{ij} \hat{S}_i \hat{S}_j$$

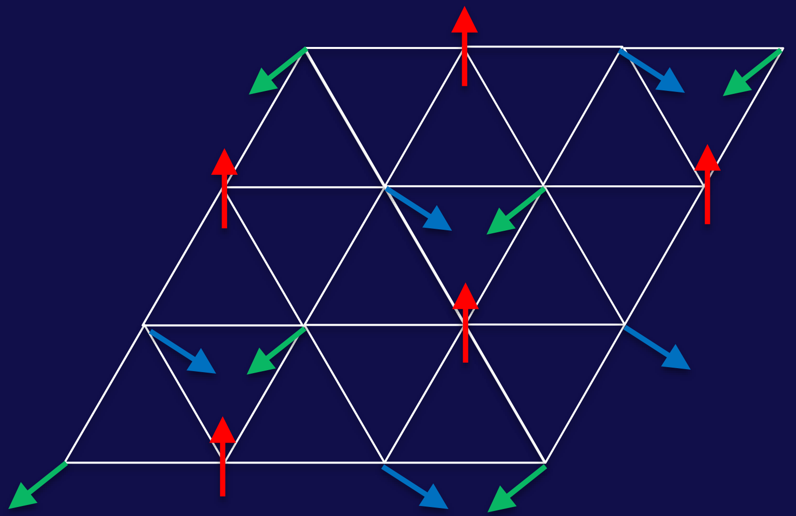
$$J_{ij} = \frac{4t_{ij}^{\;2}}{U}$$

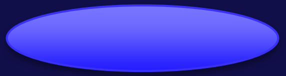


Frustration: spin pairs cannot all be simultaneously
in the lowest energy configuration

2D quantum spin liquid versus classical Neel order







$= 1/\sqrt{2}\,(\uparrow\downarrow - \downarrow\uparrow)$

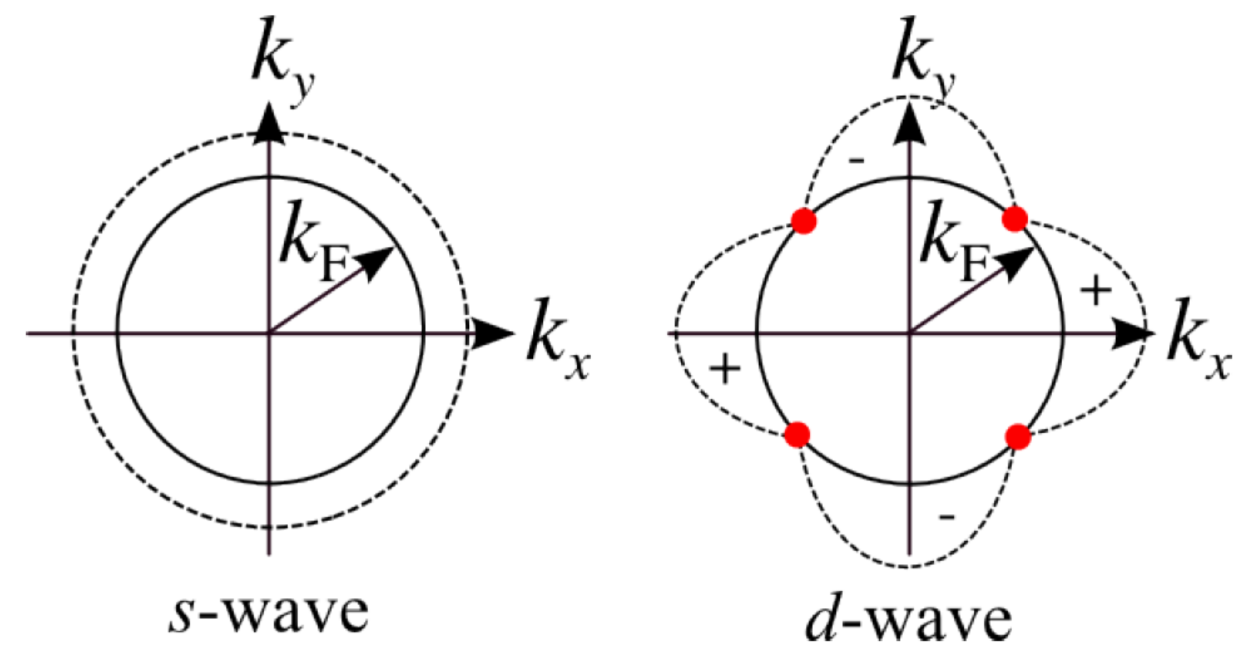
Classical Neel long-range order (120°) for large U/W

RVB: frustration, low spin, low dimensionality

Metallization or doping: route to high Tc superconductivity?

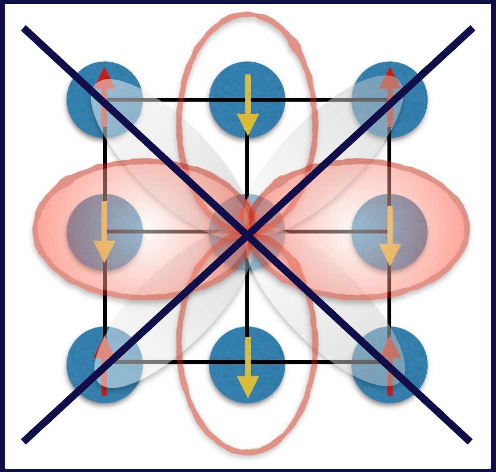
Order parameter symmetry

$$\Psi = \Delta_0 e^{i\varphi}$$



s-wave

d-wave



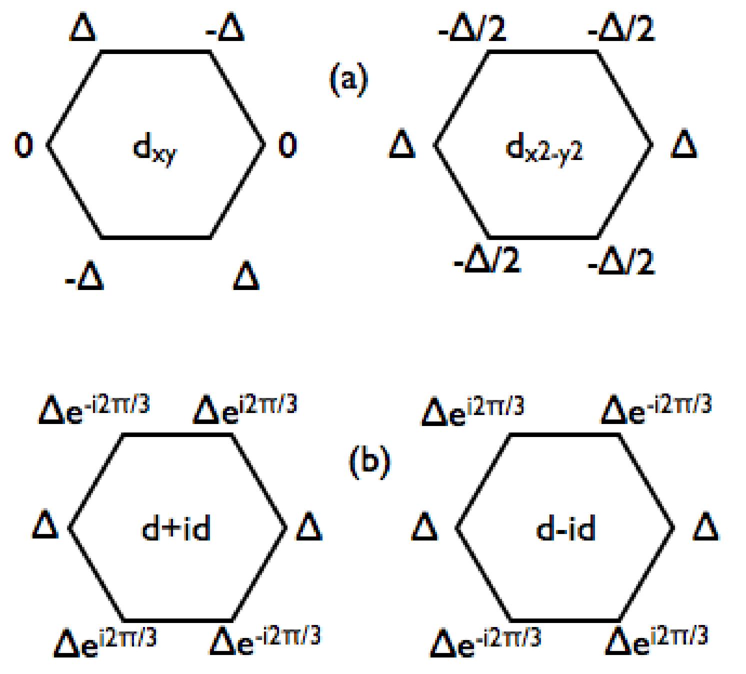
$$\Delta_{\mathbf{k}} = -\frac{1}{N}\sum_{\mathbf{k}'}\frac{V_{\mathbf{k}\mathbf{k}'}\Delta_{\mathbf{k}'}}{2E_{\mathbf{k}'}}\tanh\left(\frac{E_{\mathbf{k}'}}{2k_BT}\right)$$

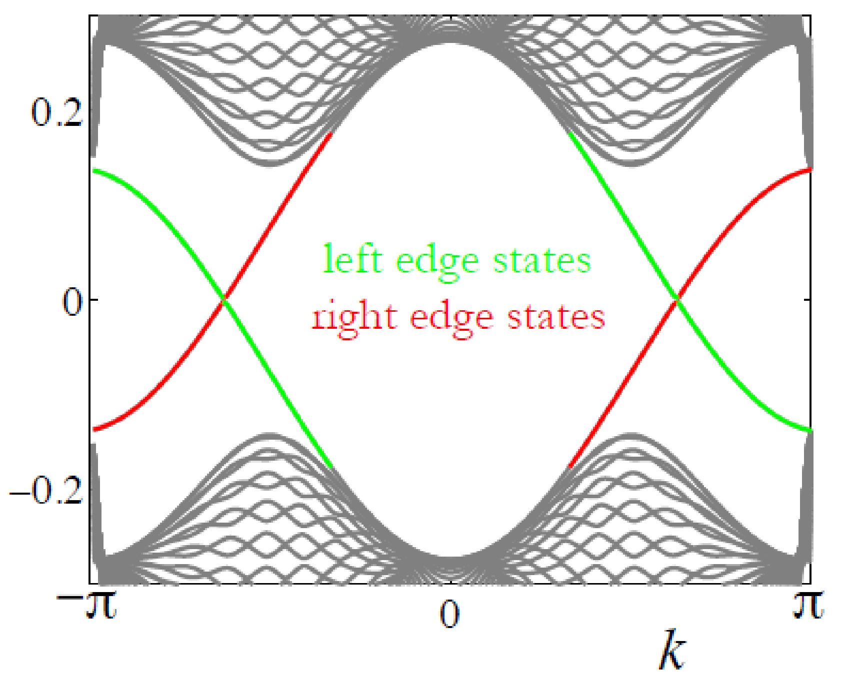
conventional
Sn, Pb, MgB_2….
phonon mediated

unconventional
high-Tc cuprates
strong Coulomb repulsion

‘Exotic’ chiral d-wave order parameter

A.M. Black-Schaffer et al., JPCM 26, 423201 (2014)



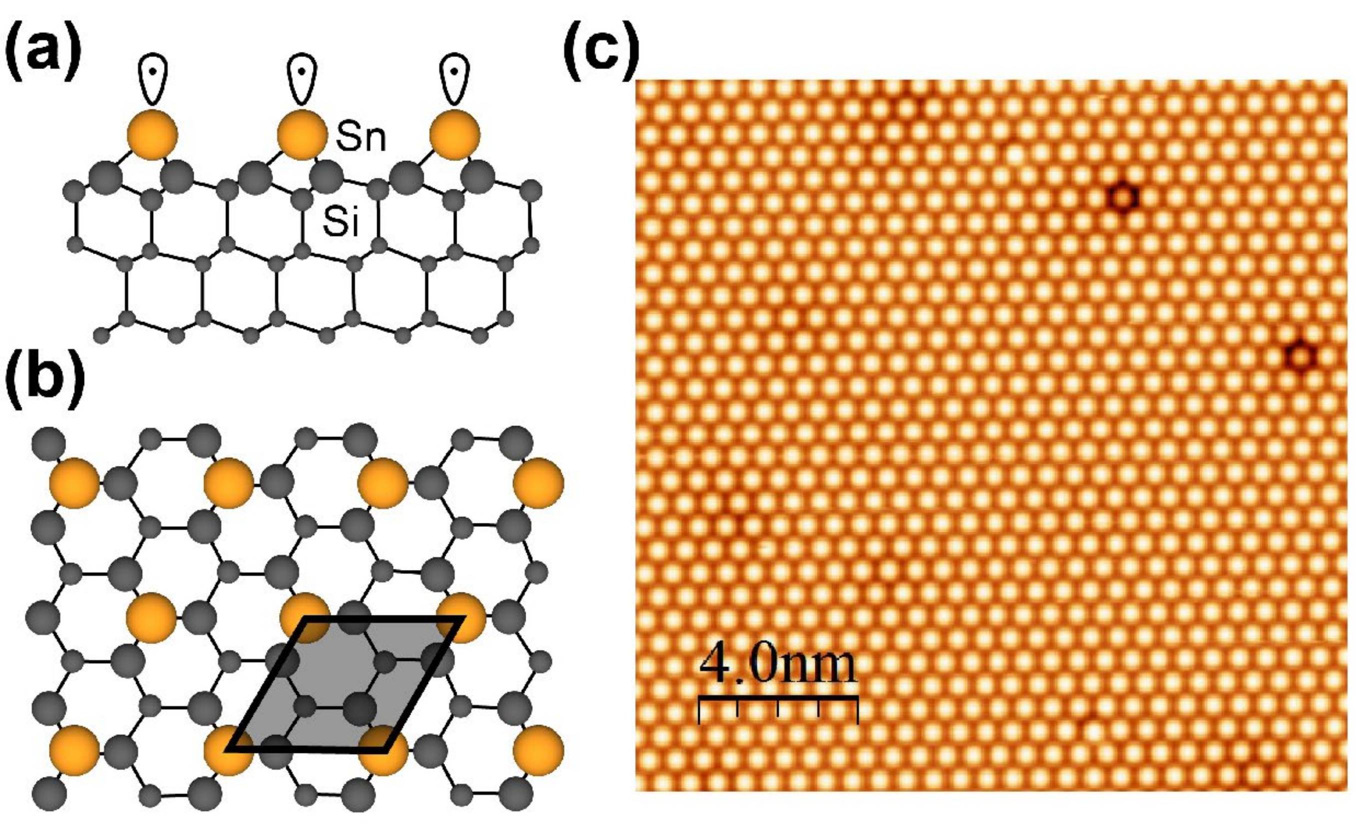


- - triangular lattice
- - d_{xy} and d_{x2-y2} order parameters 90° out of phase
- - broken TRS

- topology set by Chern/winding number of
- order parameter
- d+id wave winds twice around Γ (N = 2)
- 2 chiral copropagating edge states per edge

- $Na_xCoO_2 \cdot yH_2O$, hole-doped graphene, SrPtAs,…….

Submonolayer of Sn on Si(111)



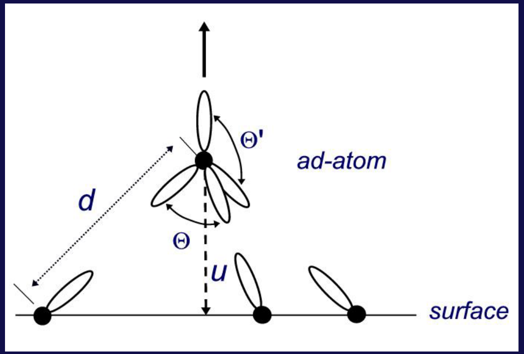
$(\sqrt{3} \times \sqrt{3})$ superlattice at 1/3 monolayer of Sn

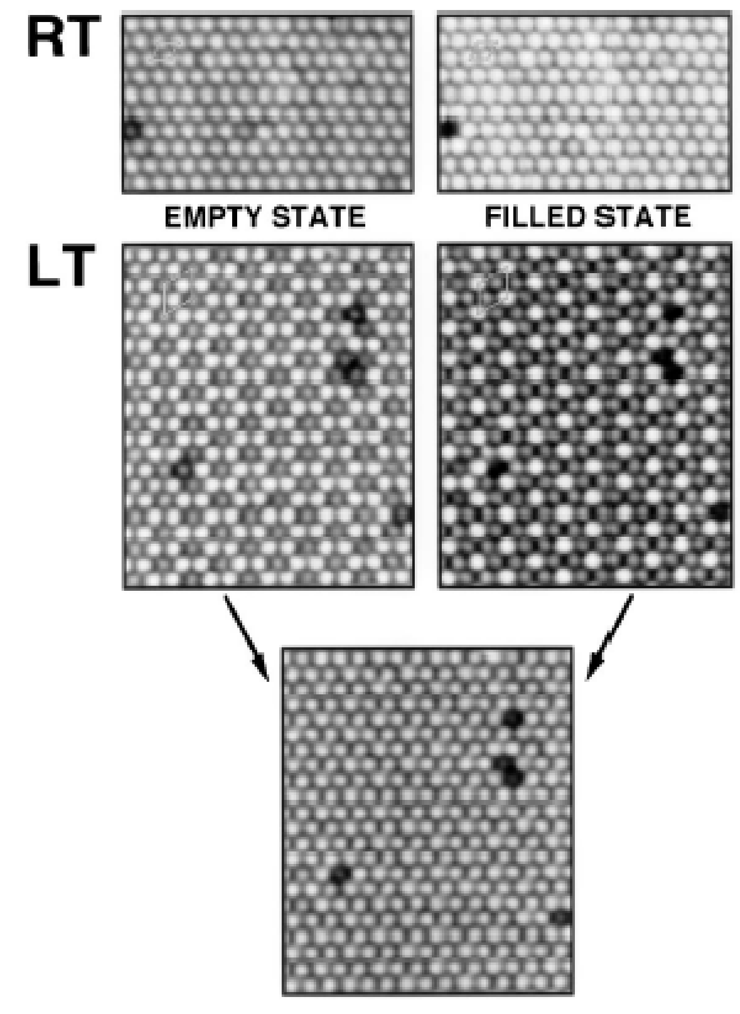
Single-band Mott insulator (T < 100 K)

Close realization of a spin ½ triangular lattice antiferromagnetic Heisenberg system

Potential for chiral superconductivity with doping $(\Delta_1 \pm i\Delta_2)$

Competing phases





3x3 charge ordered metal

3x3 charge ordered insulator

e.g., **Sn on Ge(111)**

Pb on Si(111) or Ge(111)

J.M. Carpinelli *et al*., Nature **381**, 398 (1996)
and PRL 79, 2859 (1997)

R. Cortes et al, PRB **88**,125113 (2013)

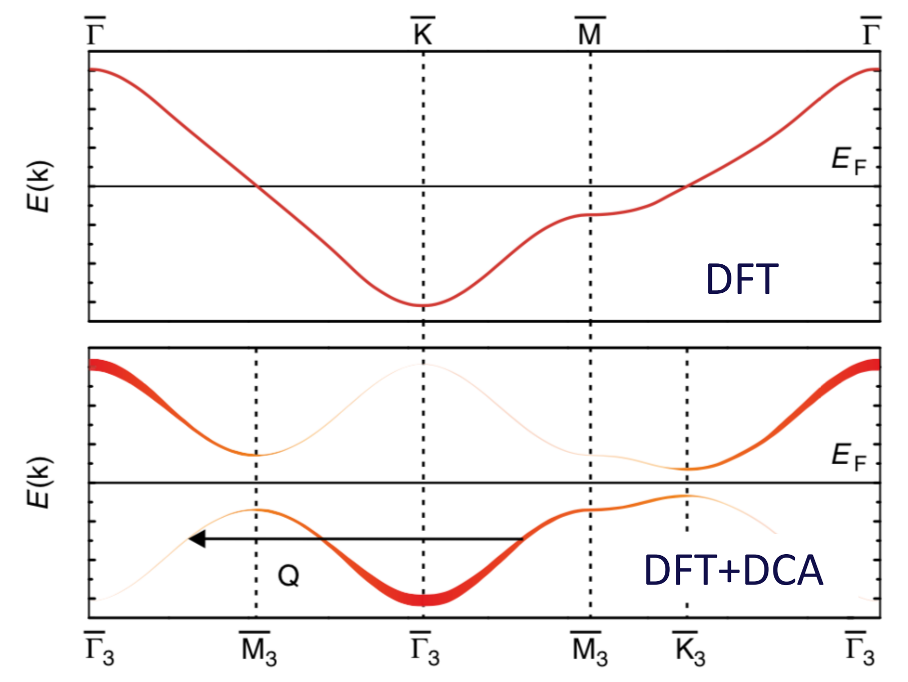
ARTICLE

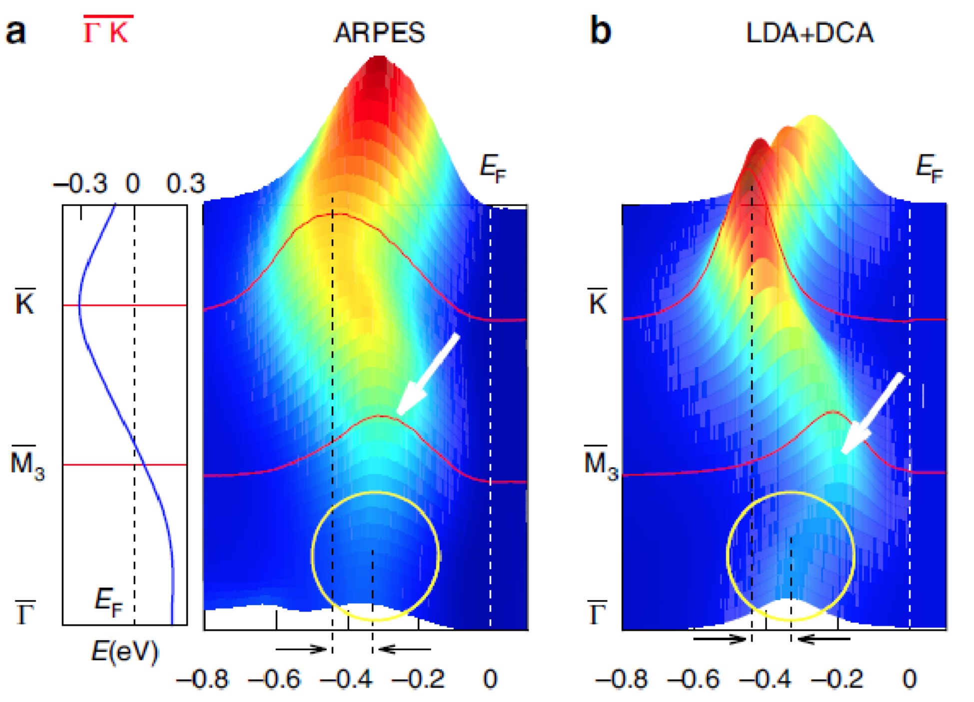
Received 26 Oct 2012 | Accepted 20 Feb 2013 | Published 27 Mar 2013

DOI: 10.1038/ncomms2617

Magnetic order in a frustrated two-dimensional atom lattice at a semiconductor surface

Gang Li[1], Philipp Höpfner[2], Jörg Schäfer[2], Christian Blumenstein[2], Sebastian Meyer[2], Aaron Bostwick[3], Eli Rotenberg[3], Ralph Claessen[2] & Werner Hanke[1]





Sn/Si(111)

ARTICLE

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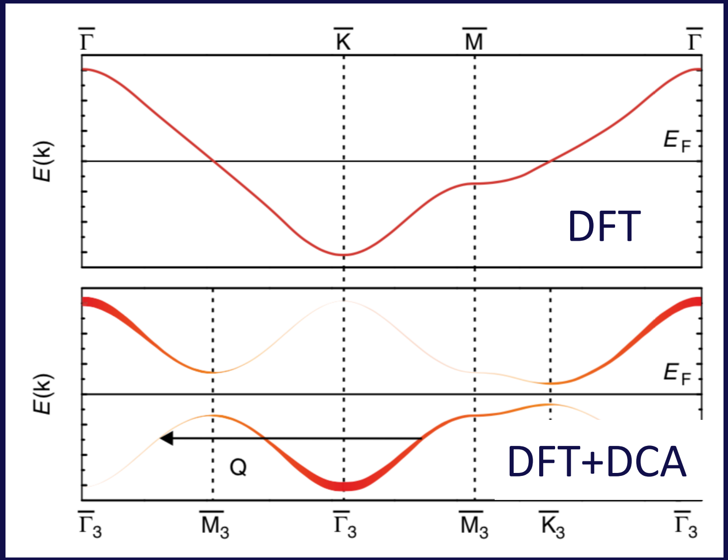
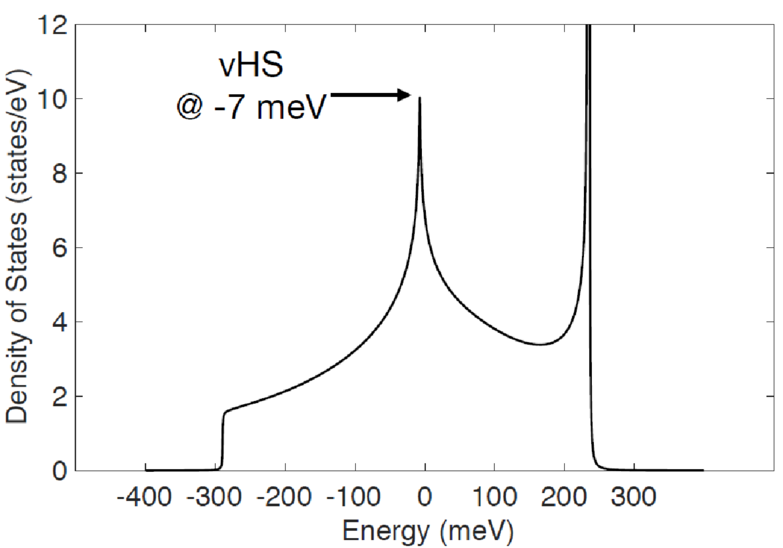
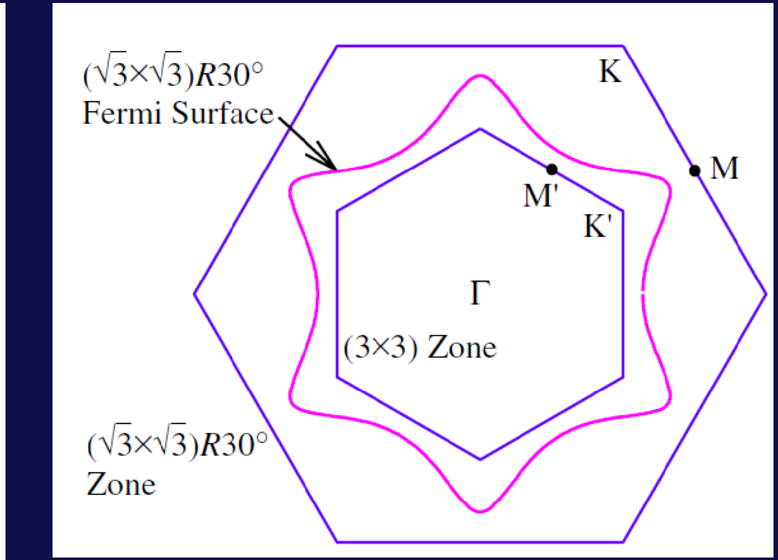


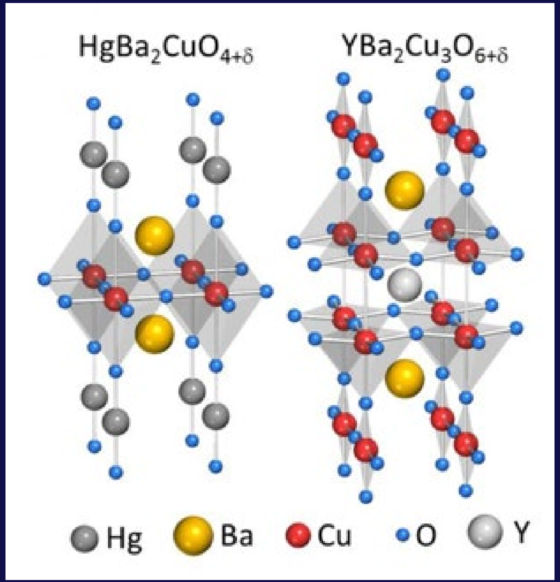




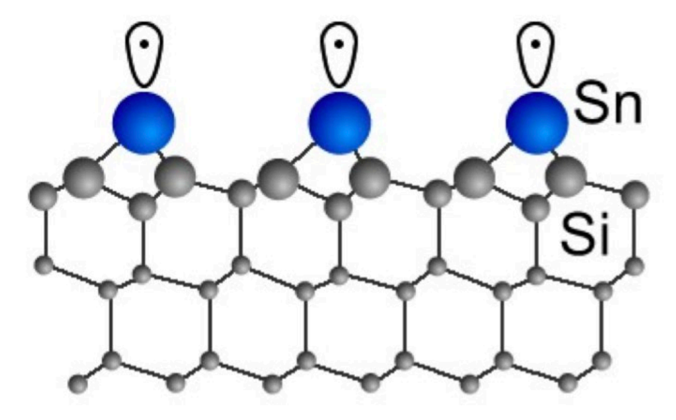
Why is this system interesting: direct access to Mottness!

Cuprates:
Complex
Oxides

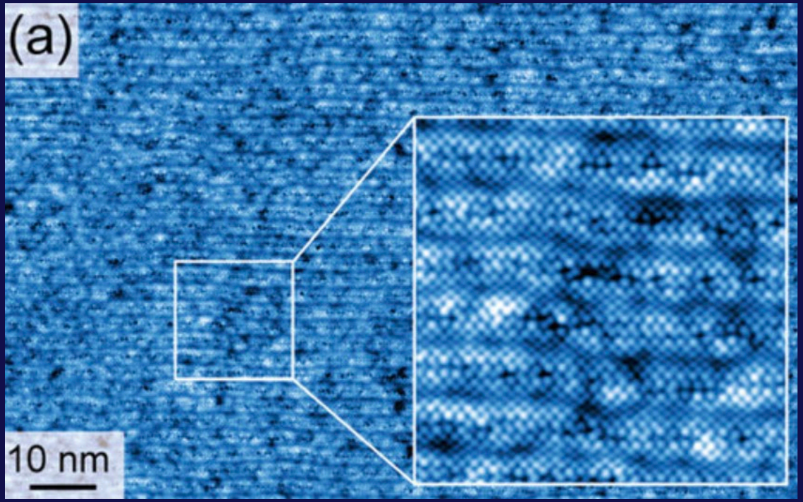
Cuprates,
Complex
oxides



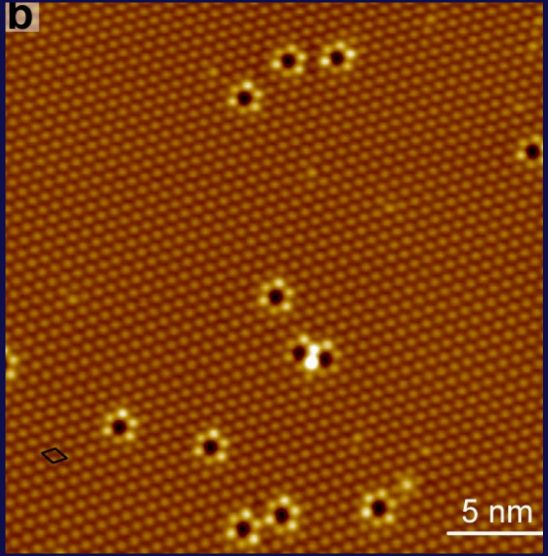
Sn/Si(111)



Sn/Si(111)

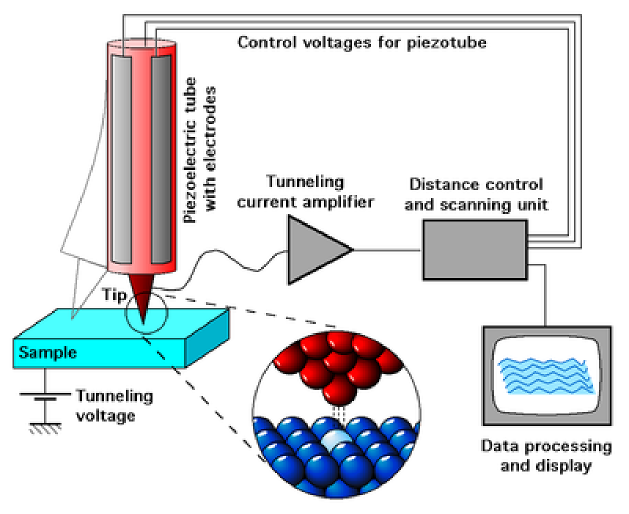


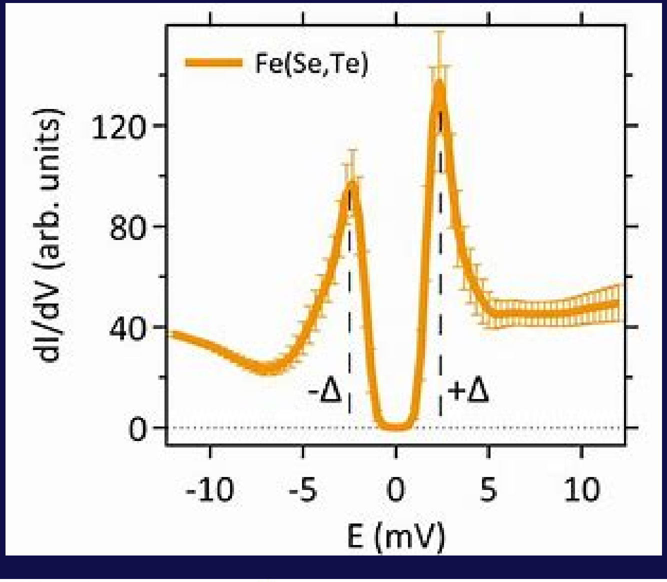
*J.-O. Jung *et al.*, Rev. Sci. Inst. **88**, 103702 (2017).



Scanning Tunneling Microscopy and Spectroscopy

STM

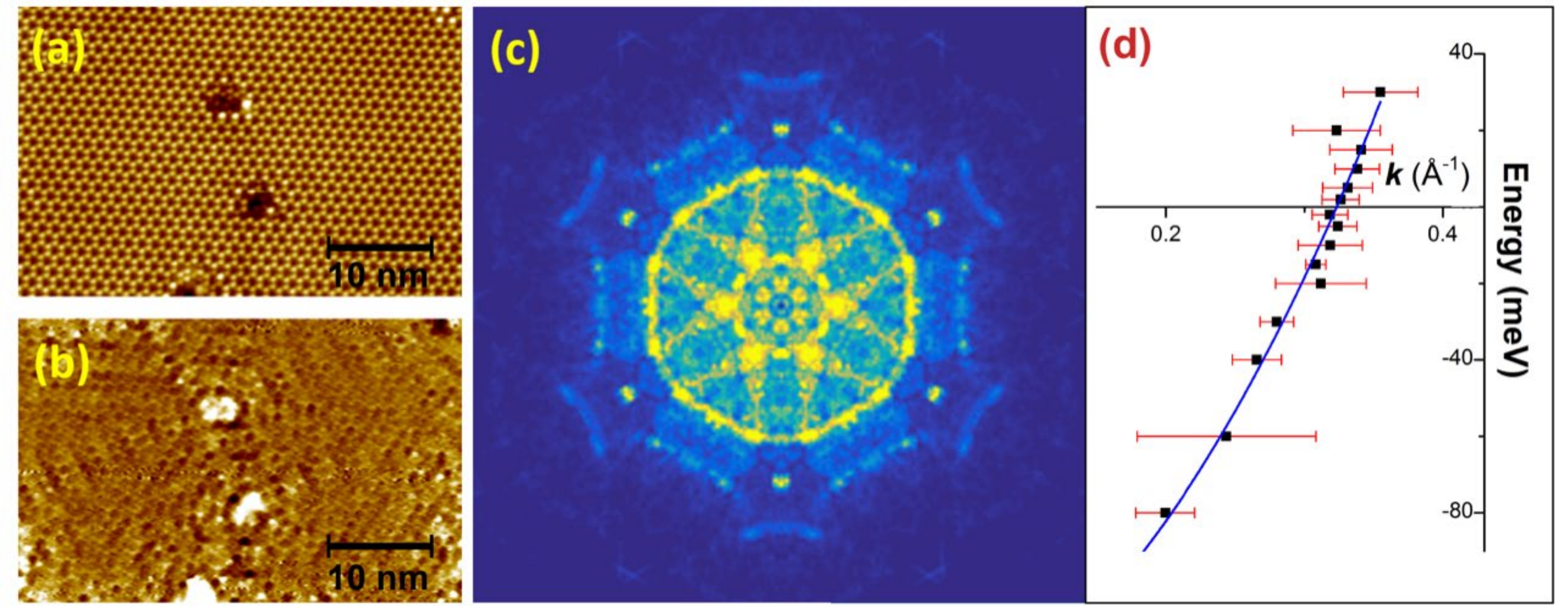




STS

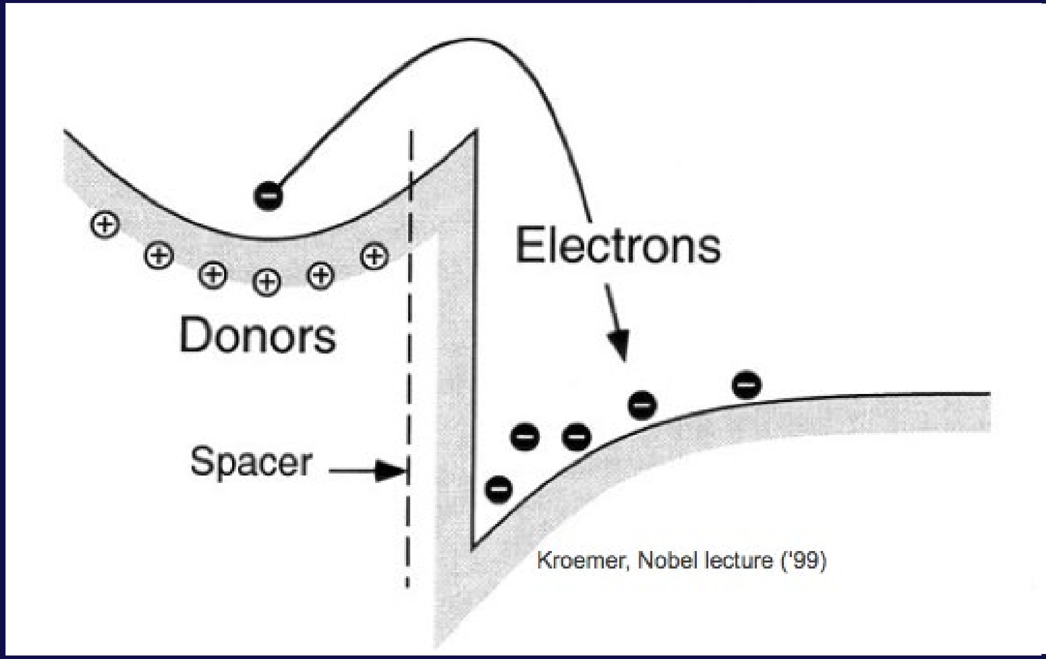
$dI/dV \propto$ LDOS

QPI

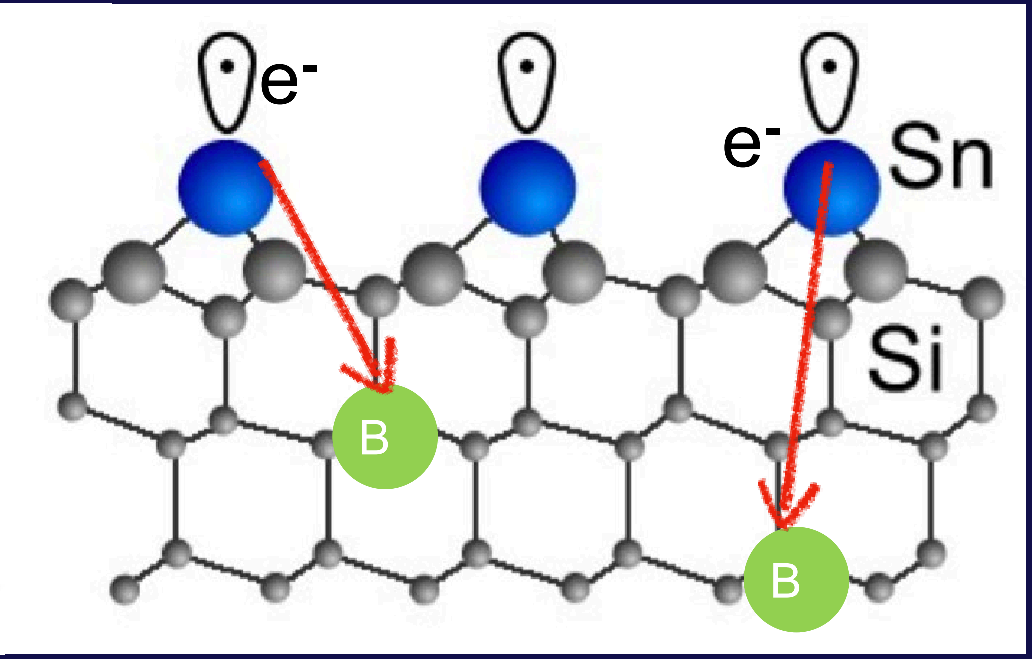


Doping the Sn terminated Si(111) surface

Modulation Doping Scheme



Kroemer, Nobel lecture ('99)



Material A

Material B

Heavily B-doped (p-type) Si substrates

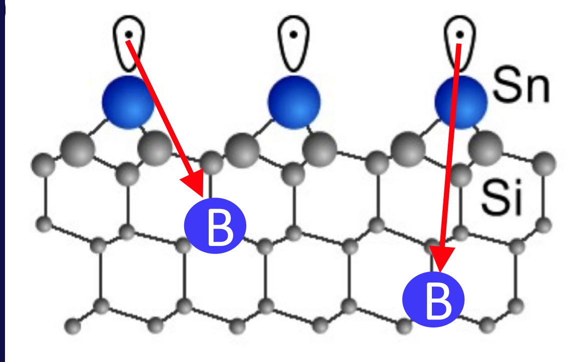
Ming et al., PRL **119**, 266802 (2017)

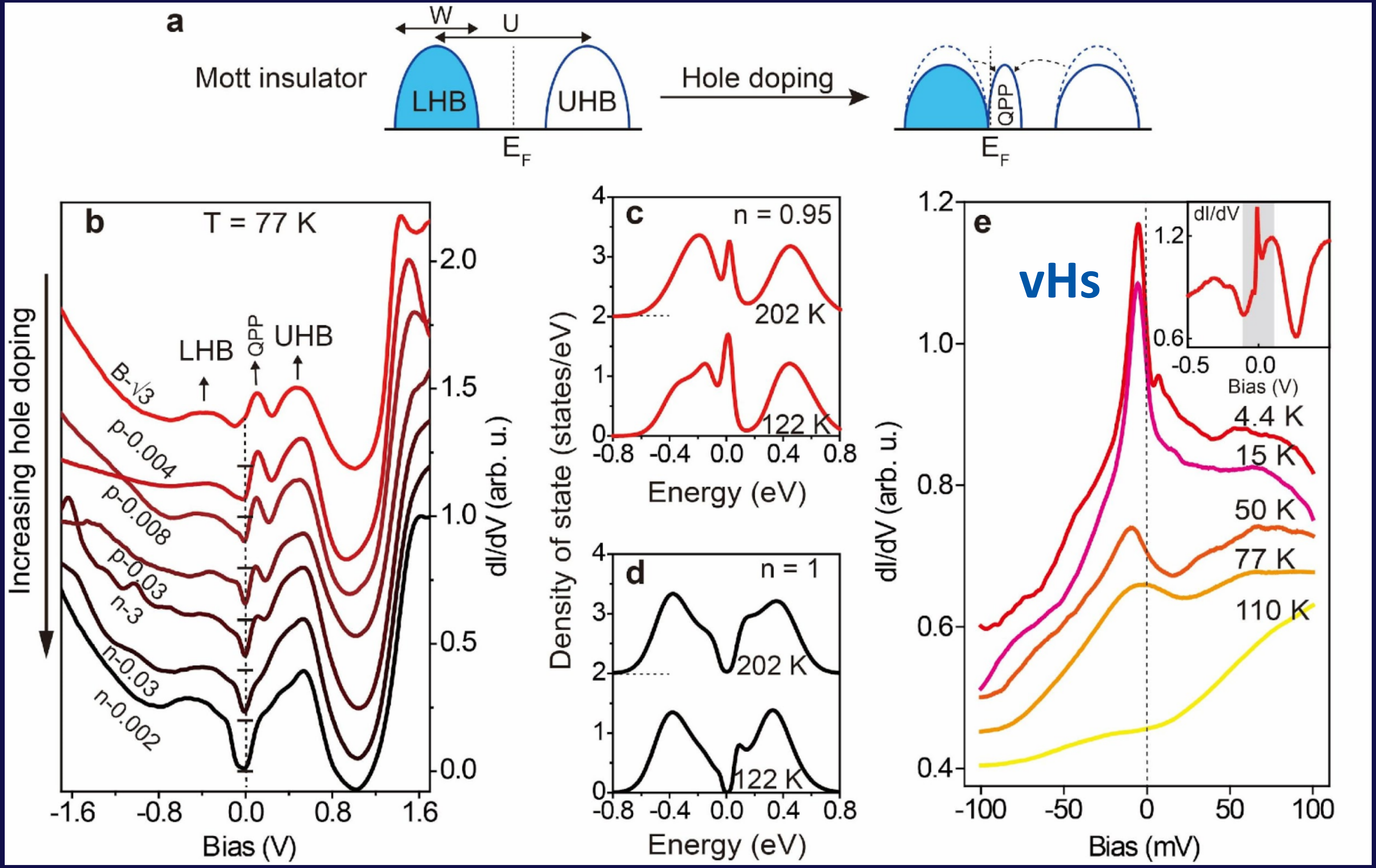
Doped Mott insulator, spectral weight transfer and *van Hove* singularity

U $\cong$ 0.6 – 0.7 eV

W $\cong$ 0.6 eV

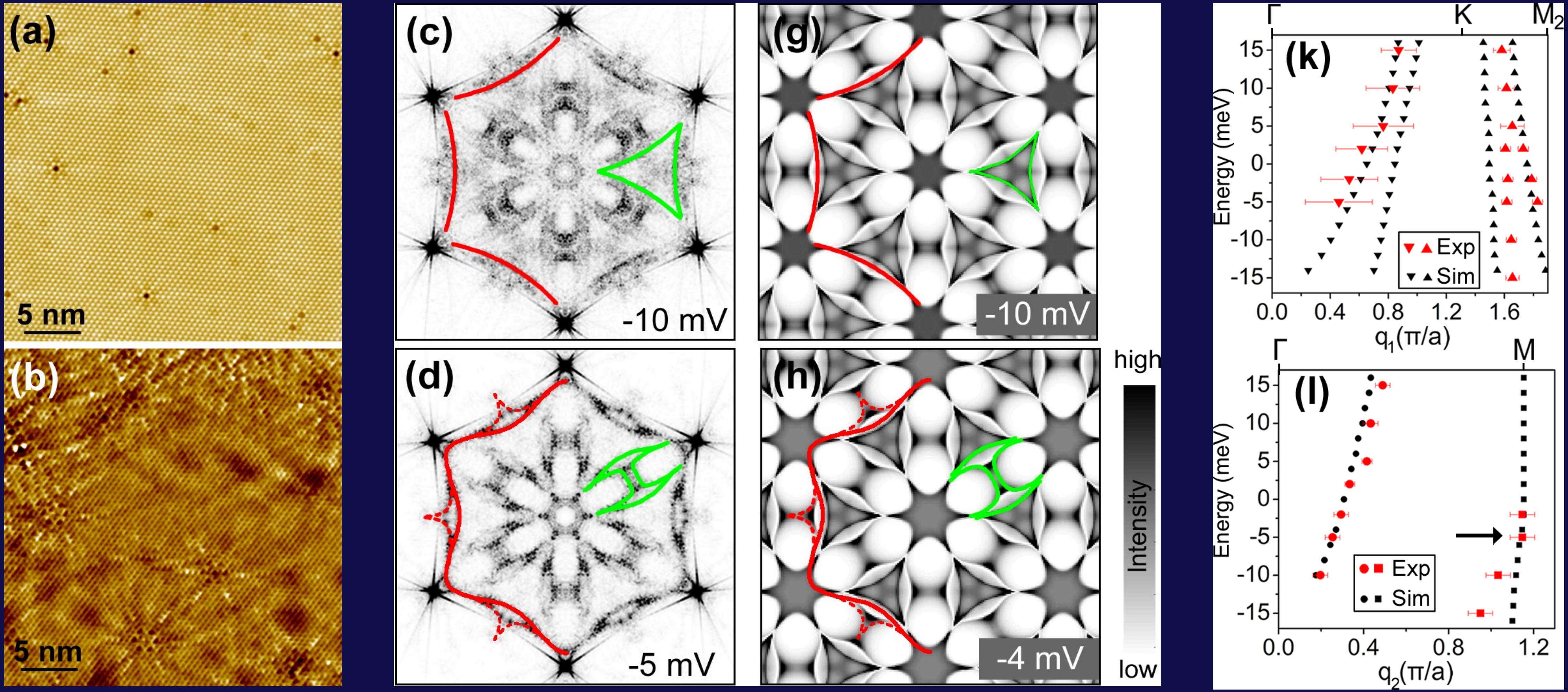
10% hole doping
(p = 0.1)





Ming et al., PRL 199, 266802 (2017)

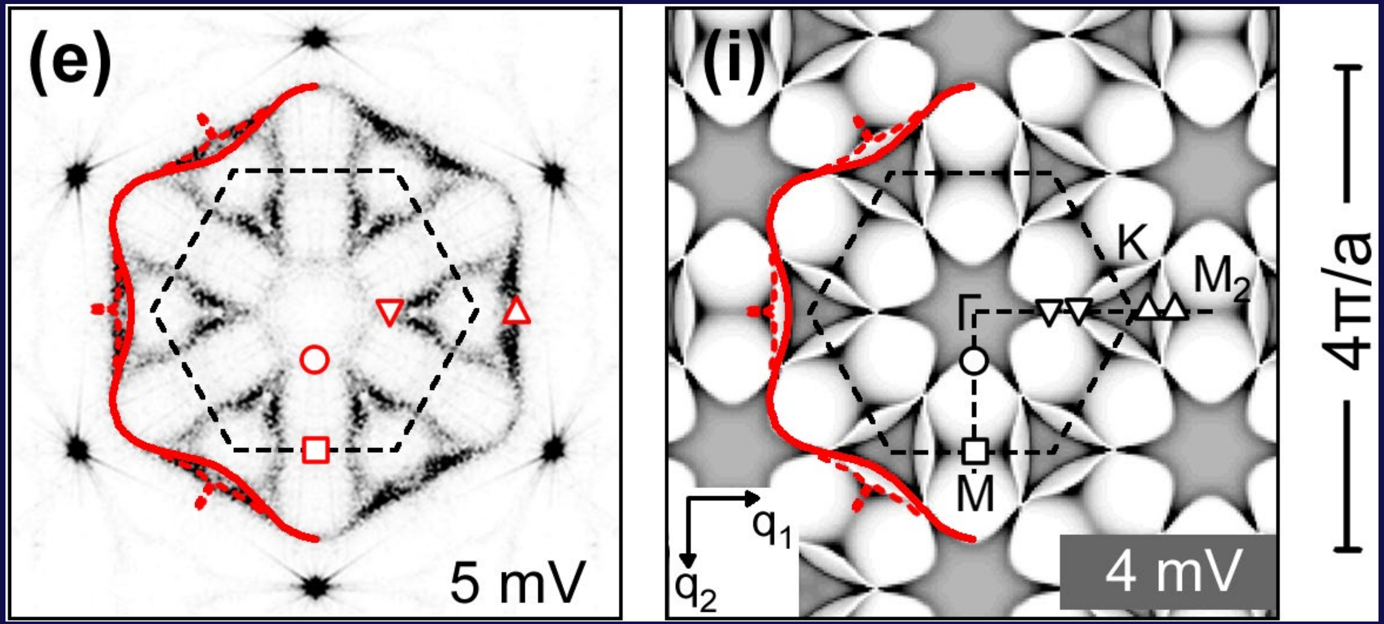
Quasi Particle Interference Imaging Sn on Si(111)



Ming et al., PRL 199, 266802 (2017)

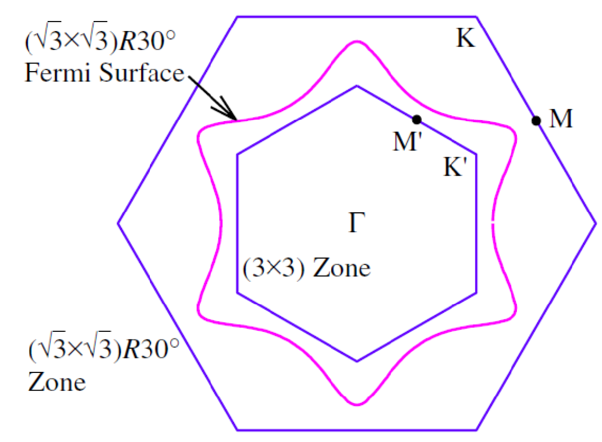
Fermi surface

QPI

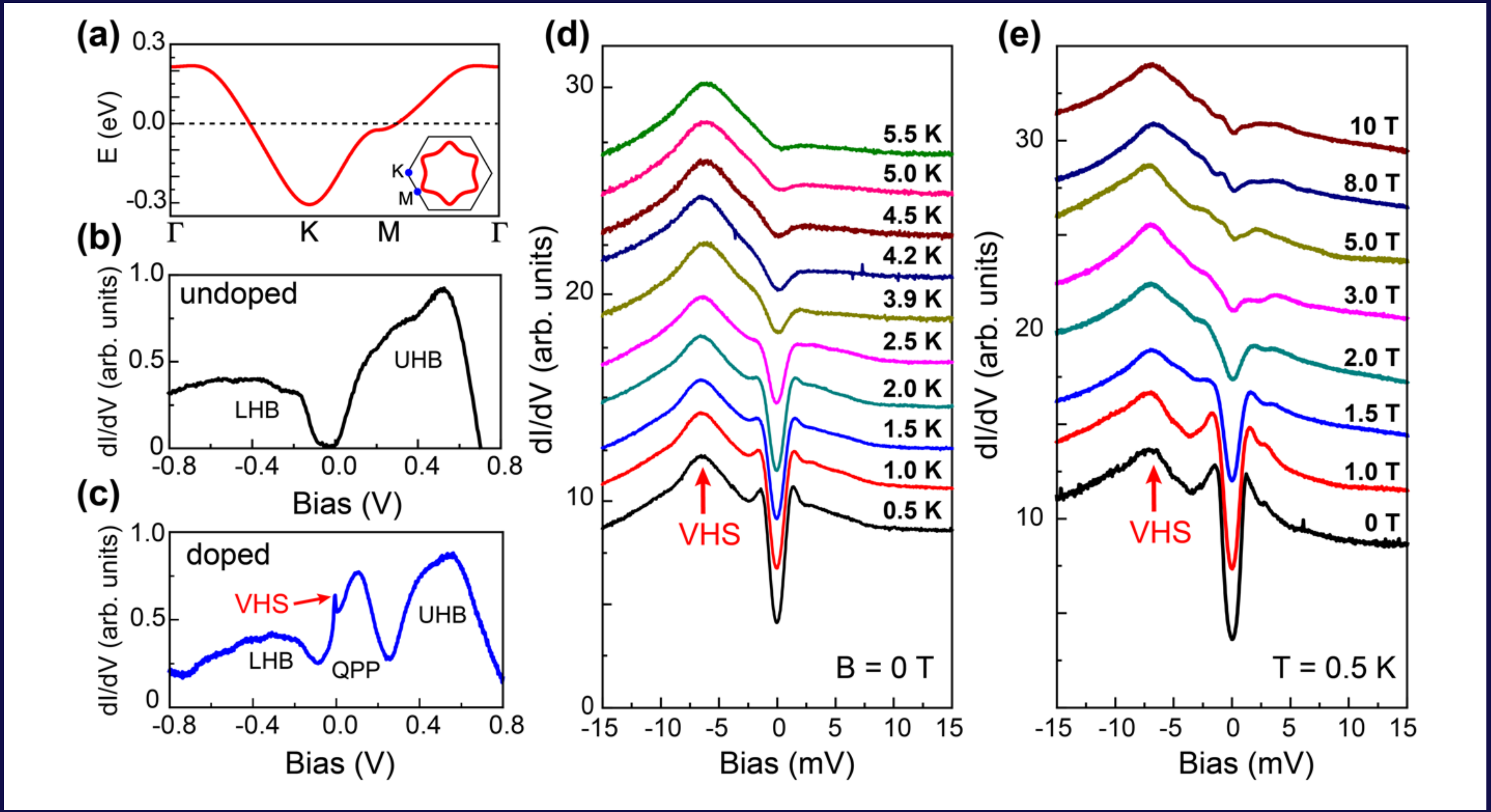


T-matrix simulation

DFT



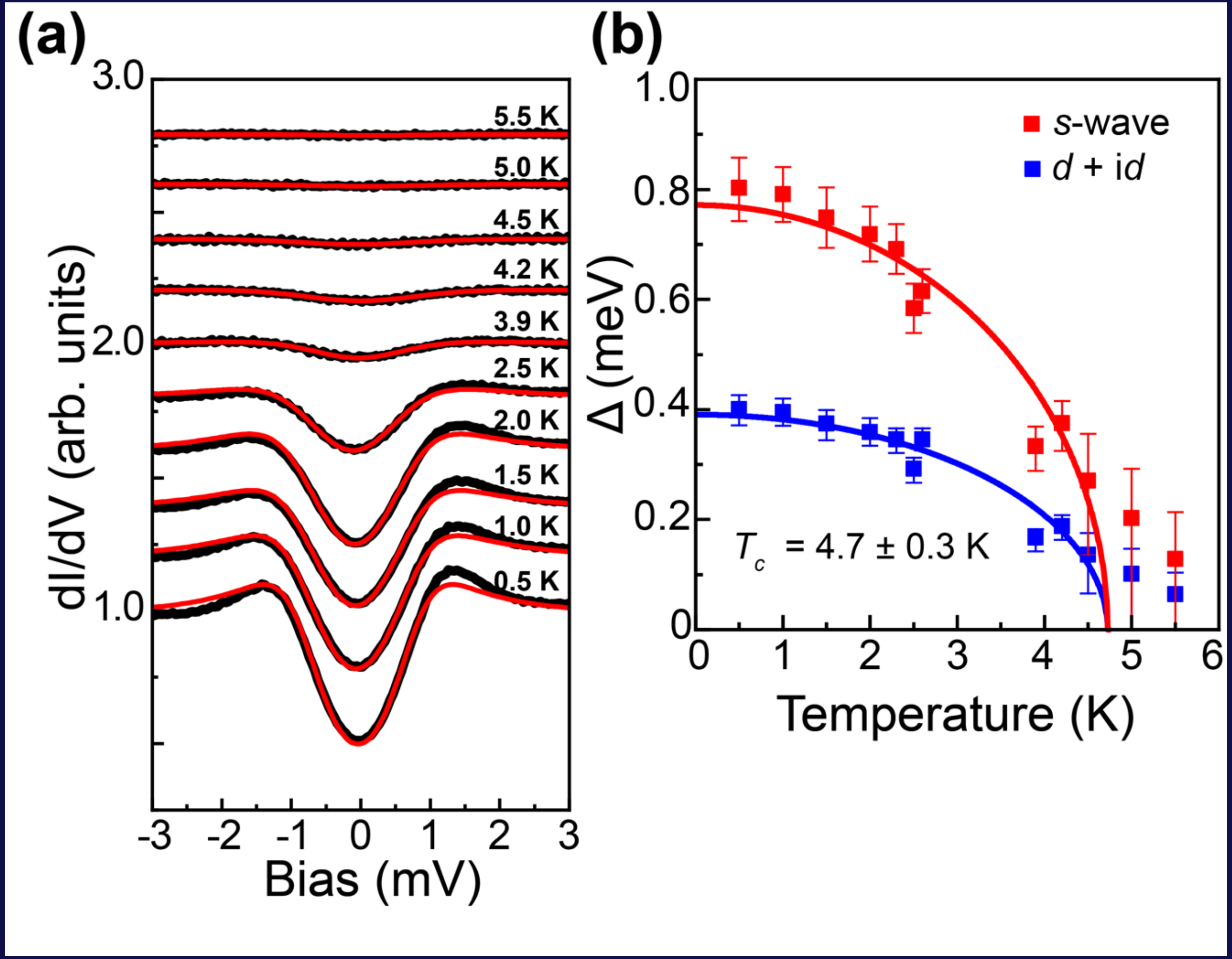
Superconductivity



p = 0.1

Wu et al., PRL 125, 117001 (2020)

Superconductivity



p = 0.1

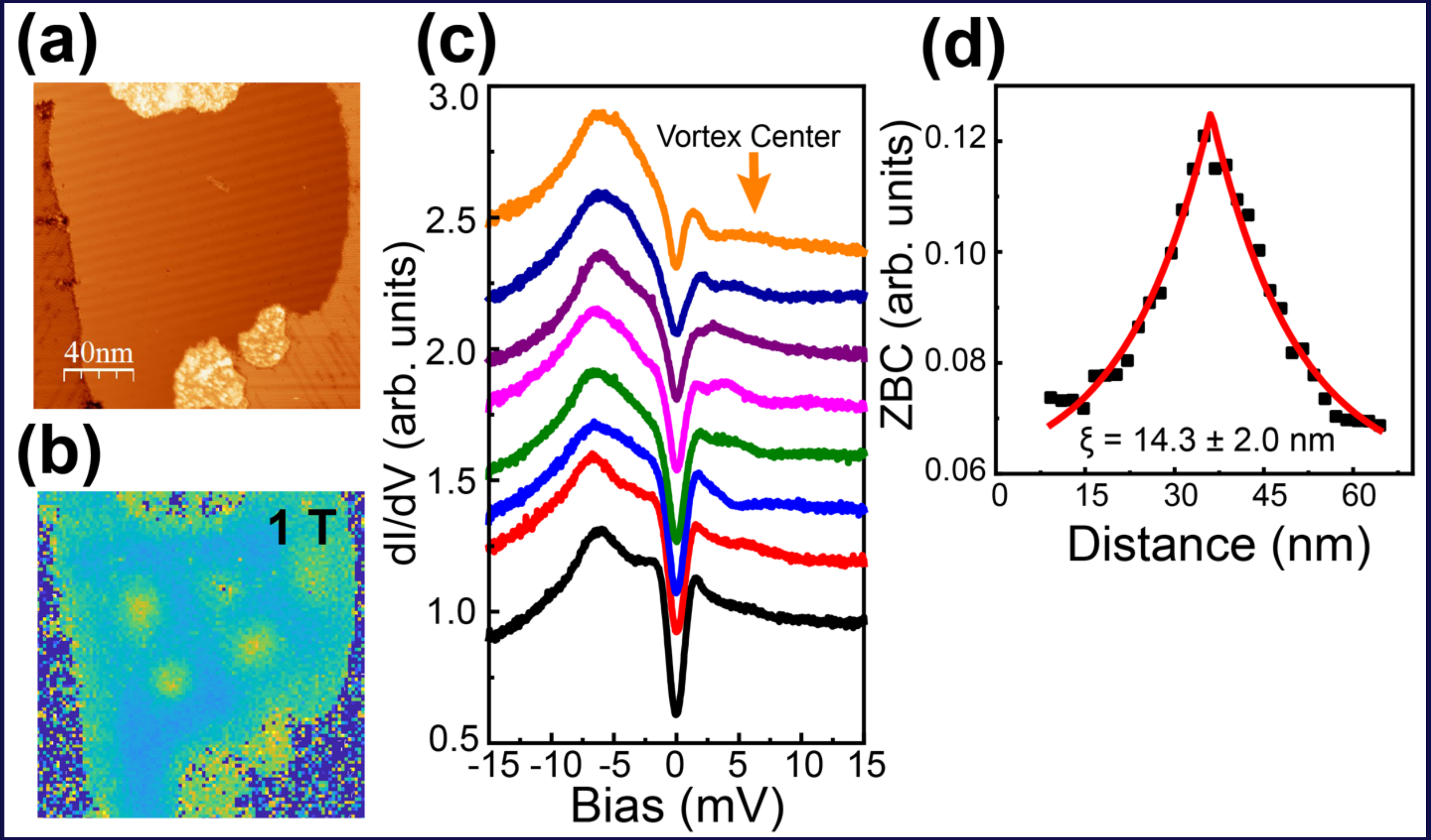
T_c = 4.7 ± 0.3 K

T_C similar to that of $Na_xCoO_2 \cdot yH_2O$

Nature **422**, 53 (2003)
Nature **424**, 527 (2003)

Wu et al., PRL 125, 117001 (2020)

Vortices



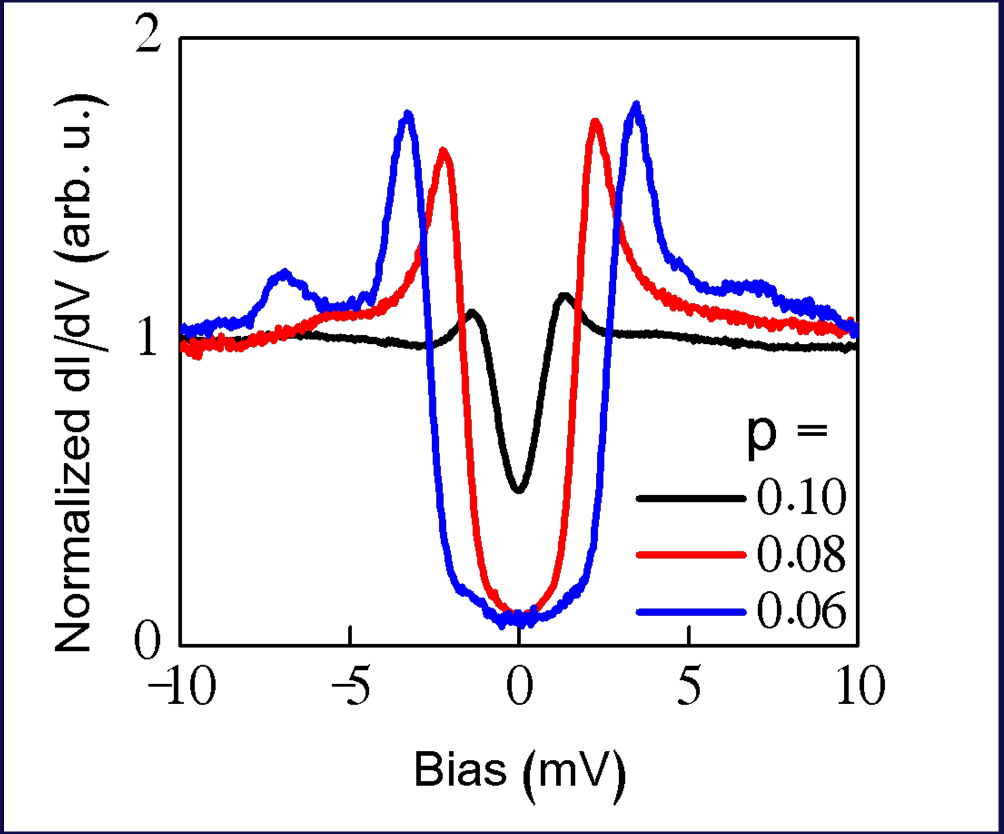
$p = 0.1$

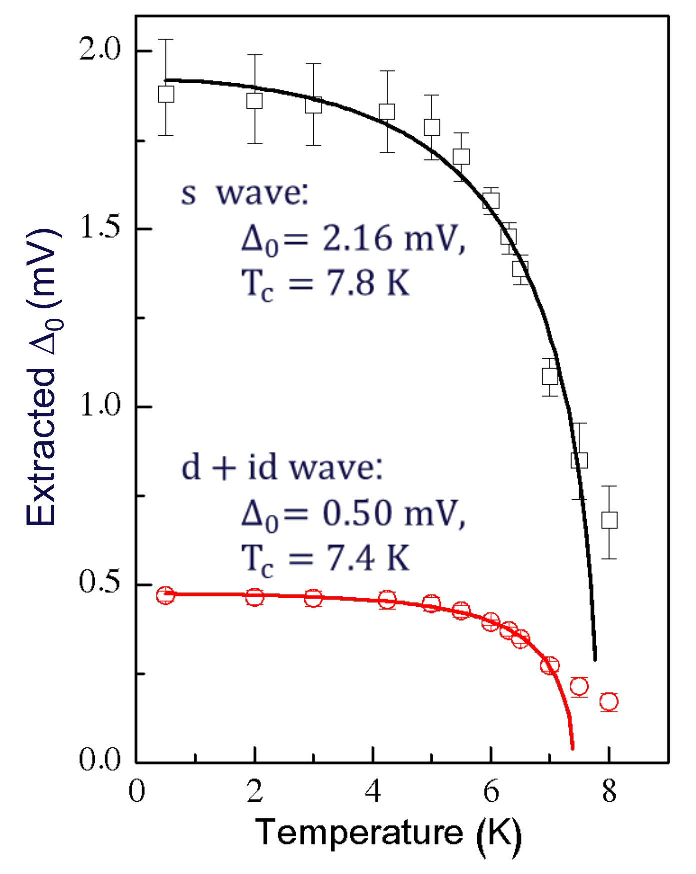
$H_{c2}(0\ \mathrm{K}) = 3\ \mathrm{T}$

$T_c = 4.7 \pm 0.3\ \mathrm{K}$

Wu et al, PRL 125, 117001 (2020)

Doping dependence

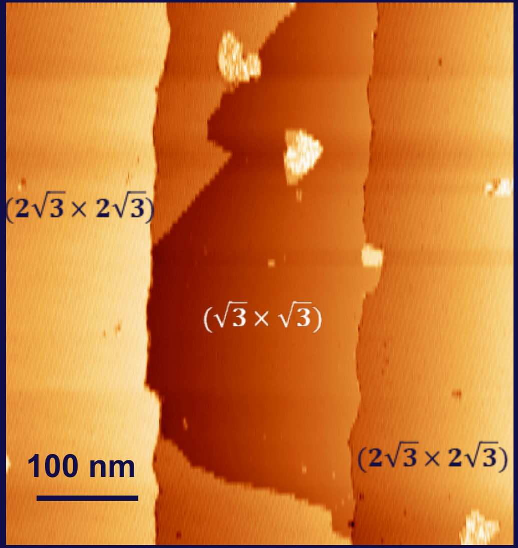


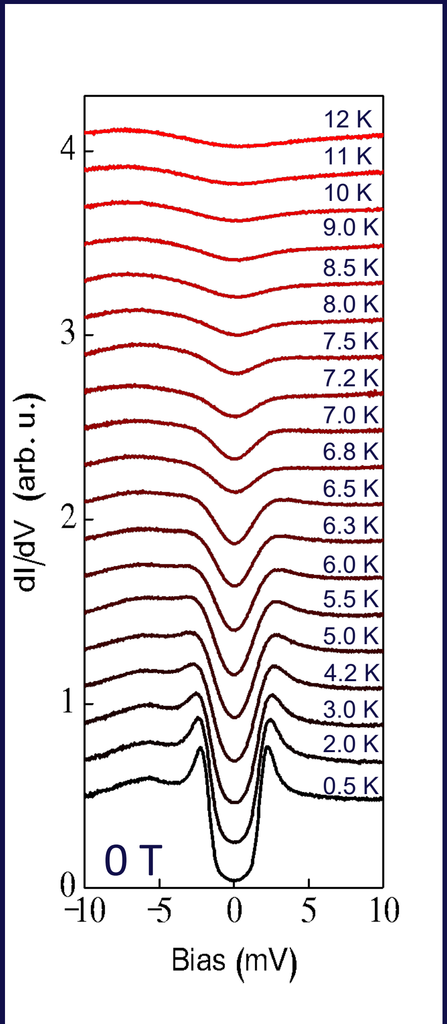


p = 0.08

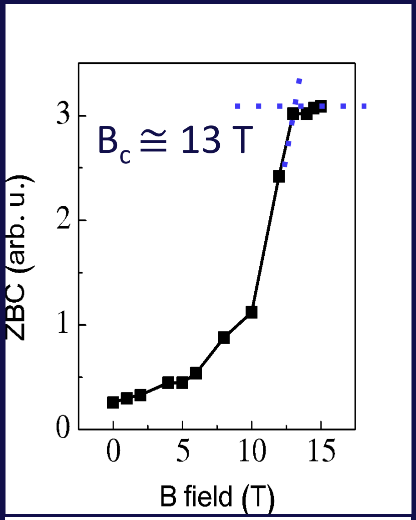
T_c = 7.6 ± 0.2 K

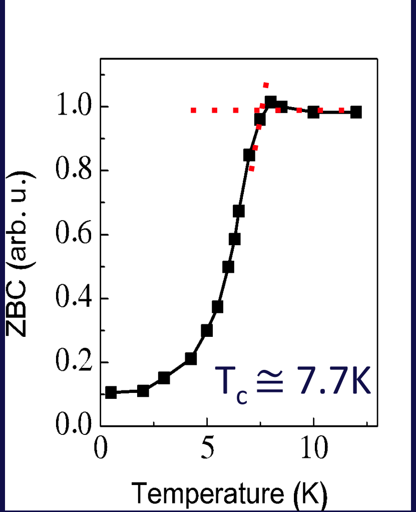
Doping dependence



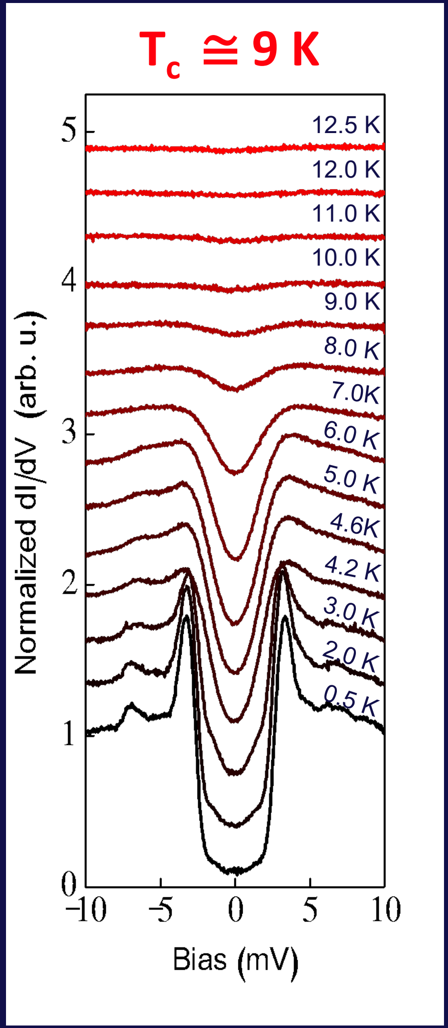


p=0.08

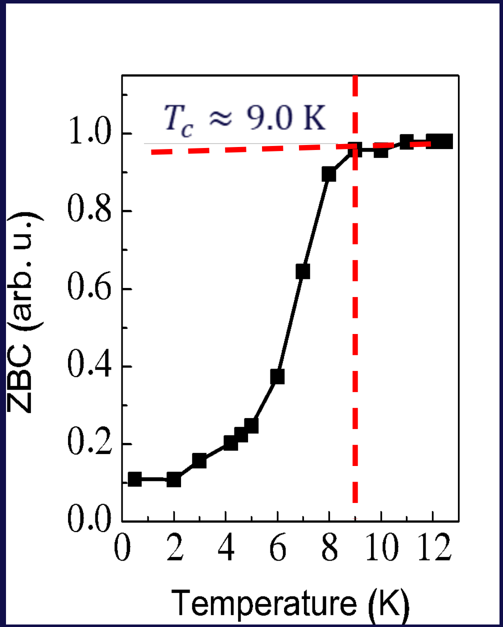




p=0.08

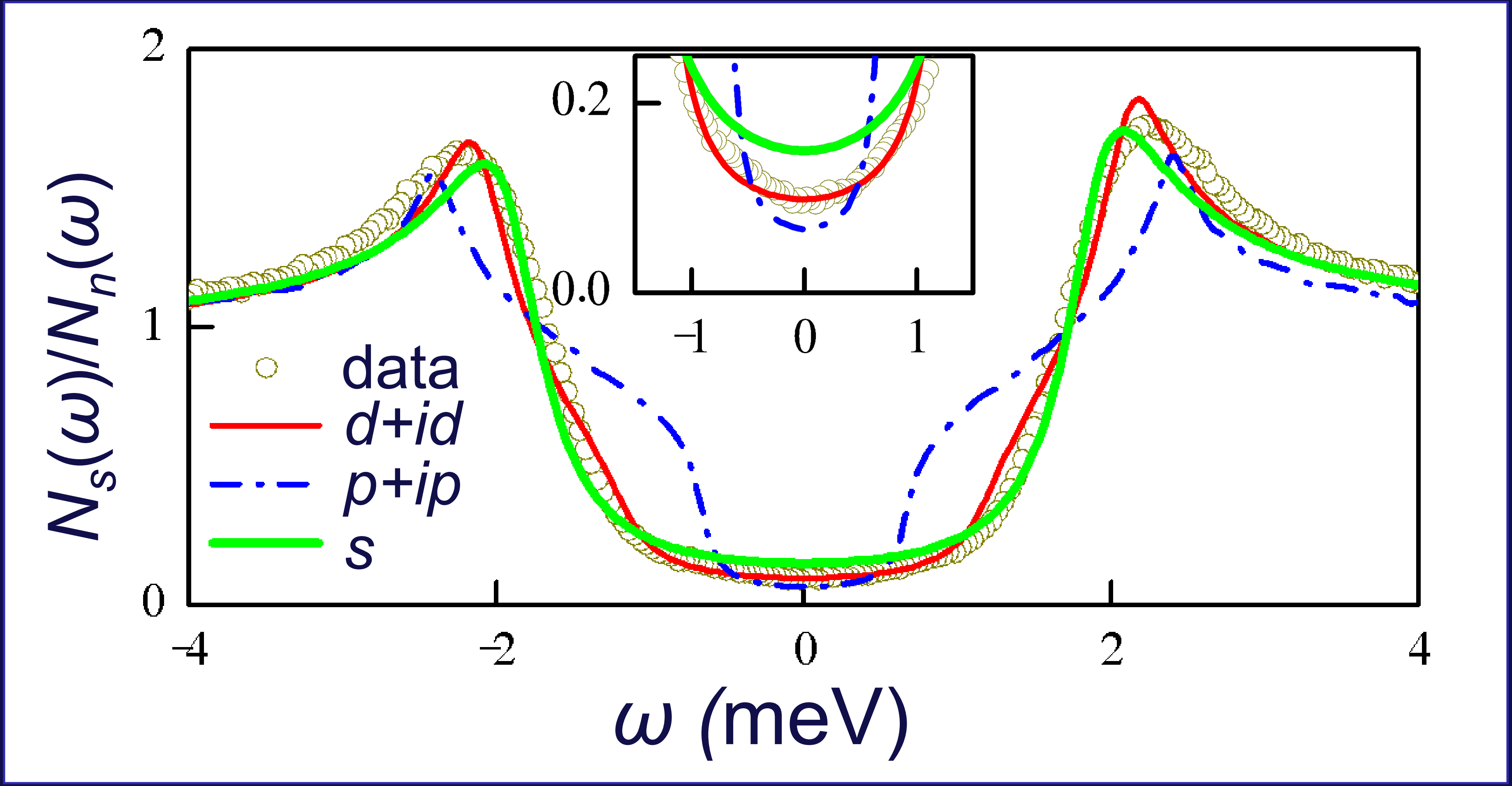


p=0.06



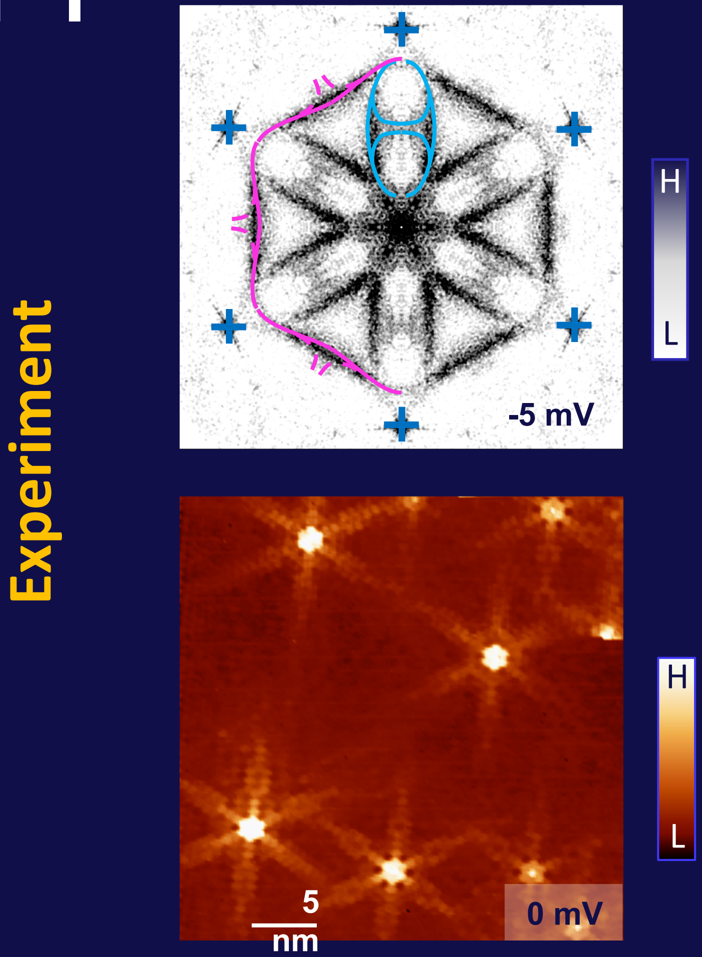
p=0.06

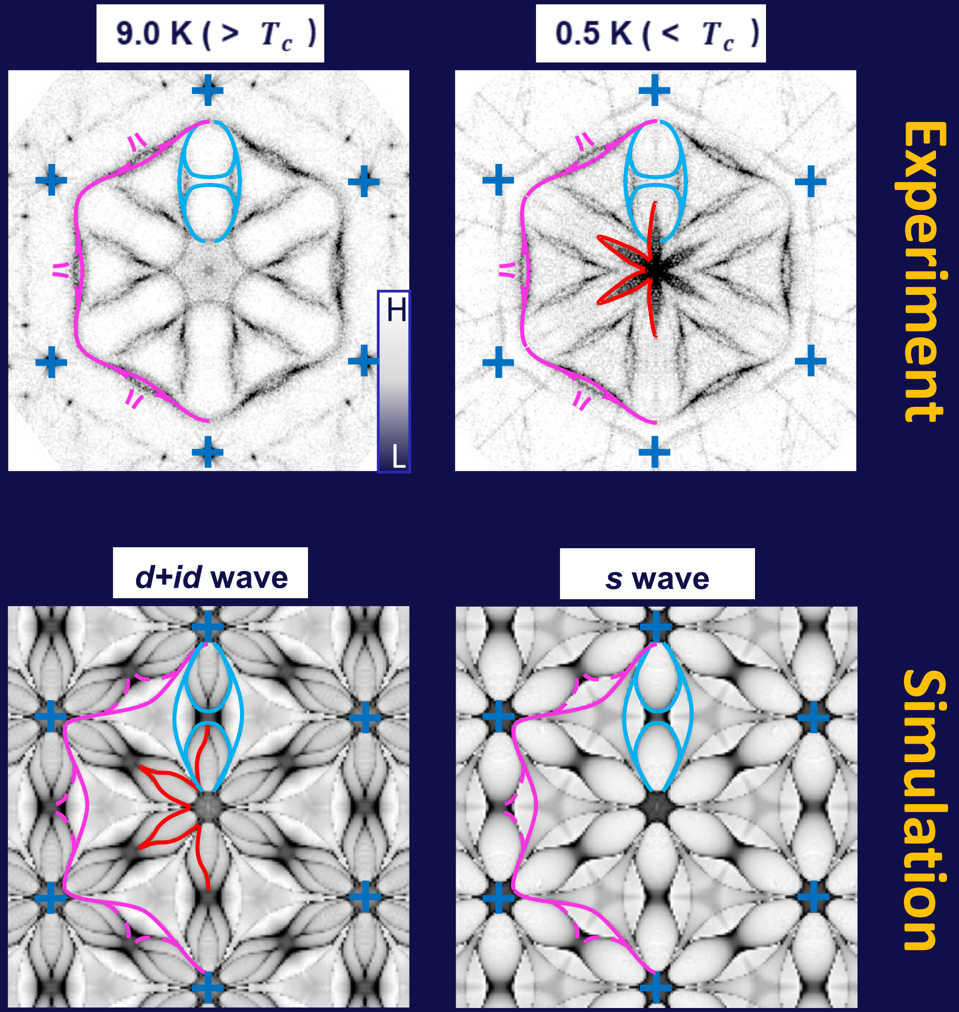
Fitting the superconducting gap



Chiral d-wave versus (anisotropic) s-wave

QPI

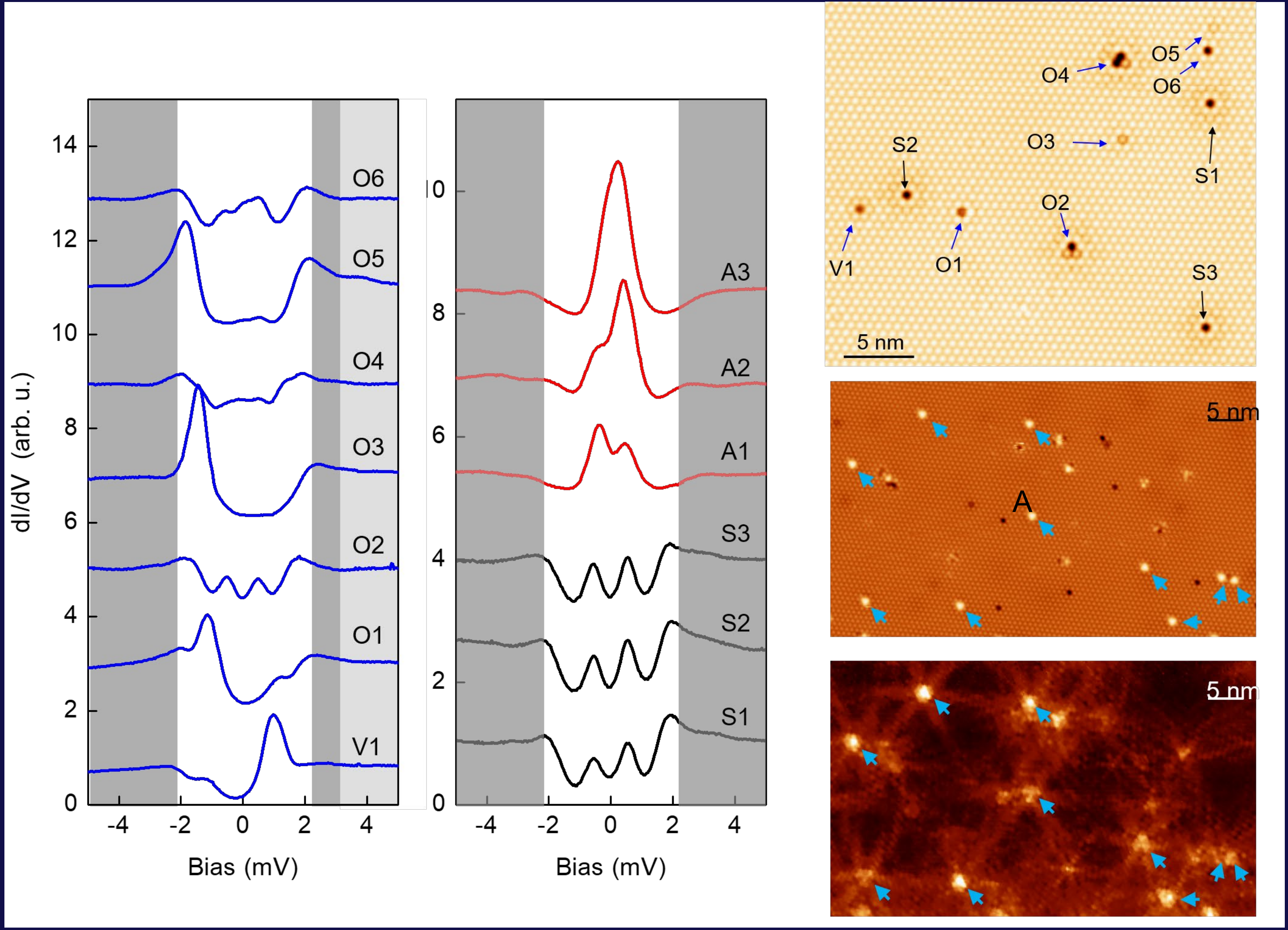




Central ‘flower’ feature from time-reversal symmetry breaking due to

magnetic defect scattering *versus* **chiral order parameter** (e.g. $d_{x^2-y^2} \pm id_{xy}$)

Defect scattering



V = adatom vacancy

S = substitutional Si atom

A = extra Sn adatoms

O_n (1-6) = unknown defects

All types of defect produce a pair of gap states. Either they are all magnetic or the superconductor breaks time-reversal symmetry (as in d+id or p+ip).

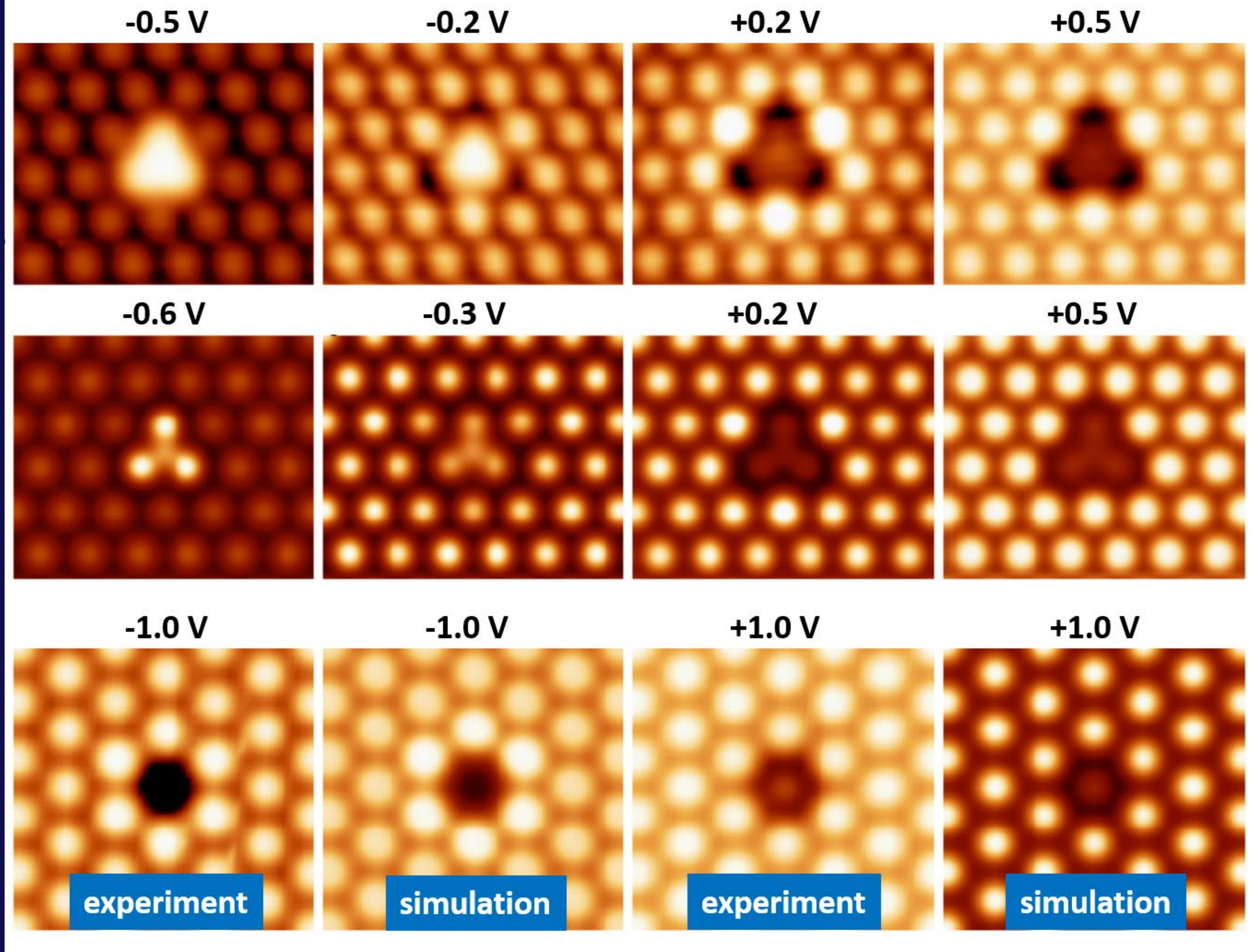
Magnetic defects?

Sn adatom defect

Experiment

simulation

Substitutional defect



Simulations by Cesar Gonzalez and Jose Ortega (Madrid)

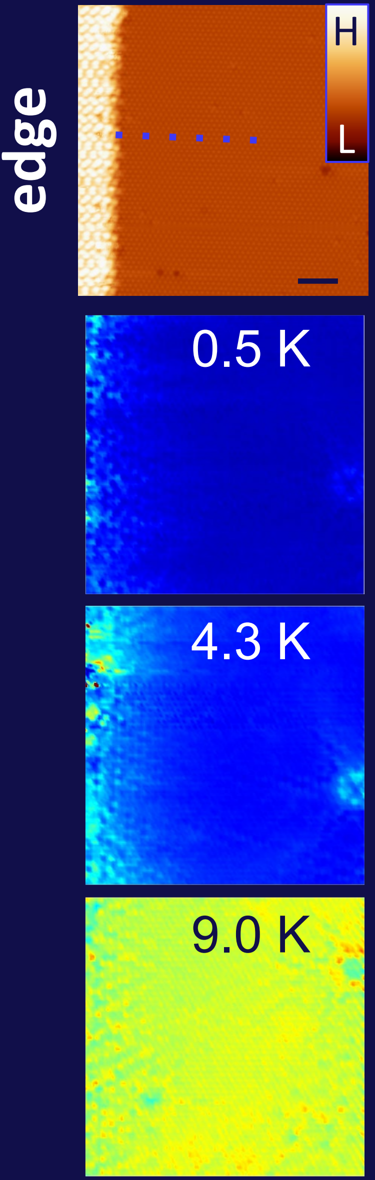
Spin-polarized DFT indicates that defects are non-magnetic

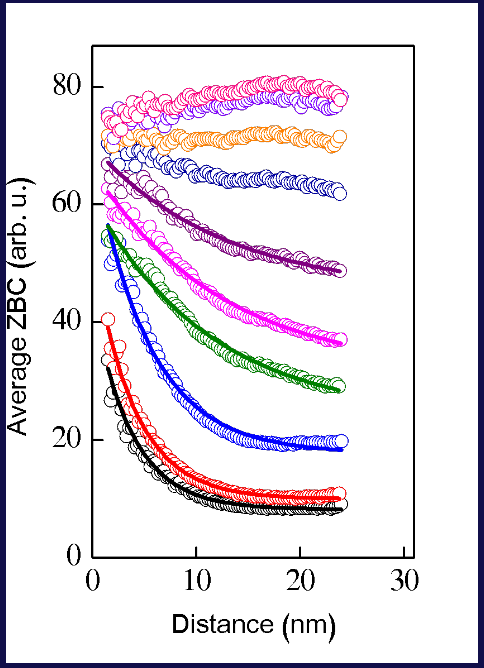
Good agreement between experimental and simulated STM images from DFT gives confidence that the DFT results are correct

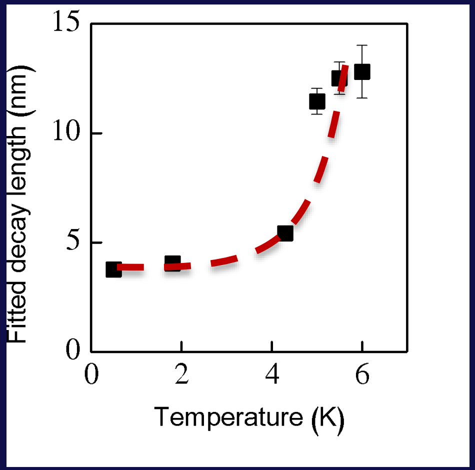
Chiral order parameter most likely interpretation (d+id or p+ip)

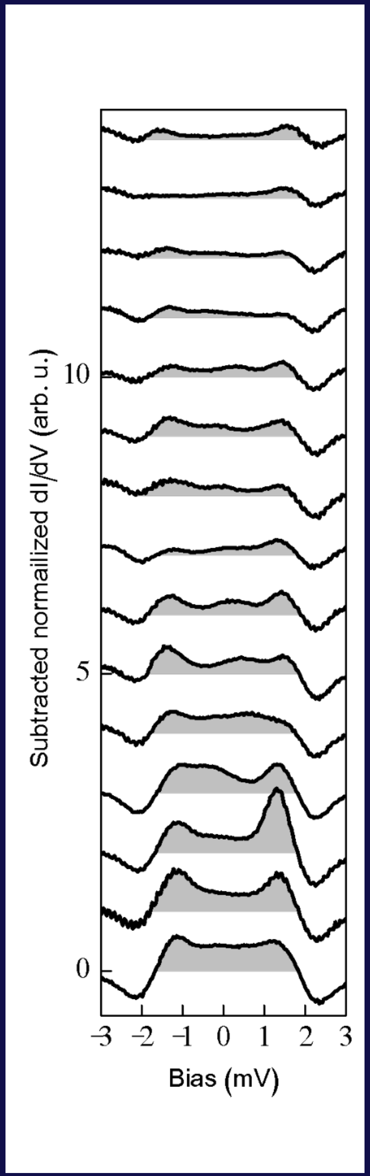
p+ip ruled out

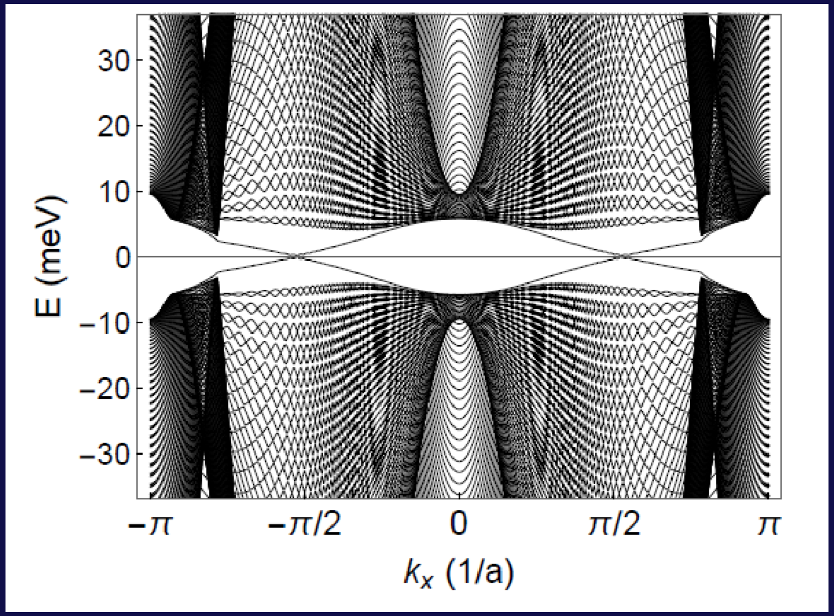
Sanity check











Near-edge ZBC in STM is consistent with existence of chiral edge states

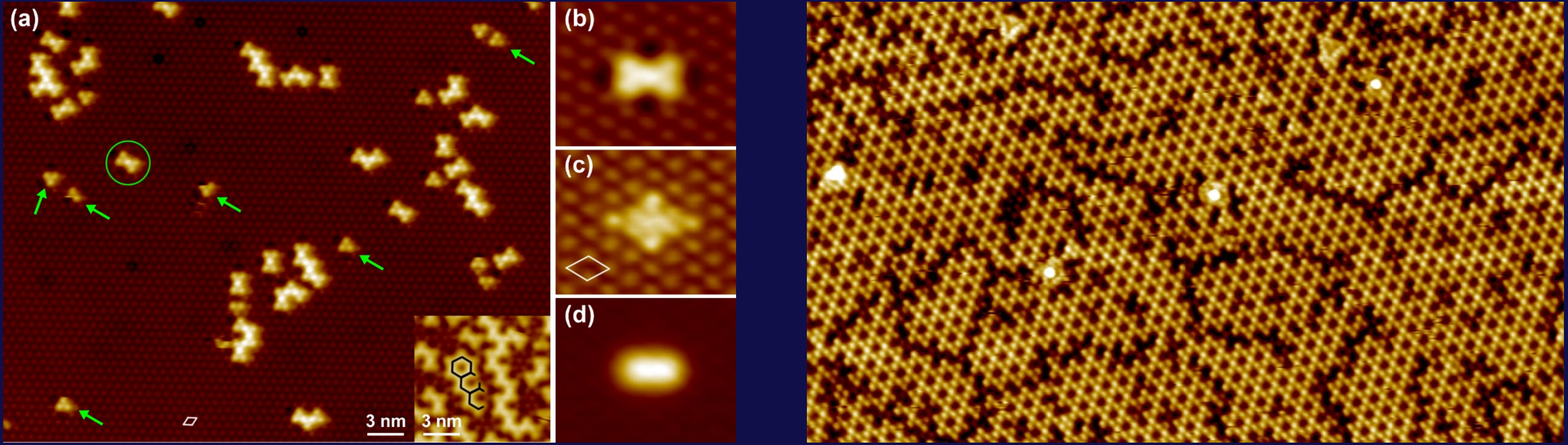
J. Strockoz and J. Venderbos (Drexel)

Conclusions and Outlook

- 1/3 ML of Sn transforms a hole-doped Si(111) semiconductor surface into a superconductor. Exceptionally clean and simple materials system
- Evidence points to Mott physics and chiral d-wave pairing
- Consistent with theoretical predictions for Sn/Si(111), e.g., Cao et al., PRB 97, 155145 (2018) and Wolf et al., PRL 128, 167002 (2022), as well as our own DCA results

- Semiconductor surfaces may be ideal test bed for studying and exploiting correlated topological states of matter
- Superconductor can possibly be altered or engineered using standard semiconductor processing or surface science approaches

Electron doping via K deposition



T.S. Smith et al, PRL **124**, 097602 (2020)

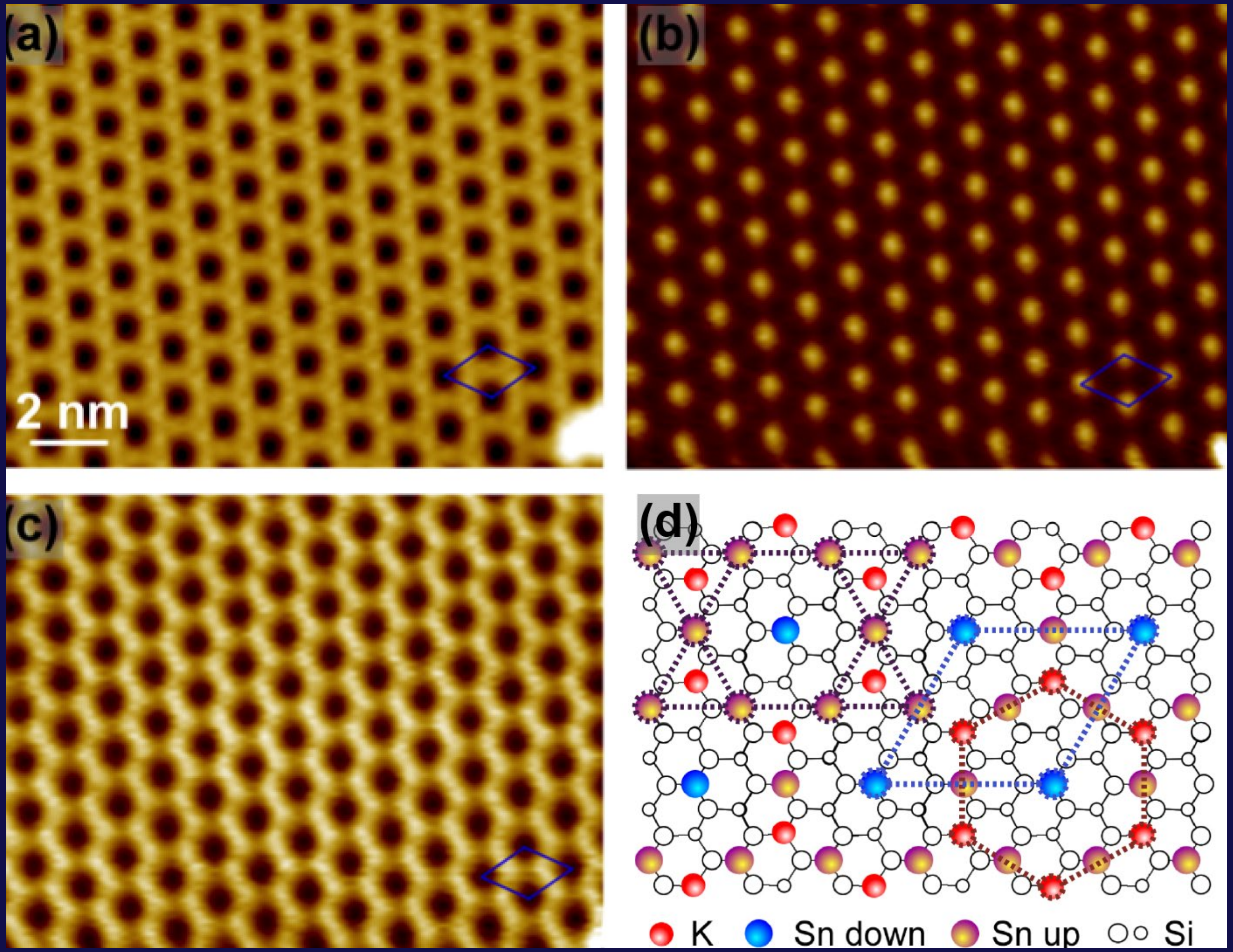
(2√3×2√3)*R*30° charge-ordered insulator

-1.5 V

Sn ‘up’

+2.5 V

K adatoms

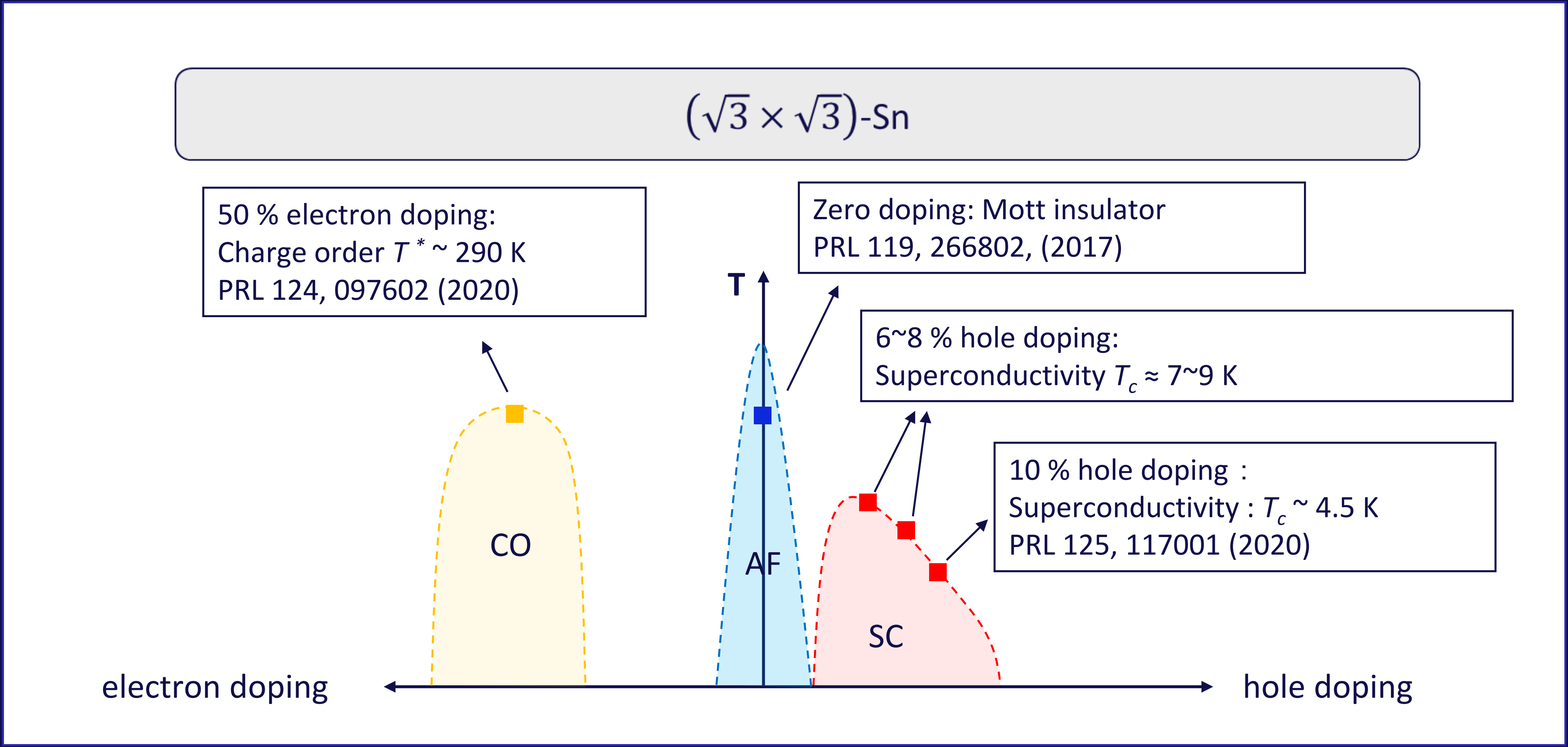


+1.0 V

Sn ‘down’

PRL **124**, 097602 (2020)

Back-up Slides



$(\sqrt{3}\times\sqrt{3})$-Sn

50 % electron doping:
Charge order T^{*} ~ 290 K
PRL 124, 097602 (2020)

Zero doping: Mott insulator
PRL 119, 266802, (2017)

6~8 % hole doping:
Superconductivity $T_c \approx$ 7~9 K

10 % hole doping :
Superconductivity : T_c ~ 4.5 K
PRL 125, 117001 (2020)

THANK YOU

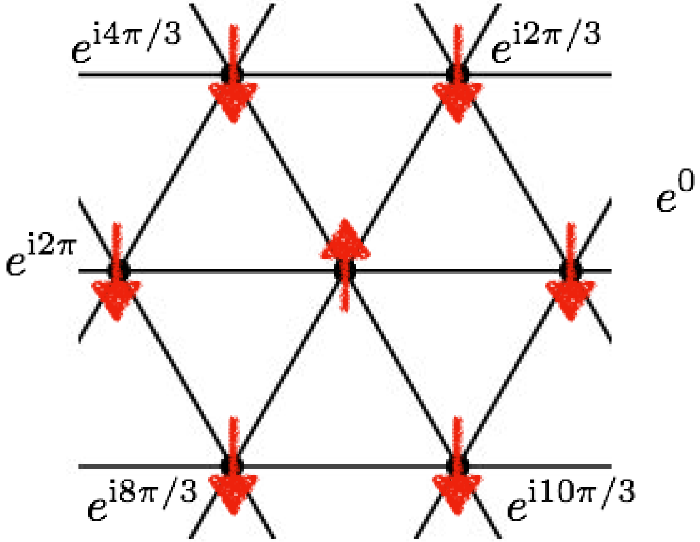
Structure of the Cooper pair

Real Space Picture

$$\Delta_{ij} = \Delta_{ln} e^{il\theta_{ij}}$$

$$\theta_{i,j} = \vec{r}_{ij} \cdot \hat{x},\ \ \vec{r}_{ij} = \vec{r}_i - \vec{r}_j$$

l = 2, n = 1



*S. Zhou & Z. Wang, Phys. Rev. Lett. **100**, 217002 (2008).

Structure of the Cooper pair

Real Space Picture

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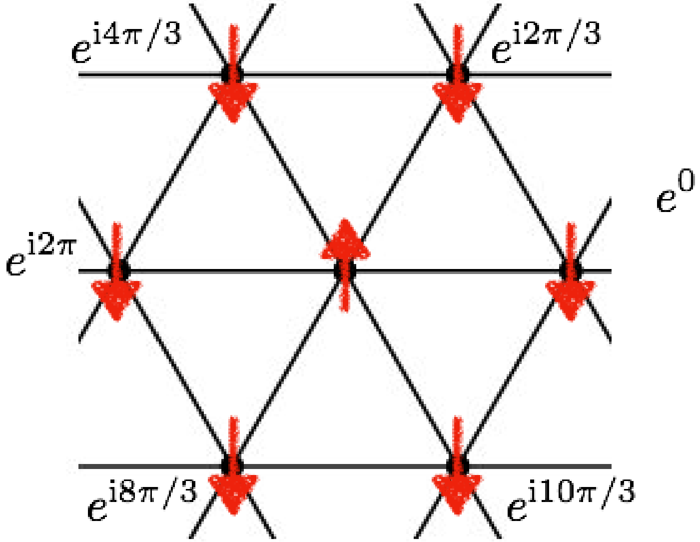


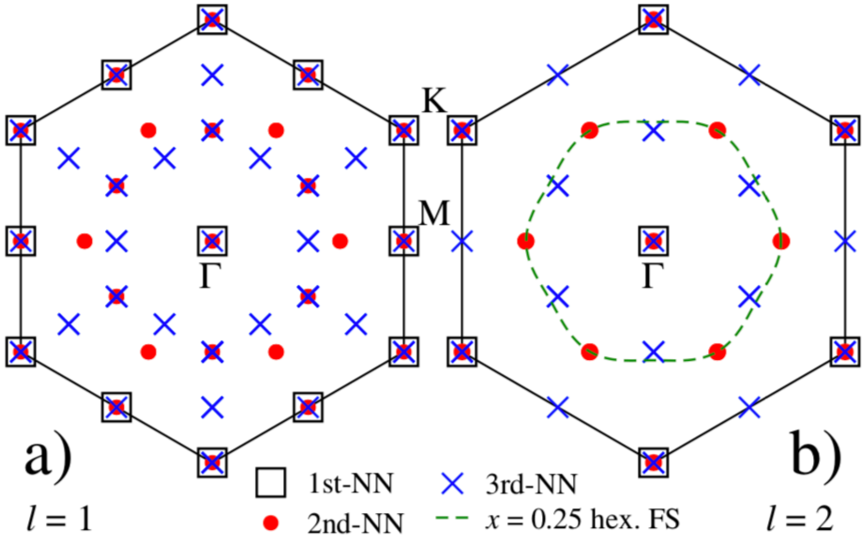
*S. Zhou & Z. Wang, Phys. Rev. Lett. **100**, 217002 (2008).

Momentum Space Picture

$$\Delta_{ln}(\mathbf{k}) = 2\left[\beta'_{ln}(\mathbf{k}) + i\beta''_{ln}(\mathbf{k})\right]$$

Chiral d-wave pairing ($\ell = 2$)

n	$\beta'_{2,n}(k)$	$\beta''_{2,n}(k)$
1	$\cos k_y - \cos\frac{\sqrt{3}}{2}k_x \cos\frac{1}{2}k_y$	$\sqrt{3}\sin\frac{\sqrt{3}}{2}k_x \sin\frac{1}{2}k_y$
2	$\cos\sqrt{3}k_x - \cos\frac{3}{2}k_y \cos\frac{\sqrt{3}}{2}k_x$	$-\sqrt{3}\sin\frac{3}{2}k_y \sin\frac{\sqrt{3}}{2}k_x$
3	$\cos 2k_y - \cos\sqrt{3}k_x \cos k_y$	$\sqrt{3}\sin\sqrt{3}k_x \sin k_y$



* Note: Zhou & Wang's (k_x,k_y) is rotated 90 degrees relative to my real space picture.

Competing explanations

- States at the Fermi level are related to a boron impurity band

  Coherent QPI scattering produces Fermi surface of $\left(\sqrt{3}\times\sqrt{3}\right)$ *Sn adatom reconstruction*

- Gap states are conventional YSR states associated with magnetic defects in an s-wave superconductor

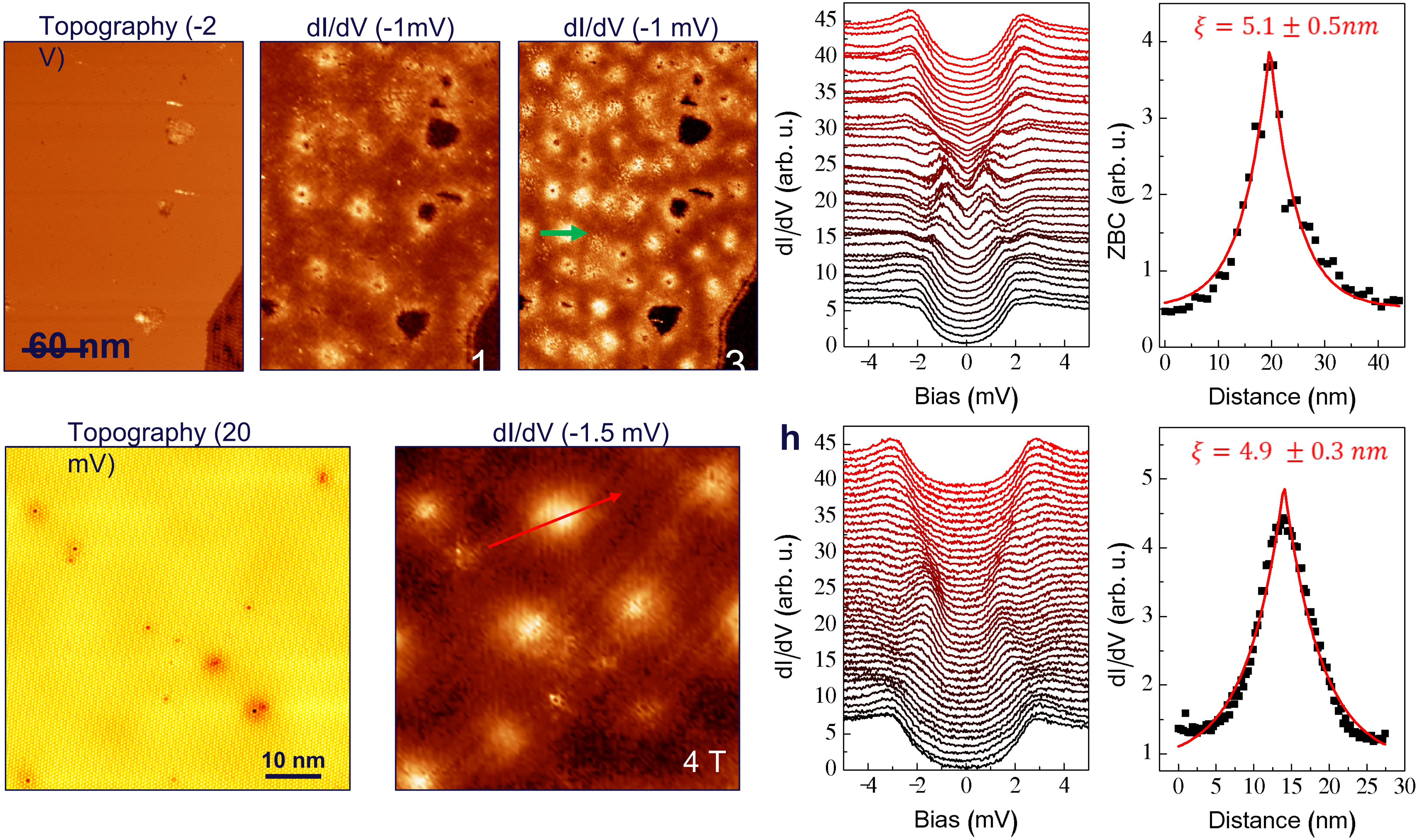
  One would have to assume that all defects, including the extra Sn adatoms and substitutional Si atoms are magnetic. DFT results are in excellent agreement with the STM data of substitutional and Sn adatom defects, and thus likely correct

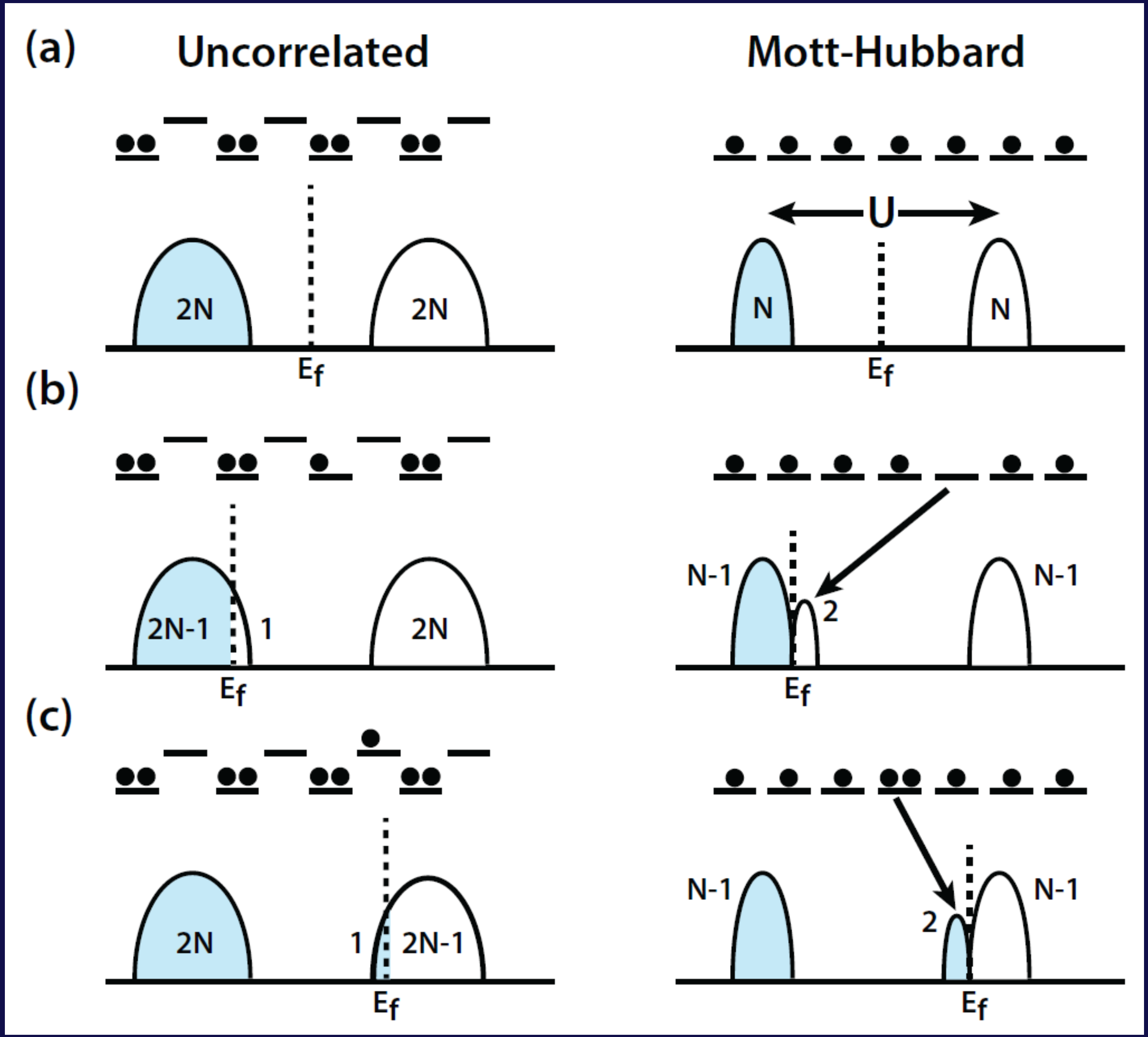
- Edge states are merely the result of an inverse proximity effect.

  Possibly true. More investigations are needed.

- Pairing symmetry is p+ip or higher angular momentum pairing channels

  Gap fitting doesn't work for p-wave. DCA calculations indicate d+id is the leading pairing instability


Topography (-2 V)
60 nm
dI/dV (-1mV)
1
dI/dV (-1 mV)
3
dI/dV (arb. u.)
Bias (mV)
ZBC (arb. u.)
Distance (nm)
$\xi = 5.1 \pm 0.5 nm$
Topography (20 mV)
10 nm
dI/dV (-1.5 mV)
4 T
h
dI/dV (arb. u.)
Bias (mV)
dI/dV (arb. u.)
Distance (nm)
$\xi = 4.9 \pm 0.3 \, nm$


(a)
Uncorrelated
Mott-Hubbard
2N
E_f
2N
U
N
E_f
N
(b)
2N-1
1
E_f
2N
N-1
2
E_f
N-1
(c)
2N
1
2N-1
E_f
N-1
2
E_f
N-1