

# VISUALIZING THE ORIGIN OF BULK METALLICITY IN $\text{SMB}_6$

Harris Pirie (Oxford)



Richard Liu

Pengcheng Chen

Christian Matt

Mohammad Hamidian

Eric Mascot

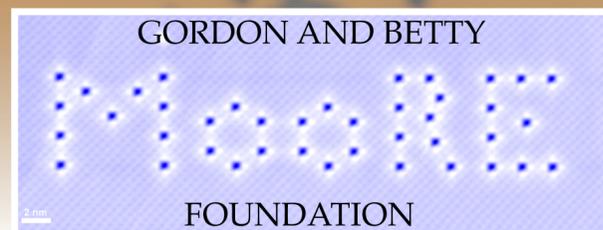
Dirk Morr

Johnpierre Paglione

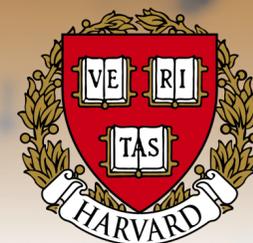
Cyrus Hirjibehedin

Séamus Davis

Jenny Hoffman

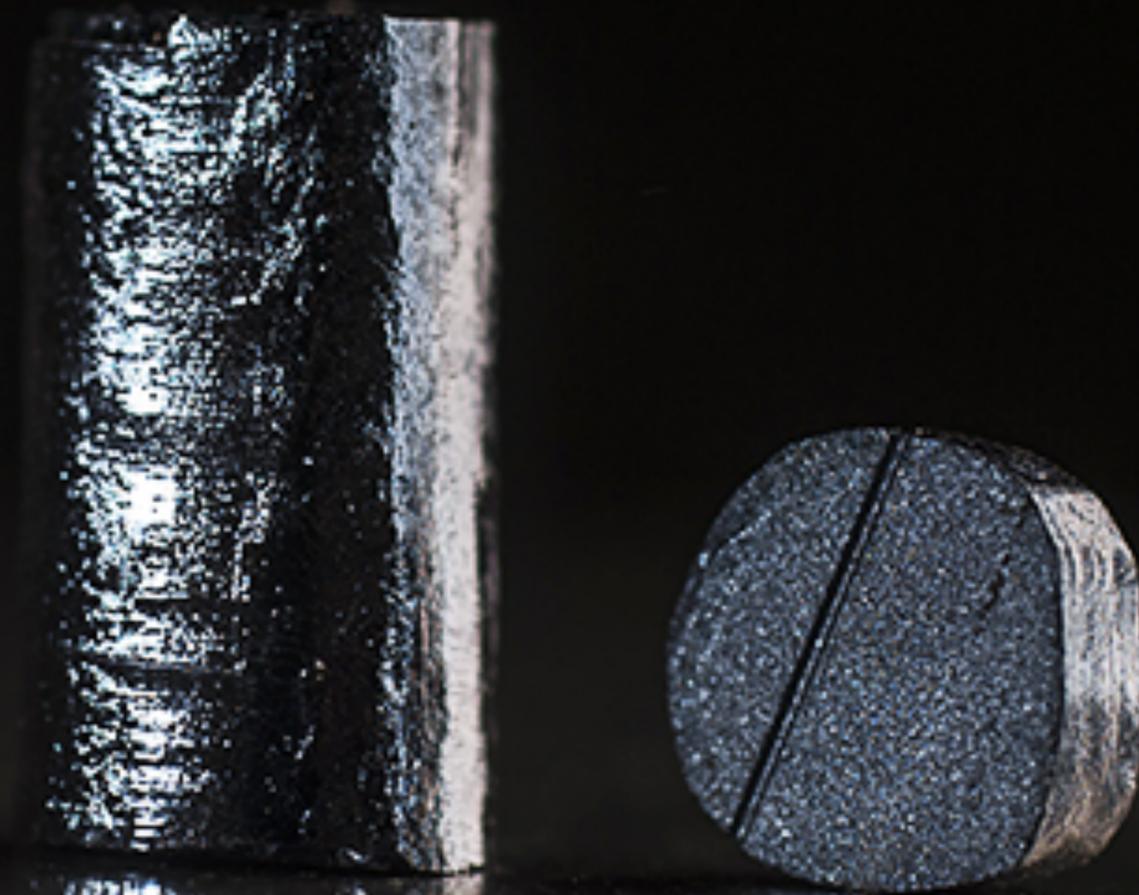


Funded by the Horizon 2020 Framework Programme of the European Union



UNIVERSITY OF OXFORD

# Samarium hexaboride ( $\text{SmB}_6$ ) is controversial



## $\text{SmB}_6$ timeline:

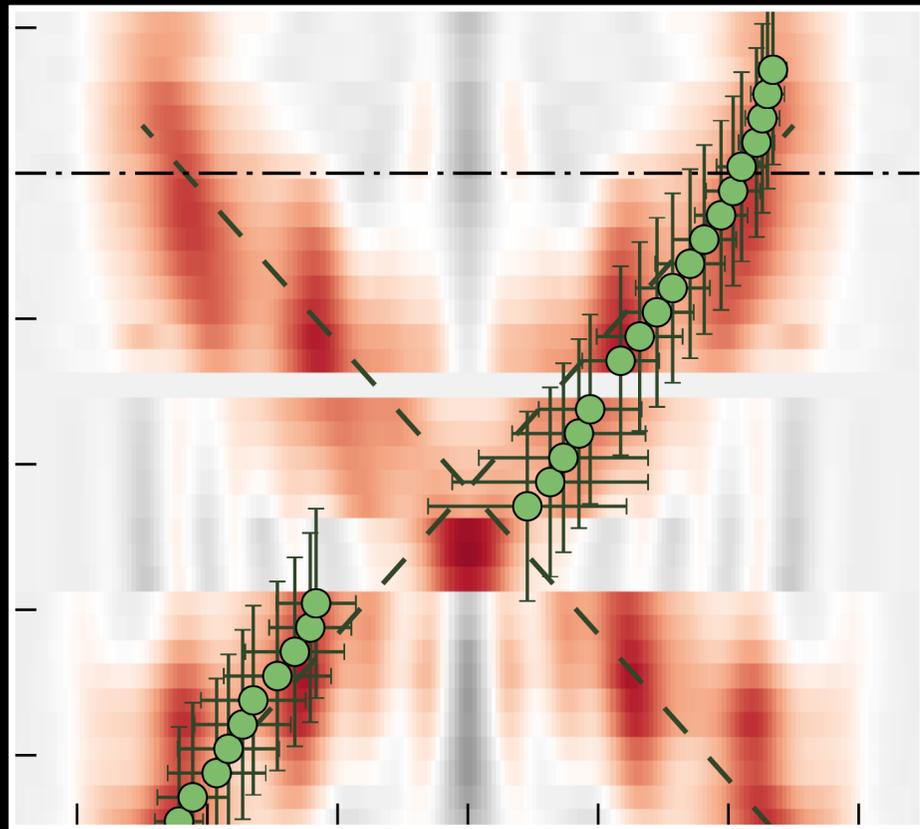
1955 - 2010 — Impurity states vs surface states

2010 - 2020 — Topological or not?

2015 - 2023 — Insulating or not?

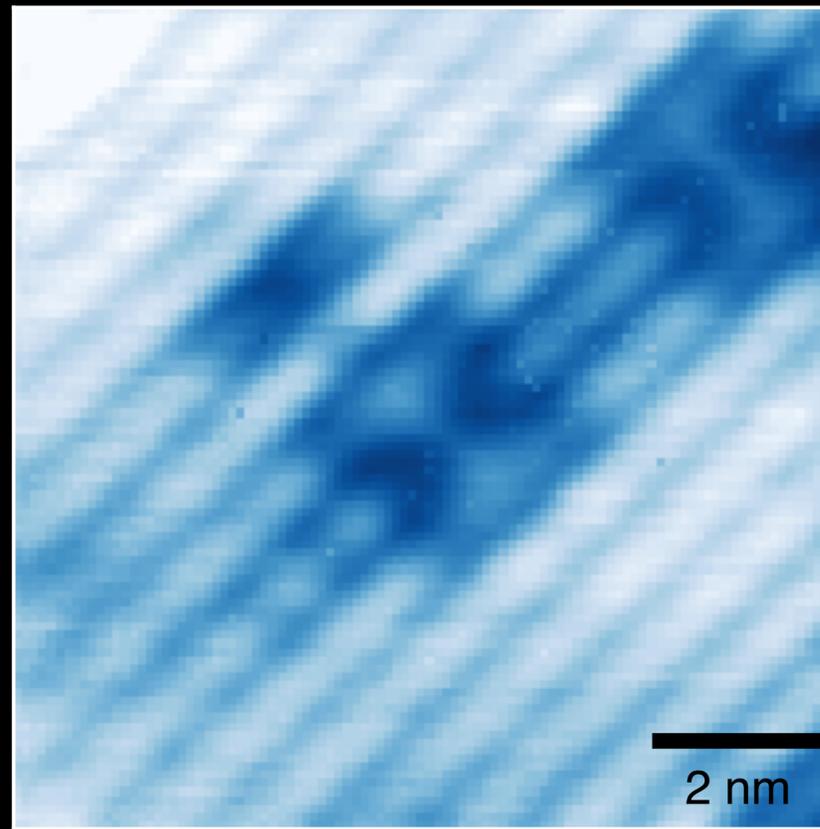
# Outline

**(1) Dirac fermions 410 times heavier than a bare electron!**



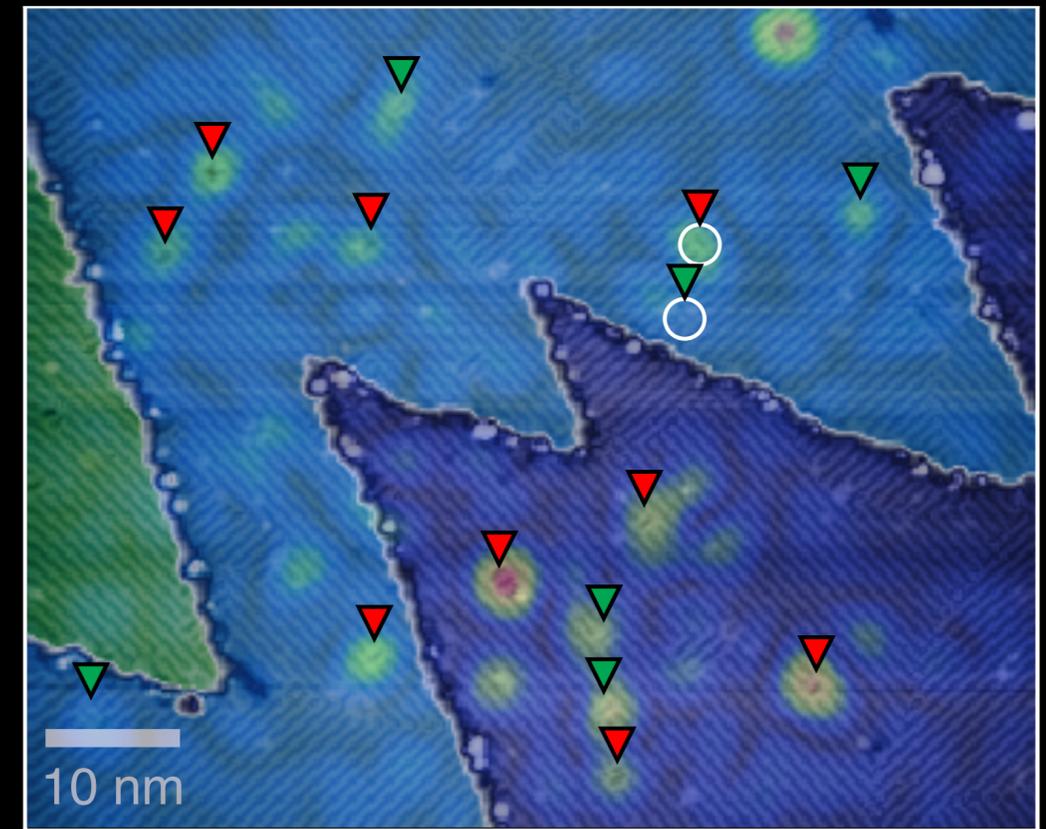
Pirie et al., *Nat. Phys.* **16**, 52 (2020)  
Matt et al., *Phys. Rev. B* **101**, 085142 (2020)

**(2) Metallicity in an insulator!**



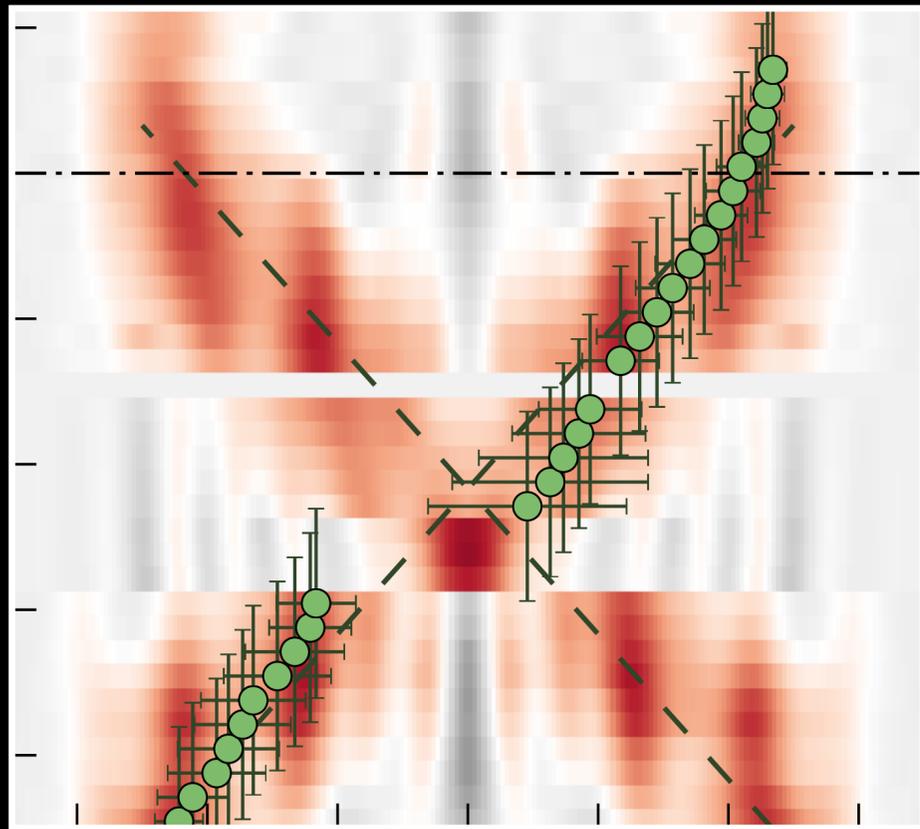
Pirie et al., *Science* **379**, 1214 (2023)

**(3) Breakdown of topological protection!**



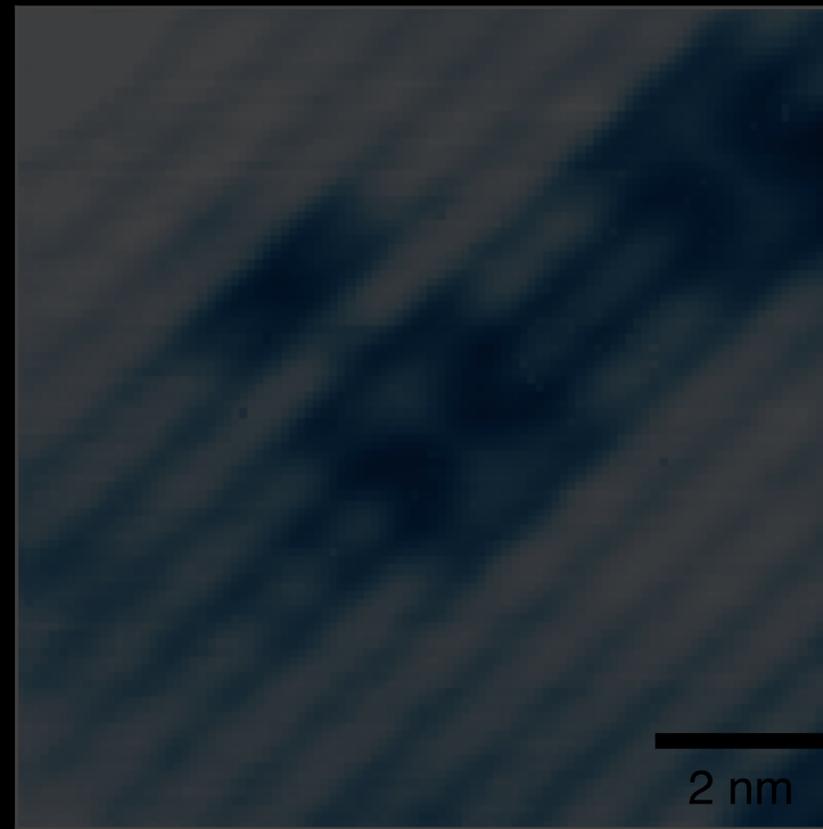
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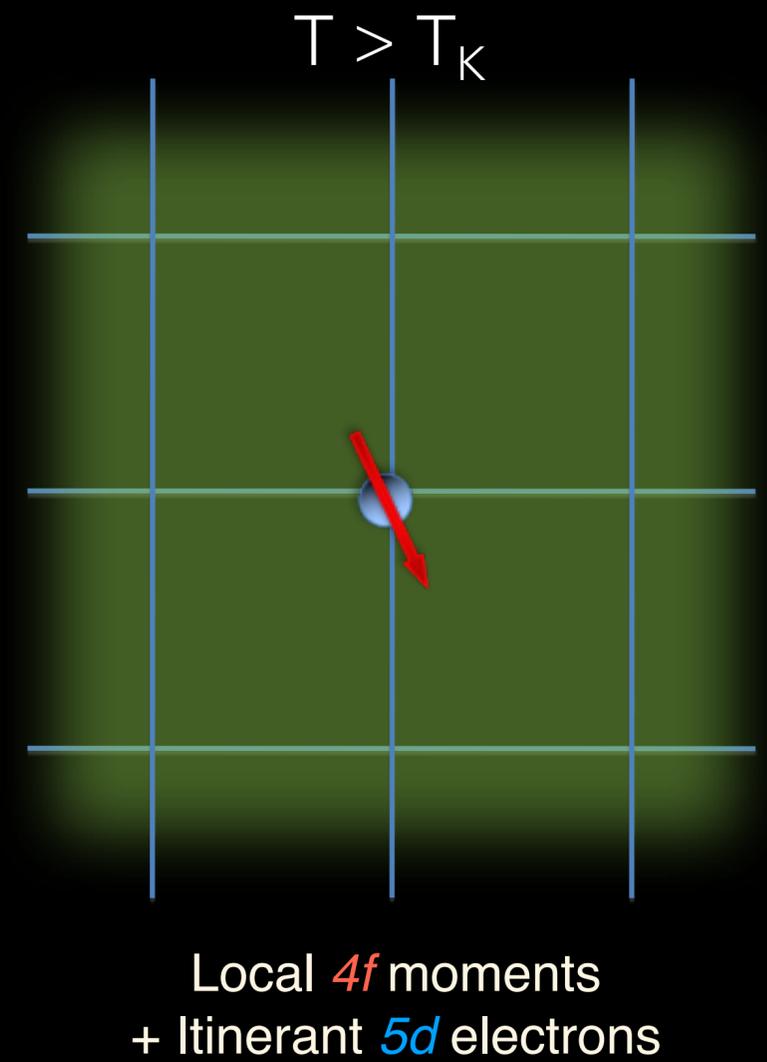
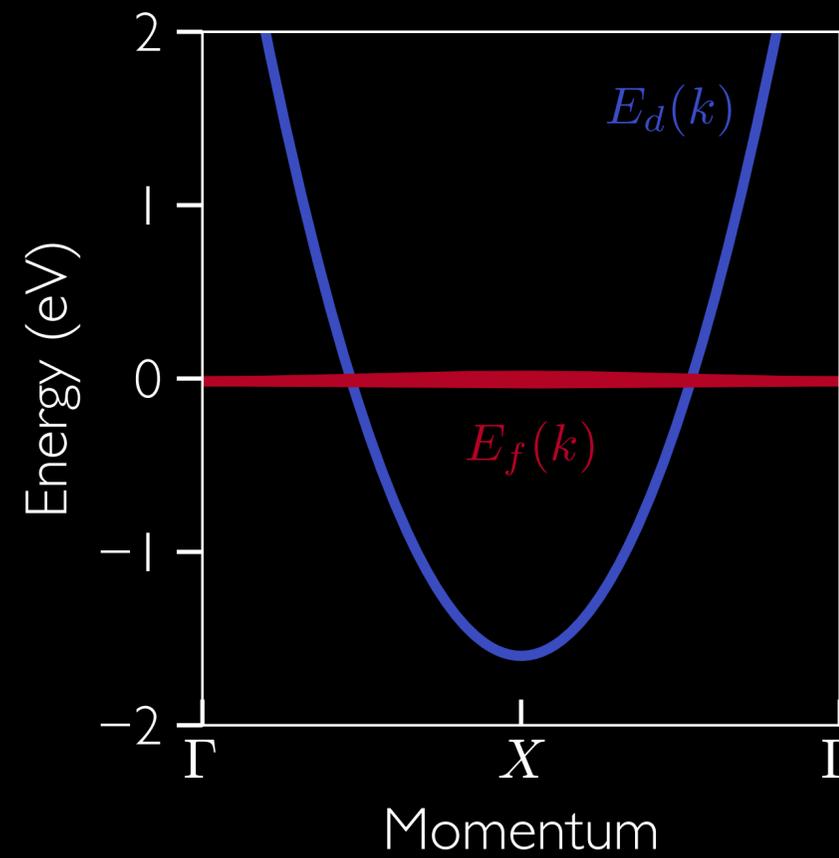
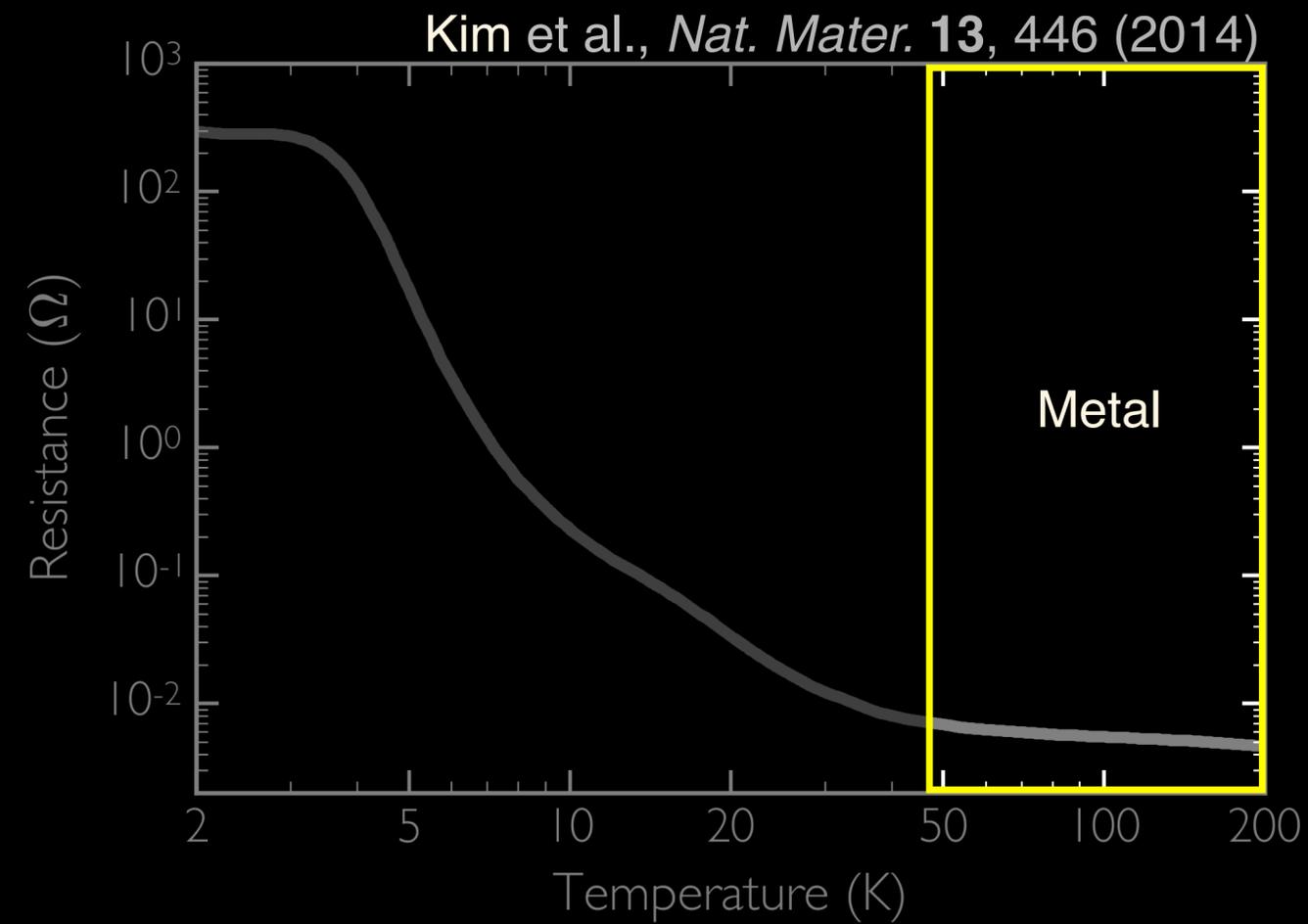


Pirie et al., *Science* **379**, 1214 (2023)

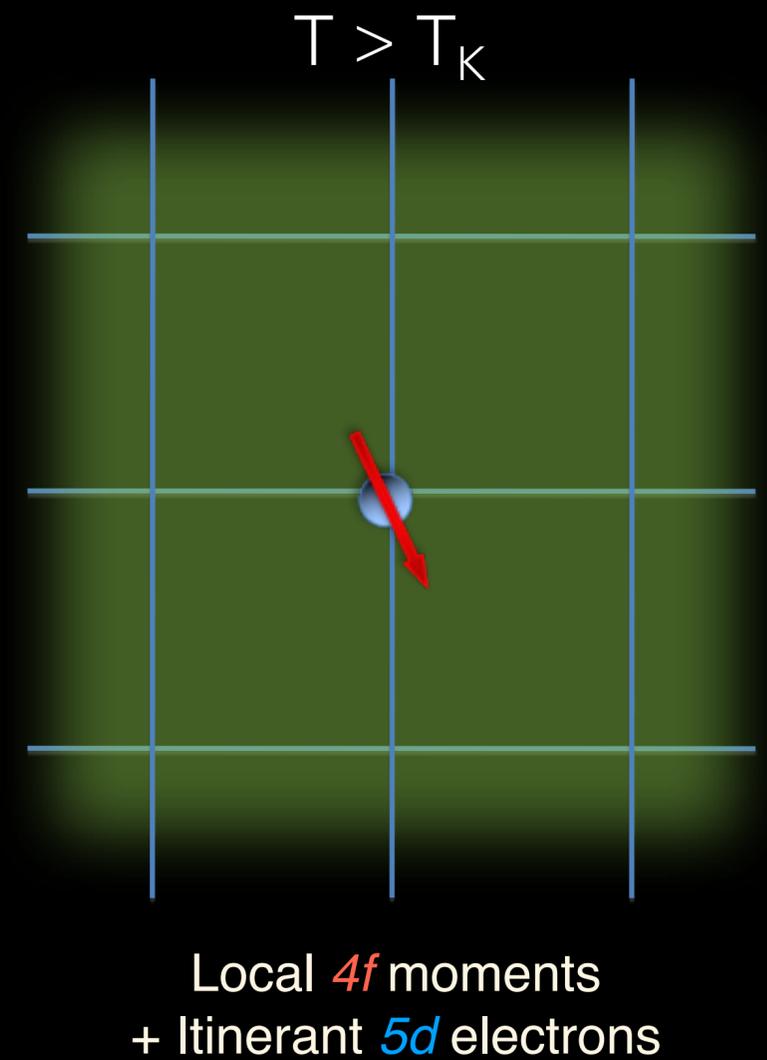
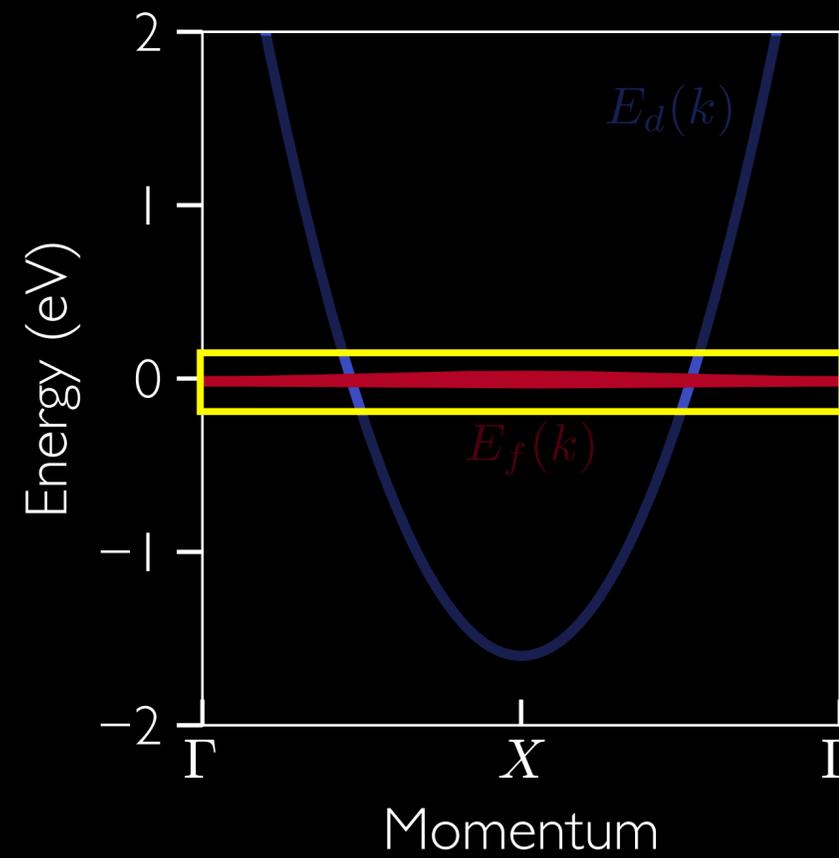
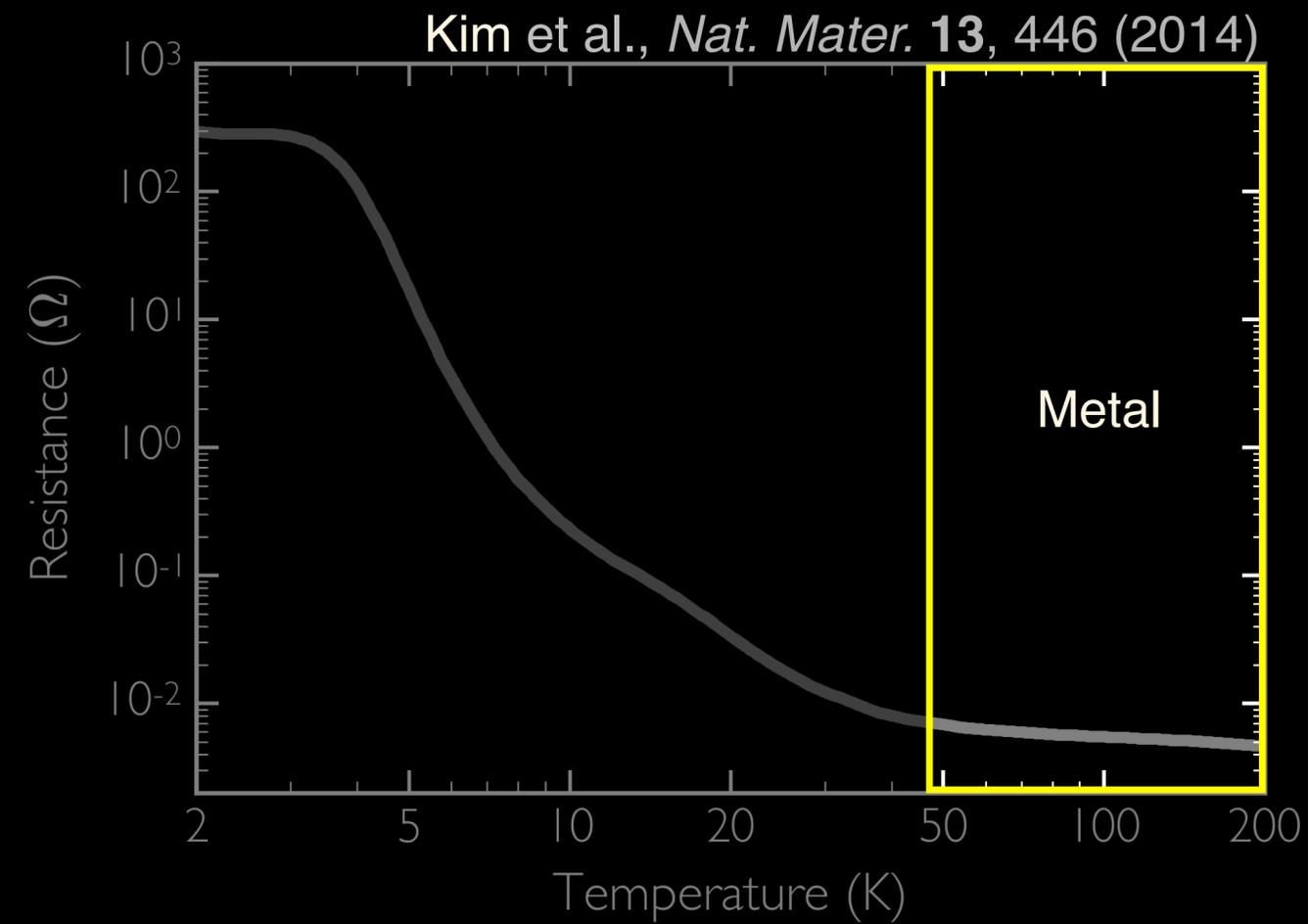
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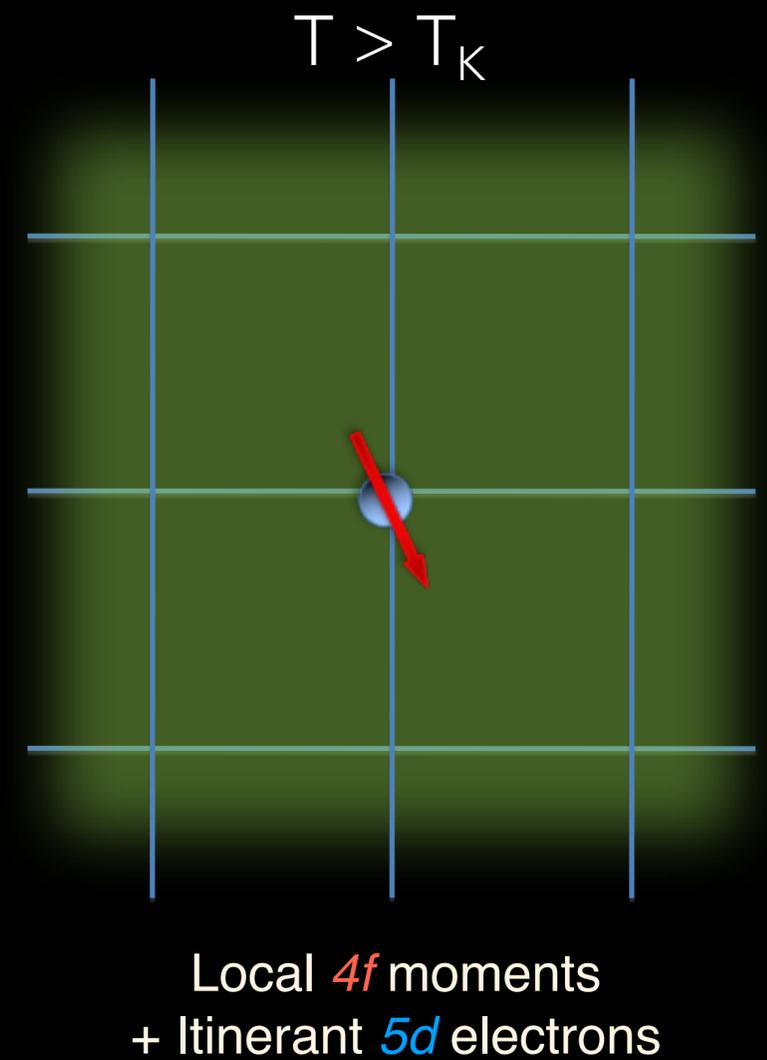
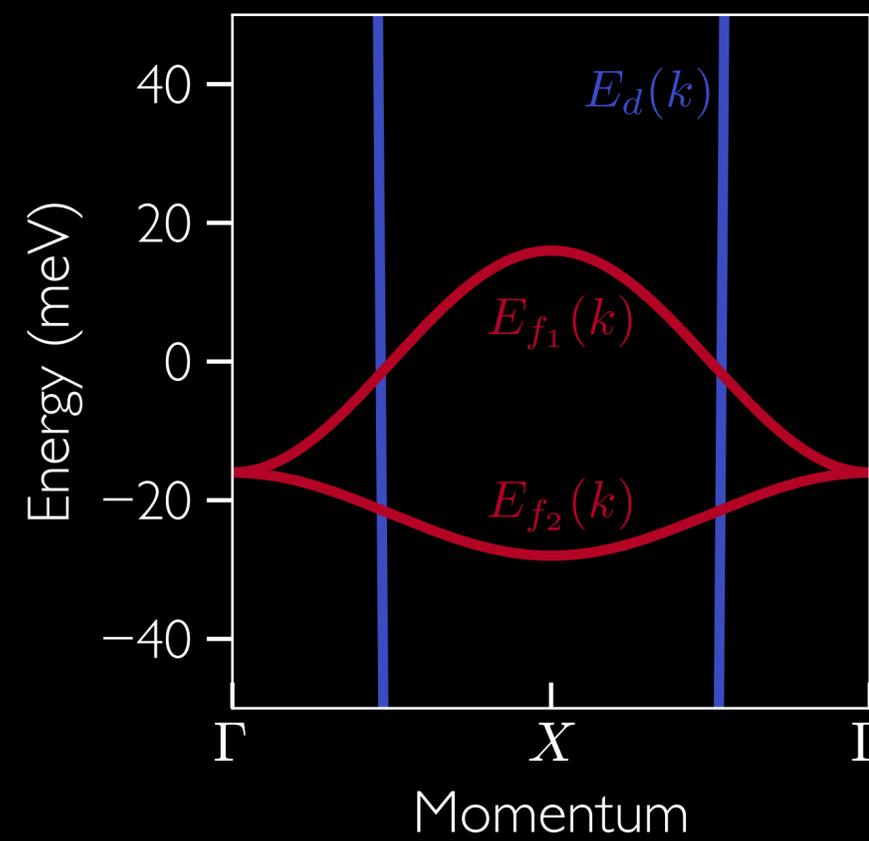
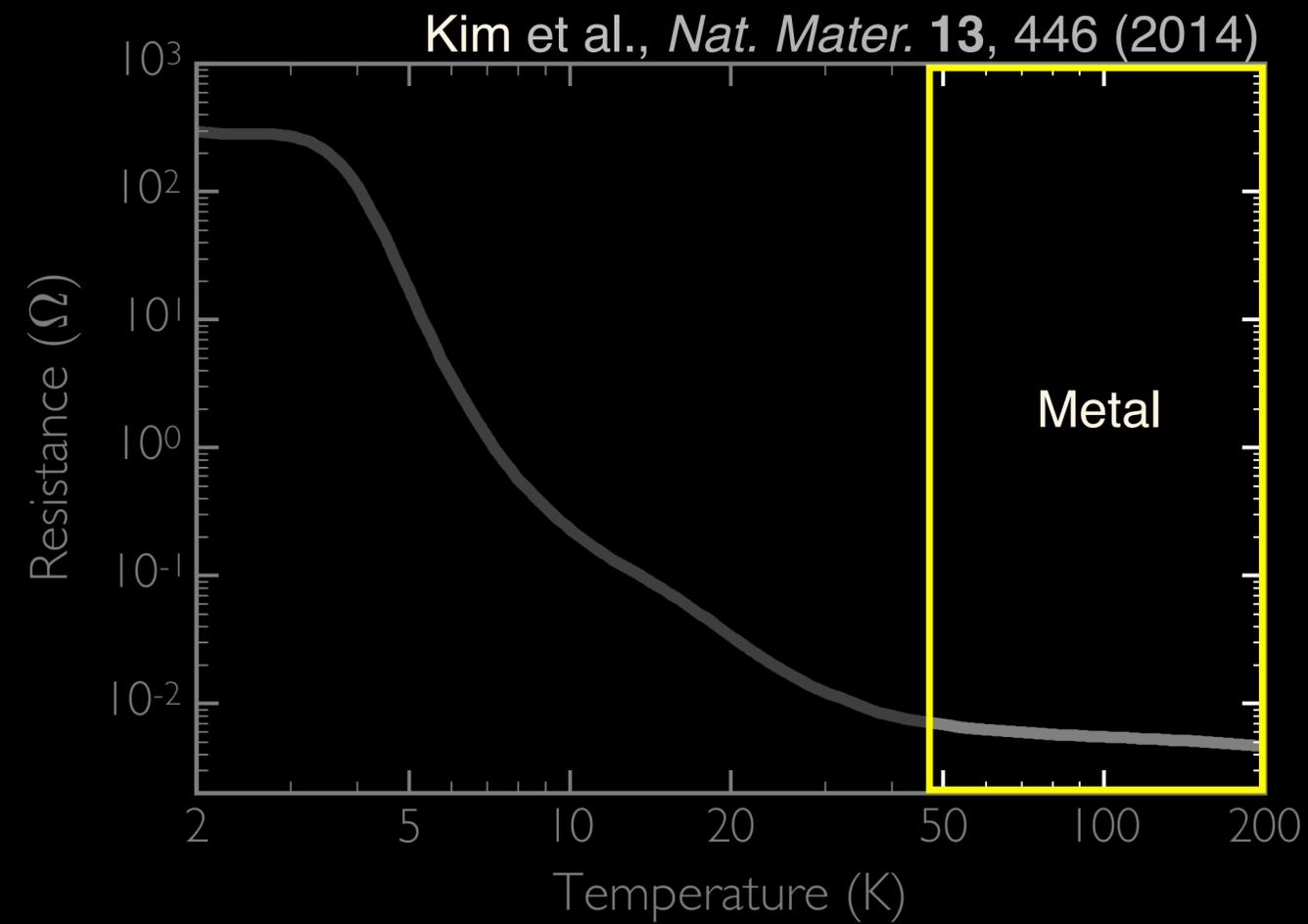
# SmB<sub>6</sub> is a high-temperature metal



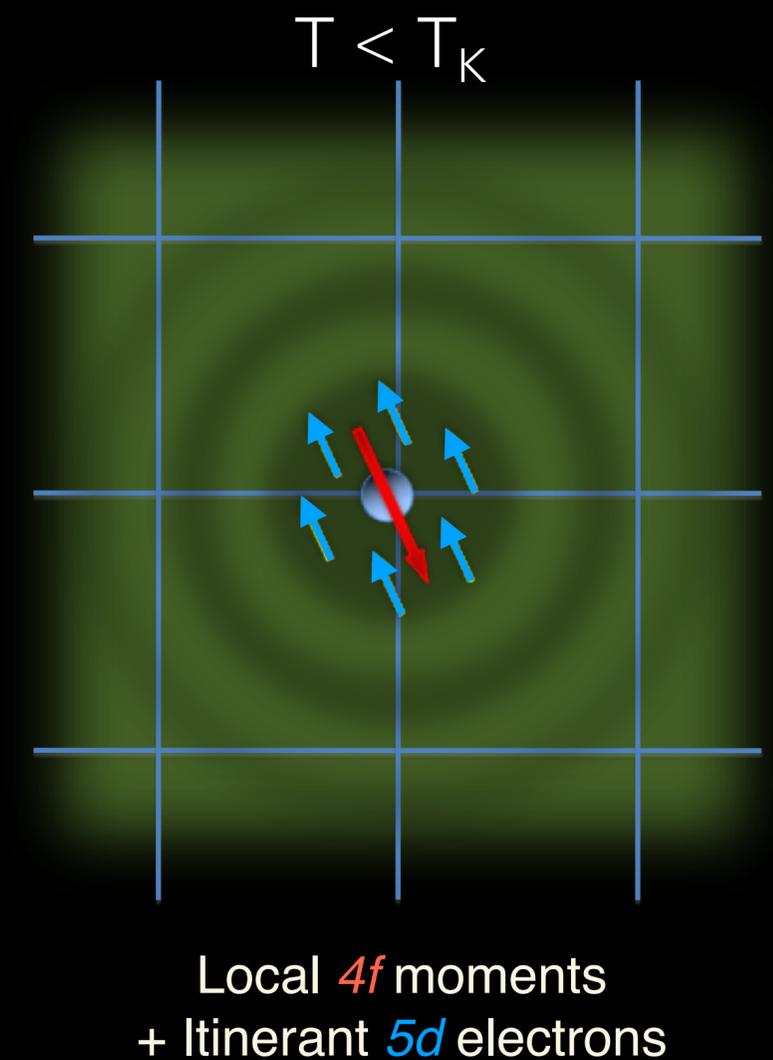
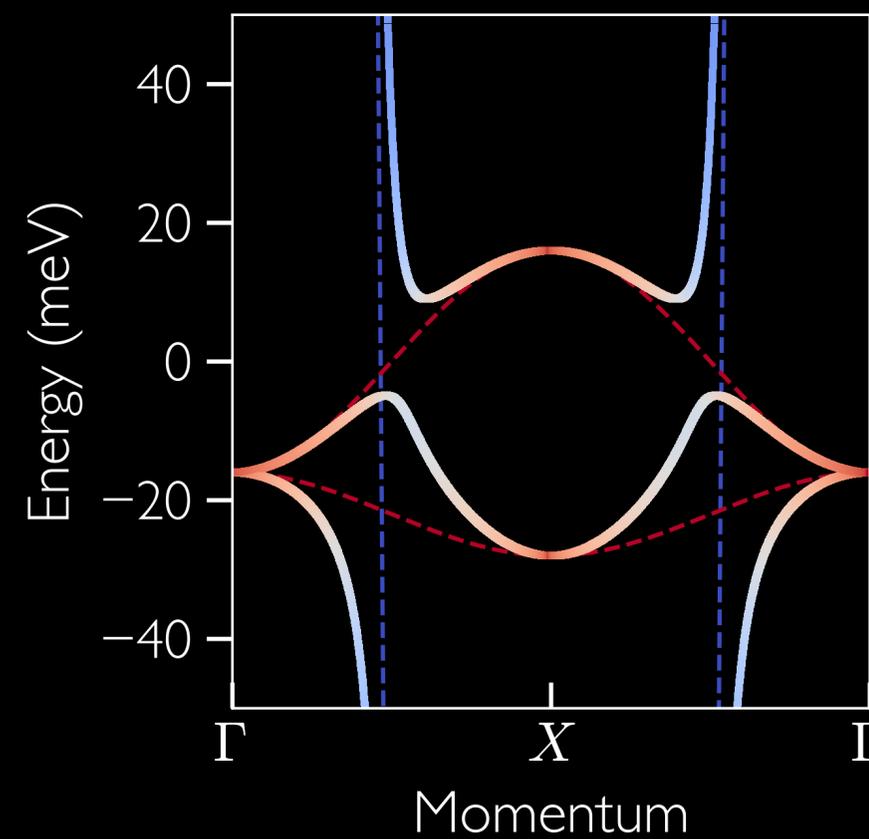
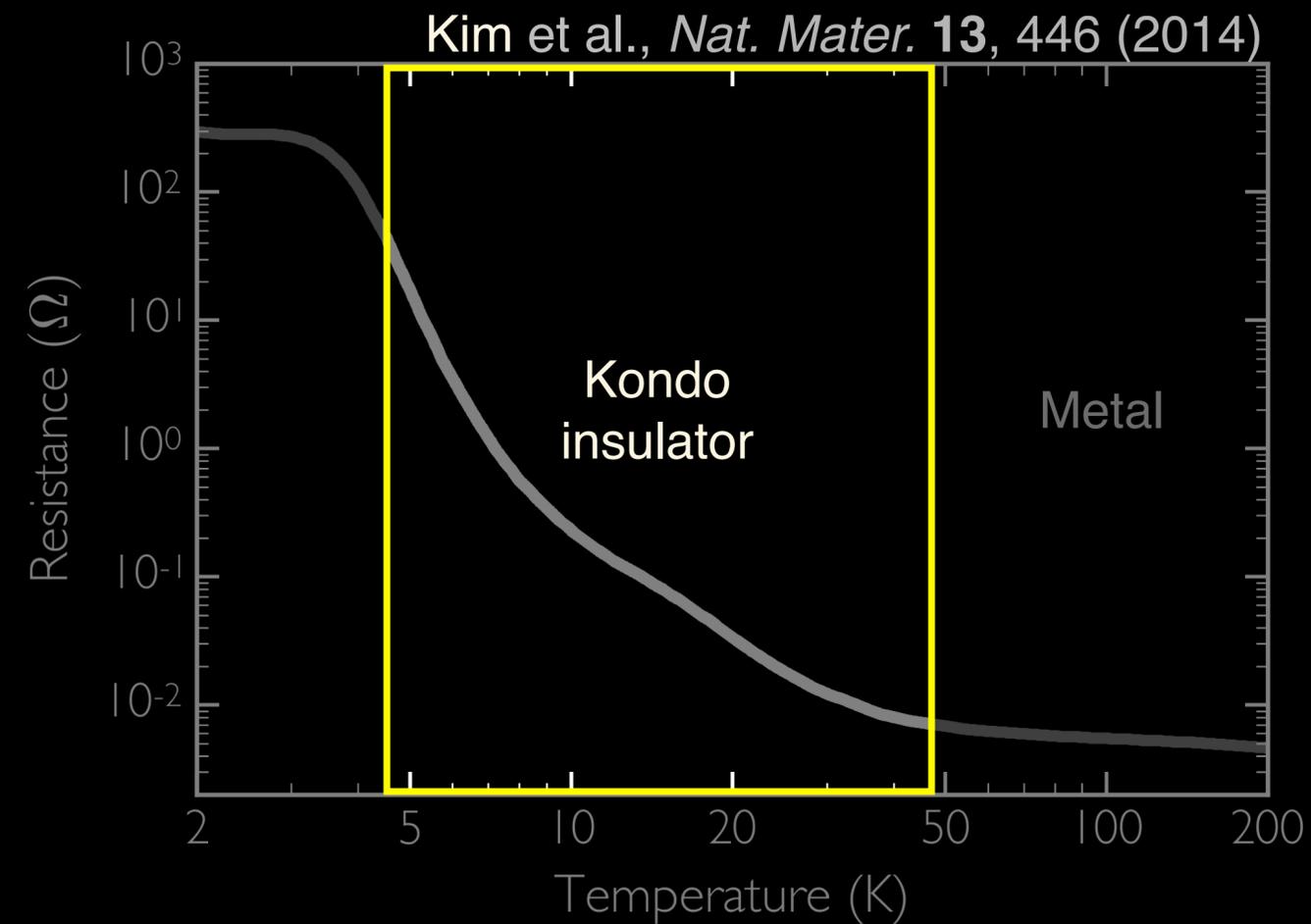
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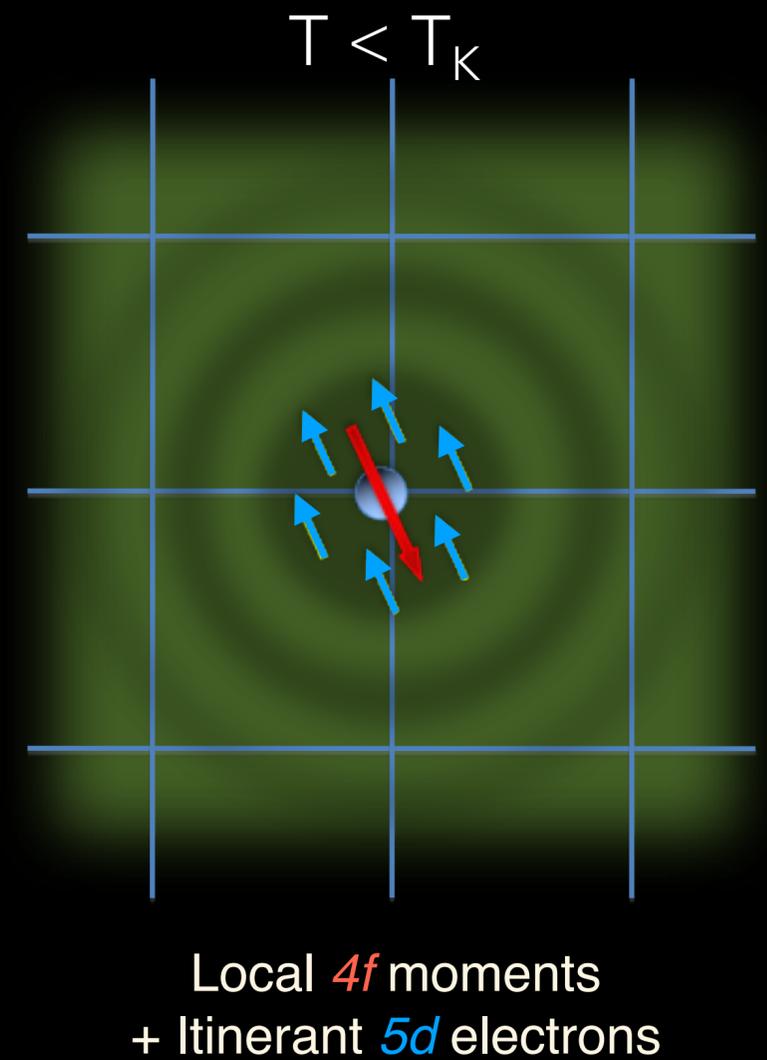
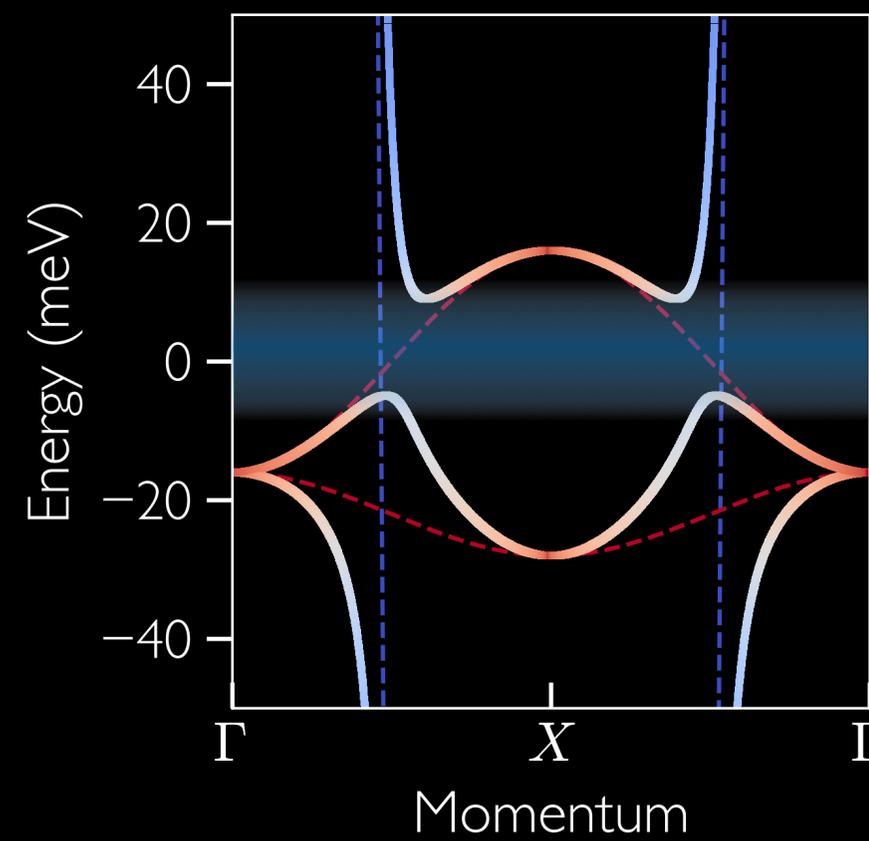
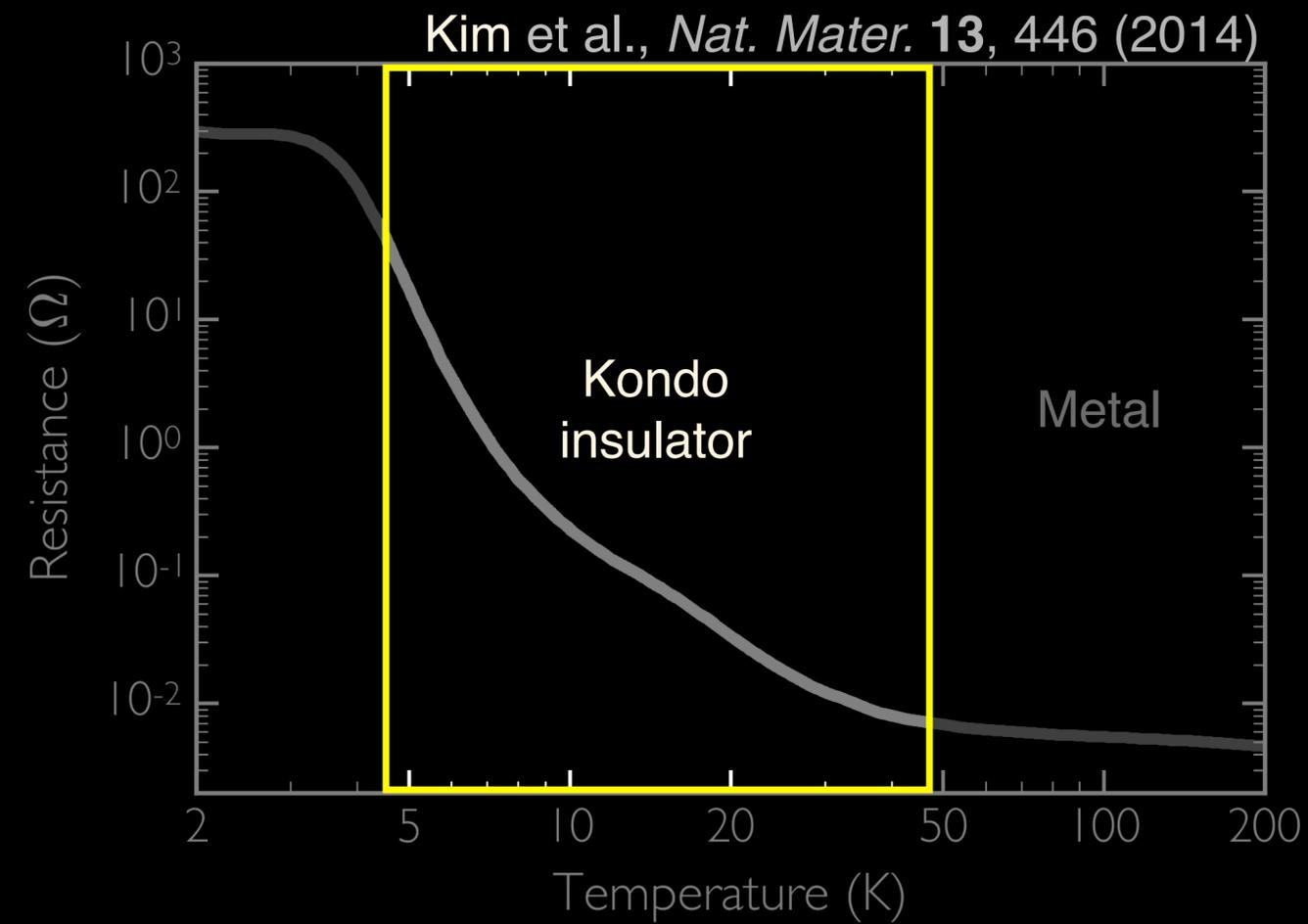
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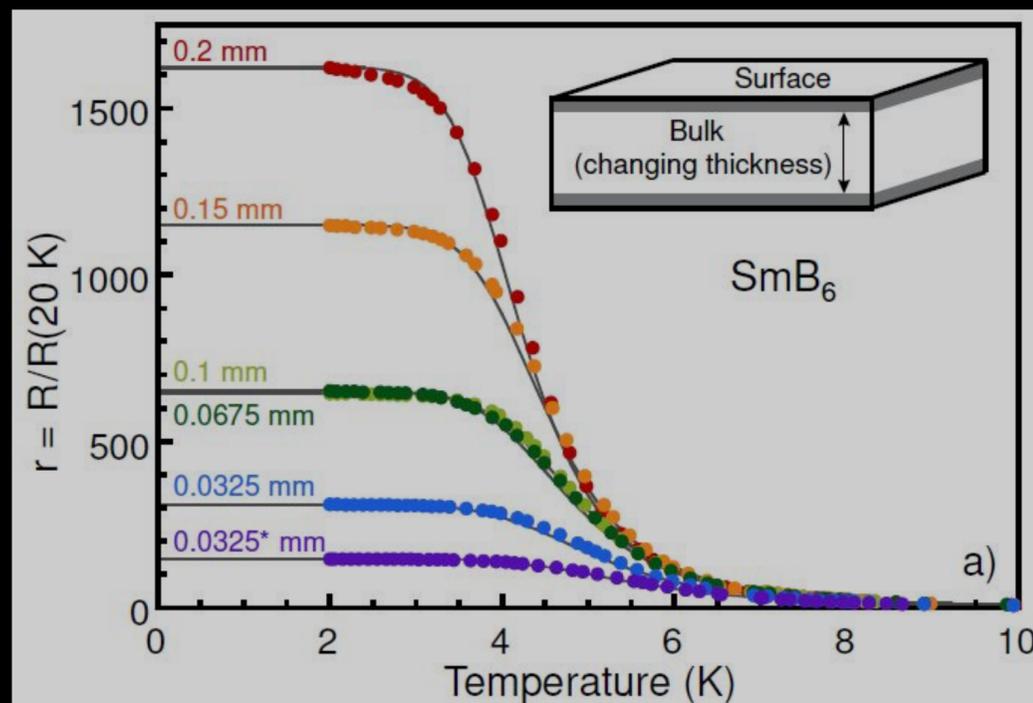
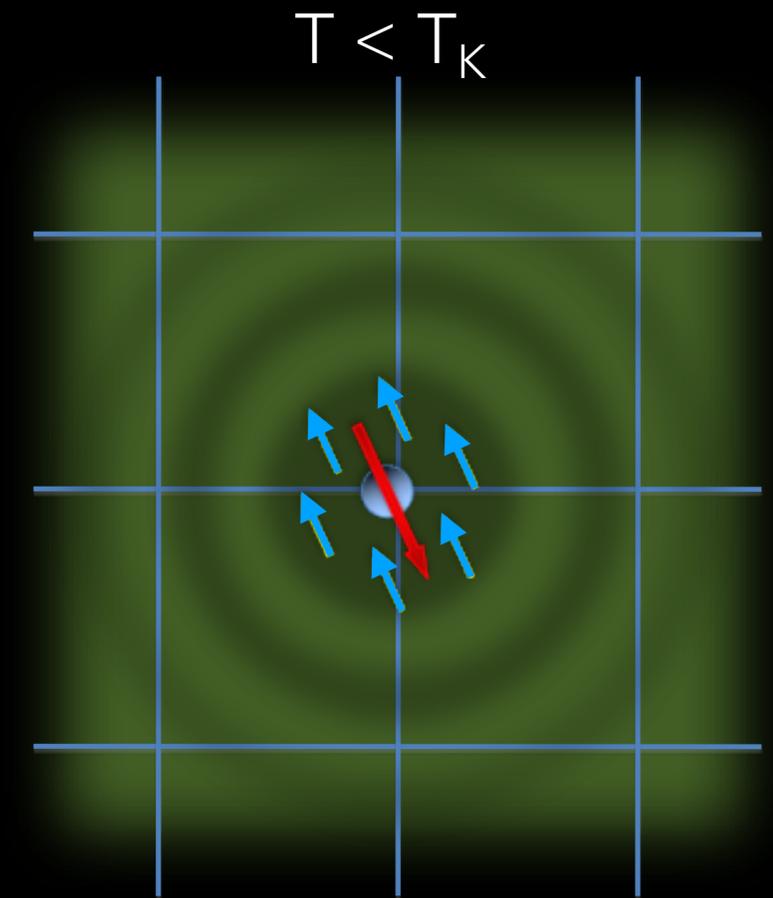
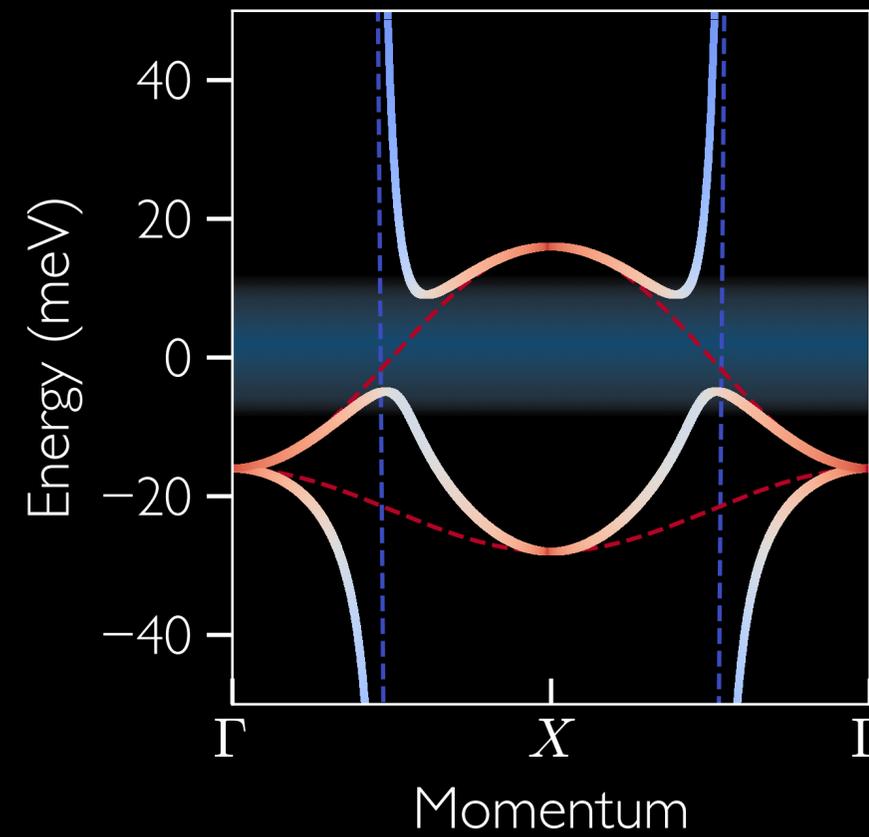
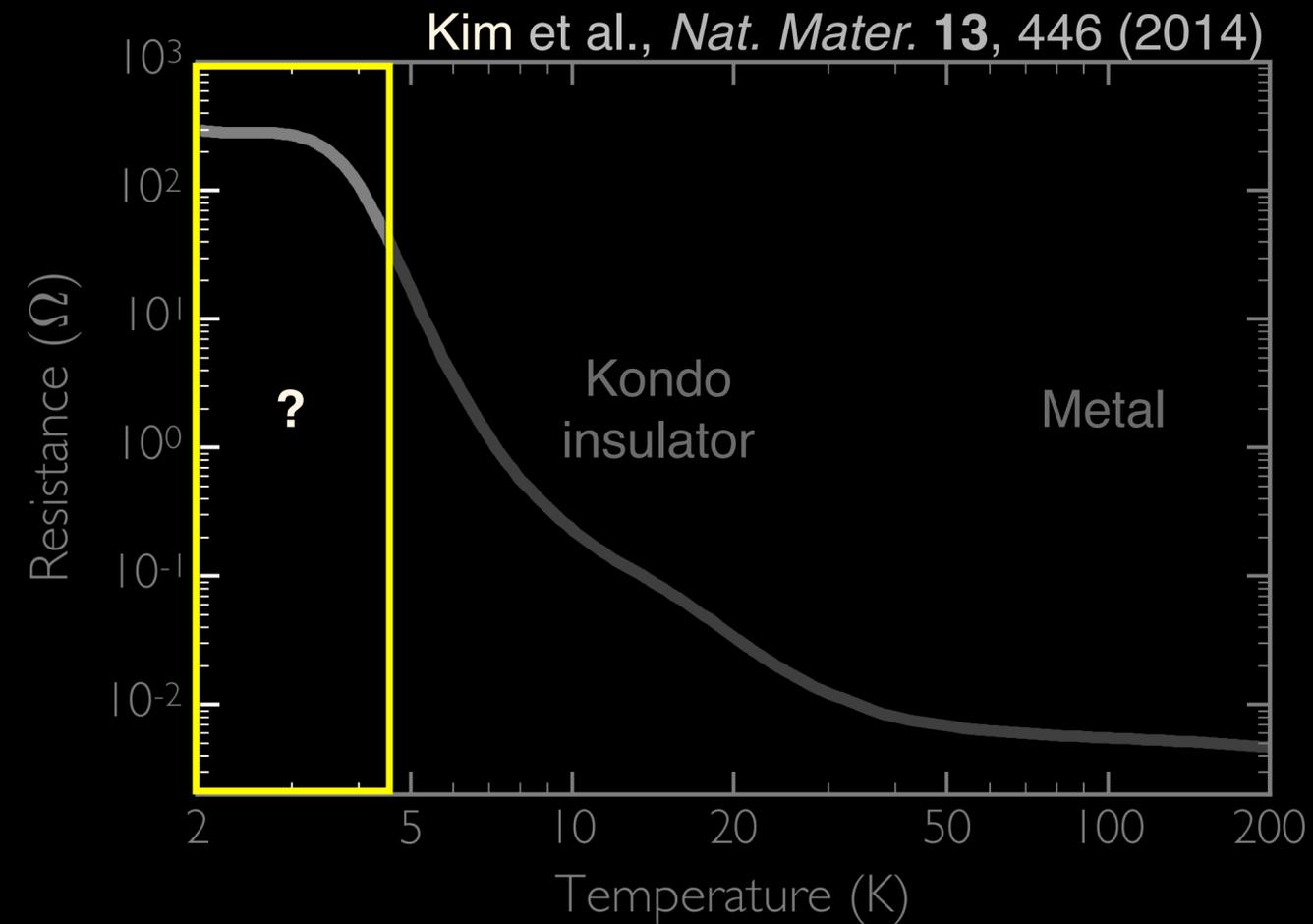
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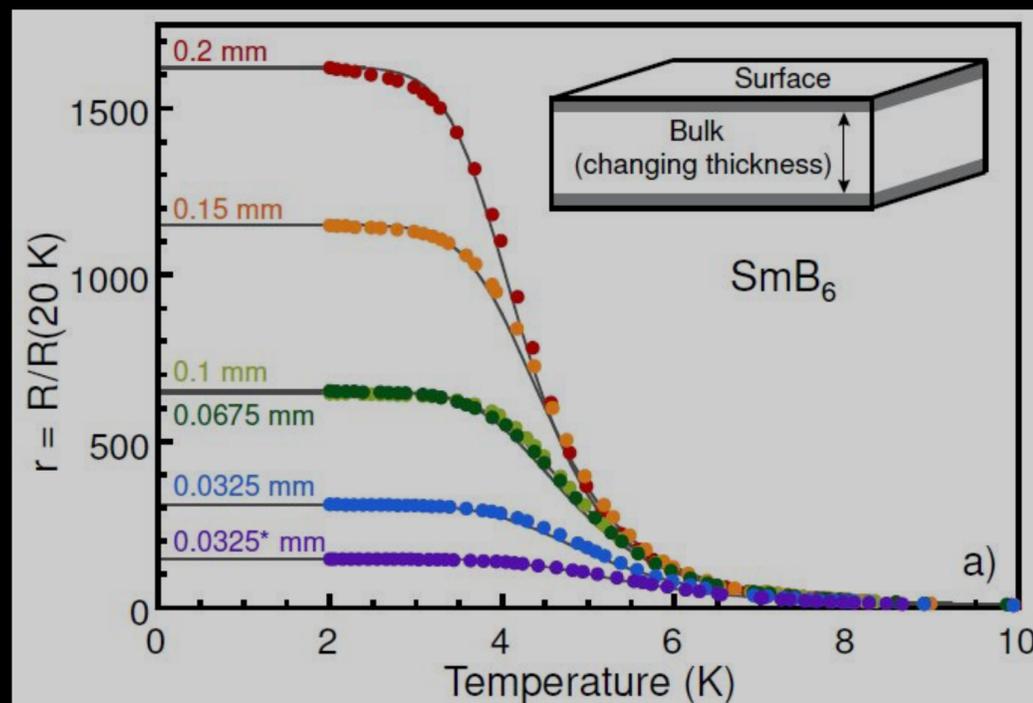
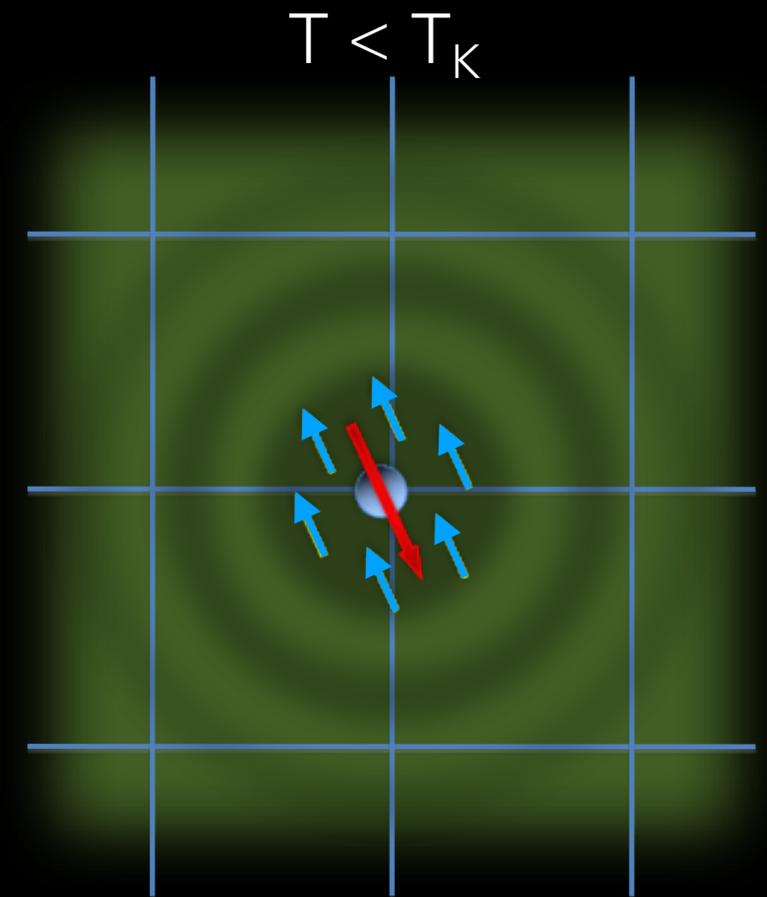
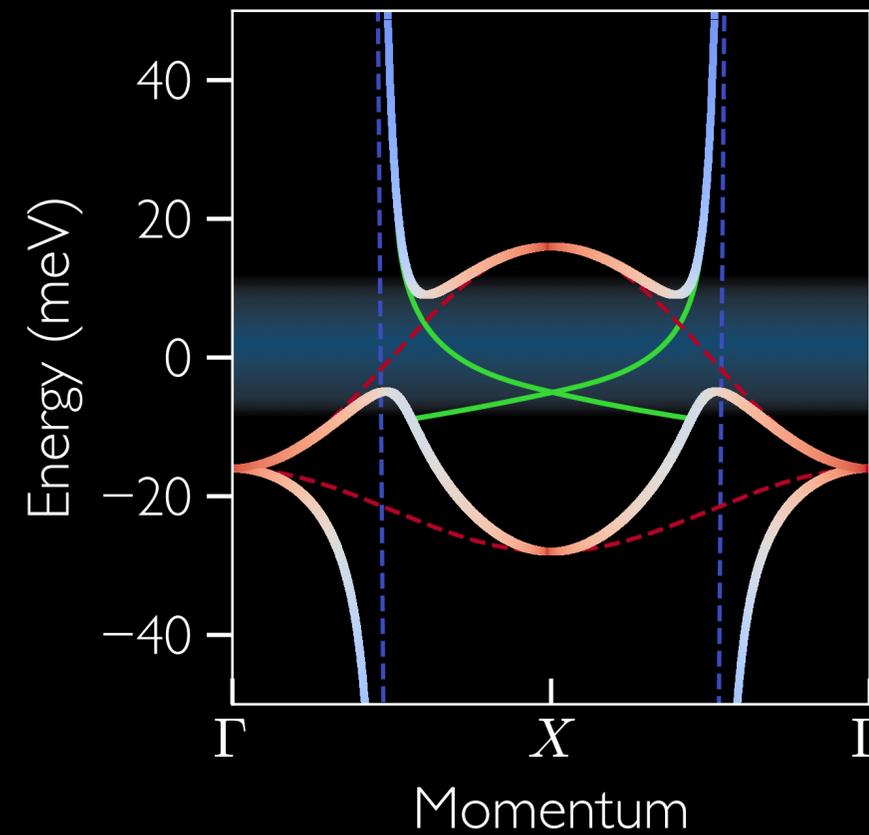
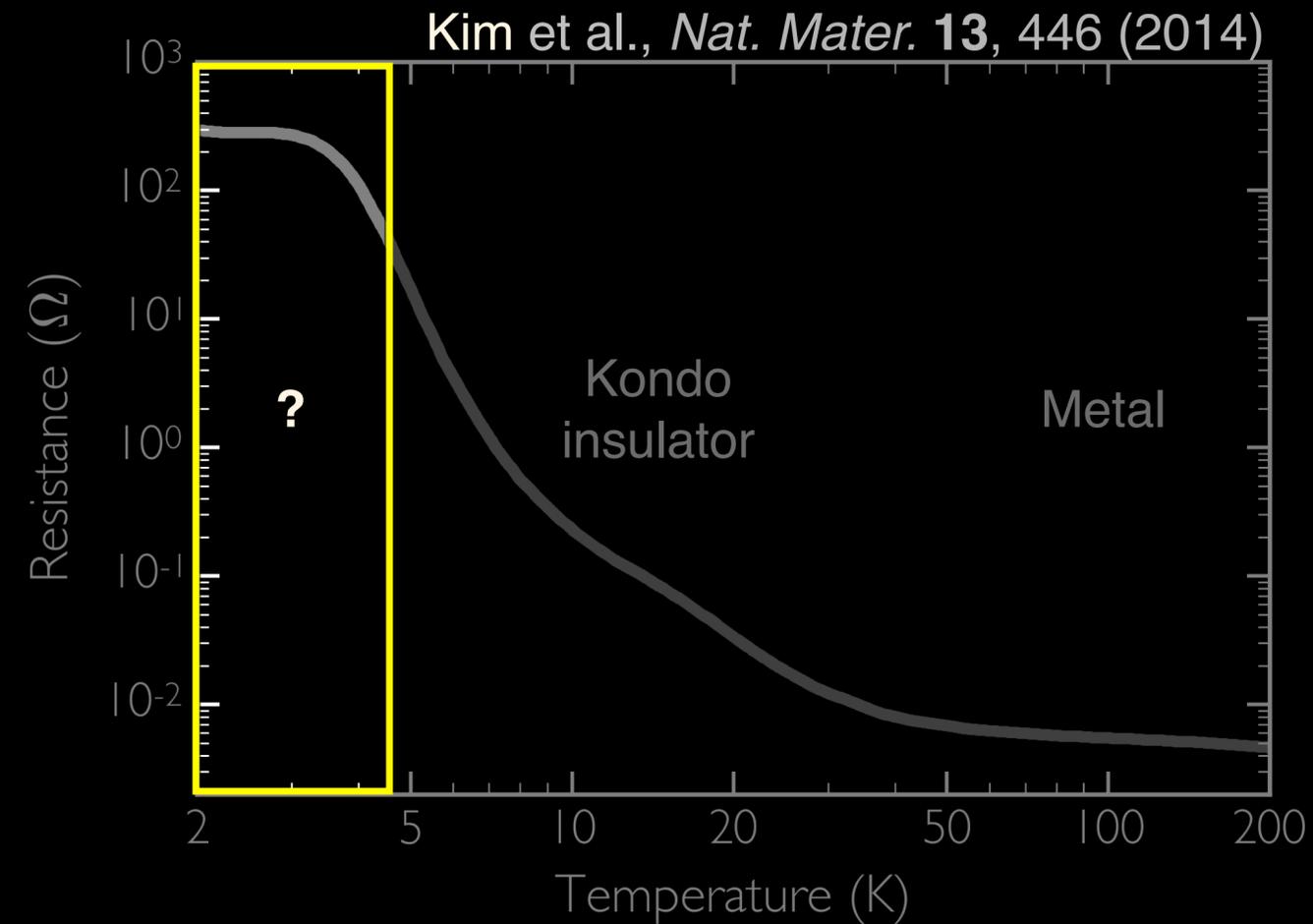


# Low- $T$ conductivity dominated by surface



Syers et al., *Phys. Rev. Lett.* **114**, 096601 (2015)

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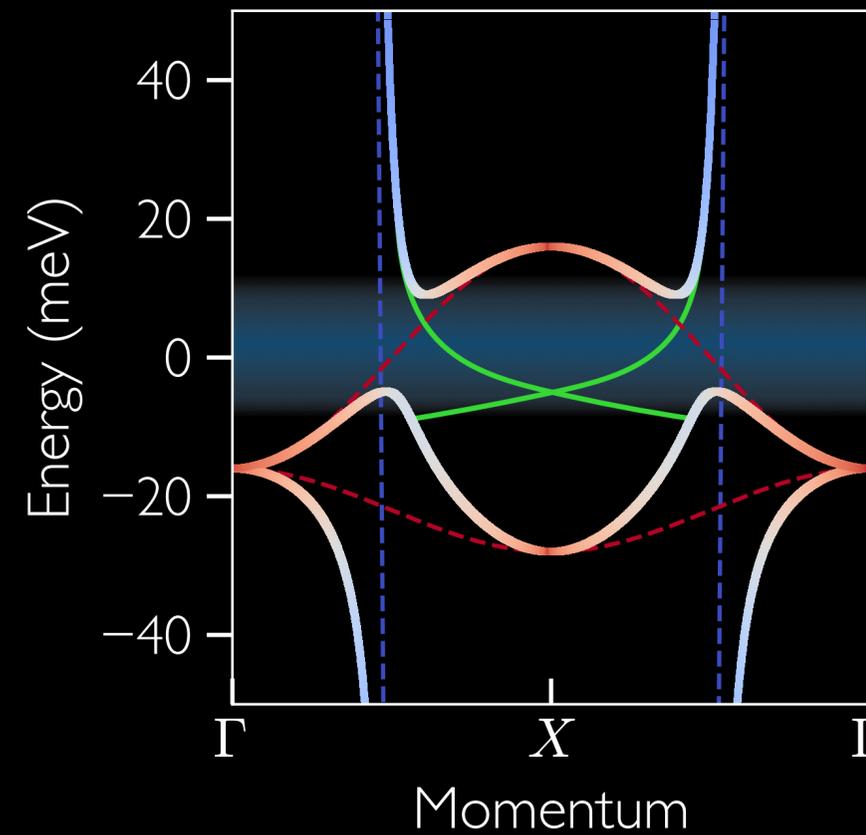
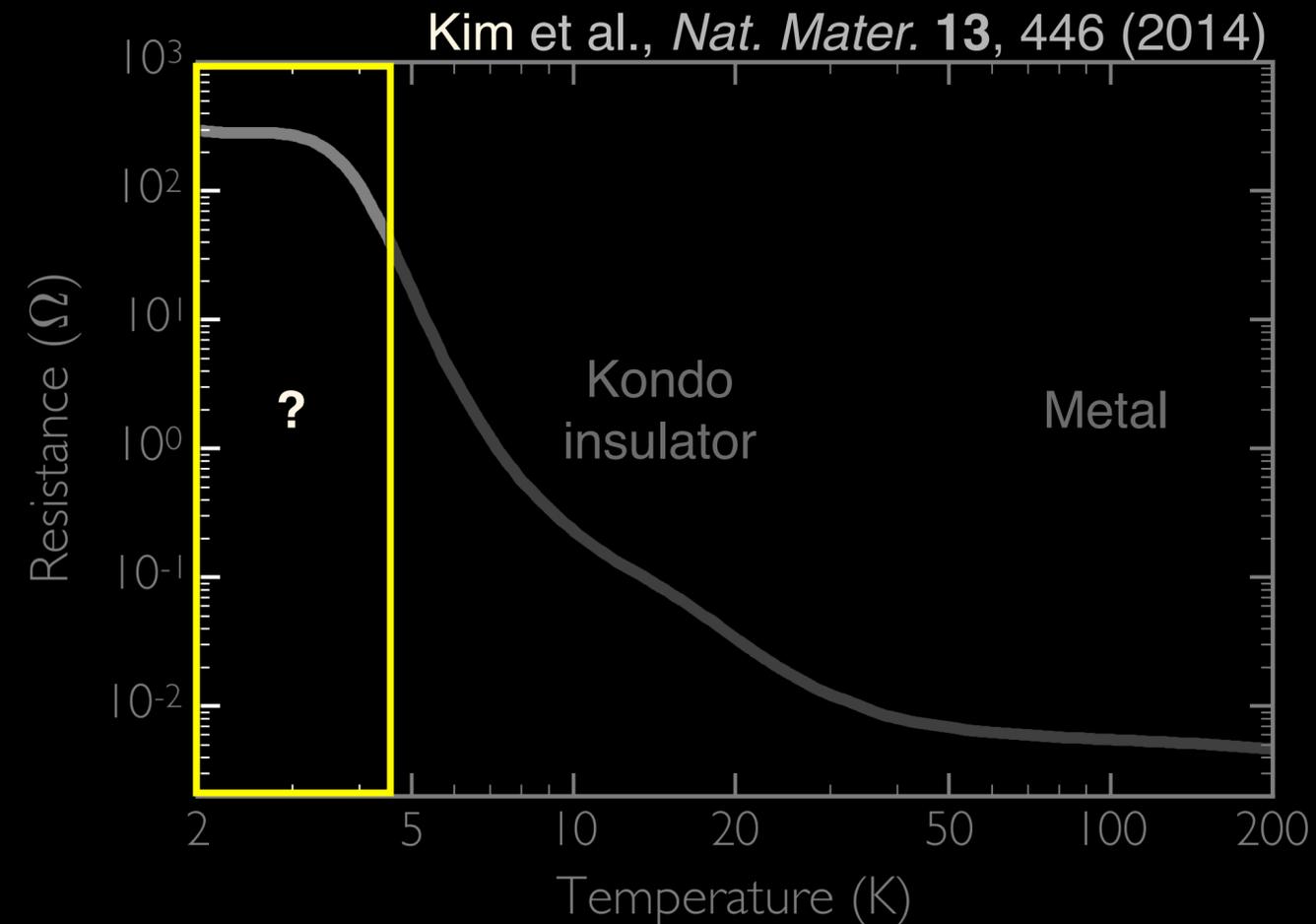
Prediction: **Strong topological insulator**

Dzero et al., *Phys. Rev. Lett.* (2010)

Takimoto et al., *JSPJ* (2011)

Lu et al., *Phys. Rev. Lett.* (2013)

# Low- $T$ conductivity dominated by surface



## Transport—can't measure velocity!

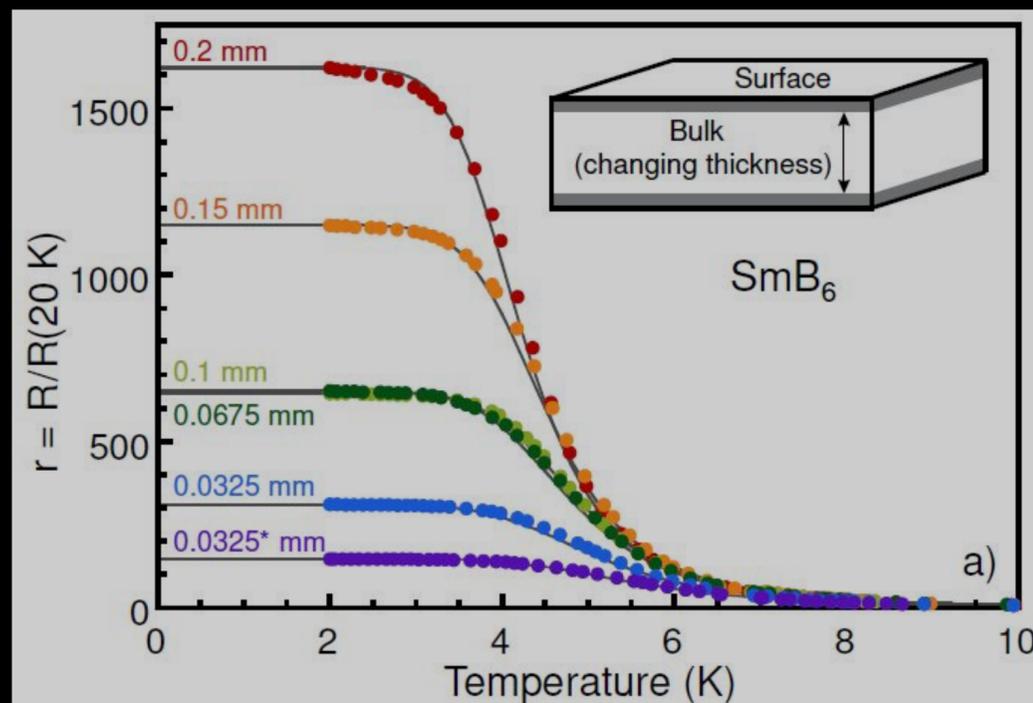
Zhang *et al*, PRX 3, 011011 (2013)  
 Wolgast *et al*, PRB 88, 180405 (2013)  
 Kim *et al*, Nat Mat 13, 3913 (2014)  
 Luo *et al*, PRB 91, 075130 (2014)  
 Syers *et al*, PRL 114, 096601 (2015)

## Photoemission (ARPES)—10x too fast!

Miyazaki *et al*, PRB 86, 075105 (2012)  
 Xu *et al*, PRB 88, 121102 (2013)  
 Jiang *et al*, Nat Com 4, 3010 (2013)  
 Neupane *et al*, Nat Com 4, 2991 (2013)  
 Denlinger *et al*, arXiv:1312.6637 (2013)  
 Suga *et al*, JPSJ 83, 014705 (2014)  
 Xu *et al*, Nat Com 5, 5566 (2014)

## Quantum oscillations (dHvA)—100x too fast!

Li *et al*, Science 346, 6214 (2014)

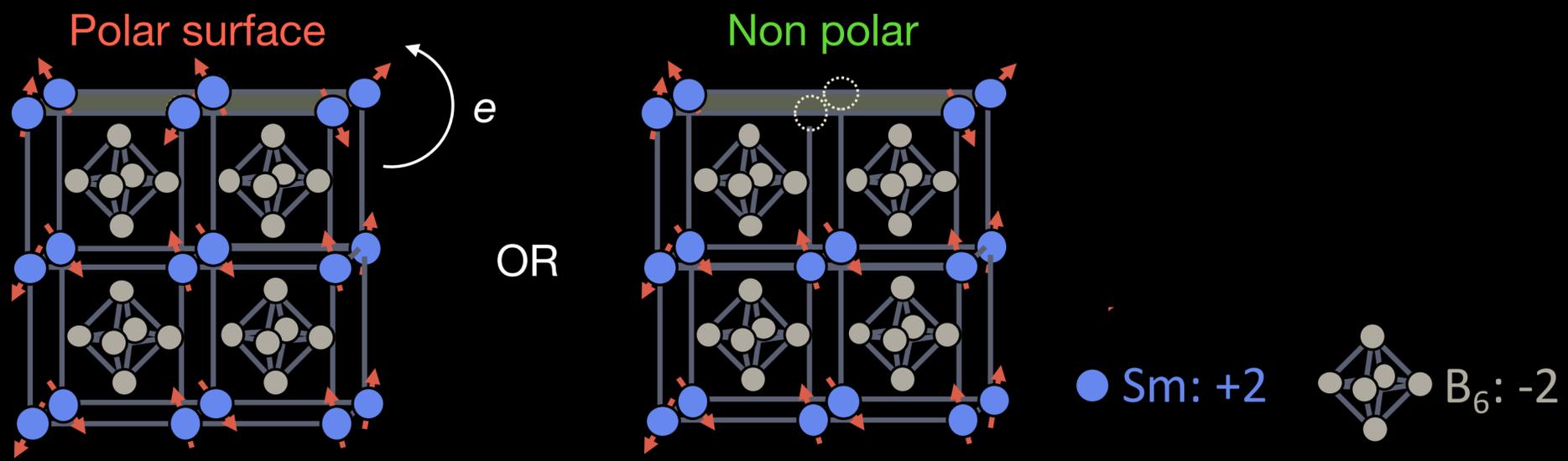
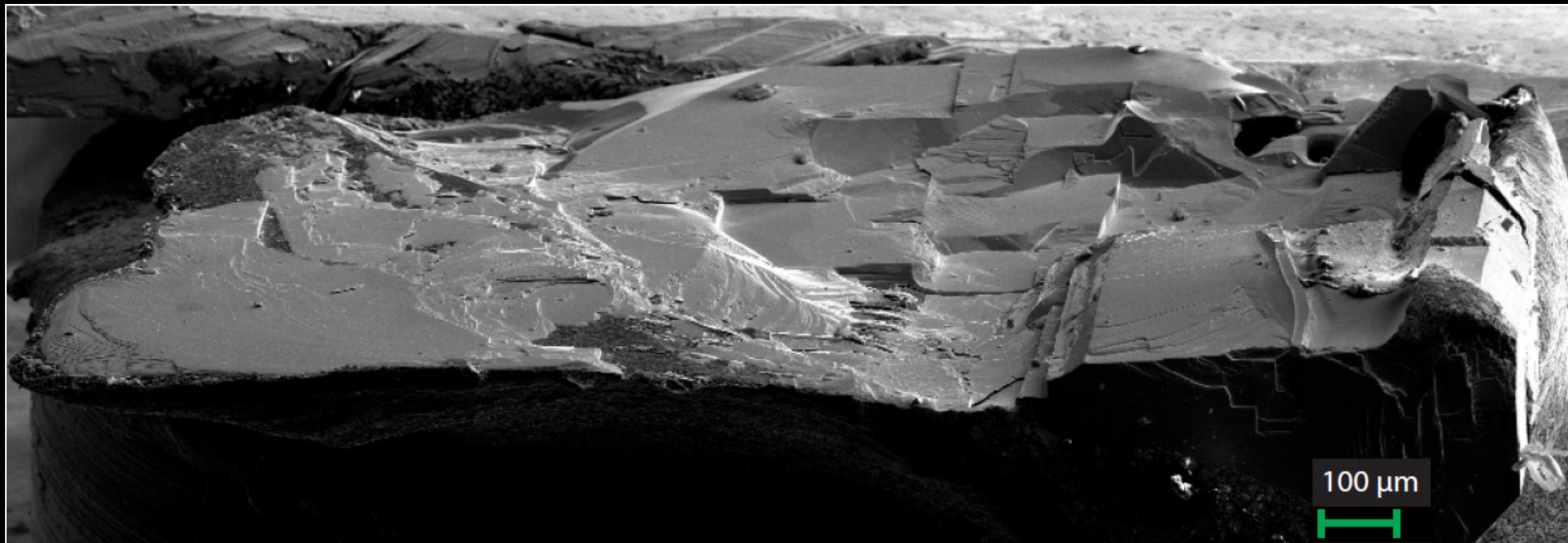


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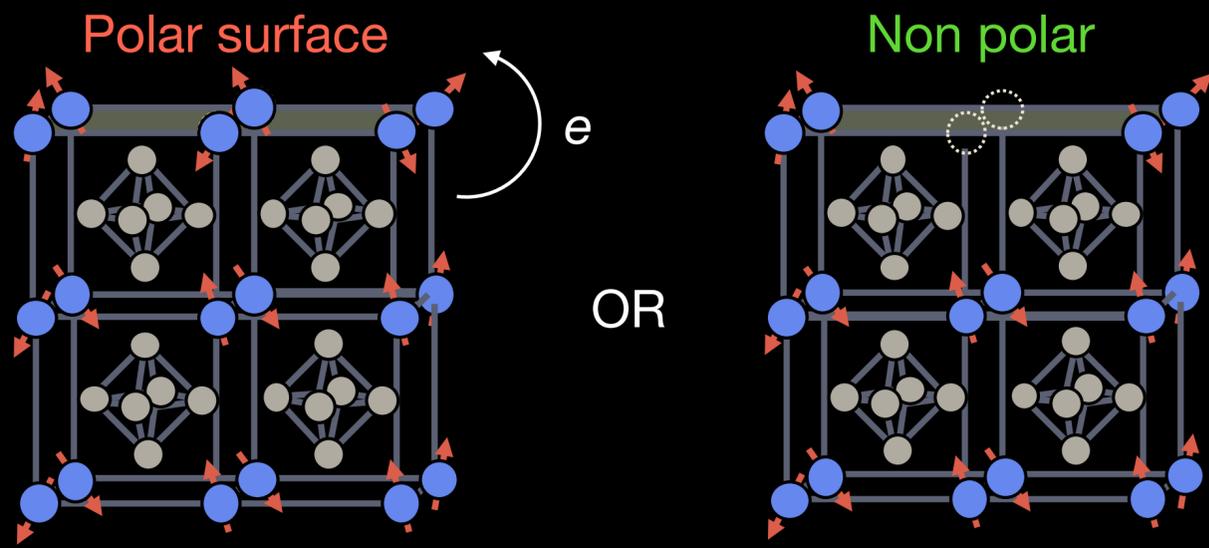
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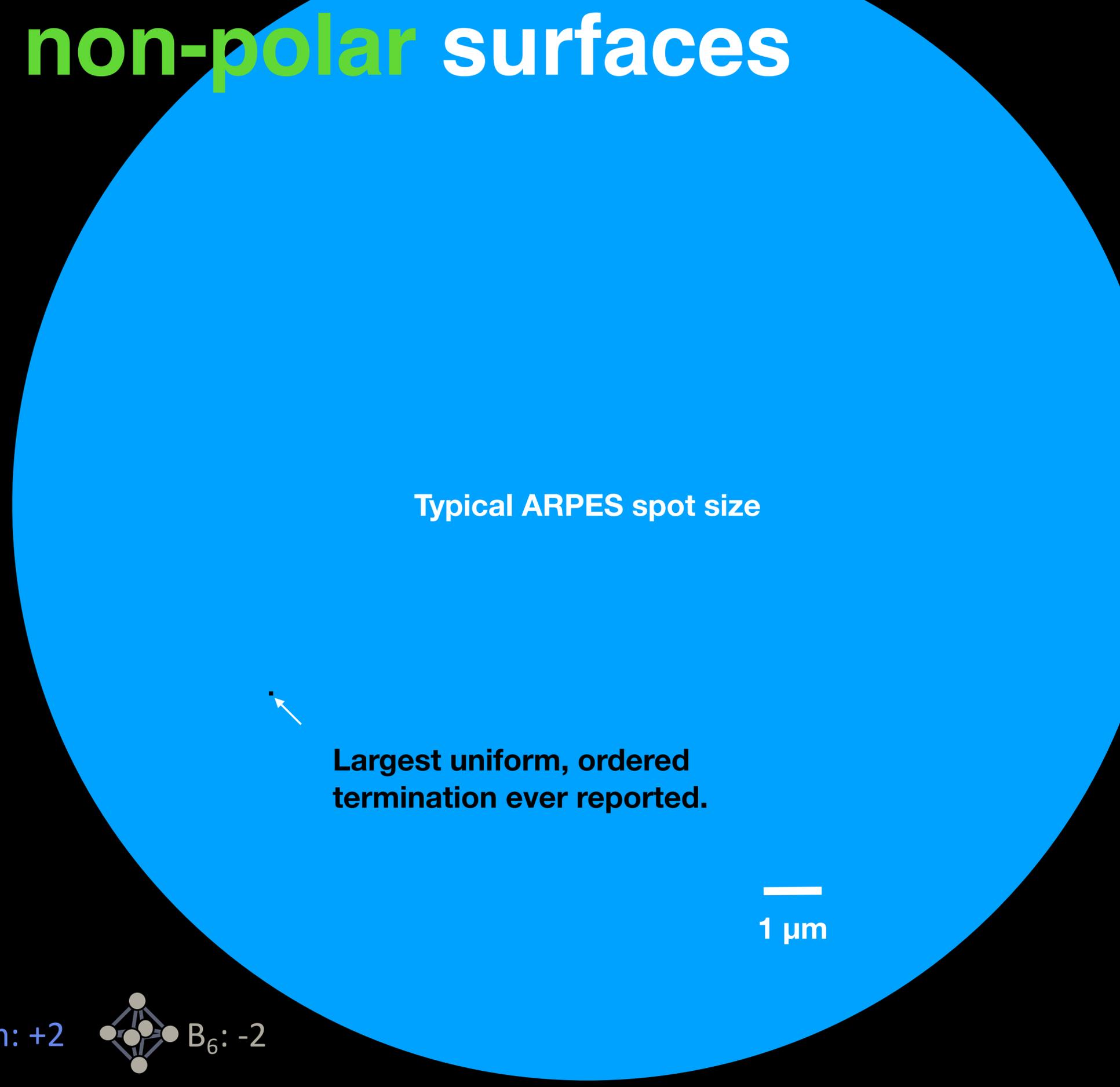
# SmB<sub>6</sub> has **polar** and **non-polar** surfaces



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● Sm: +2     B<sub>6</sub>: -2

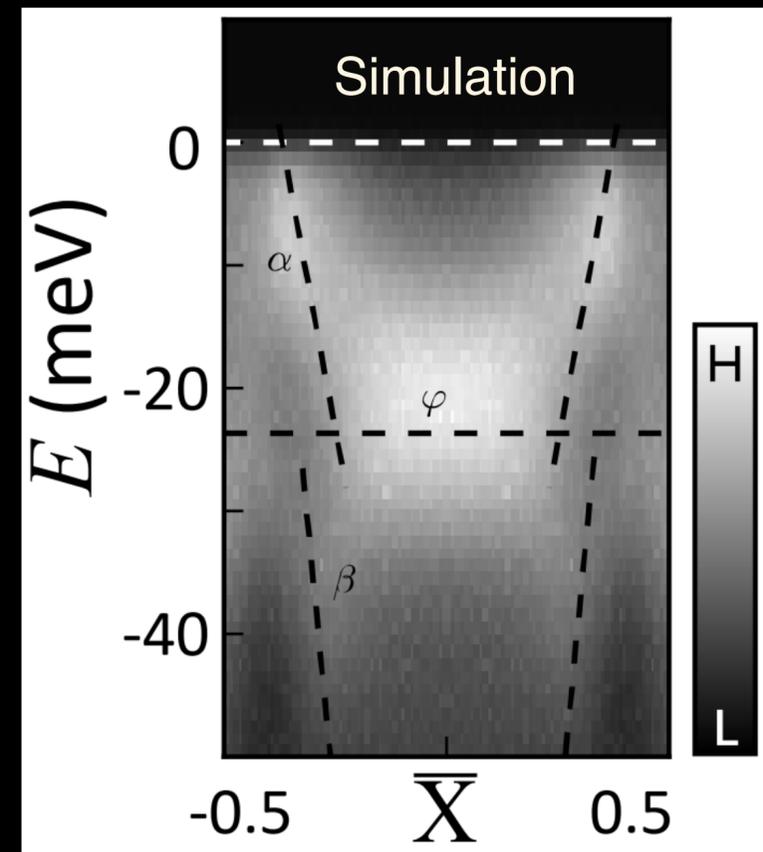


Typical ARPES spot size

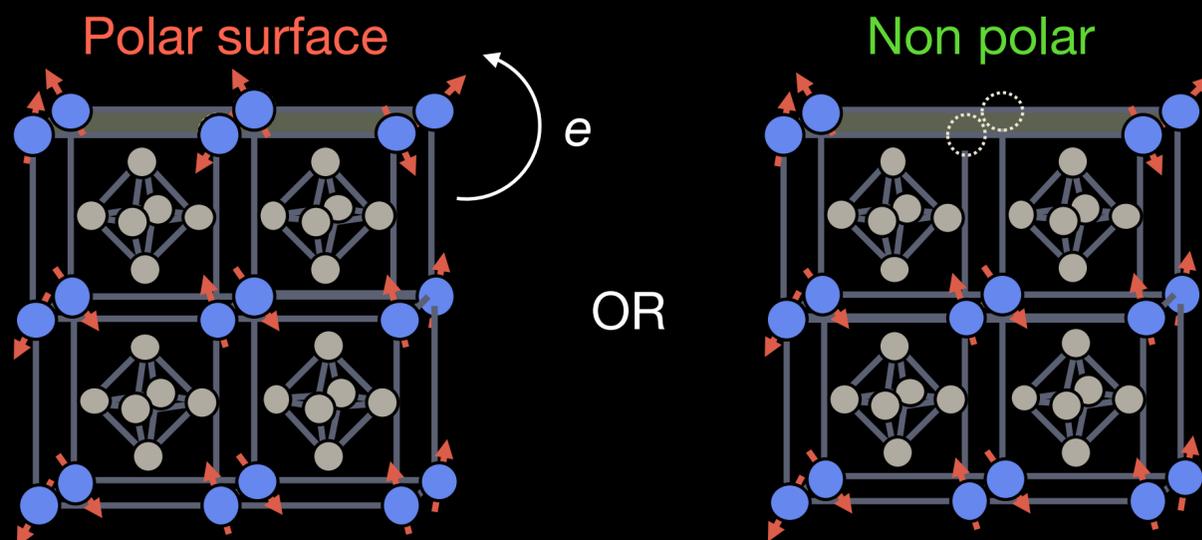
Largest uniform, ordered termination ever reported.

1 μm

# Simulated APRES has *light* surface states



Matt et al., *Phys. Rev. B* 101, 085142 (2020)



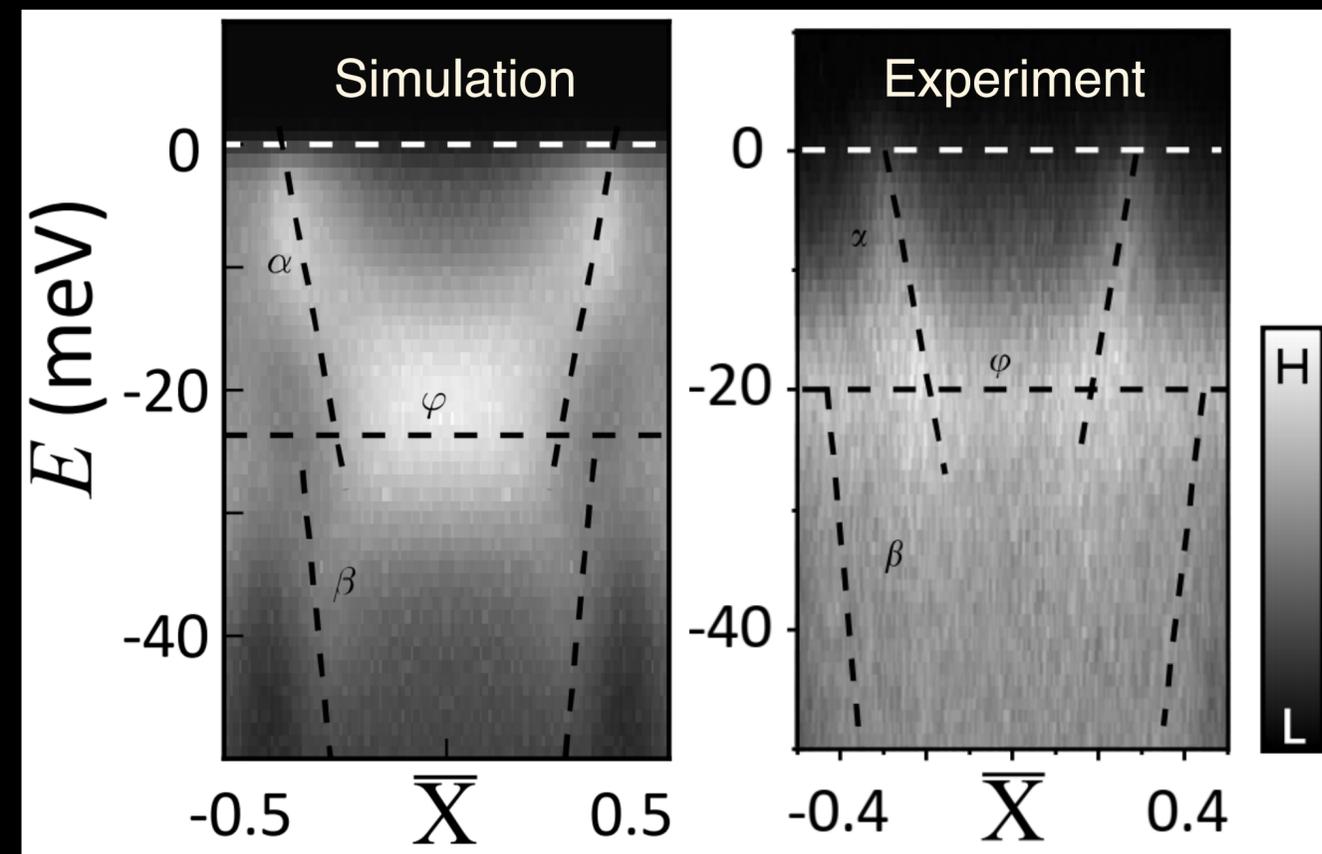
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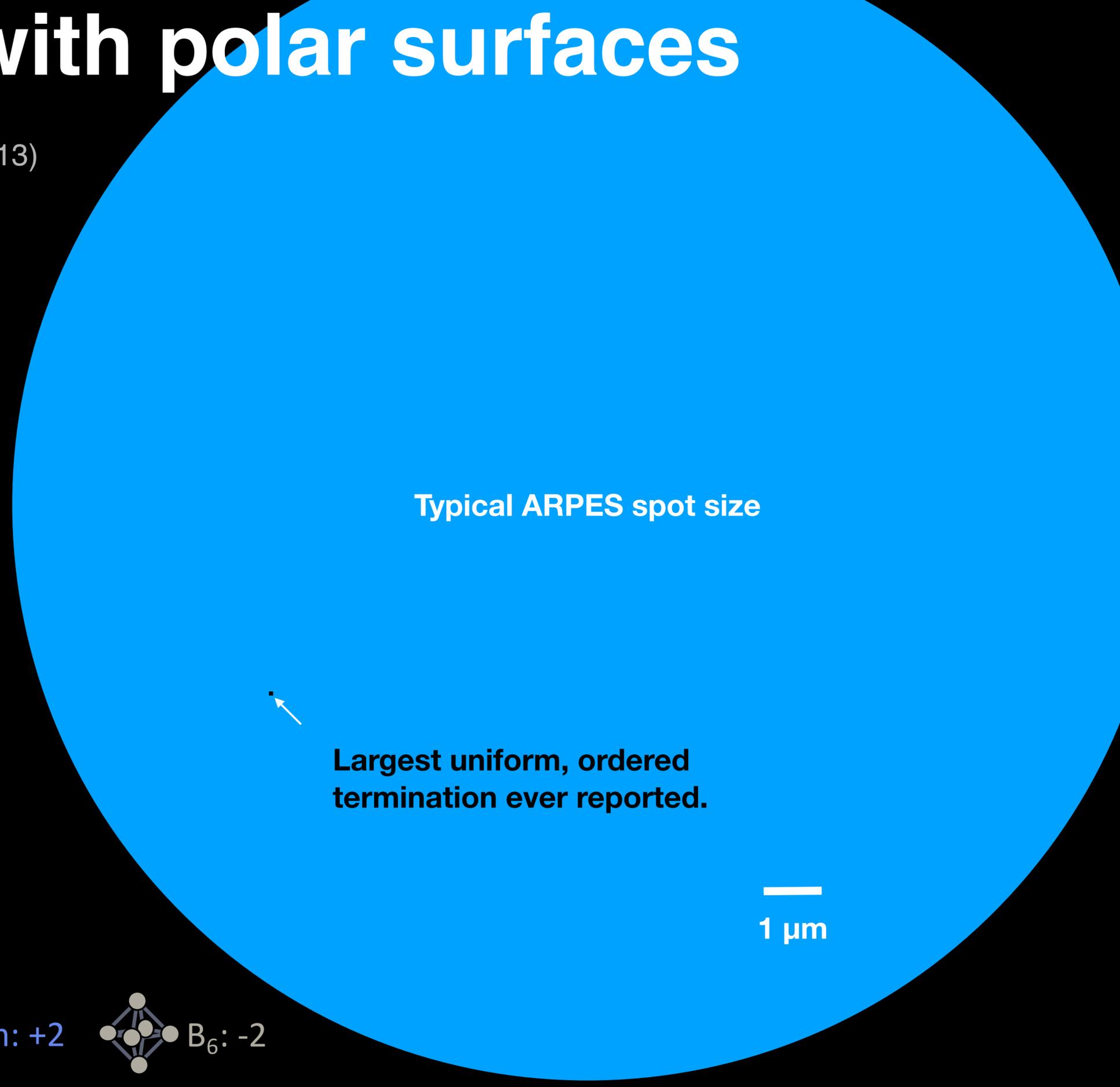
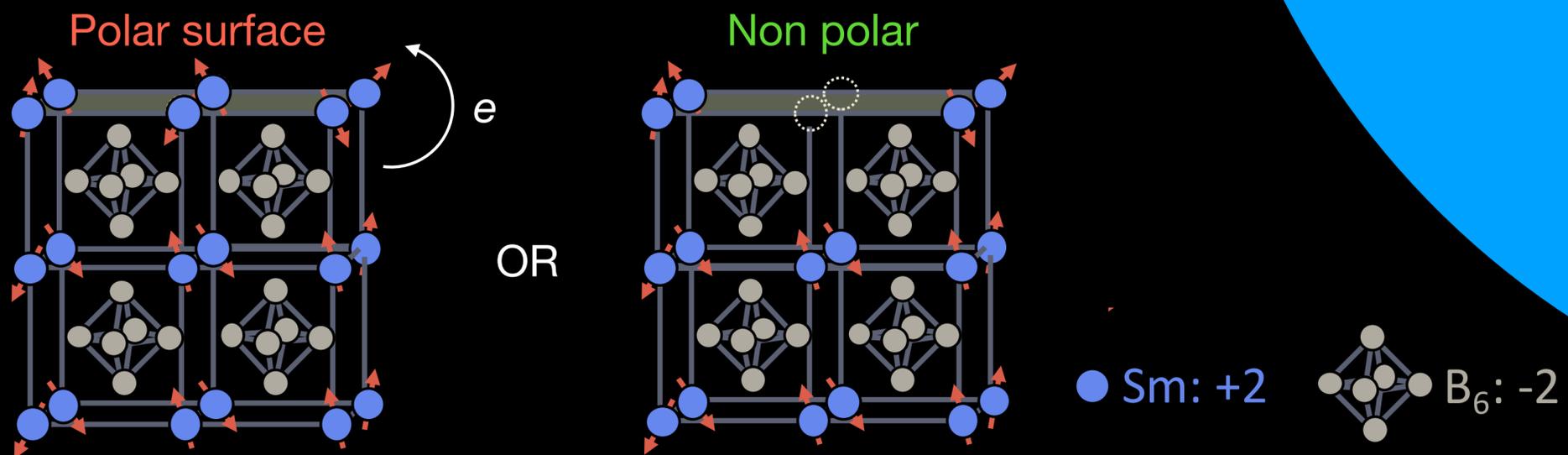
1  $\mu\text{m}$

# ARPES consistent with polar surfaces

Jiang et al., *Nat. Commun.* 4, 3010 (2013)



Matt et al., *Phys. Rev. B* 101, 085142 (2020)



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1  $\mu\text{m}$

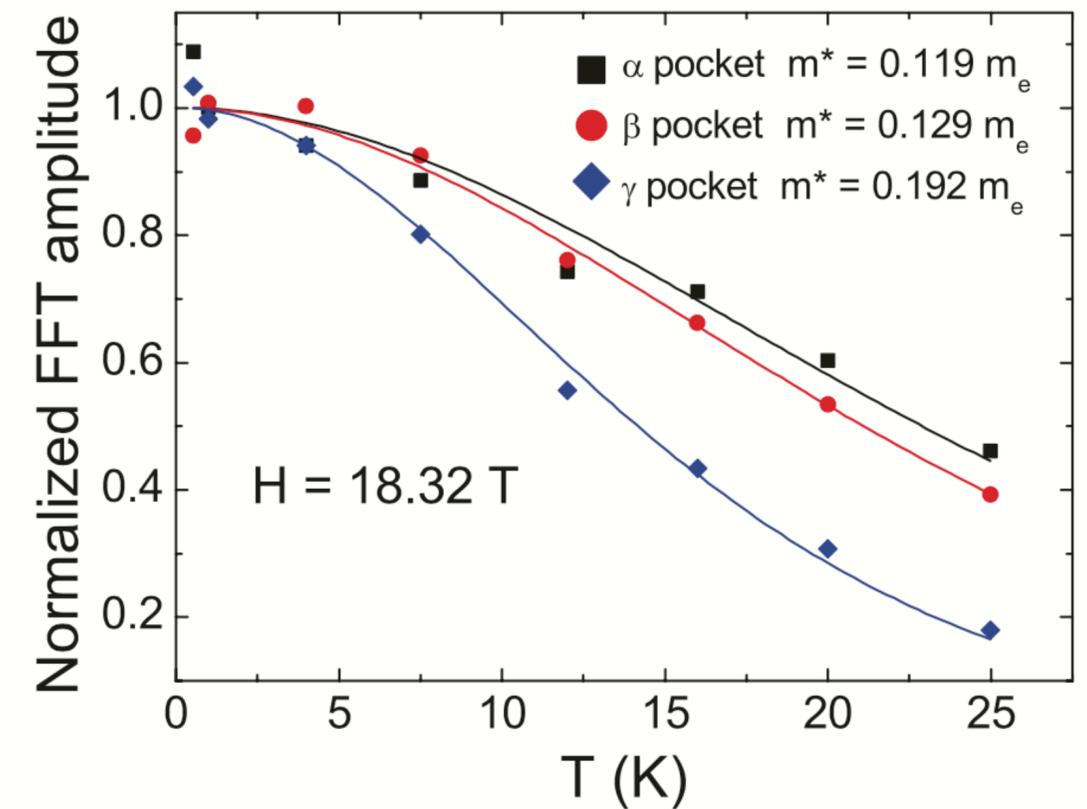
# dHvA measures *light* Dirac fermions?

Li et al., *Science*. 346, 1208 (2014)

## HEAVY FERMIONS

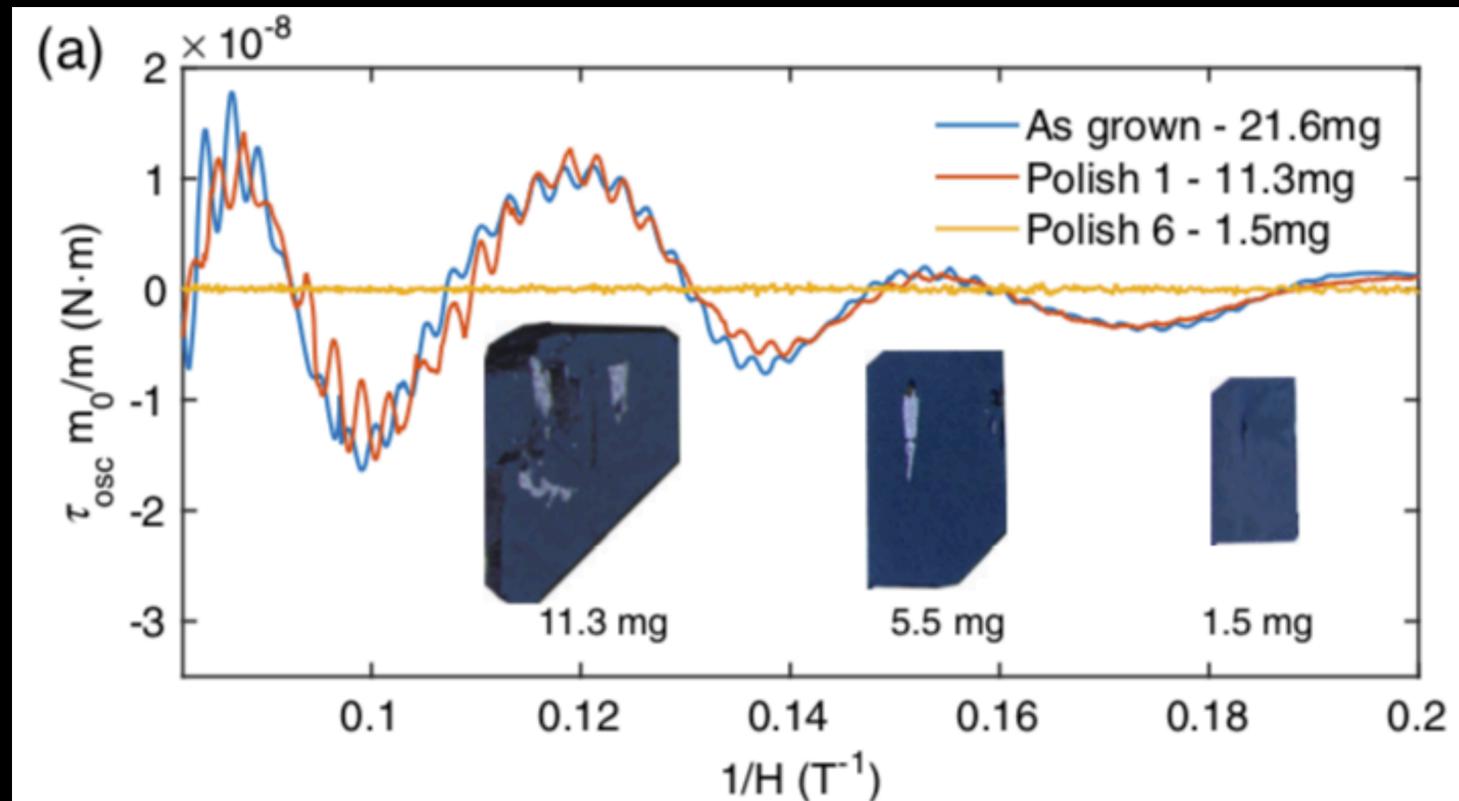
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# dHvA measures aluminum inclusions?

Thomas et al., *Phys. Rev. Lett.* **122**, 166401 (2019)



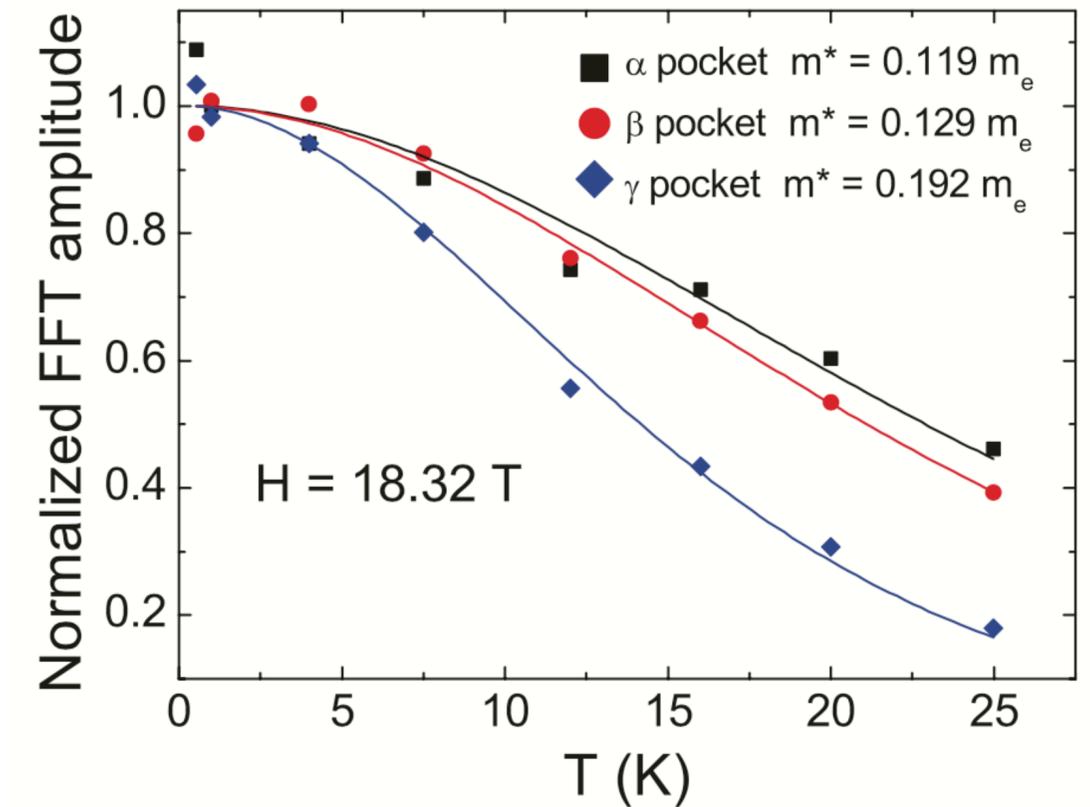
This  $\text{SmB}_6$  is grown with **aluminum flux**

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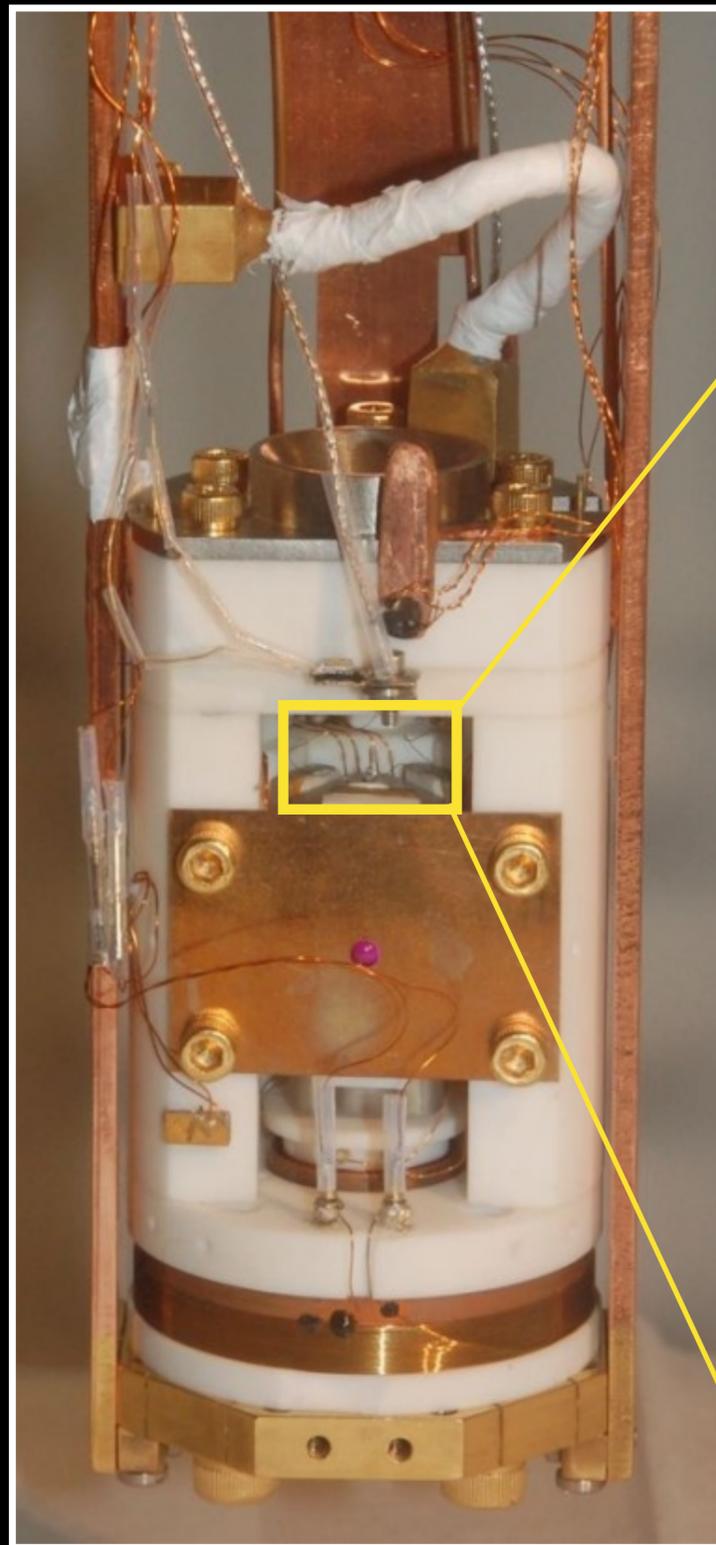
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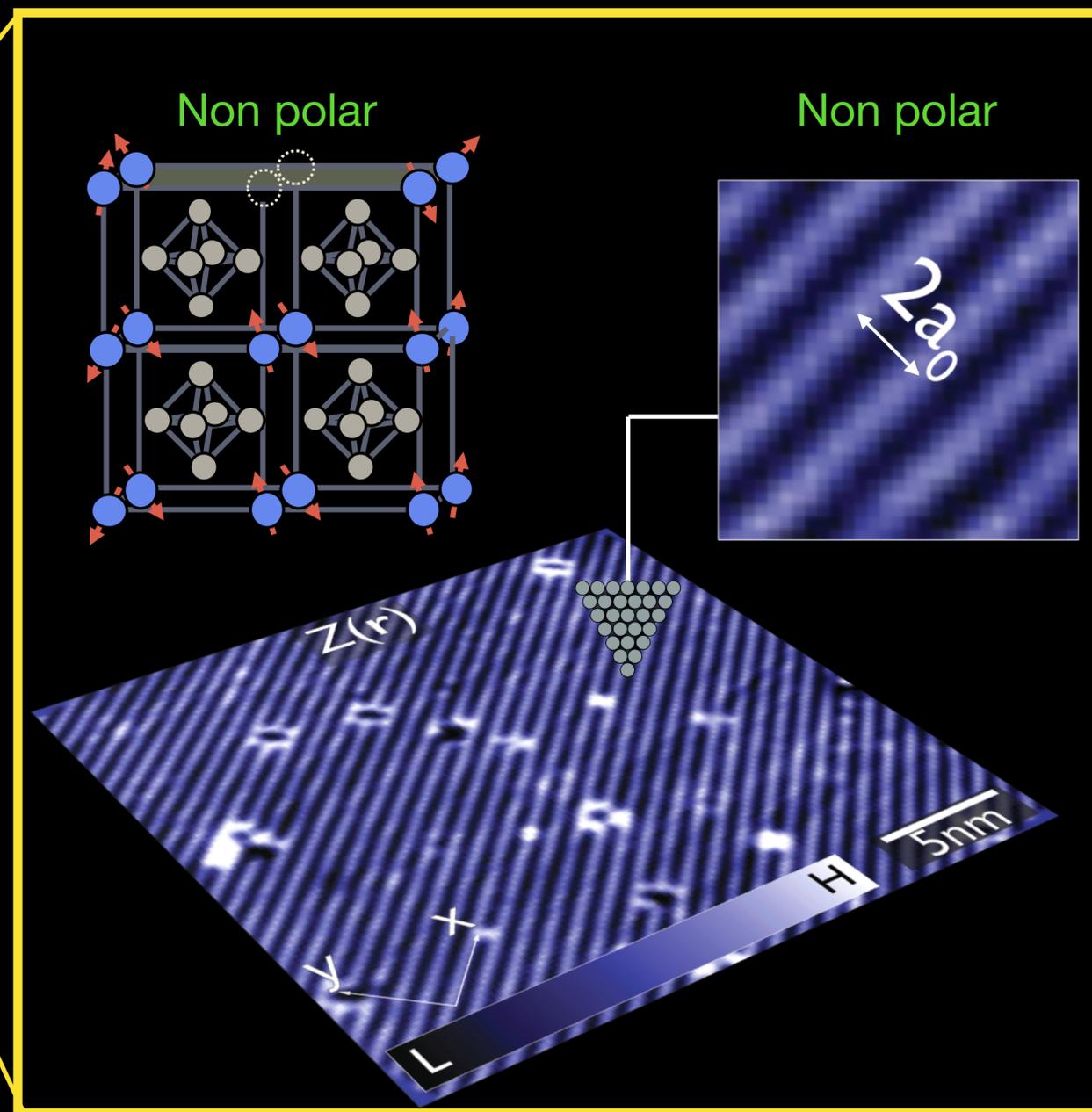


# Requirements to image heavy Dirac states

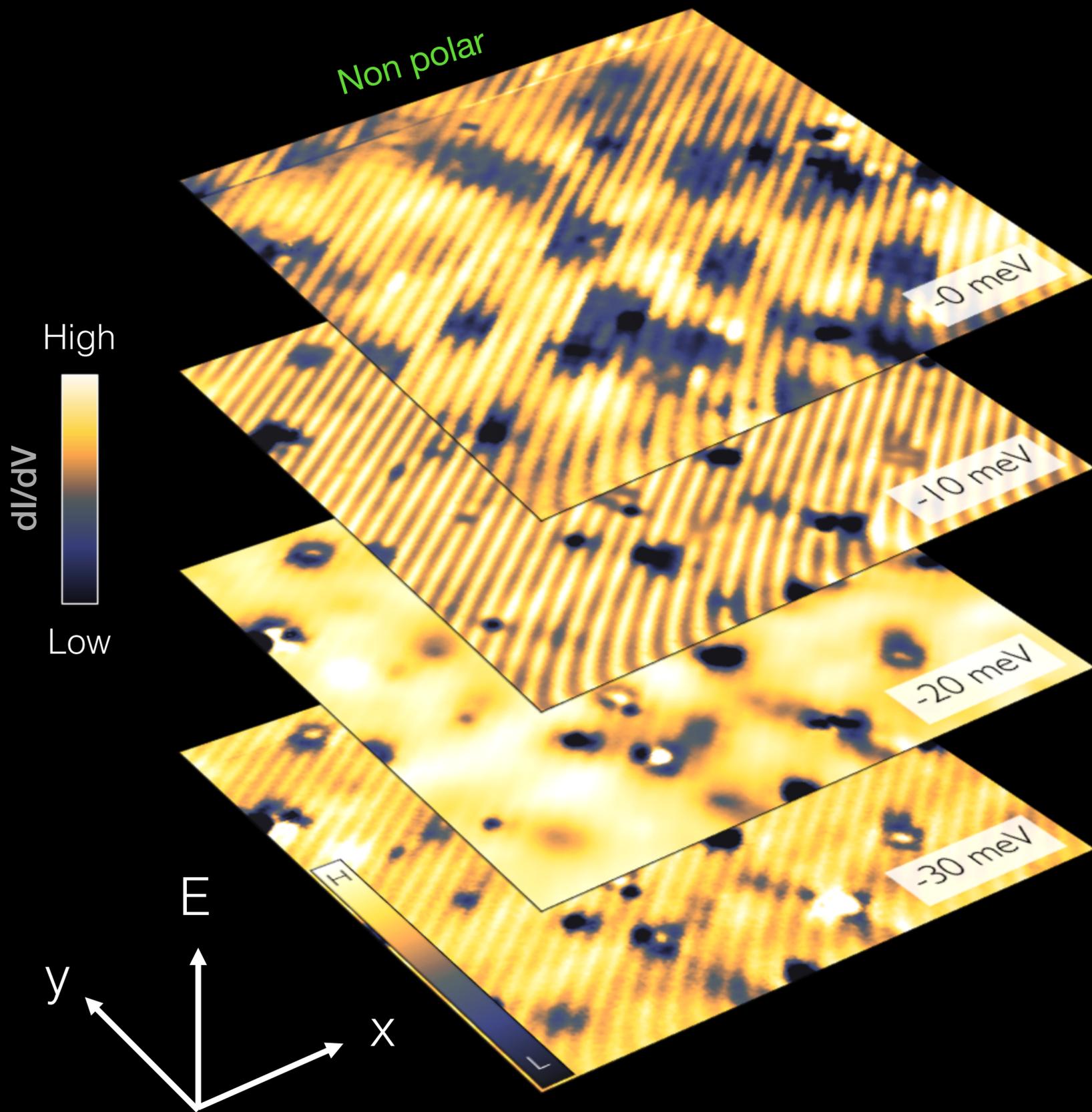
- ◎ Small polar and non-polar terminations
  - Need **nanometer** spatial resolution
- ◎ Gap and most of the surface state is above  $E_F$ 
  - Need to access both **filled & empty** states
- ◎ Heavy fermion bands are extremely flat
  - Need **sub-meV** energy resolution
- ◎ Heavy fermions are deeply entangled
  - Need **real & momentum** space imaging



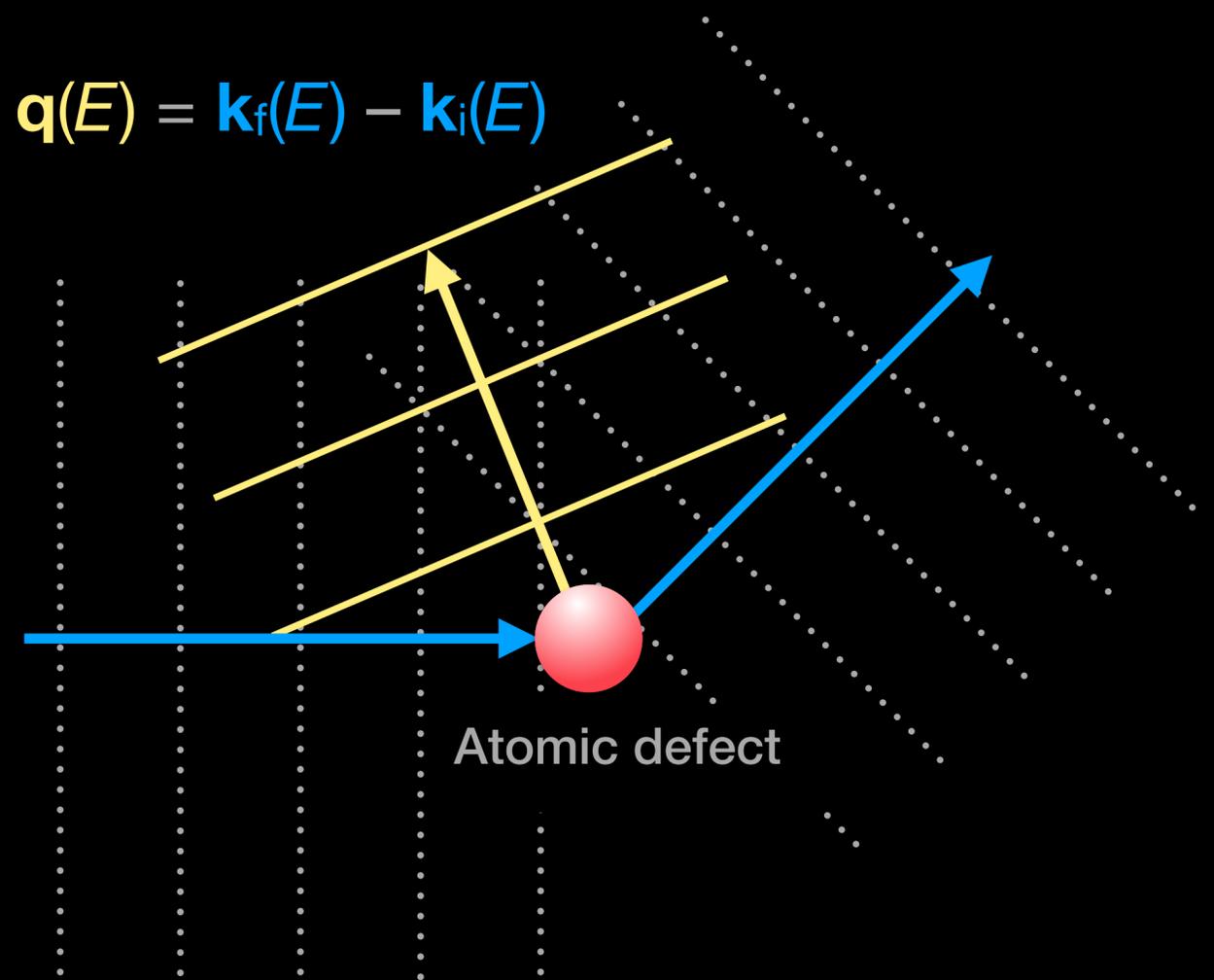
Scanning tunneling microscopy



# Quasiparticle interference reveals $E(k)$

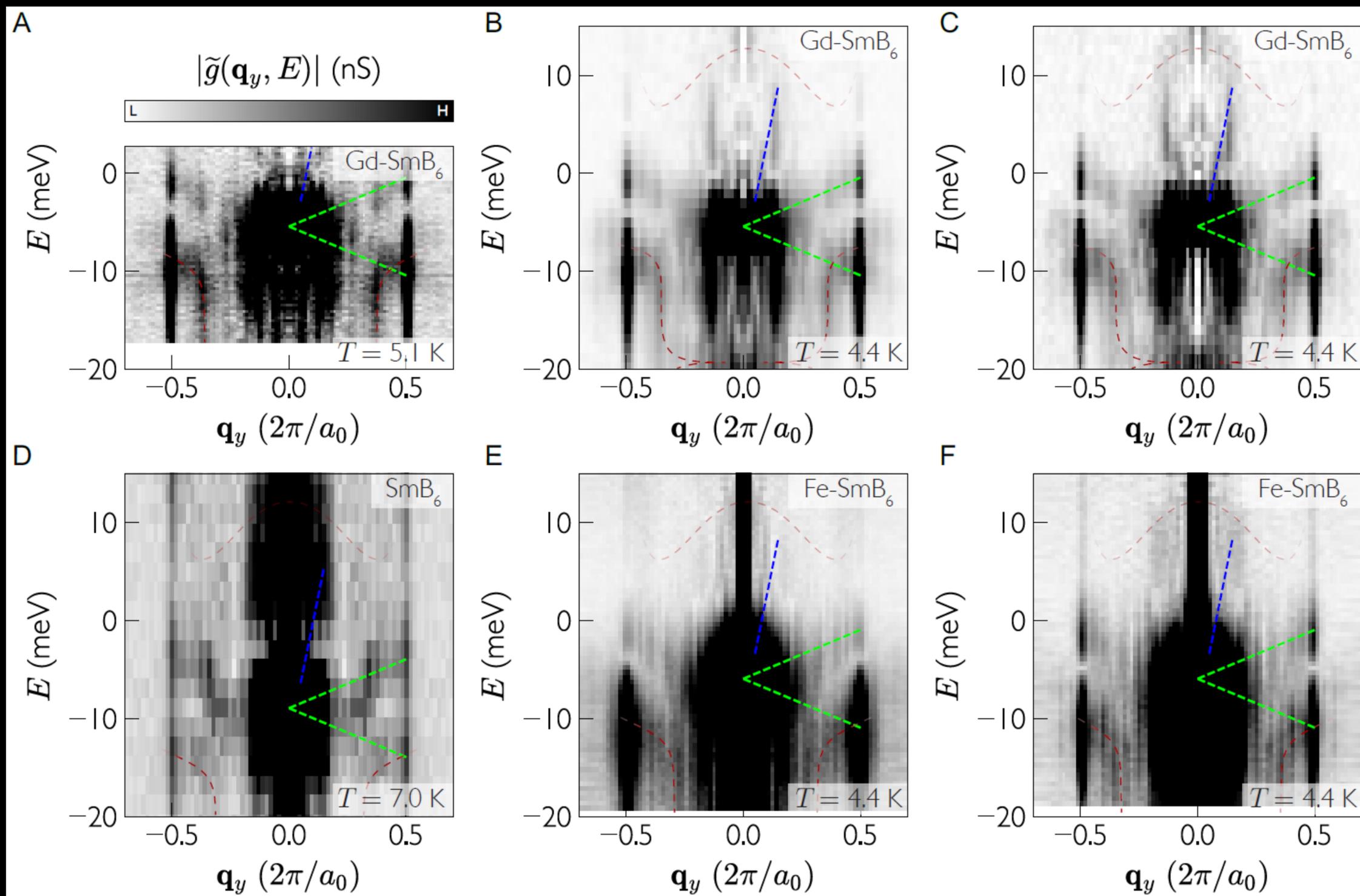


$$\mathbf{q}(E) = \mathbf{k}_f(E) - \mathbf{k}_i(E)$$



# Heavy Dirac states visible in raw data

Pirie et al., *Nat. Phys.* **16**, 52 (2020)

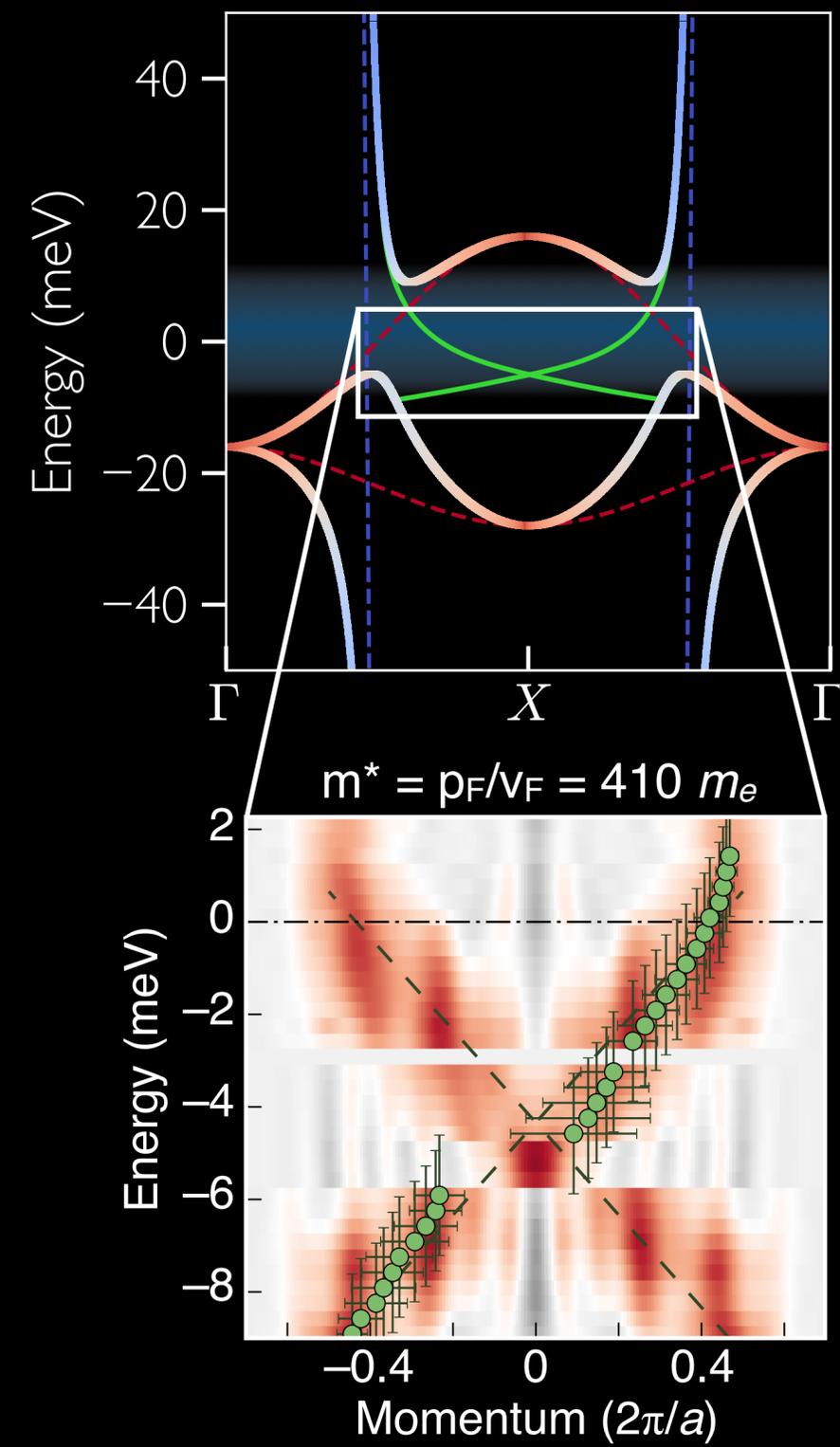
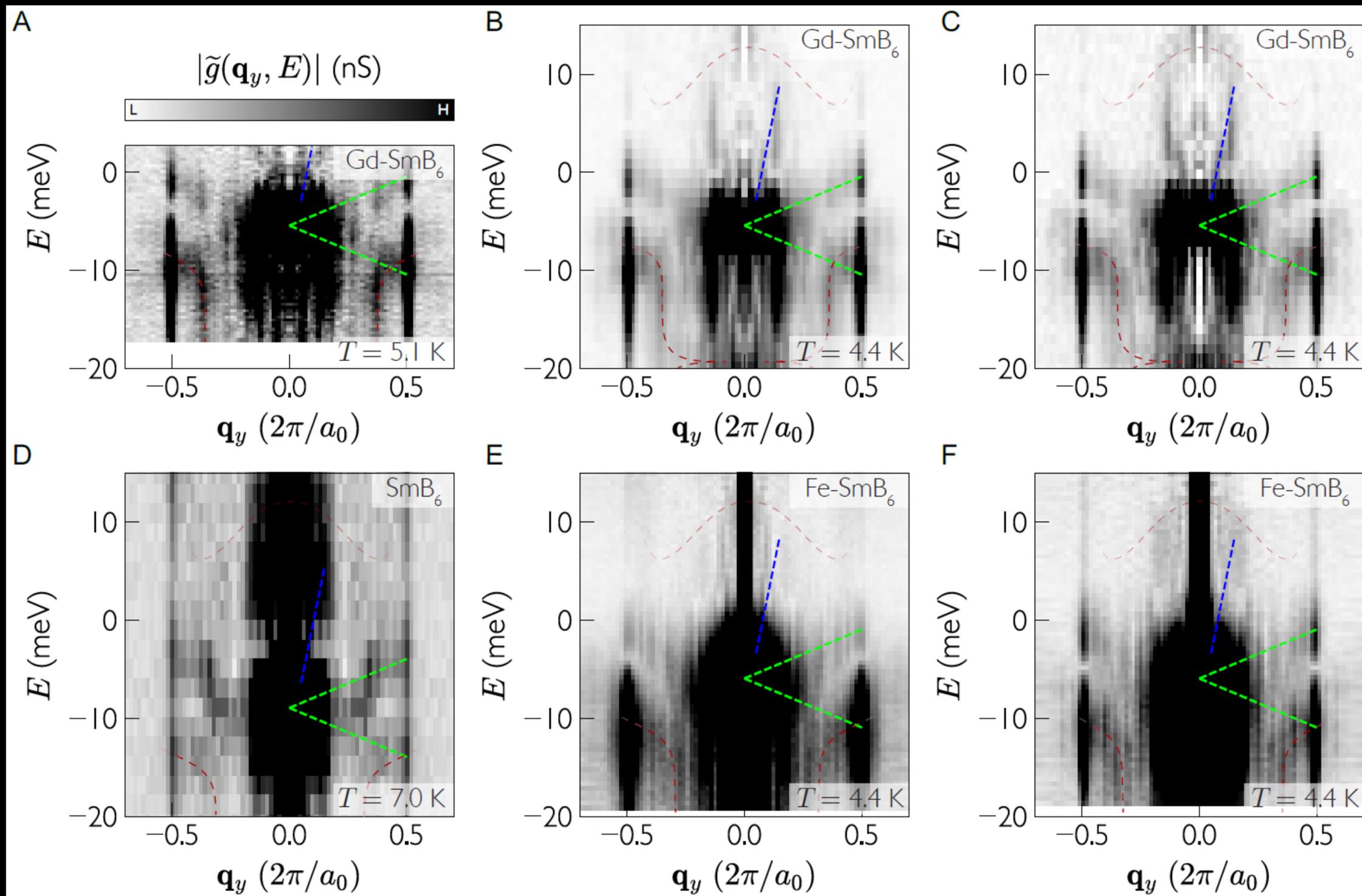


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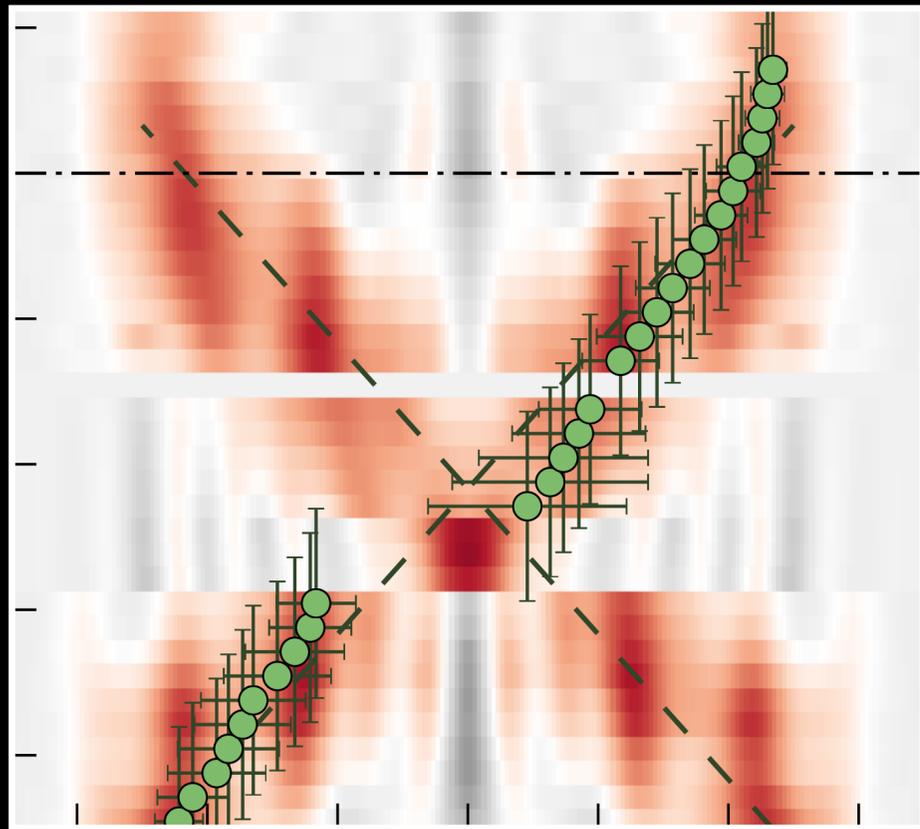
Measured velocity:  $7.6 \pm 0.3 \text{ meV}\cdot\text{\AA}$

Predicted velocity:  $16 \text{ meV}\cdot\text{\AA}$



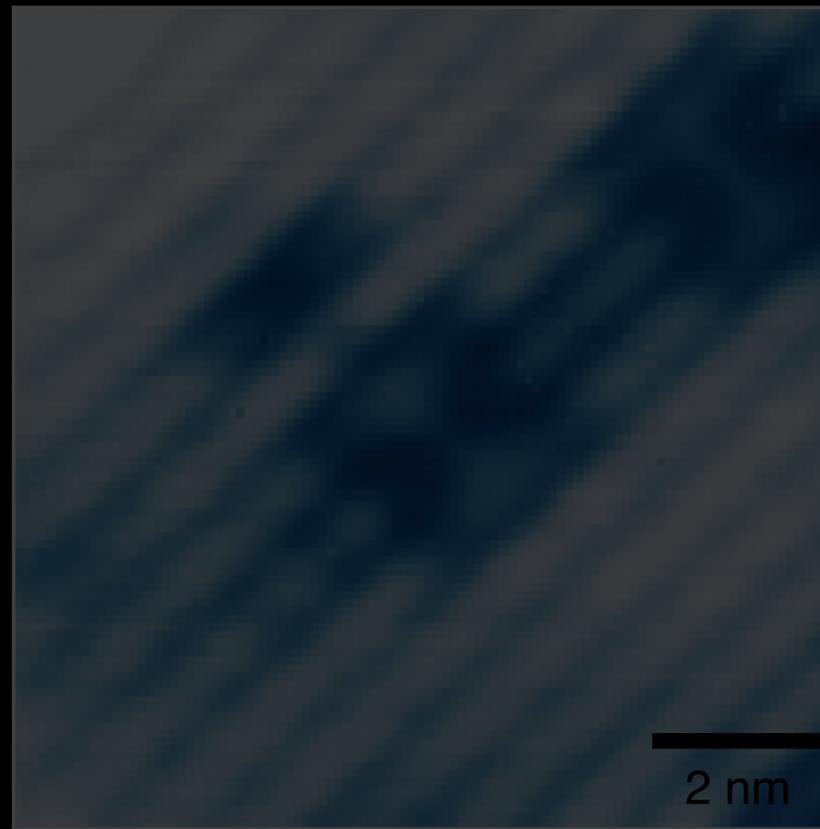
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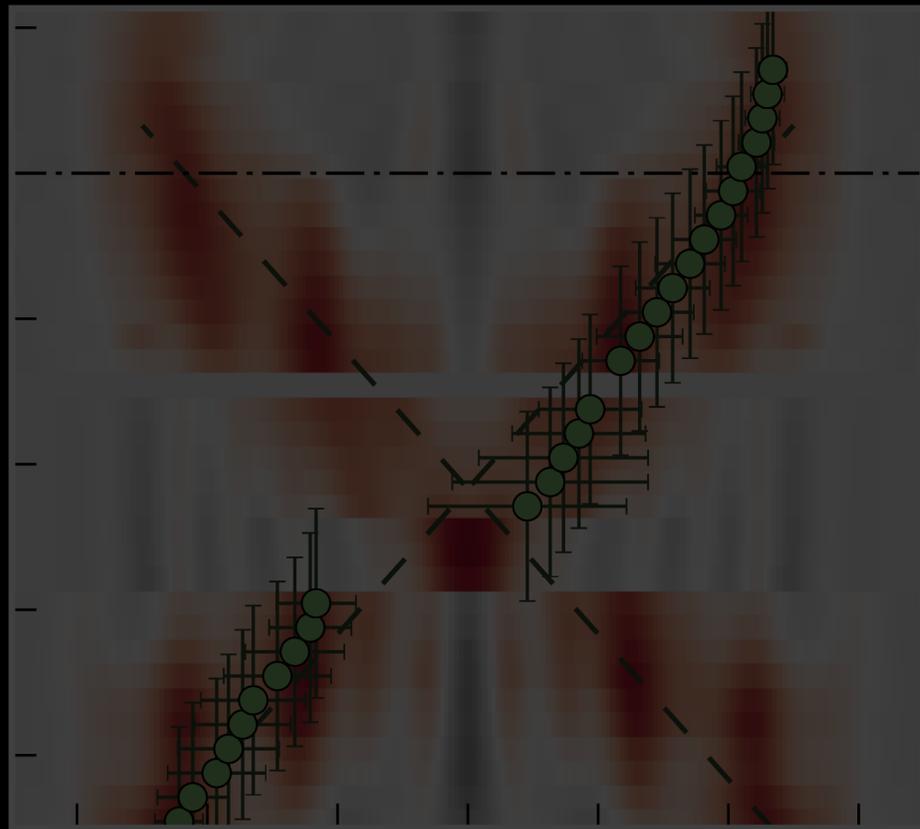
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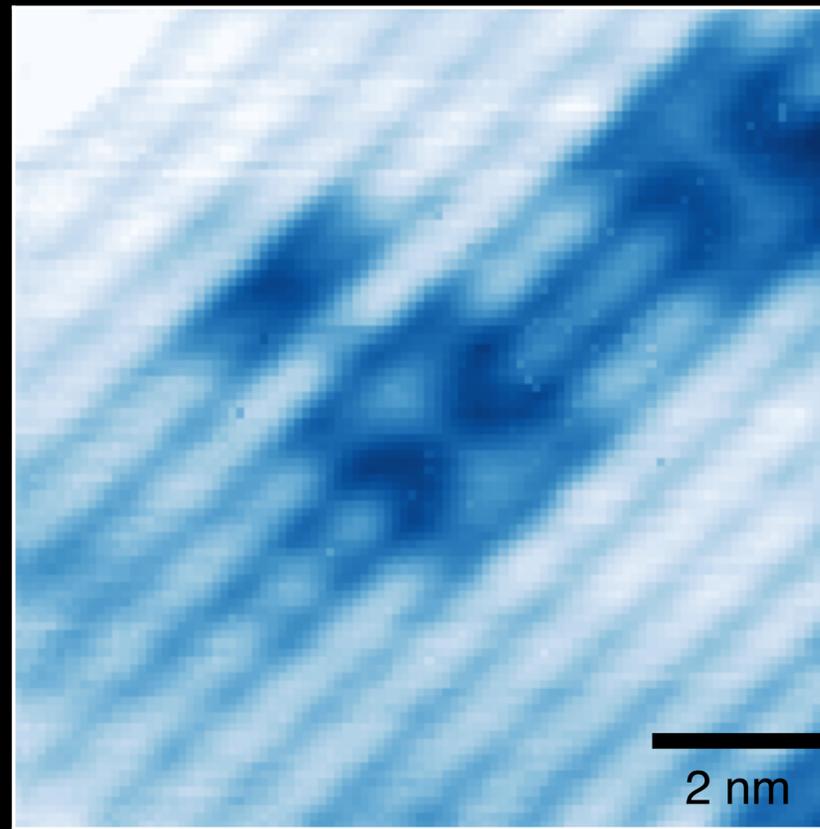
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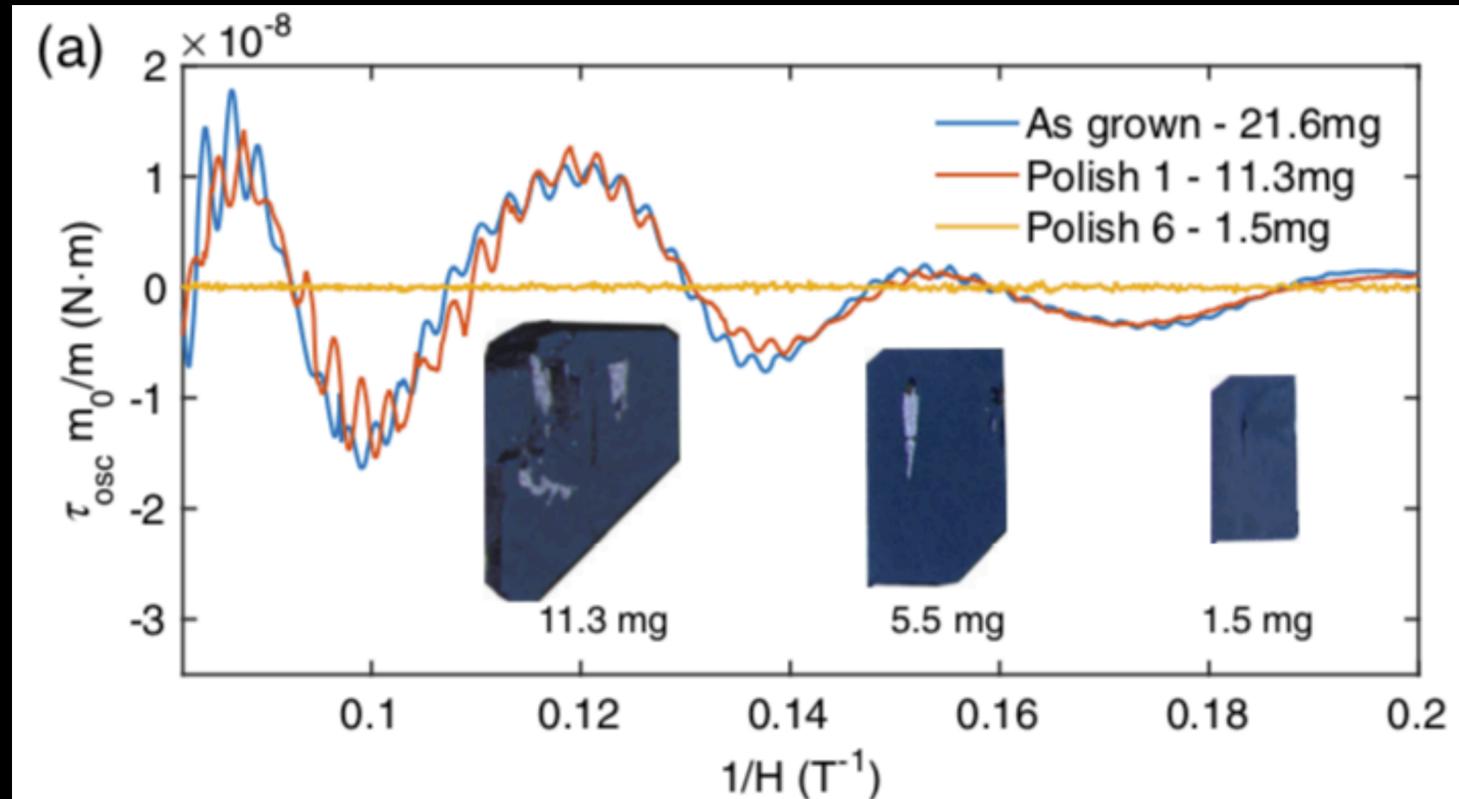
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Thomas et al., *Phys. Rev. Lett.* **122**, 166401 (2019)



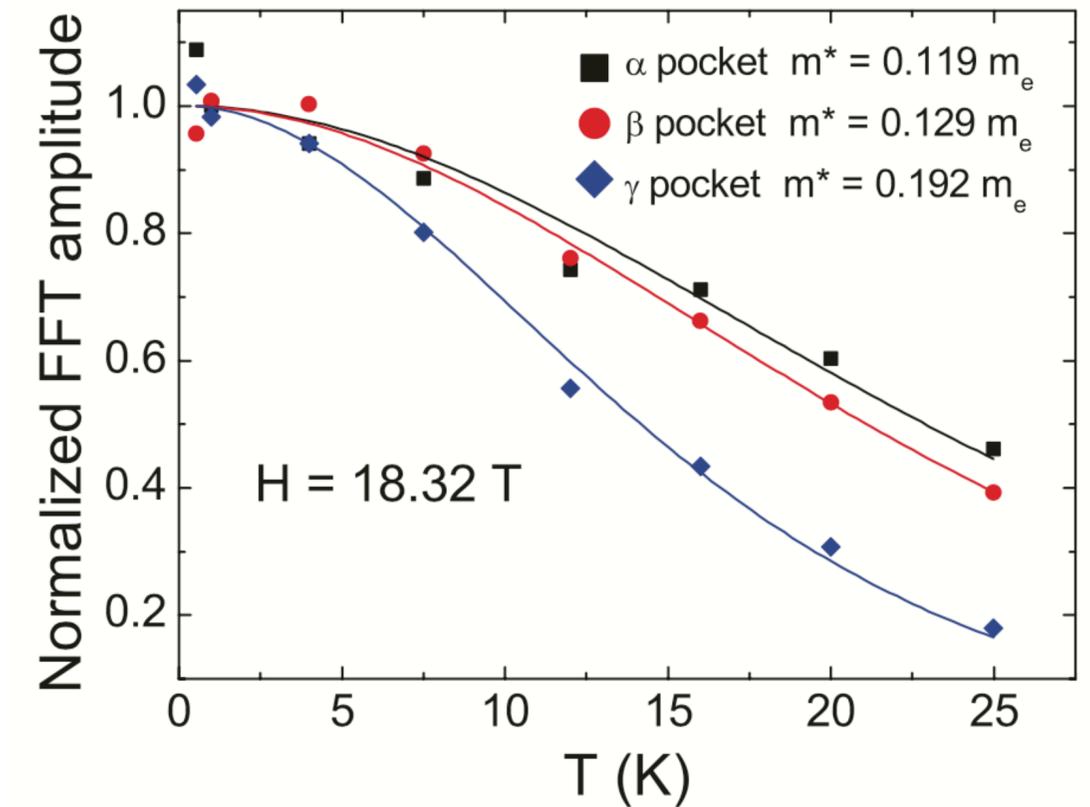
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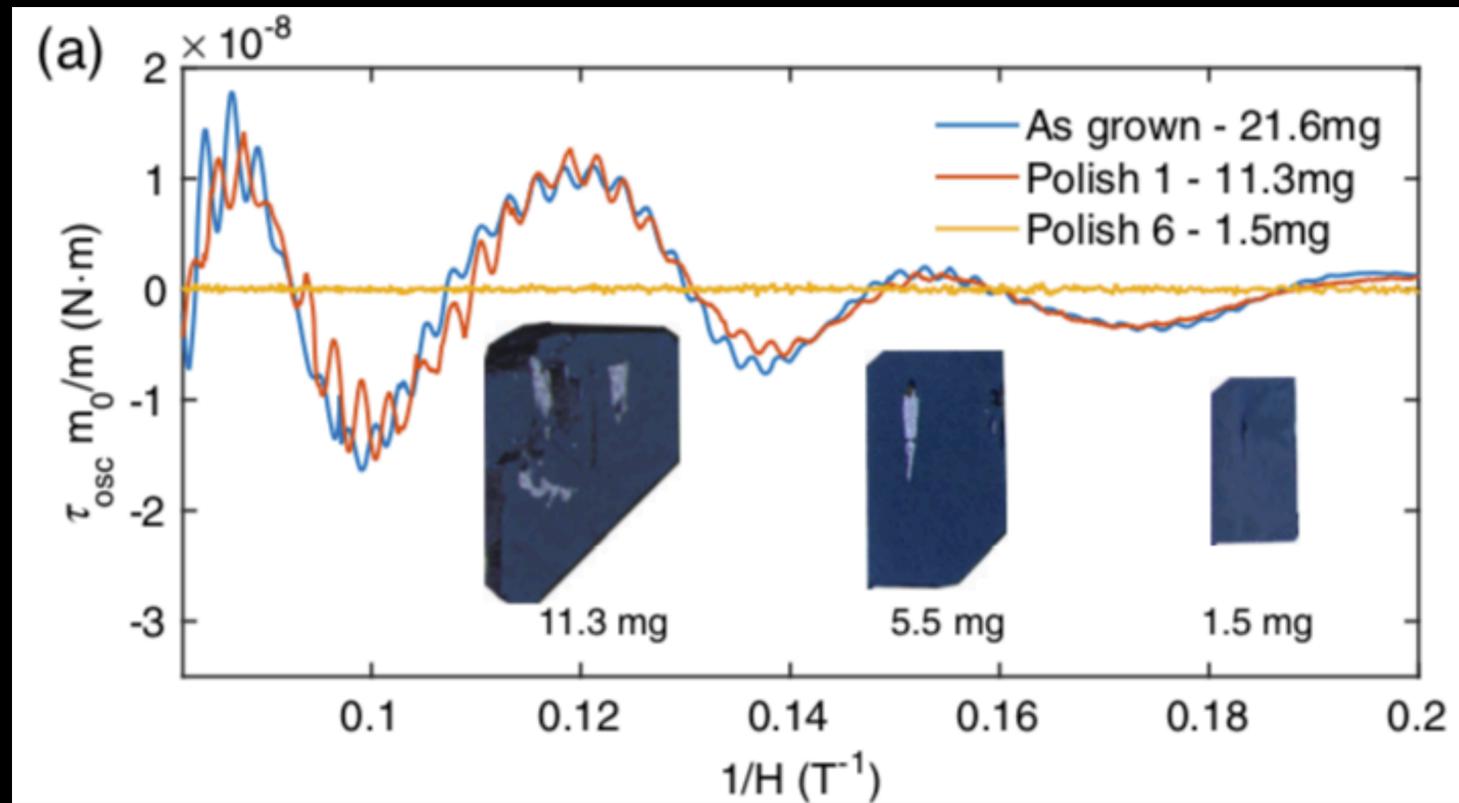
\*This is only half of the story...

# dHvA without aluminum inclusions!

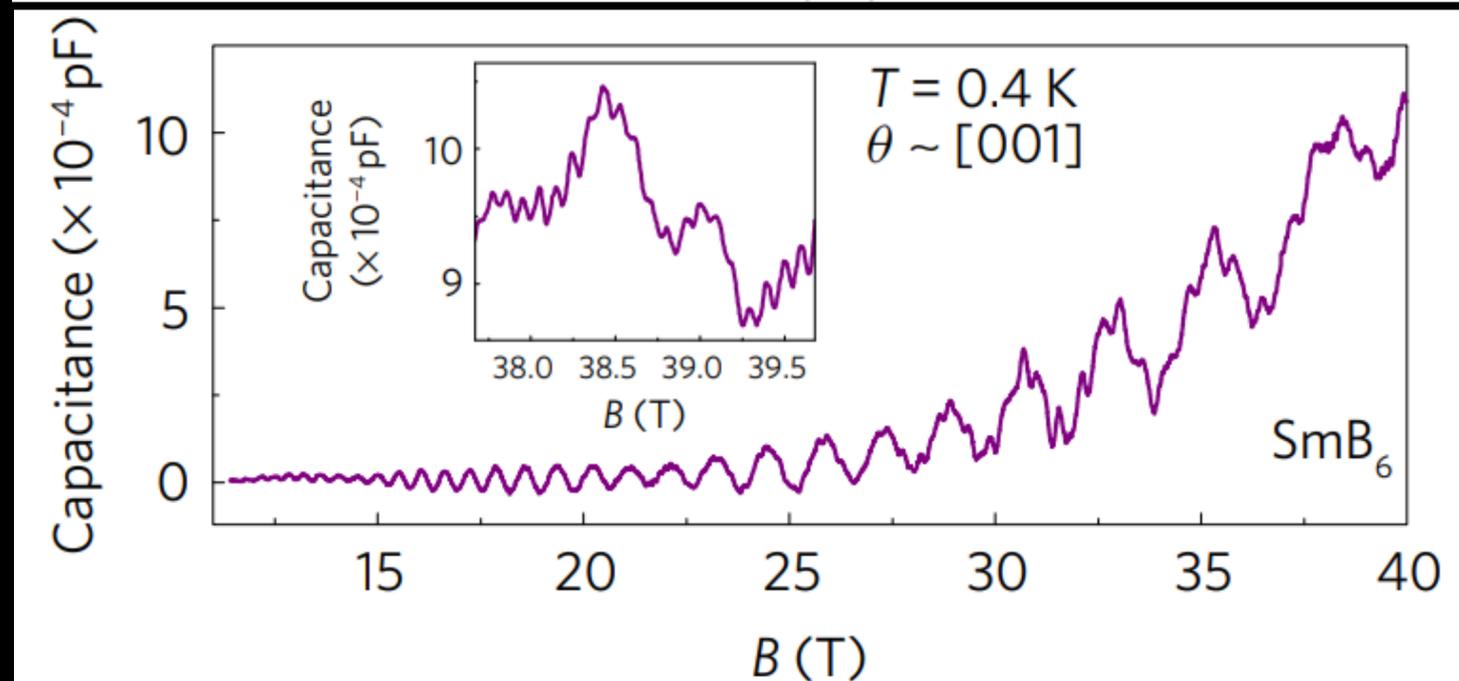
Thomas et al., *Phys. Rev. Lett.* **122**, 166401 (2019)

Li et al., *Science*. **346**, 1208 (2014)

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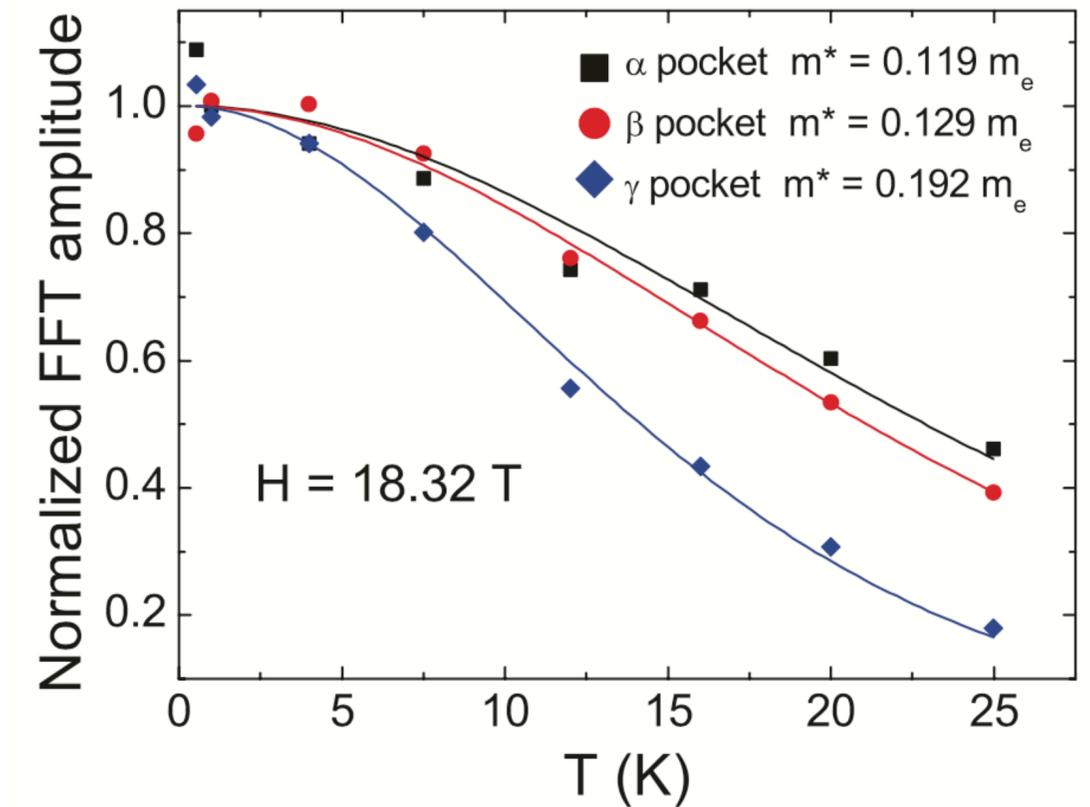
This SmB<sub>6</sub> is grown using **floating zone** technique (typically introduces  $\sim 0.1\%$  Sm vacancies)



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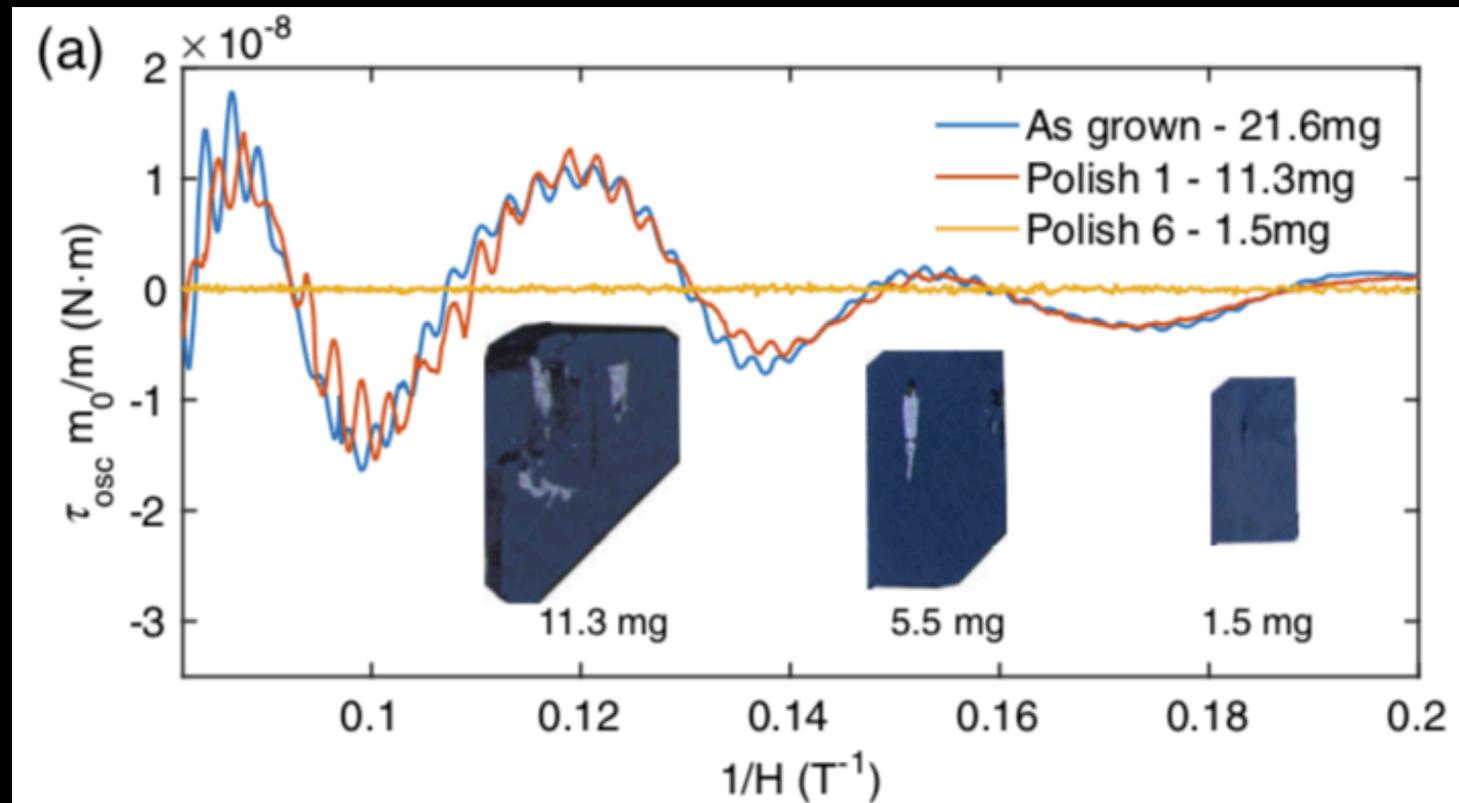


Tan et al., *Science* **349**, 287 (2015)

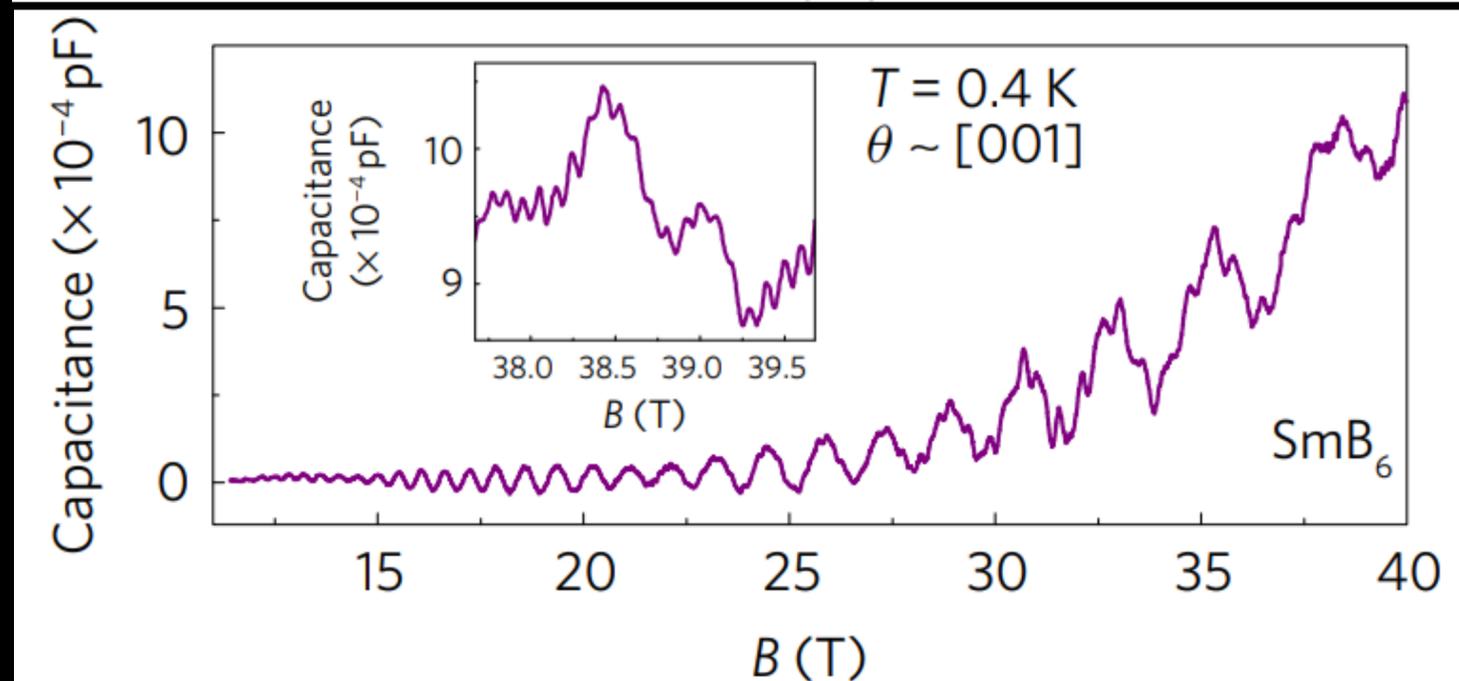
Hartstein et al., *Phys. Rev. Lett.* **14**, 166 (2018)

# dHvA without aluminum inclusions!

Thomas et al., *Phys. Rev. Lett.* 122, 166401 (2019)



This SmB<sub>6</sub> is grown with **aluminum flux**

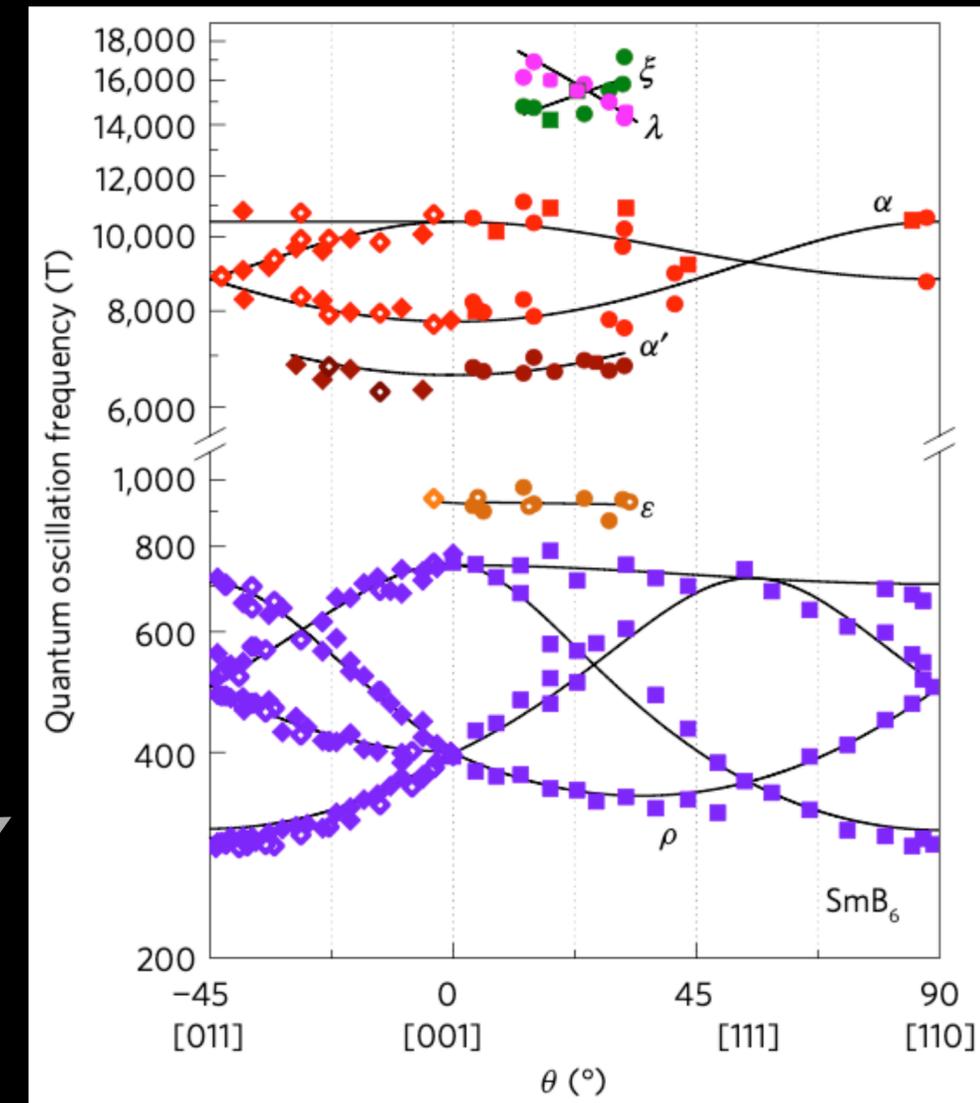


This SmB<sub>6</sub> is grown using **floating zone** technique (typically introduces  $\sim 0.1\%$  Sm vacancies)

Tan et al., *Science* 349, 287 (2015)

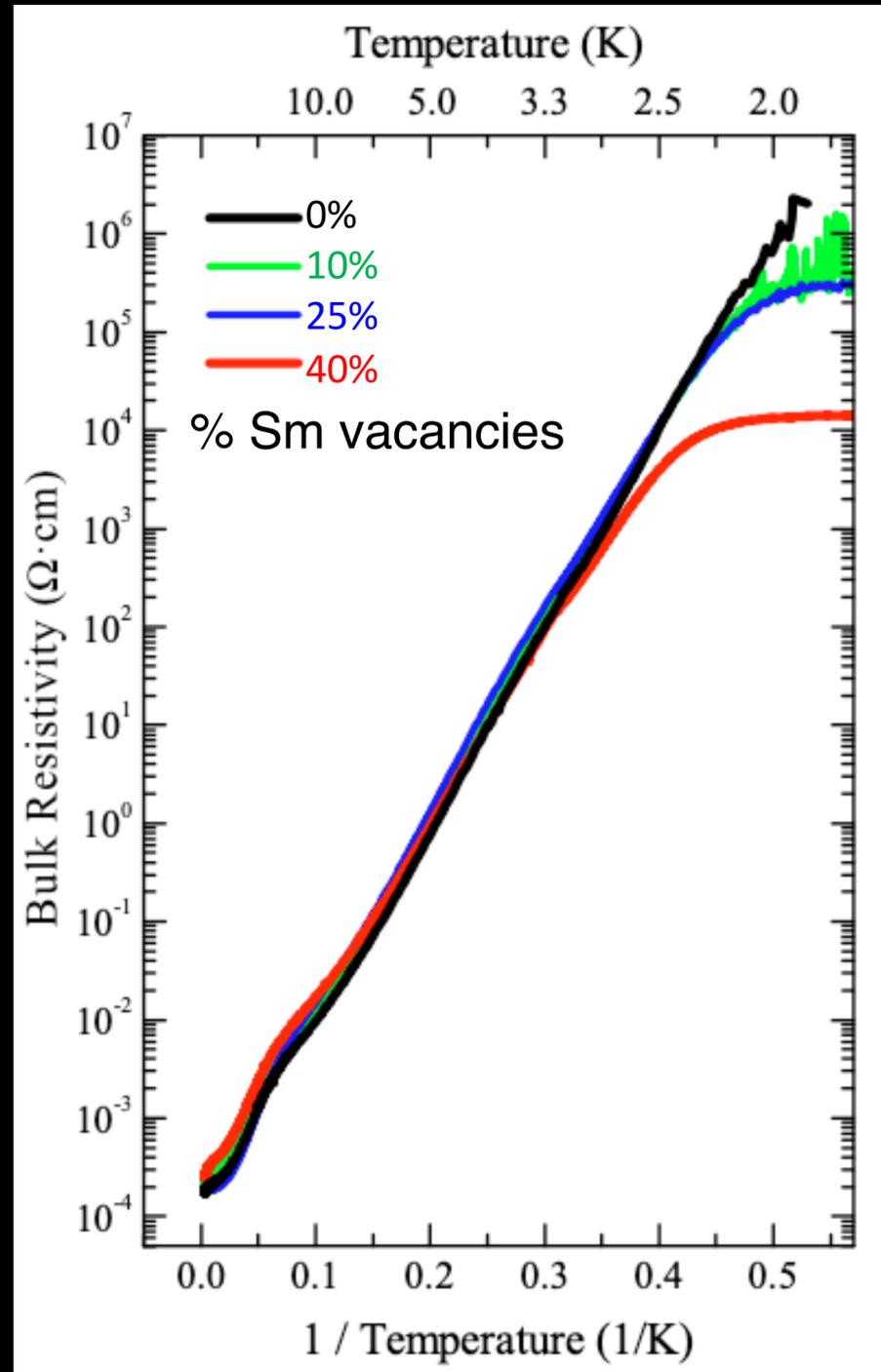
Hartstein et al., *Phys. Rev. Lett.* 14, 166 (2018)

Angular dependence of dHvA implies a **3D Fermi surface**



# But SmB<sub>6</sub> is an excellent insulator!

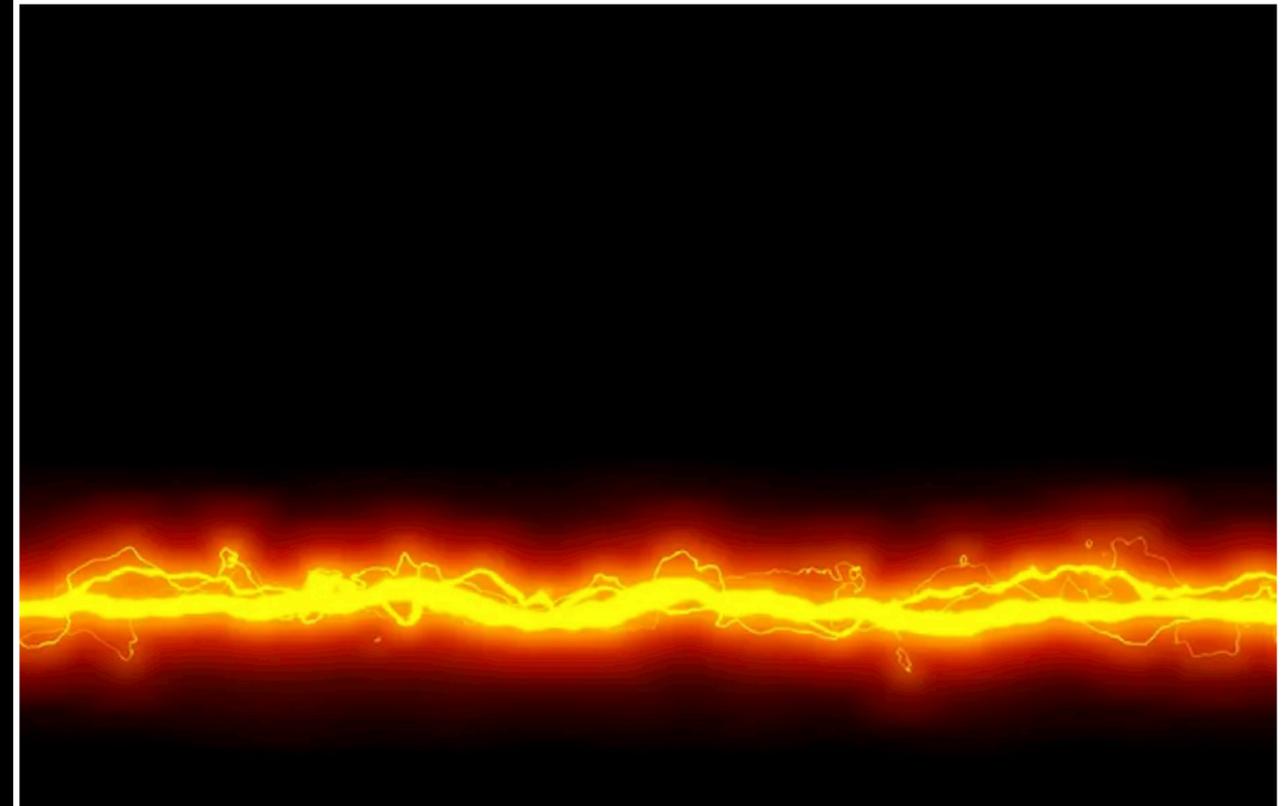
Inverted Corbino transport



Eo et al., *Proc. Nat. Acad. Sci.* 116, 26 (2019)

## How we discovered 'impossible' material that both conducts electricity – and doesn't

Published: July 10, 2015 2:17pm BST



Current + lack of current = a headache for physicists. Geralt/pixabay

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Twitter

27

Facebook

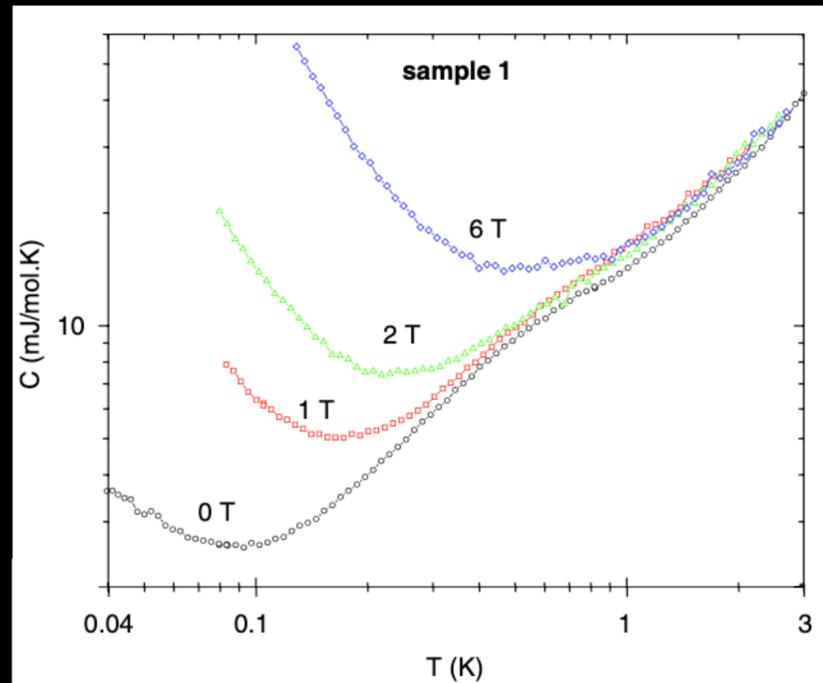
931

LinkedIn

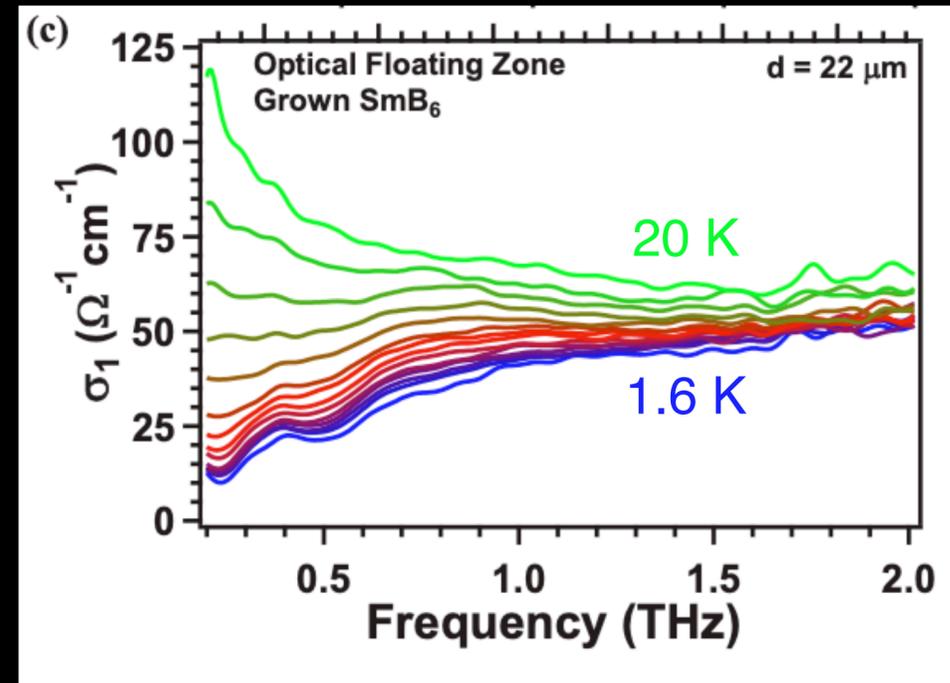
Metals, which conduct electricity, and insulators, which don't, are polar opposites. At least that's what we've believed until now. But we have discovered that a well-known insulator can simultaneously act like a conductor in certain measurements.

# Remnant metallicity in floating-zone SmB<sub>6</sub>

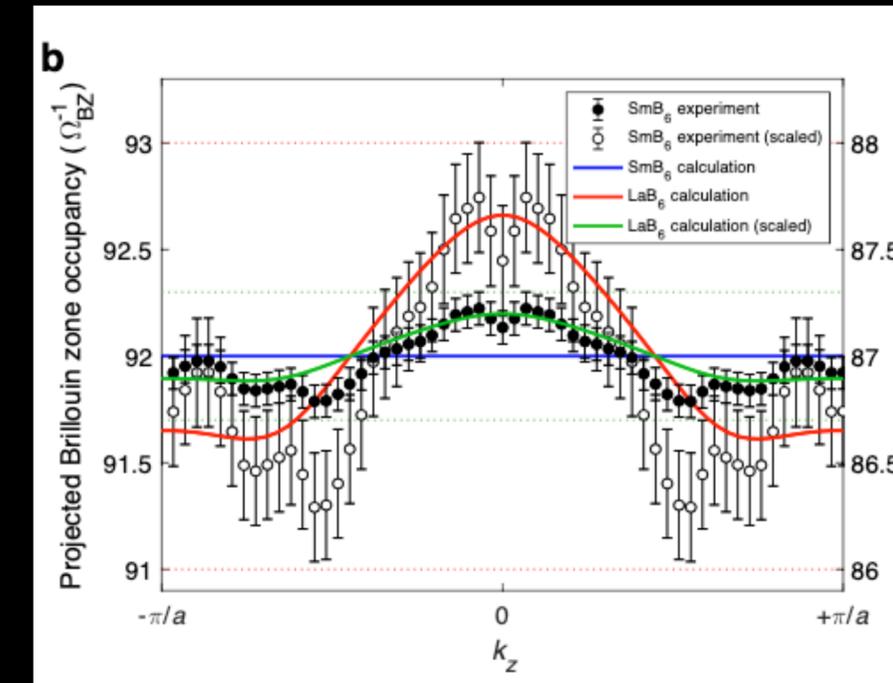
Low temperature specific heat



AC optical conductivity



Compton scattering

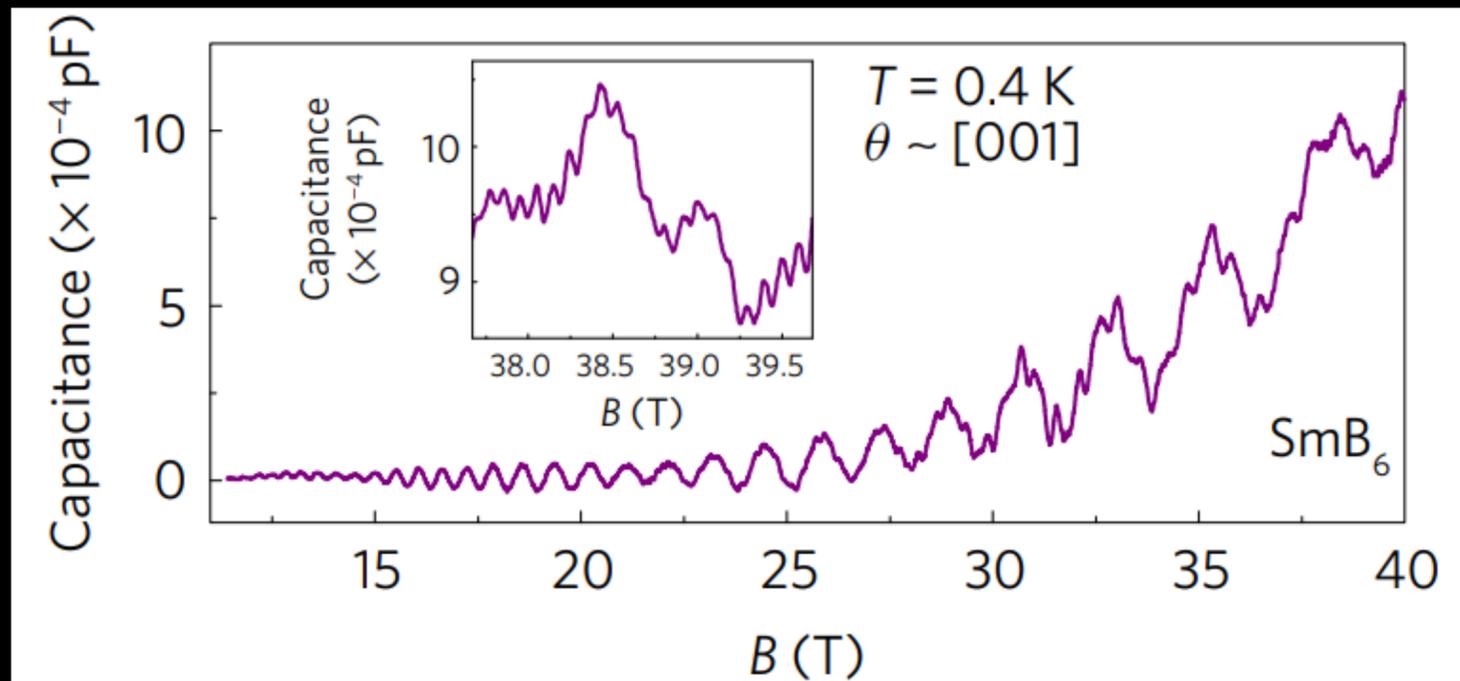


Flachbart et al., *Physica B*. **378**, 610 (2006)

Laurita et al., *Phys. Rev. B* **122**, 165154 (2016)

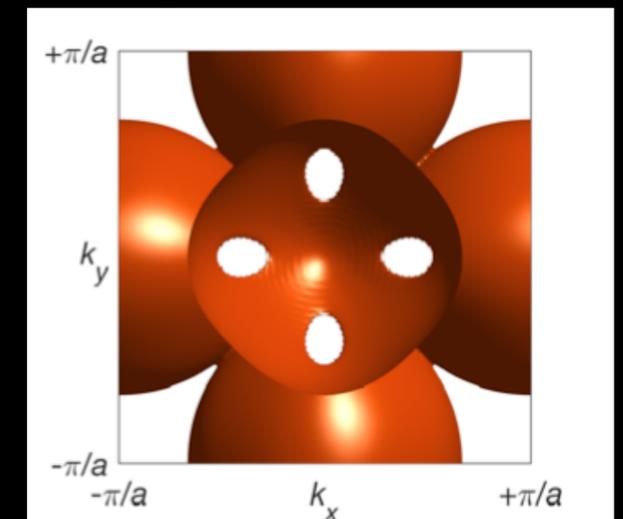
Millichamp et al., *arXiv 2011.07727* (2021)

This SmB<sub>6</sub> is grown using **floating zone** technique



Hartstein et al., *Phys. Rev. Lett.* **14**, 166 (2018)

Looks like SmB<sub>6</sub> at high temperature





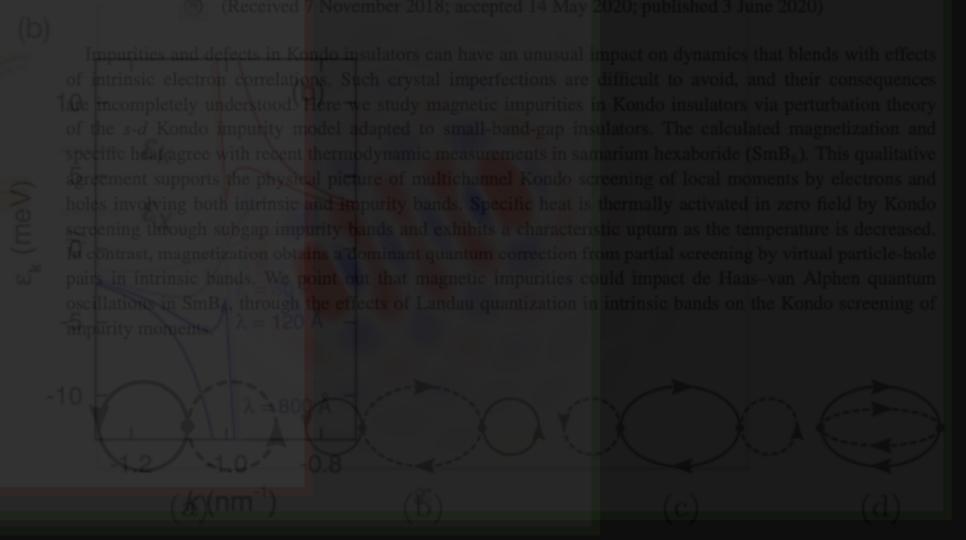
# Is the origin **intrinsic** or **extrinsic**?

We will answer this question experimentally:

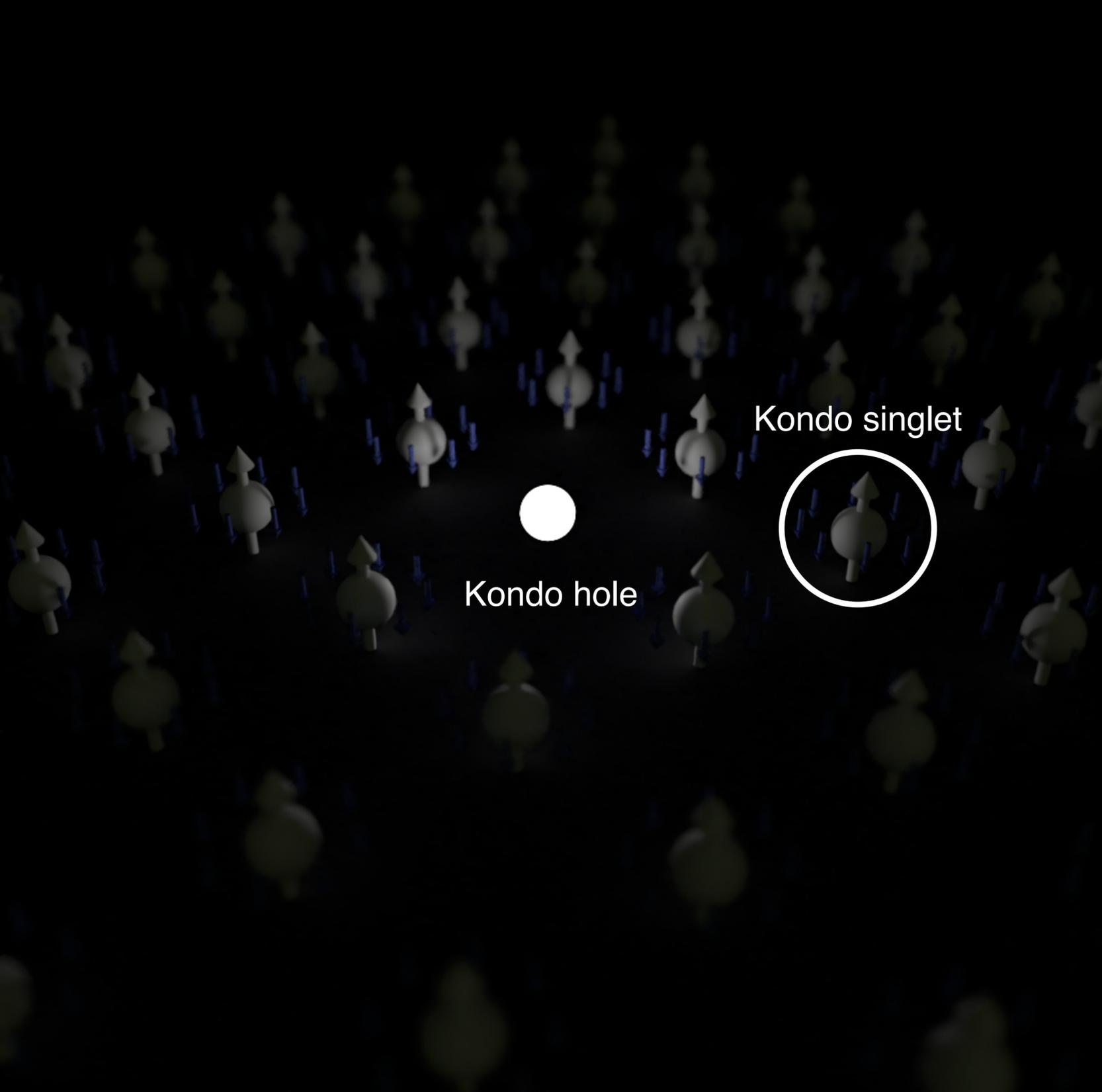
Question: Is there microscopic evidence of light electrons?

**Majorana Fermi Sea in Insulating  $\text{SmB}_6$ :**  
A proposal and a Theory of Quantum Oscillations in Kondo Insulators  
D. Baskaran  
The Institute of Mathematics, Perimeter Institute for Theoretical Physics, Waterloo, Ontario, Canada  
In an exciting development,  $\text{SmB}_6$  Kondo insulator has been shown to exhibit bulk quantum oscillations. We propose that  $\text{SmB}_6$  is a bulk scalar Majorana Fermi Liquid (MFL) with a finite Fermi surface for charge and spin excitations. In their study of Kondo insulators in 1993, Coleman, Misra, and Tsvetkiv (CMT) envisaged such a remarkable possibility, being a mean field ansatz. The CMT theory to non-zero magnetic fields and show a counter intuitive result, that the scalar fermi liquid, while remaining electrically insulating, responds to external magnetic field and exhibits Landau diamagnetism and quantum oscillations. Physics of an emergent compactified Kondo lattice physics that is behind formation of the novel scalar Majorana fermi liquid is discussed. It is also suggested that a known resistivity saturation in  $\text{SmB}_6$  as well as strong deviation of quantum oscillation amplitude from Lifshitz-Kosevich formula in  $\text{SmB}_6$  at low temperatures are due to a coherent fluctuation of charge of a neutral scalar Majorana fermion. Possible presence of 2-dimensional Majorana fermion excitations in surfaces of  $\text{SmB}_6$ , and other Topological Kondo Insulators (TKI) is pointed out.

**Critical examination of quantum oscillations in  $\text{SmB}_6$**   
Peter S. Riseborough  
Z. Fisk  
Physics Department, Temple University Philadelphia, Pennsylvania 19122, USA  
University of California, Irvine, Irvine, California 92717, USA  
We critically review the results of magnetic torque measurements on  $\text{SmB}_6$  that show quantum oscillations. Similar studies have been given two different interpretations. One interpretation is based on the existence of metallic surface states, while the second interpretation is in terms of a three-dimensional Fermi surface involving both fermionic excitations. We suggest that the low-field oscillations that are seen in both groups for  $B$  fields as small as 0 T might be due to metallic surface states. The high-field three-dimensional oscillations are only seen by one group for fields  $B > 18$  T. The phenomenon of magnetic breakthrough occurs at high fields and involves the formation of Landau orbits that produces a directional dependent suppression of Bragg scattering. We argue that the measurements performed under higher-field conditions are fully consistent with expectations of a three-dimensional semiconducting state with magnetic breakthrough.



# Are Kondo holes the culprit?



PRL 107, 066401 (2011)

PHYSICAL REVIEW LETTERS

week ending  
5 AUGUST 2011

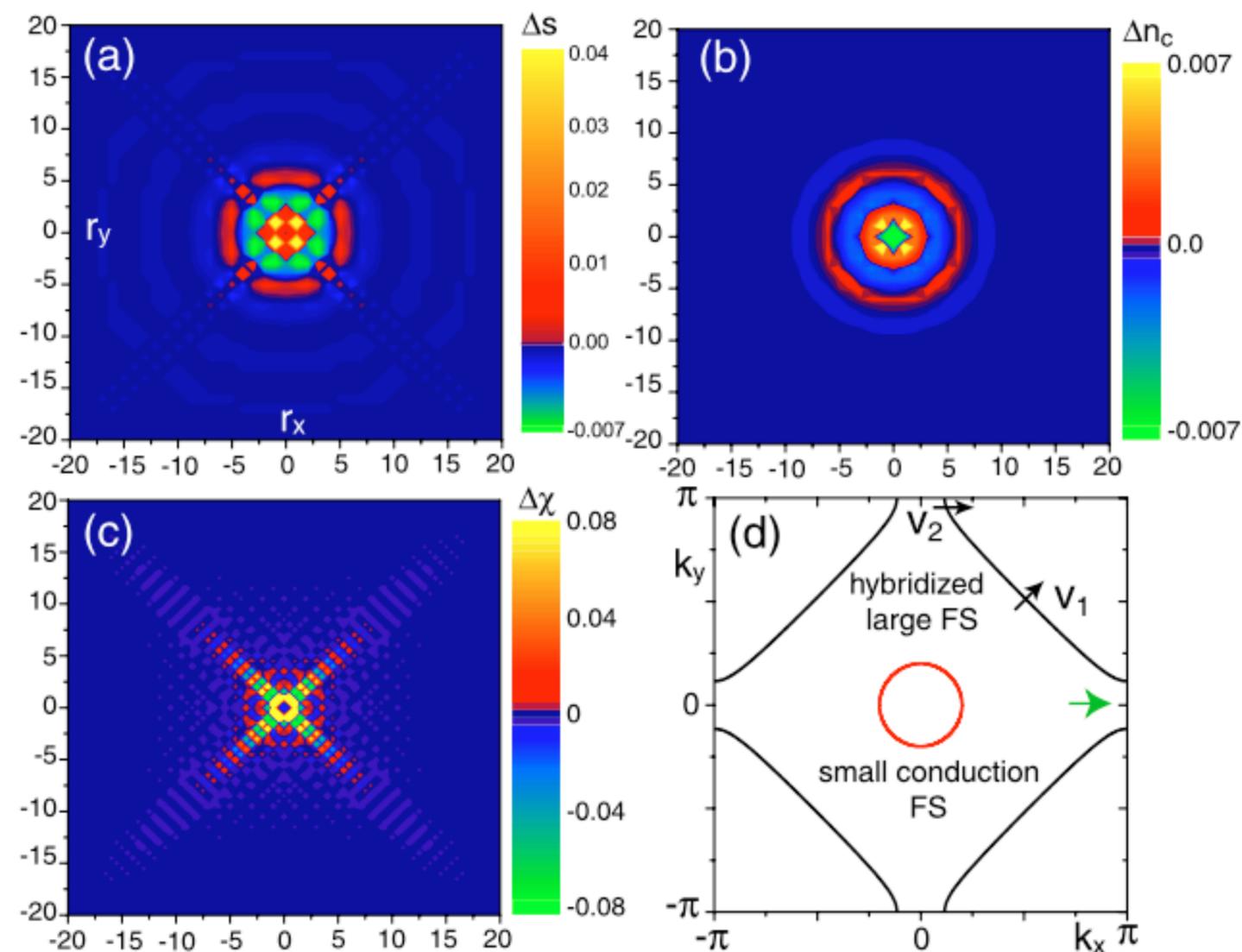
## Defects in Heavy-Fermion Materials: Unveiling Strong Correlations in Real Space

Jeremy Figgins and Dirk K. Morr

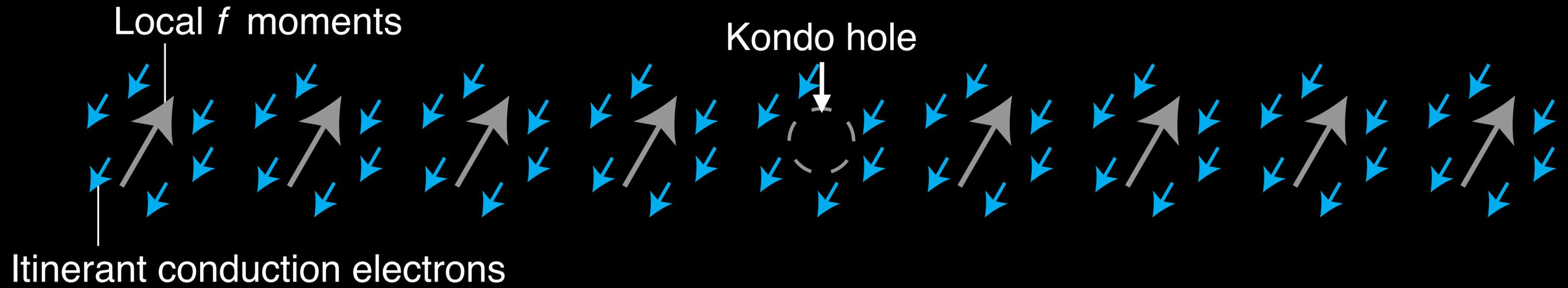
*Department of Physics, University of Illinois at Chicago, Chicago, Illinois 60607, USA*

(Received 27 October 2010; published 2 August 2011)

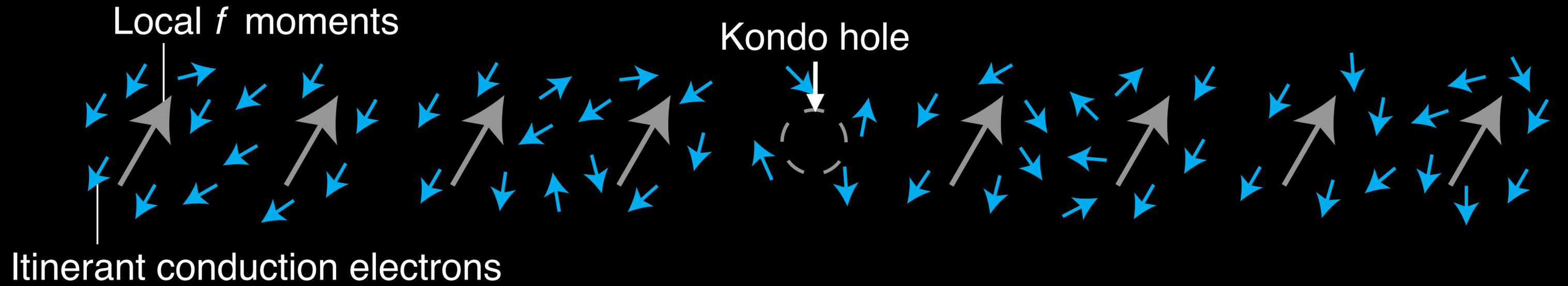
Defects provide important insight into the complex electronic and magnetic structure of heavy-fermion materials by inducing qualitatively different real-space perturbations in the electronic and magnetic correlations of the system. These perturbations possess direct experimental signatures in the local density of states, such as an impurity bound state, and the nonlocal spin susceptibility. Moreover, highly nonlinear quantum interference between defect-induced perturbations can drive the system through a first-order phase transition to a novel inhomogeneous ground state.



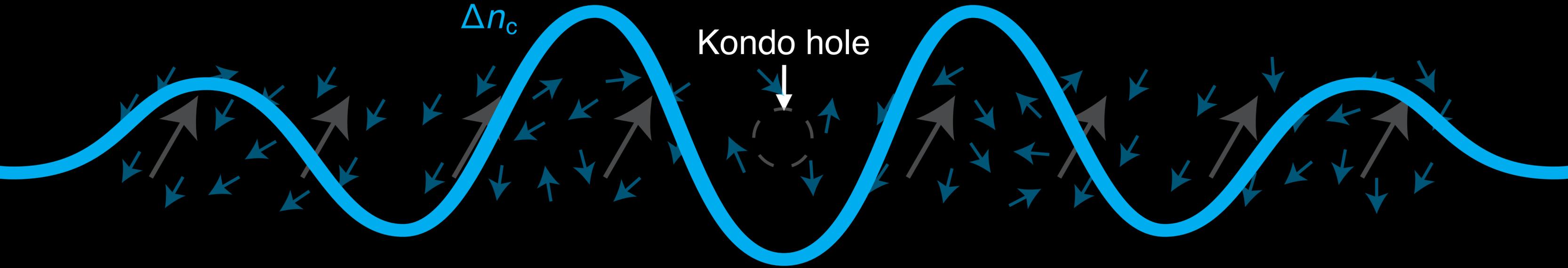
# Punching a hole in a Kondo lattice



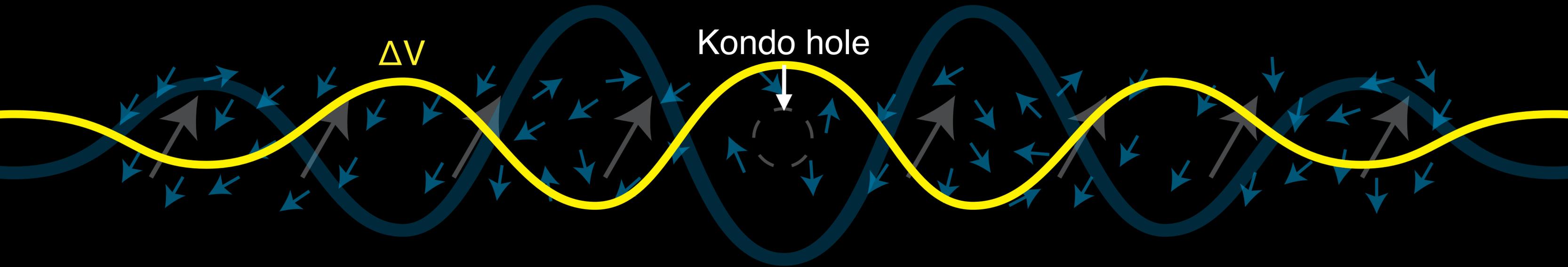
# Kondo holes redistribute the screening cloud



# Kondo holes modulate the local **charge density**



# Kondo holes modulate the **interaction strength**



Already observed in  $\text{URu}_2\text{Si}_2$

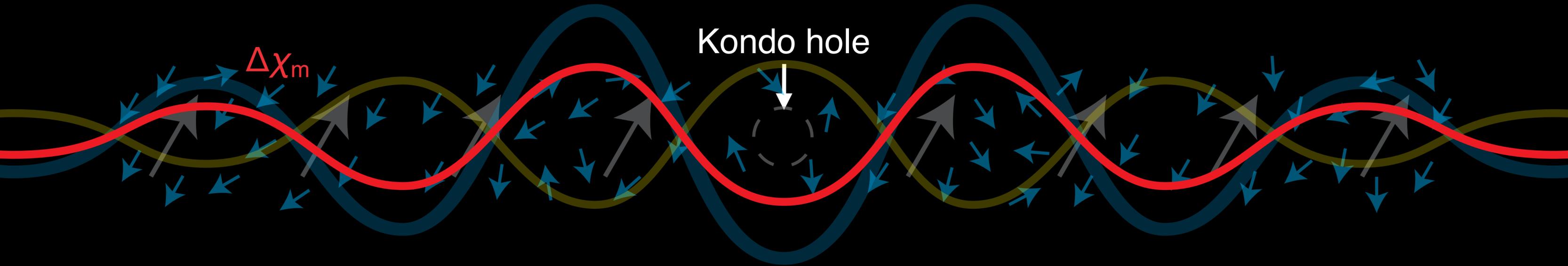
PNAS

## How Kondo-holes create intense nanoscale heavy-fermion hybridization disorder

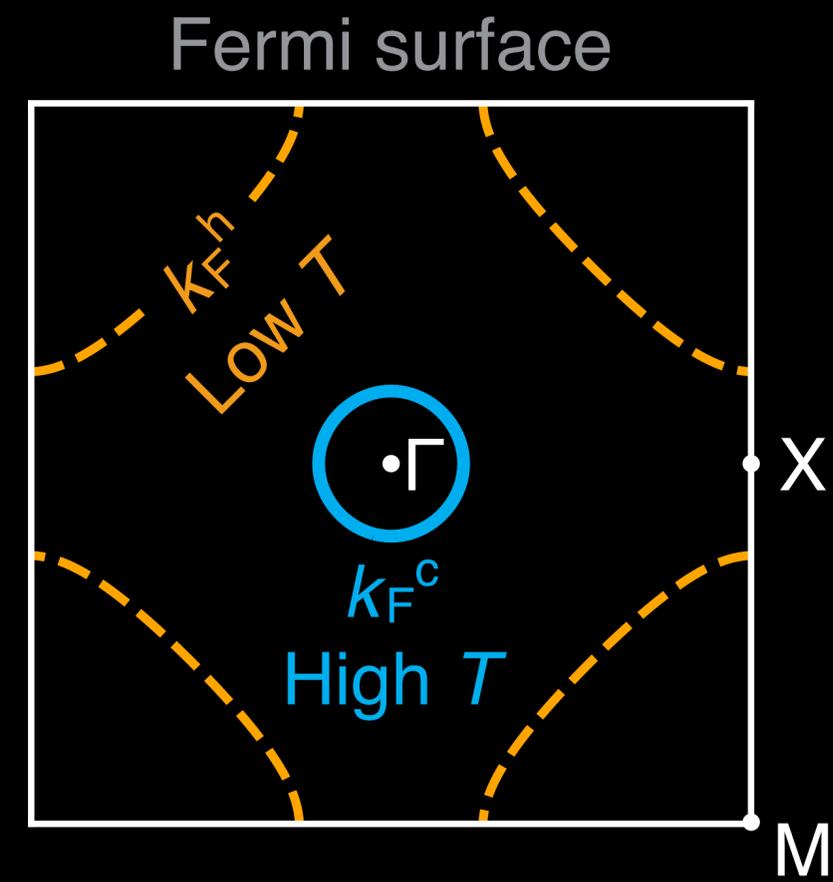
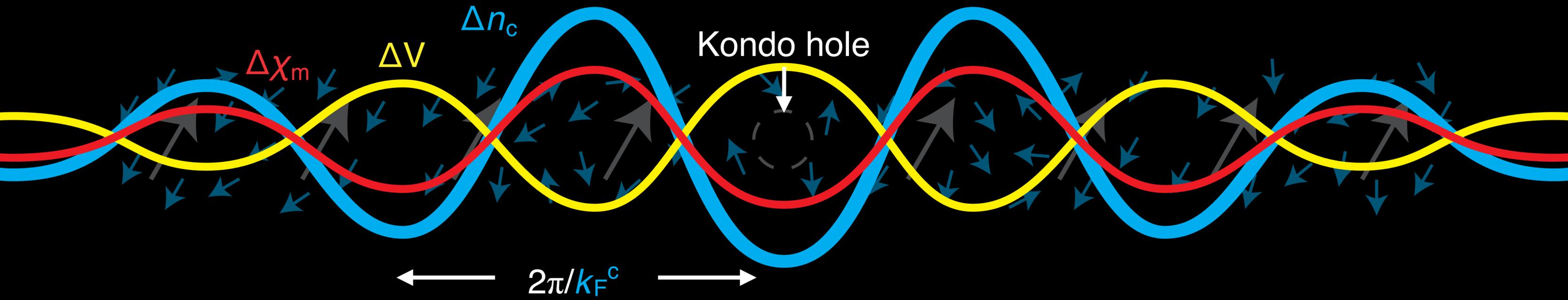
Mohammad H. Hamidian<sup>a,b,1</sup>, Andrew R. Schmidt<sup>a,b,c,1</sup>, Inês A. Firmo<sup>a,b</sup>, Milan P. Allan<sup>a,d</sup>, Phelim Bradley<sup>e</sup>, Jim D. Garrett<sup>f</sup>, Travis J. Williams<sup>g</sup>, Graeme M. Luke<sup>g,h</sup>, Yonatan Dubi<sup>i,j</sup>, Alexander V. Balatsky<sup>i</sup>, and J. C. Davis<sup>a,b,d,k,2</sup>

Contributed by J.C. Séamus Davis, September 13, 2011 (sent for review July 27, 2011)

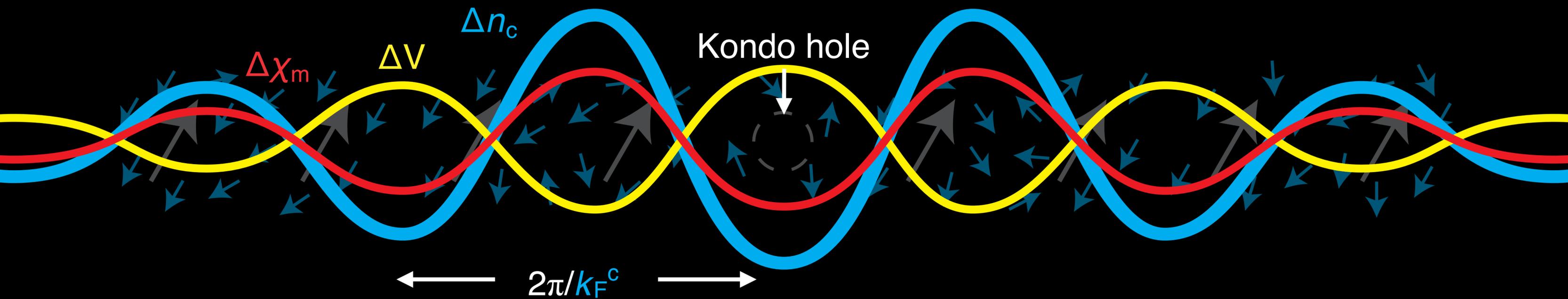
# Kondo holes modulate **magnetic susceptibility**



# Kondo holes reveal the **itinerant electrons**

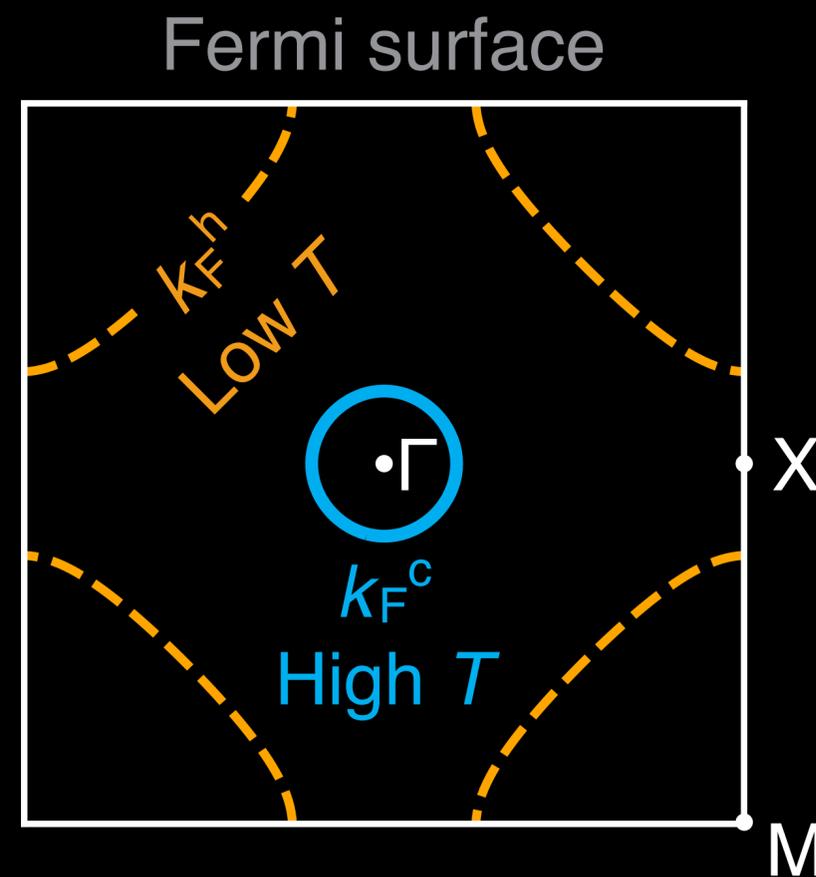


# How can we measure them?

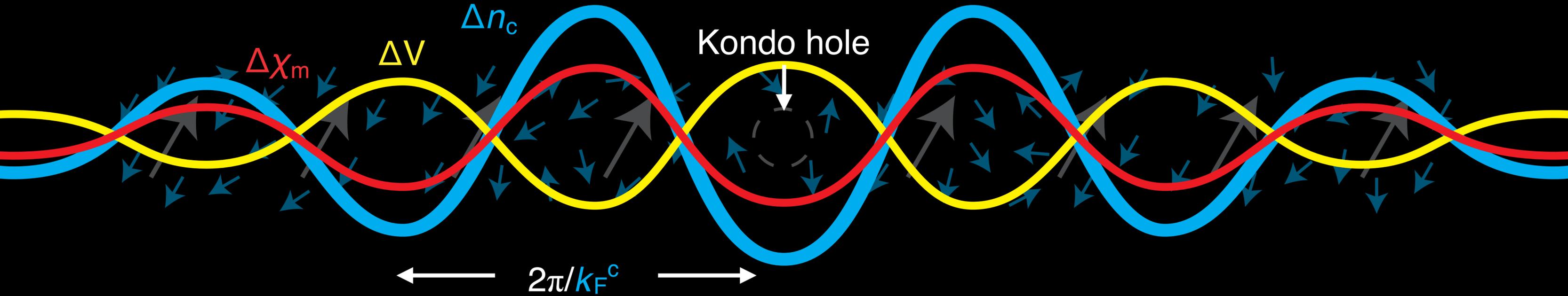


Experimental requirements:

- < nm spatial resolution
- < mV energy resolution
- $\sim$  K temperature
- Charge probe

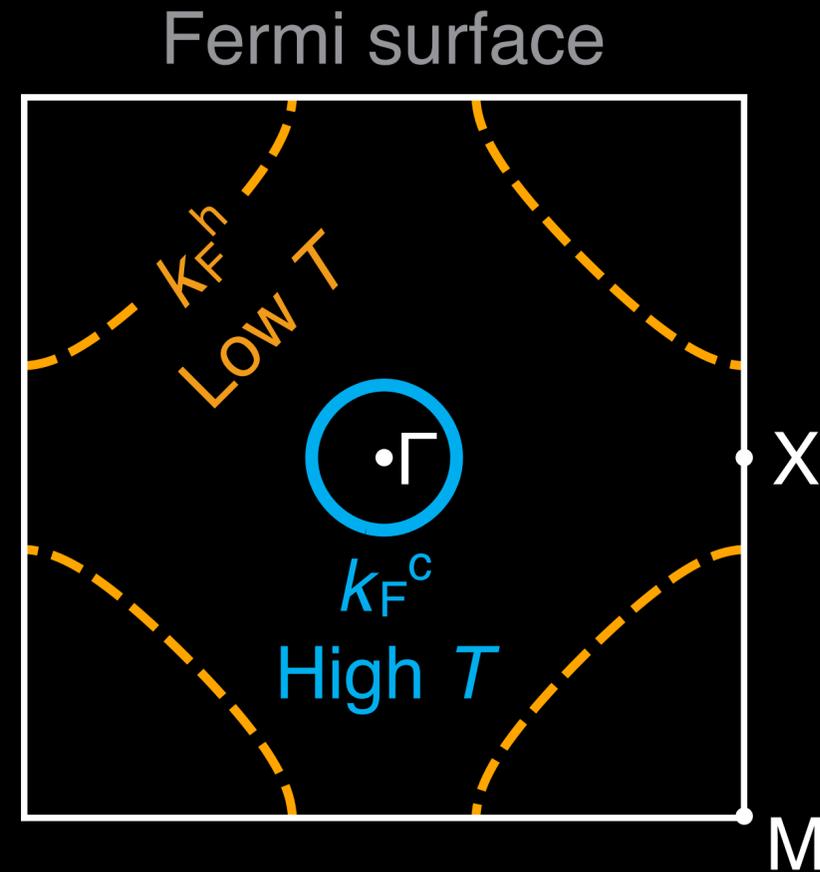


# STM could work, but needs **charge sensitivity**



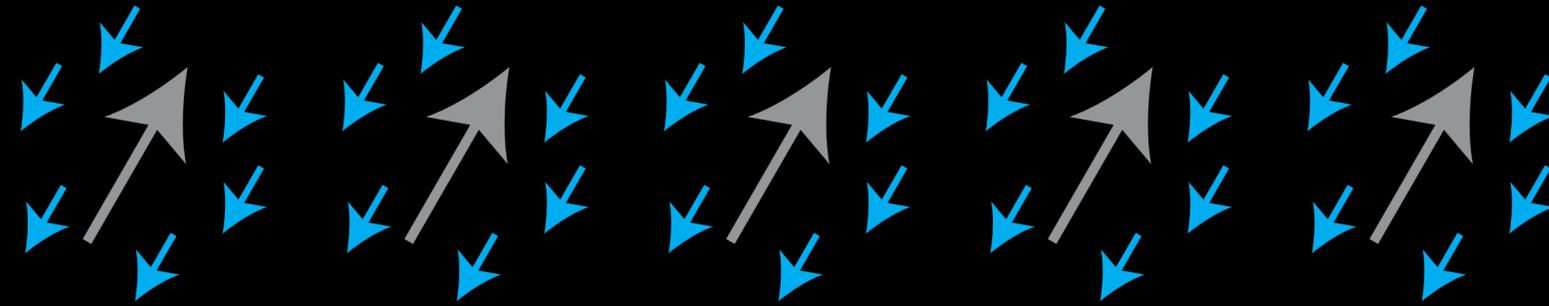
Scanning tunneling microscopy:

- ✓  $< \text{nm}$  spatial resolution
- ✓  $< \text{mV}$  energy resolution
- ✓  $\sim \text{K}$  temperature
- ✗ Charge probe



# What happens when we add charge?

Adding charge cannot change the number of  $f$  electrons,  $n_f$

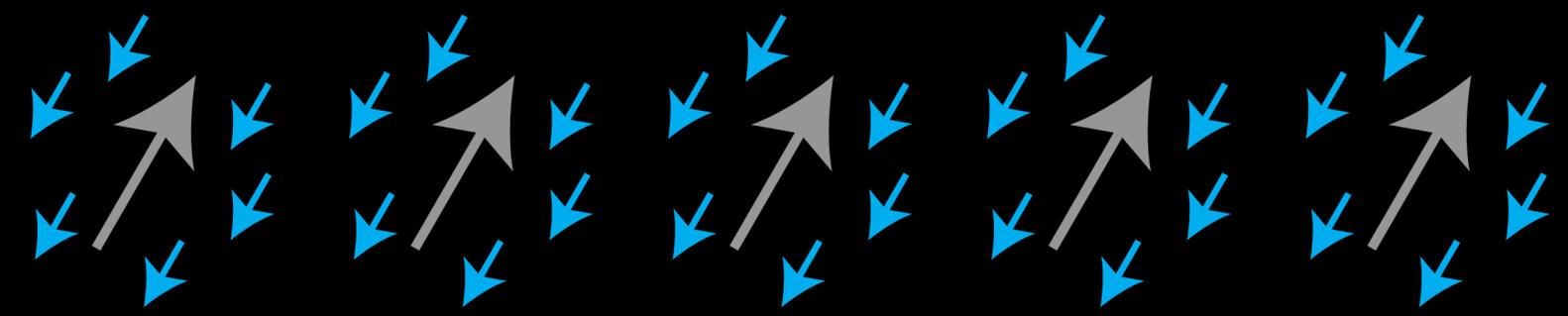


Eric  
Mascot

Dirk  
Morr

# What happens when we add charge?

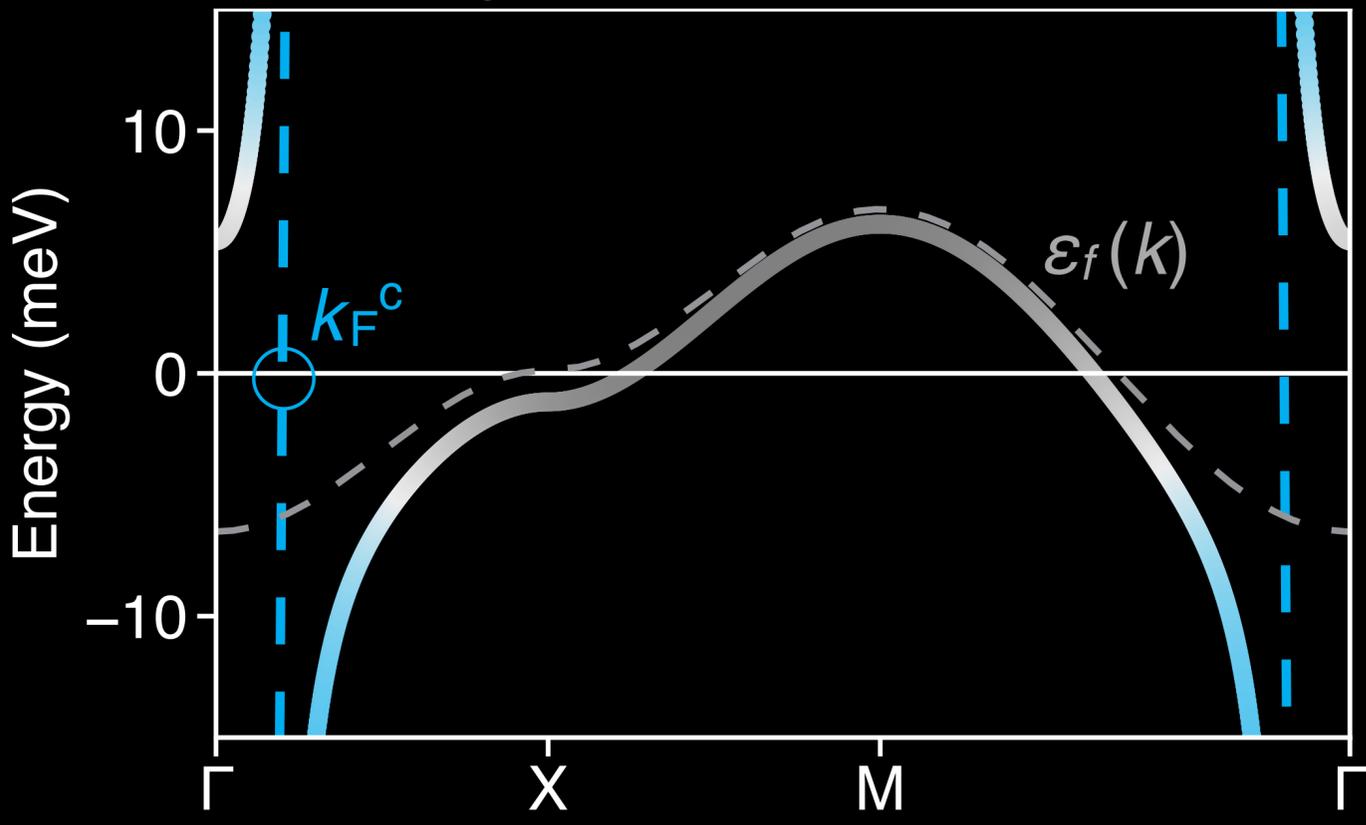
Adding charge cannot change the number of  $f$  electrons,  $n_f$



Eric Mascot

Dirk Morr

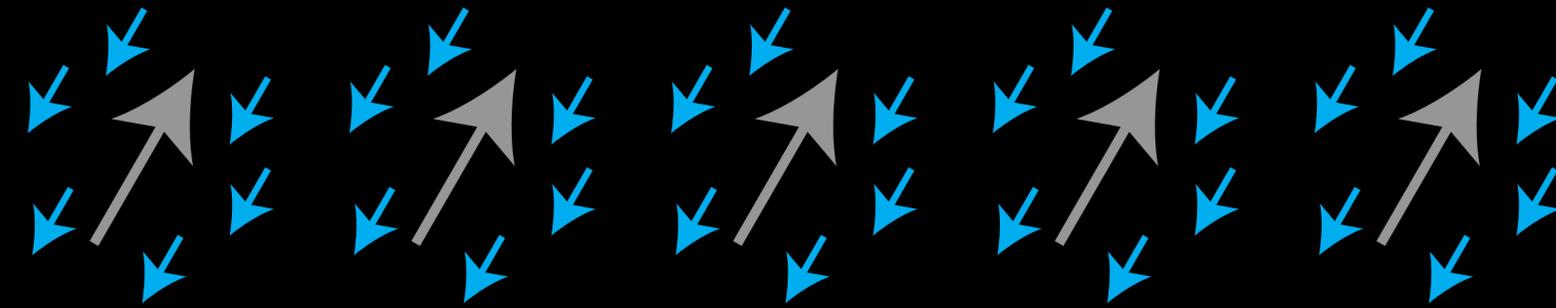
Heavy fermion band structure



integrated  
 $n_f = f$  orbital = 1  
character

# Adding charge adjusts the energy position of $\epsilon_f$

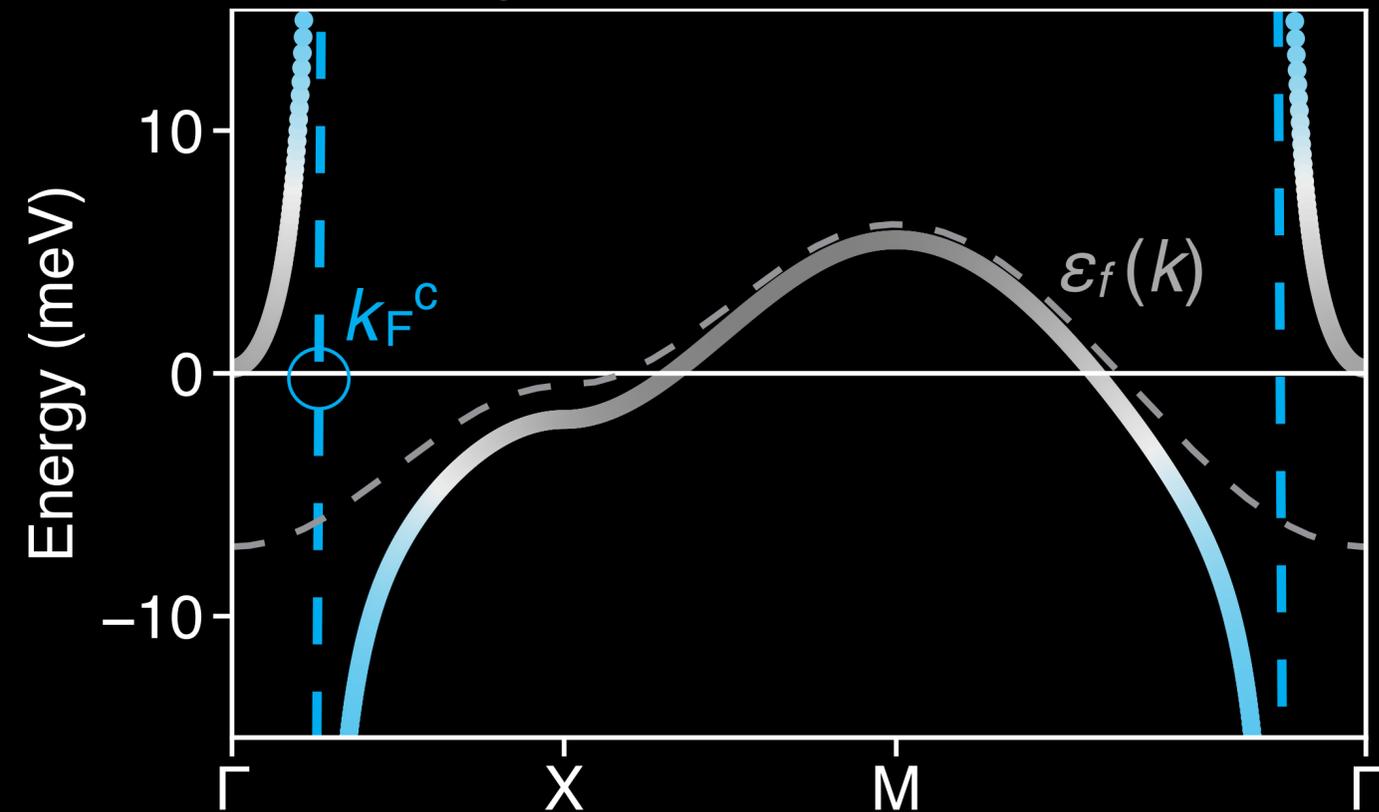
Adding charge cannot change the number of  $f$  electrons,  $n_f$



Eric Mascot

Dirk Morr

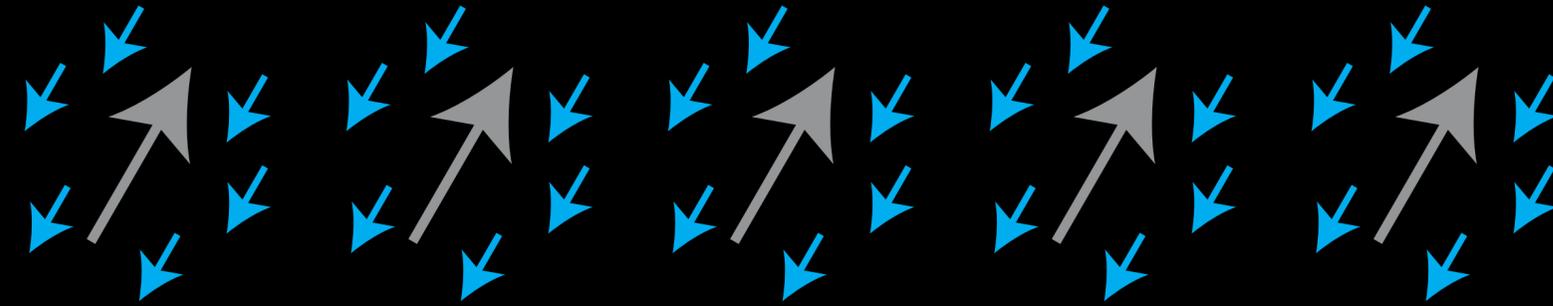
Heavy fermion band structure



integrated  
 $n_f = f$  orbital = 1  
character

# The Kondo resonance is a **charge** sensor

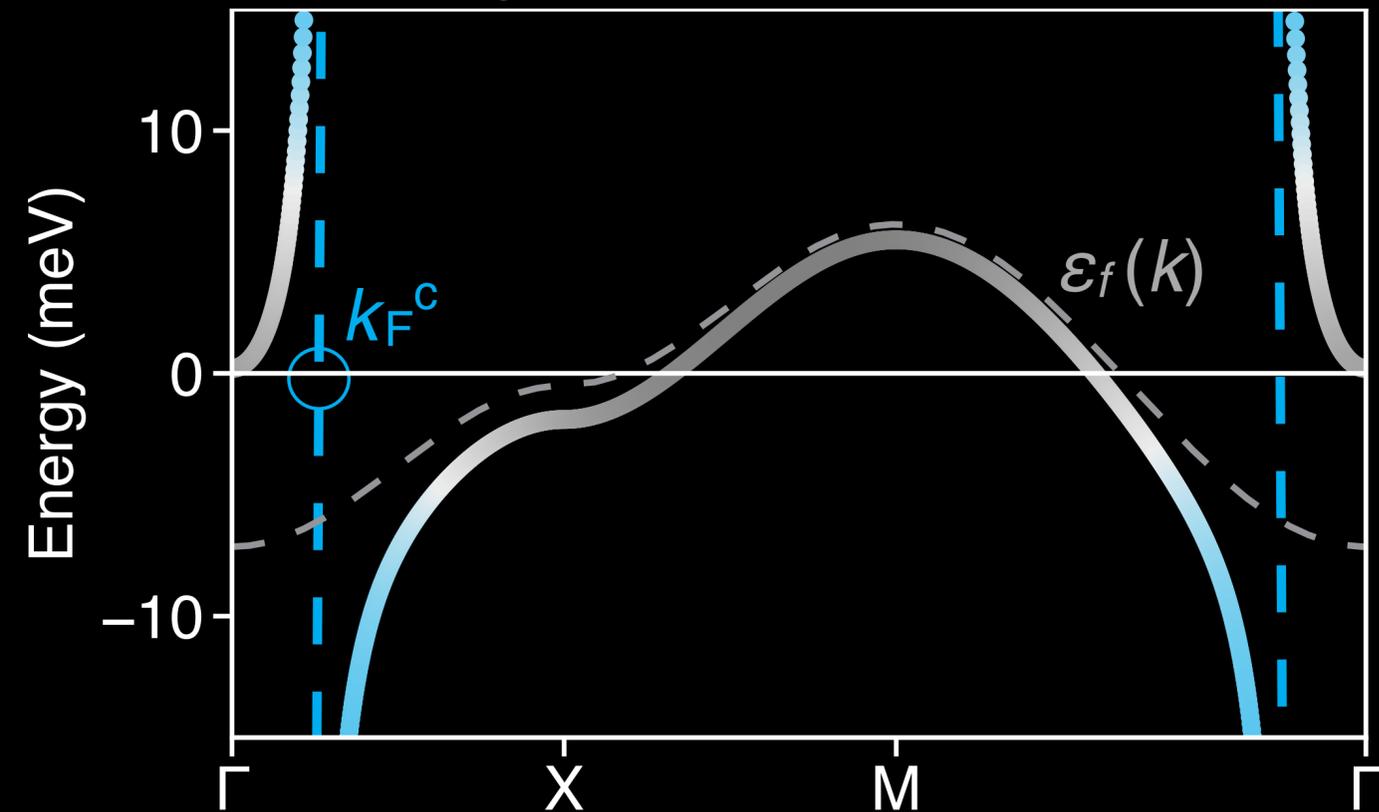
Adding charge cannot change the number of  $f$  electrons,  $n_f$



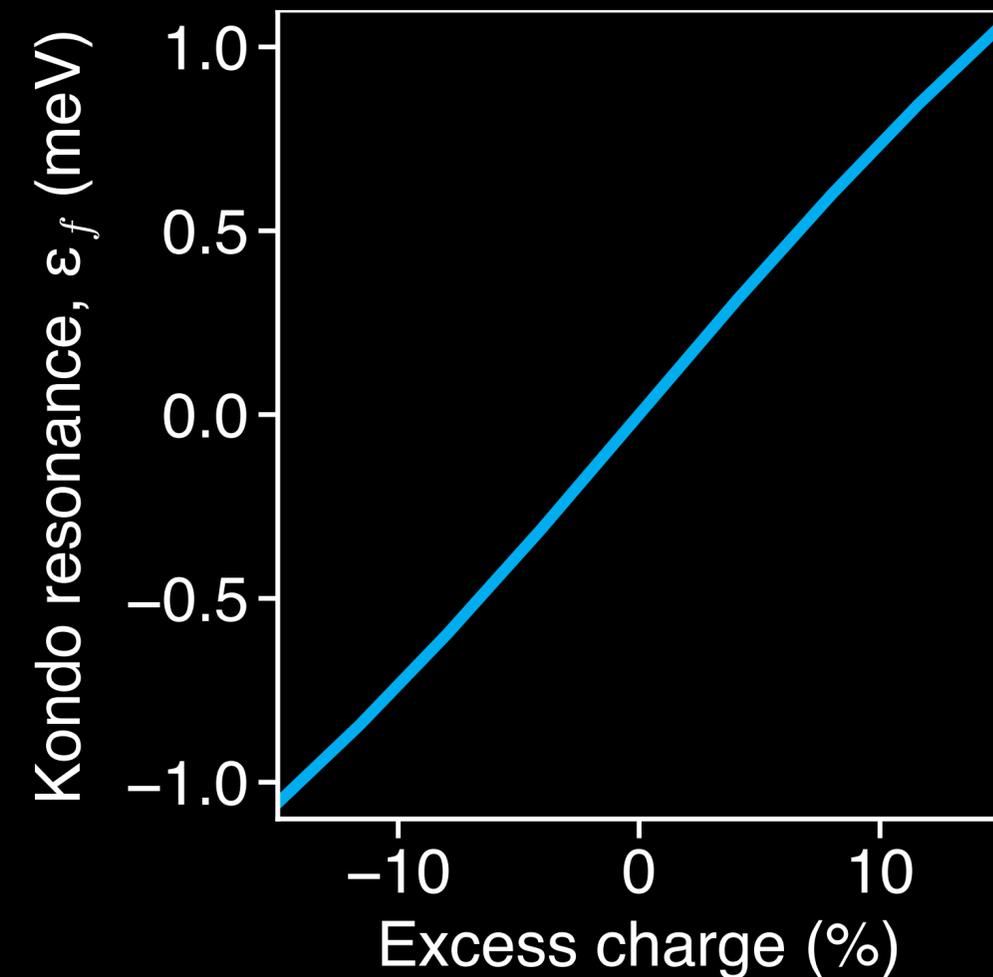
Eric Mascot

Dirk Morr

Heavy fermion band structure

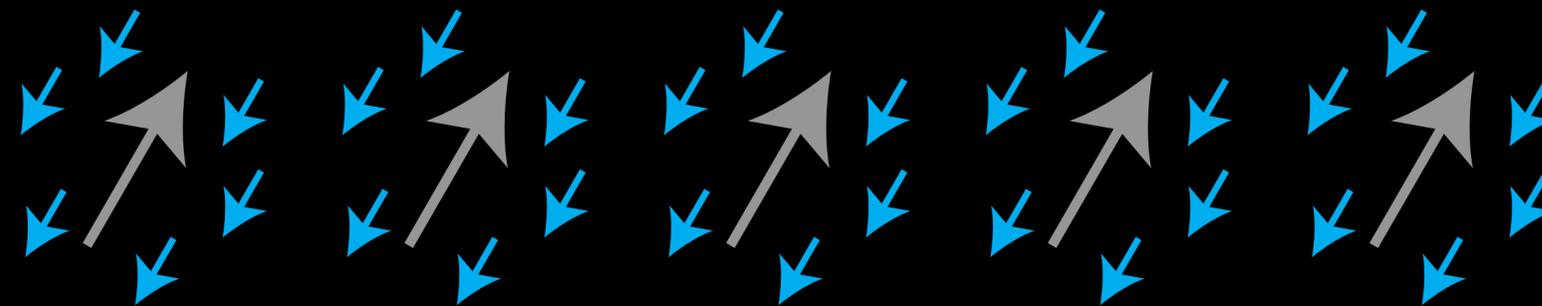


integrated  
 $n_f = f$  orbital = 1  
character



# How to image the small $\Delta\varepsilon_f$ shifts?

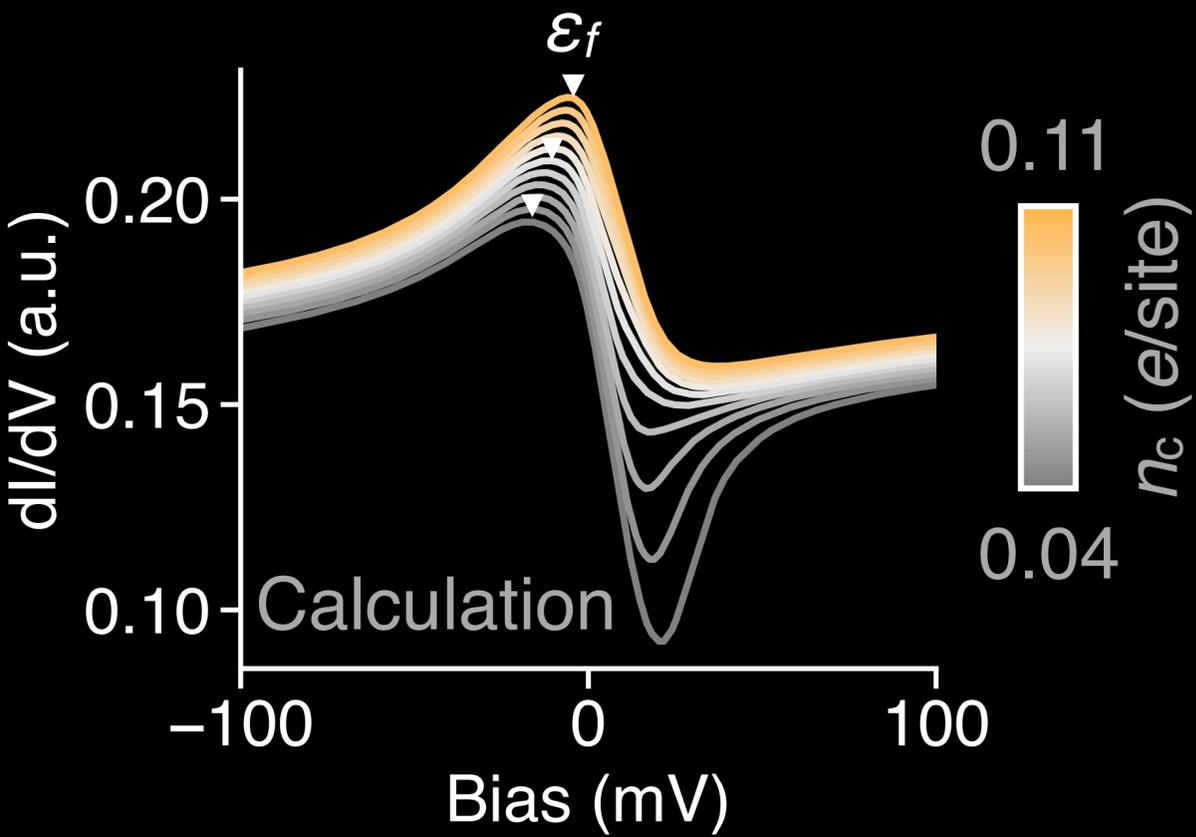
Adding charge cannot change the number of  $f$  electrons,  $n_f$



Eric  
Mascot

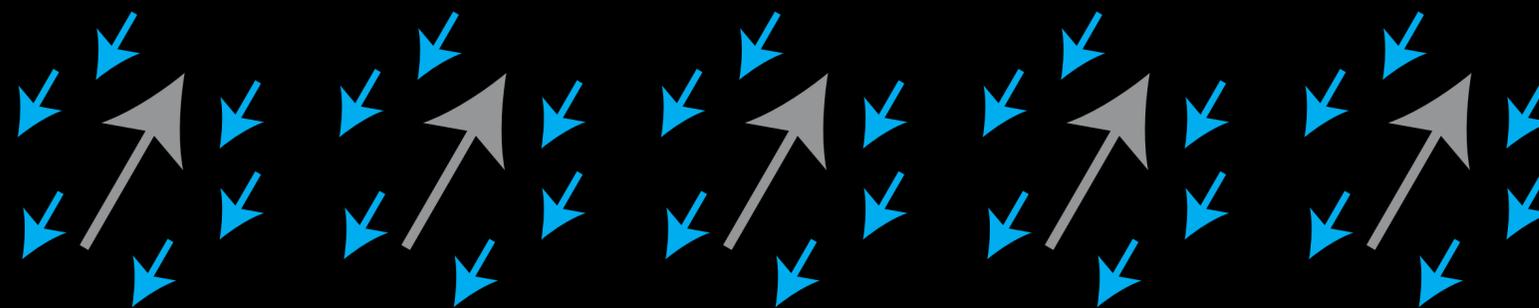
Dirk  
Morr

(1)  $\Delta\varepsilon_f$  moves  $dI/dV$  peak position

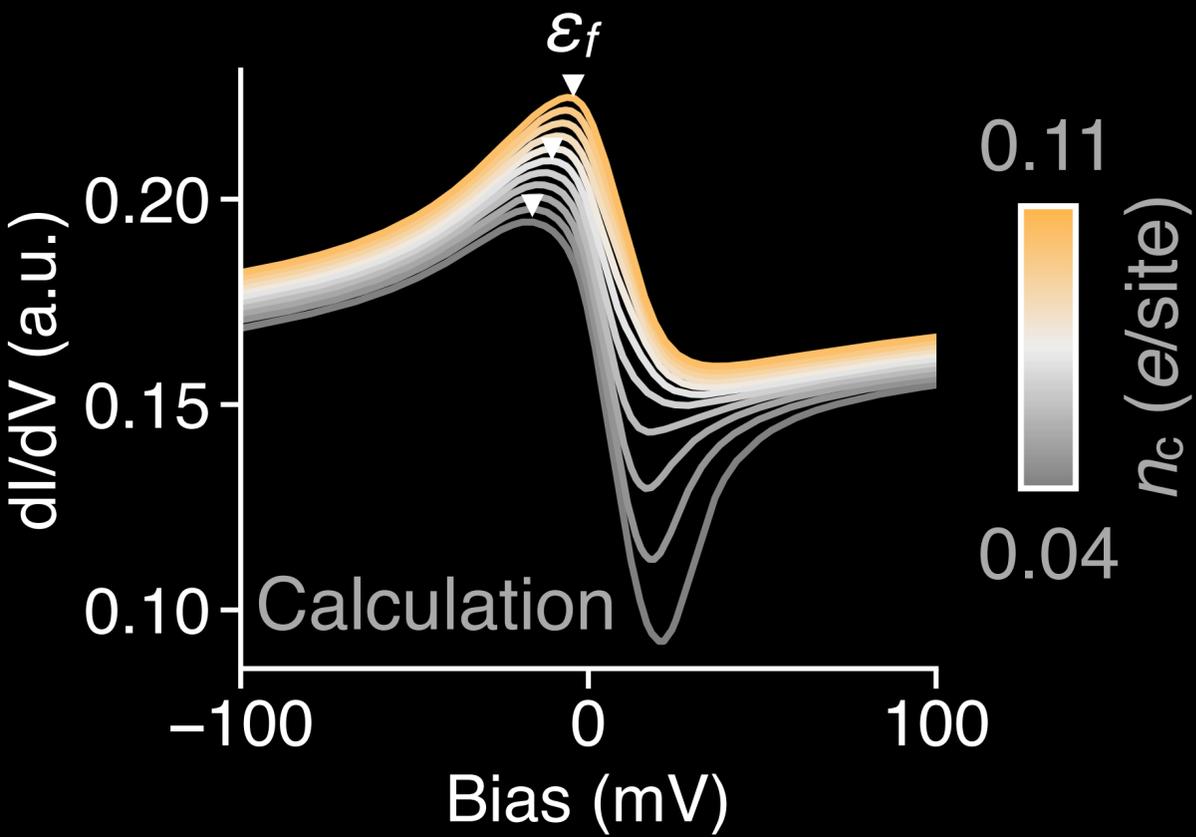


# The $\Delta\varepsilon_f$ shifts are magnified in $R(\mathbf{r}, V)$

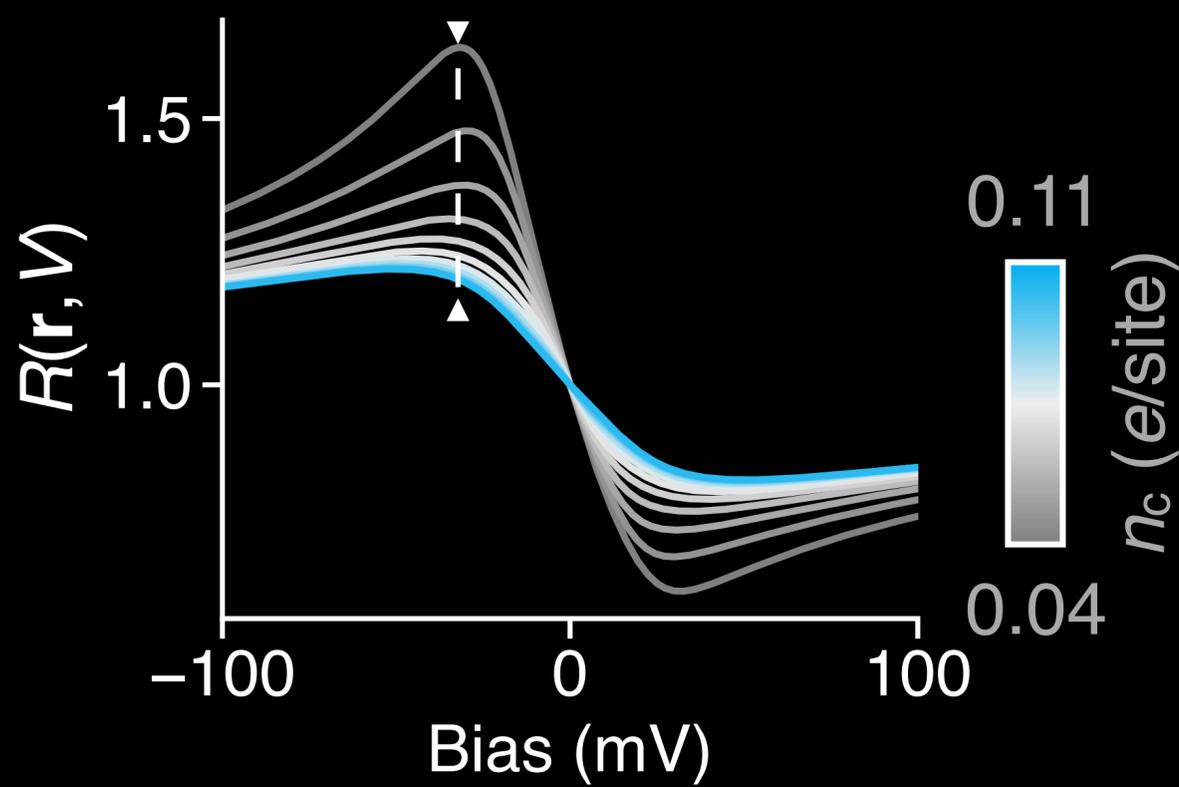
Adding charge cannot change the number of  $f$  electrons,  $n_f$



(1)  $\Delta\varepsilon_f$  moves  $dI/dV$  peak position



(2) Changes  $I(+V)$  compared to  $I(-V)$

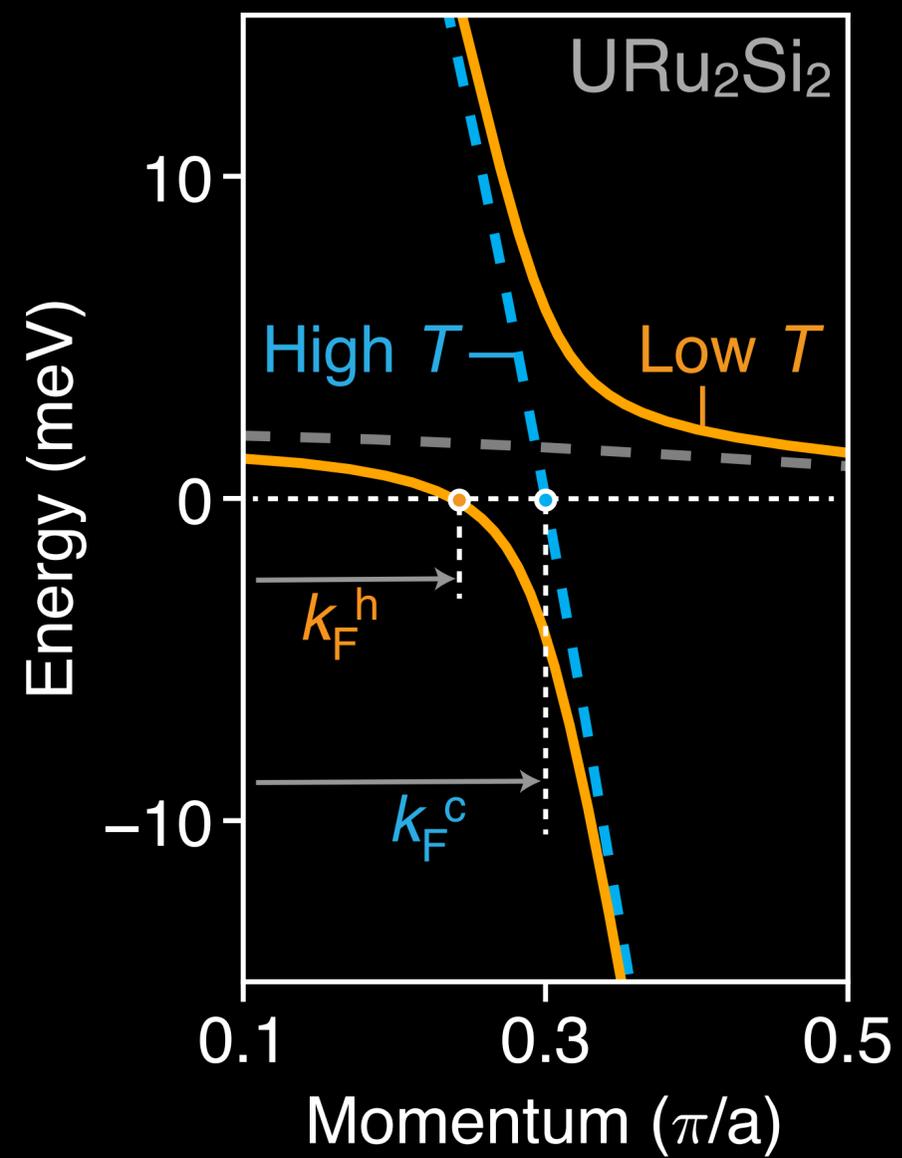


Rectification:

$$R(\mathbf{r}, V) = \frac{I(\mathbf{r}, +V)}{I(\mathbf{r}, -V)}$$

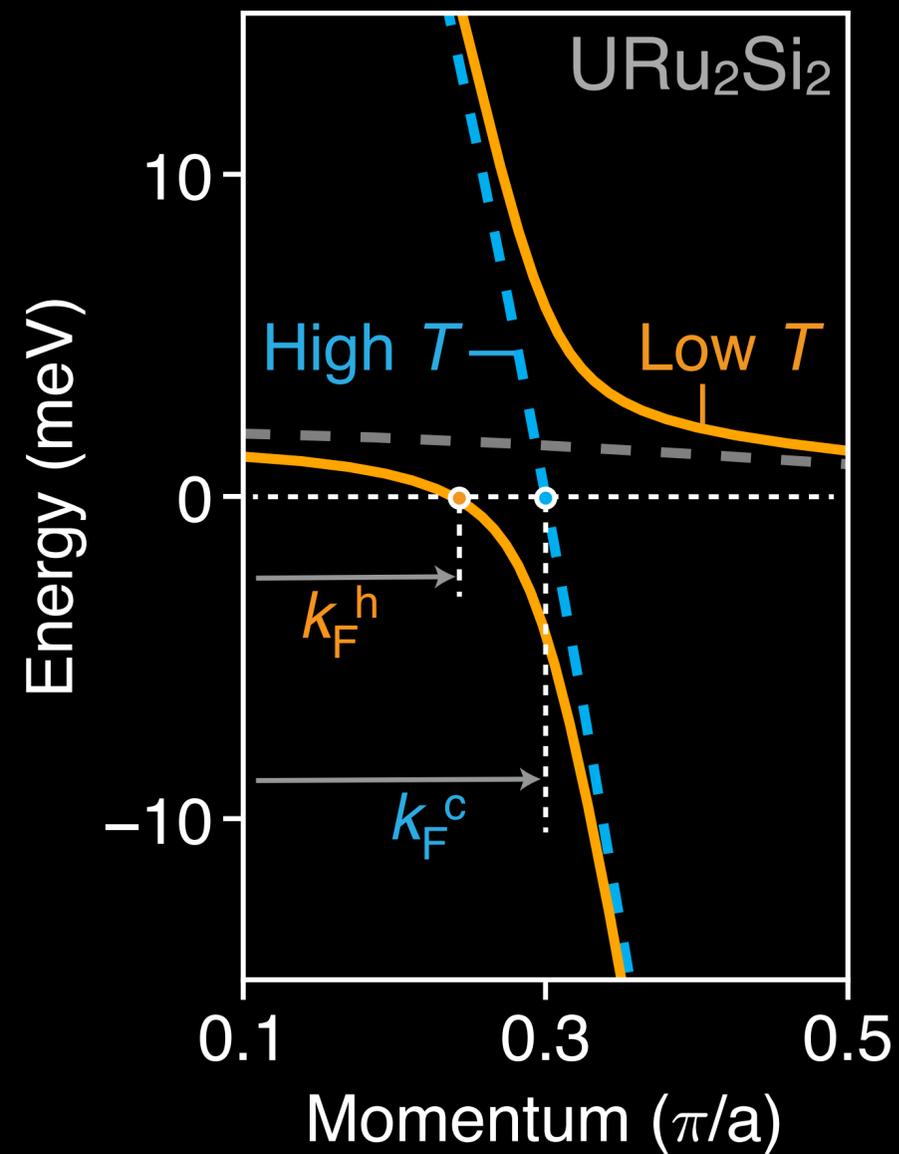
# Test case: URu<sub>2</sub>Si<sub>2</sub>

Heavy fermions at  $T < 16\text{K}$

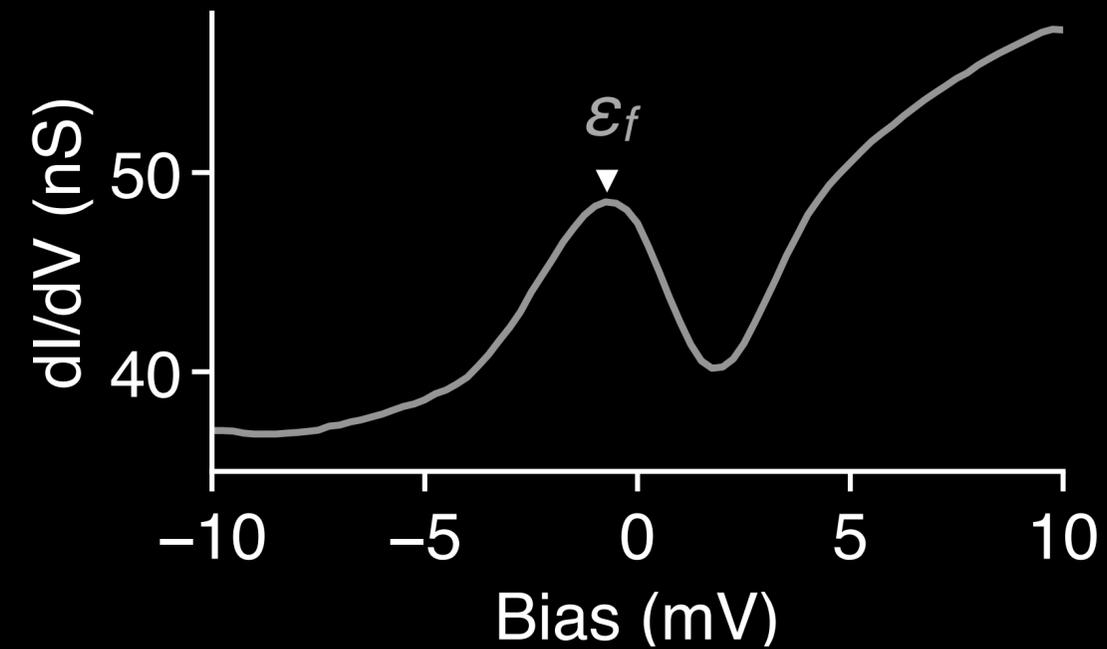


# Kondo resonance appears as Fano-like $dI/dV$

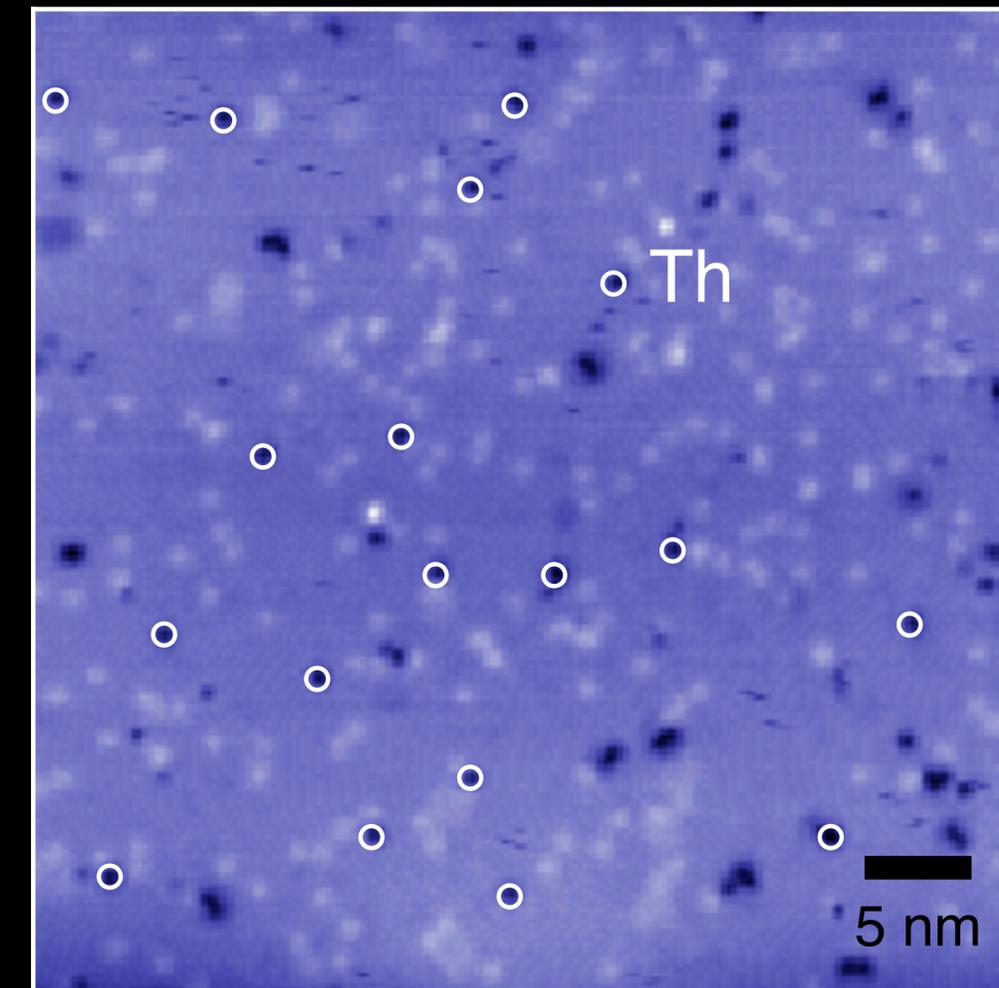
Heavy fermions at  $T < 16\text{K}$



Measured at  $T = 5.9\text{K}$

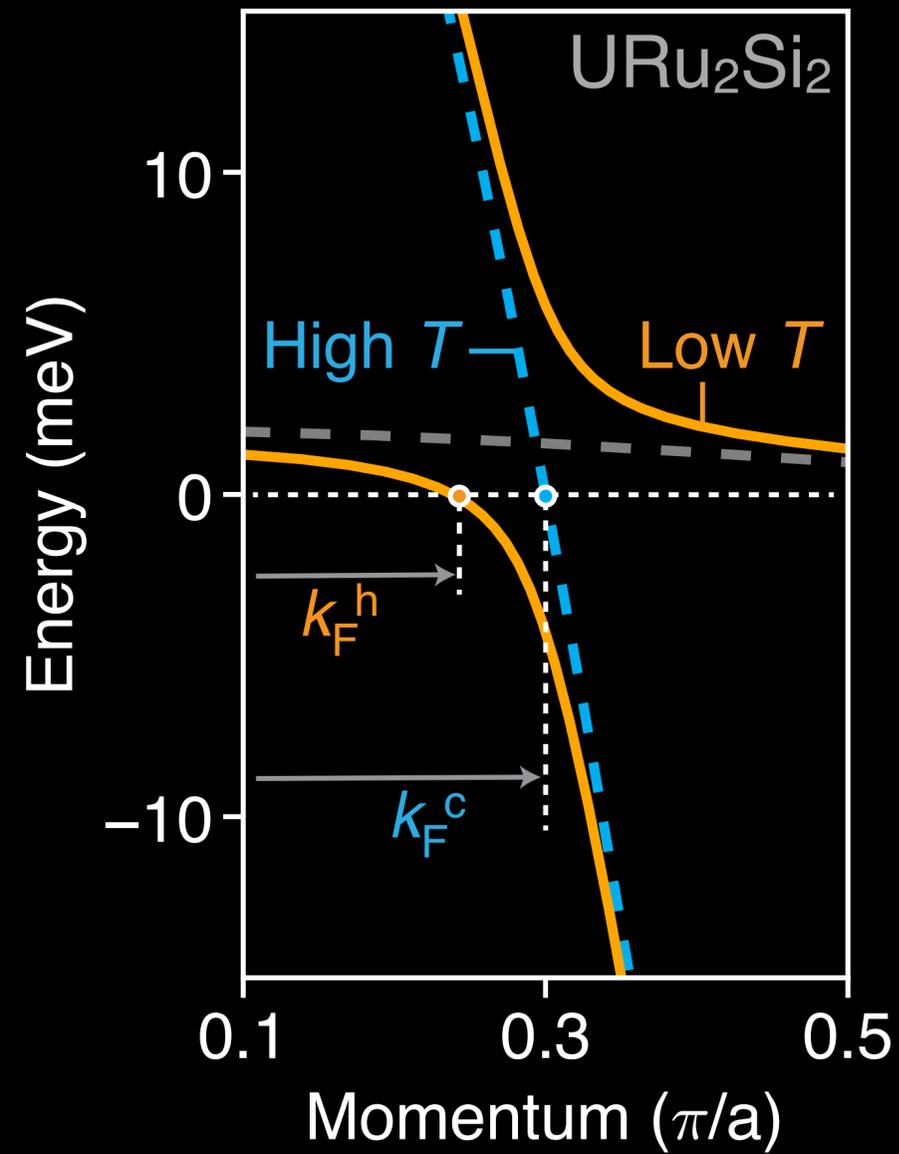


Topography  
0 pm  40 pm

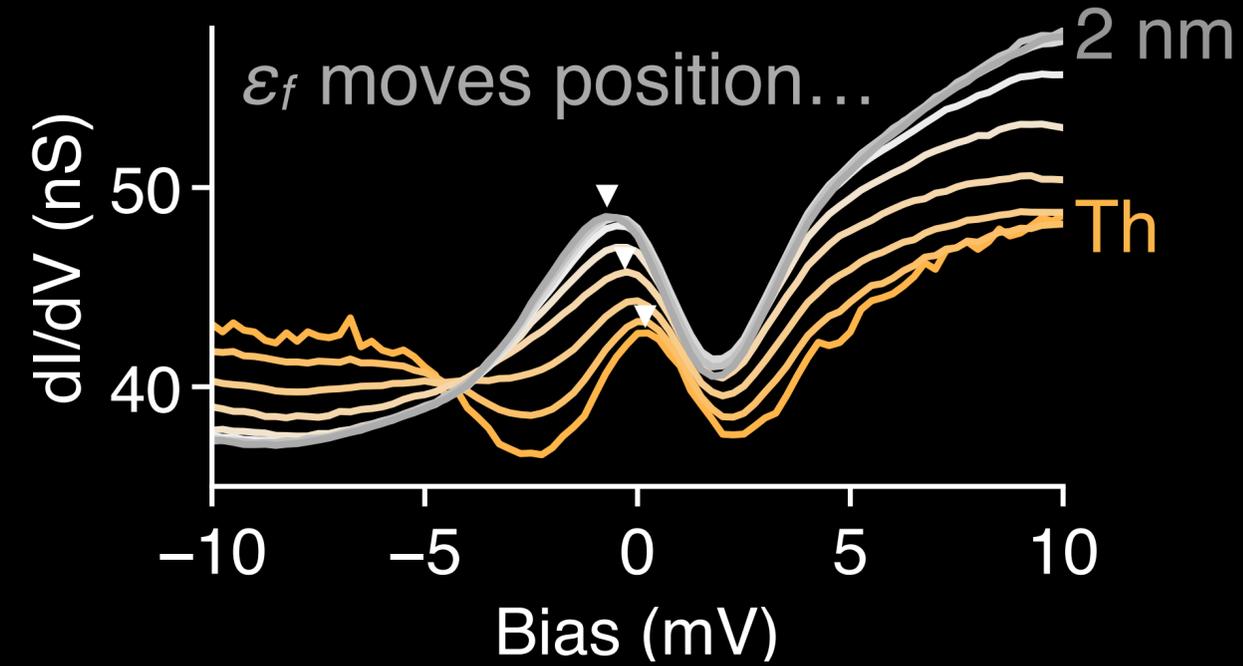


# $\epsilon_f$ shifts energy position around Th dopants

Heavy fermions at  $T < 16\text{K}$

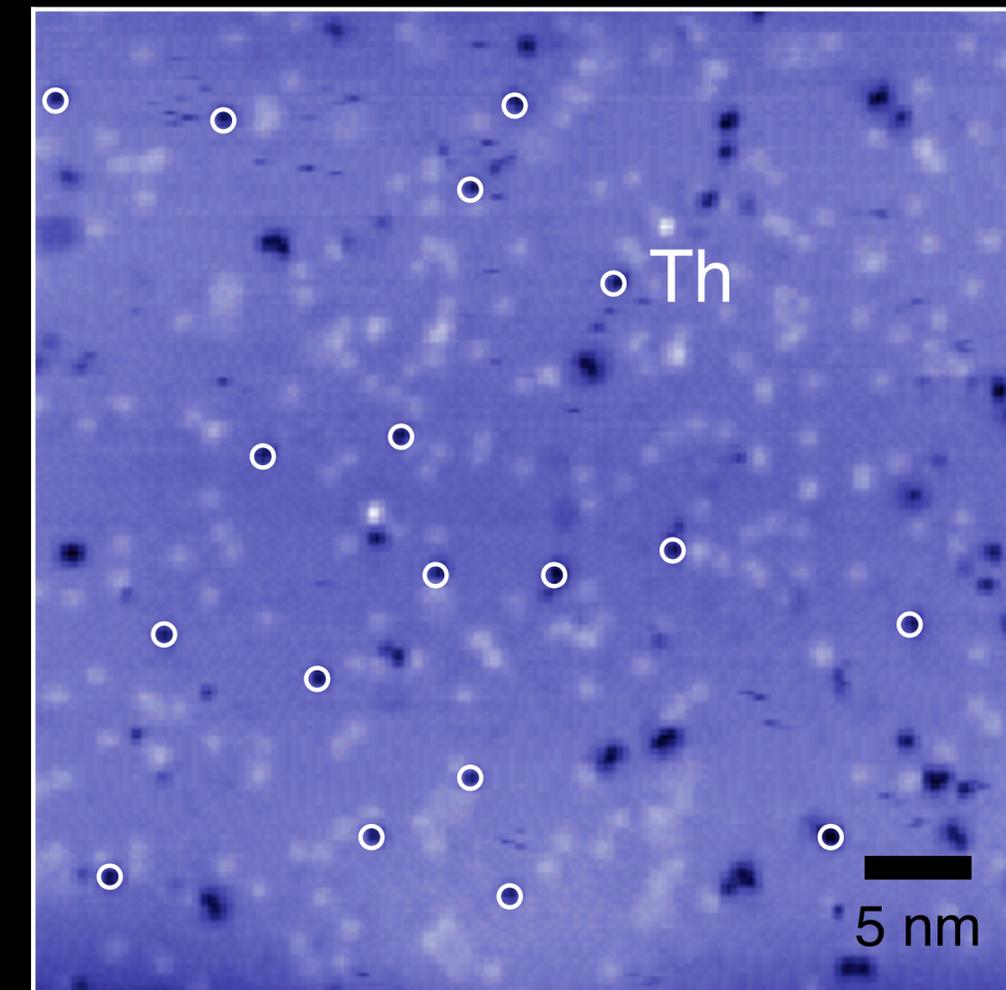


Measured at  $T = 5.9\text{ K}$



Topography

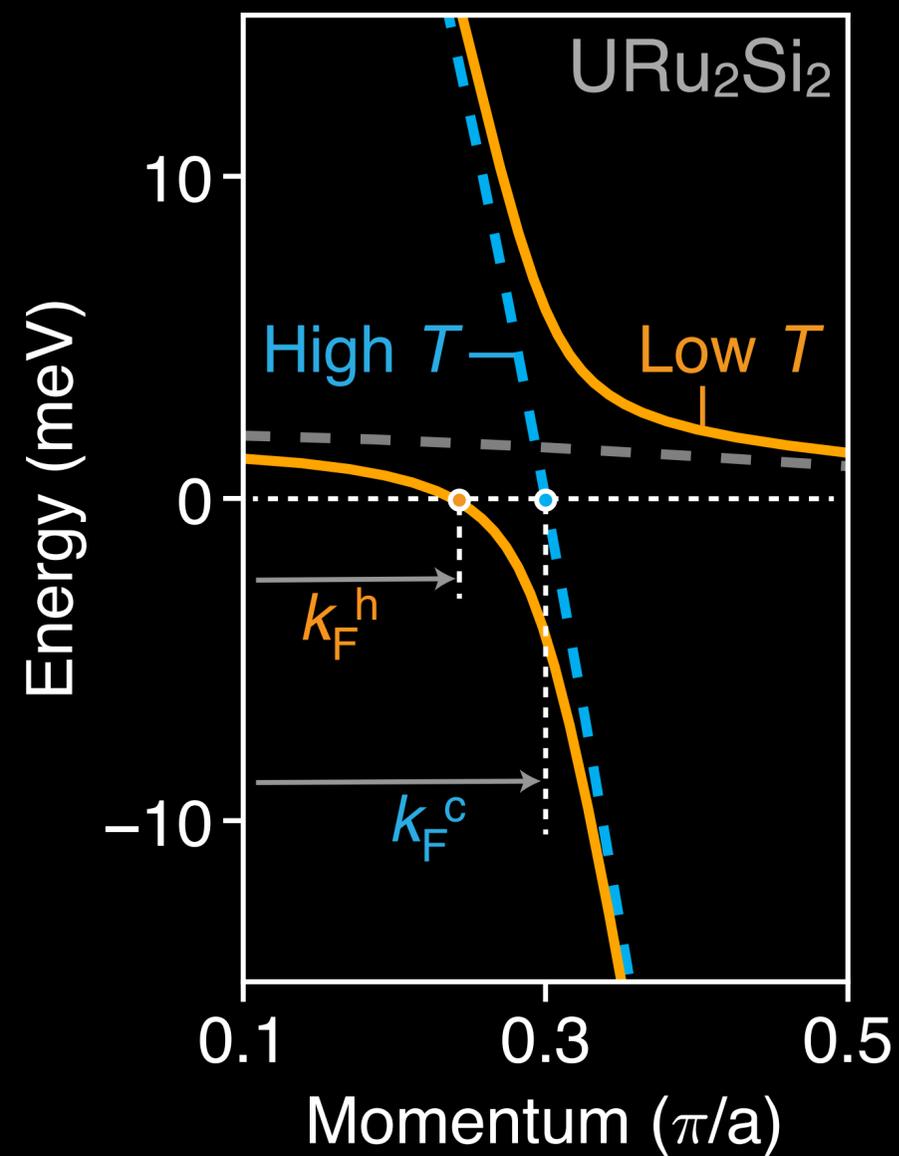
0 pm 90 pm



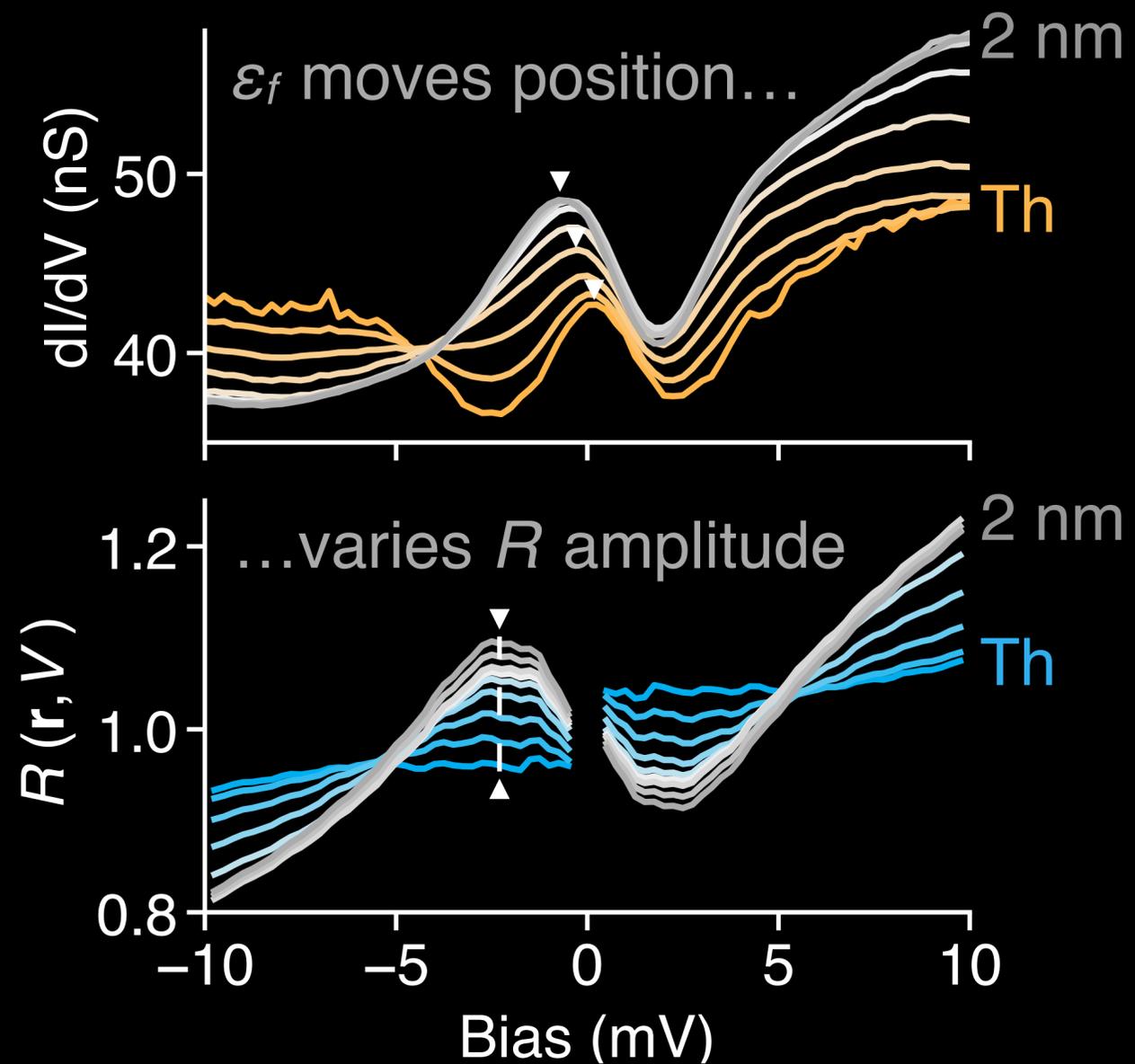
Measured at  $T = 5.9\text{ K}$

# These $\varepsilon_f$ shifts are amplified in $R(\mathbf{r}, V)$

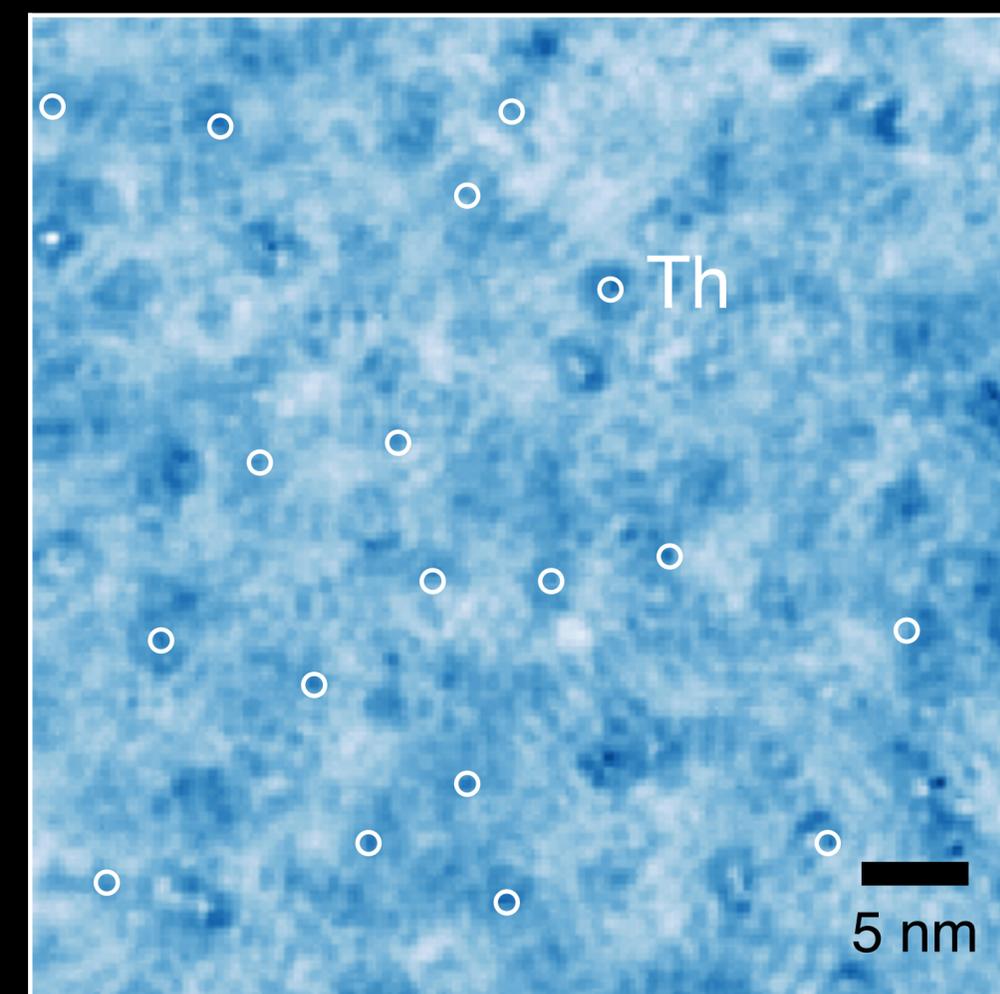
Heavy fermions at  $T < 16\text{K}$



Measured at  $T = 5.9\text{K}$



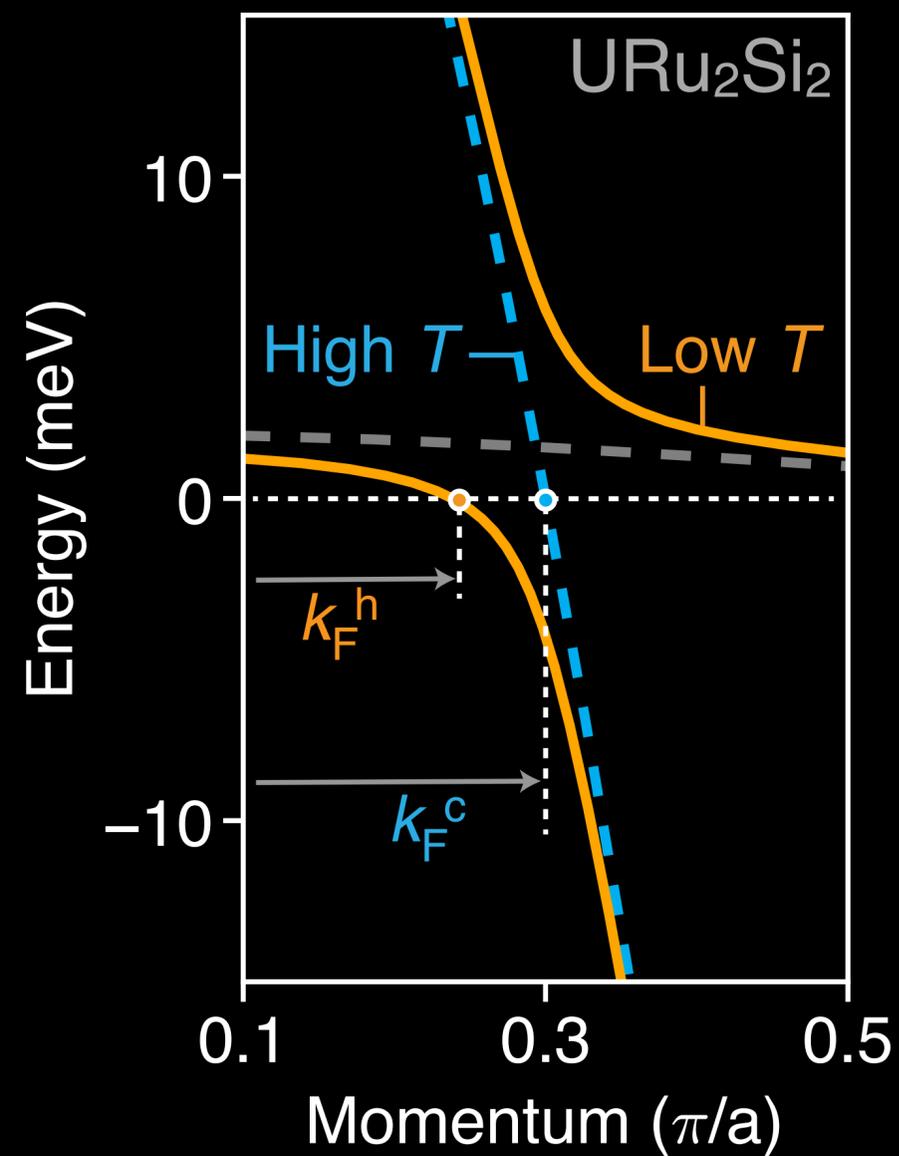
$R(\mathbf{r}, V = -3\text{ meV})$   
0.8  1.3



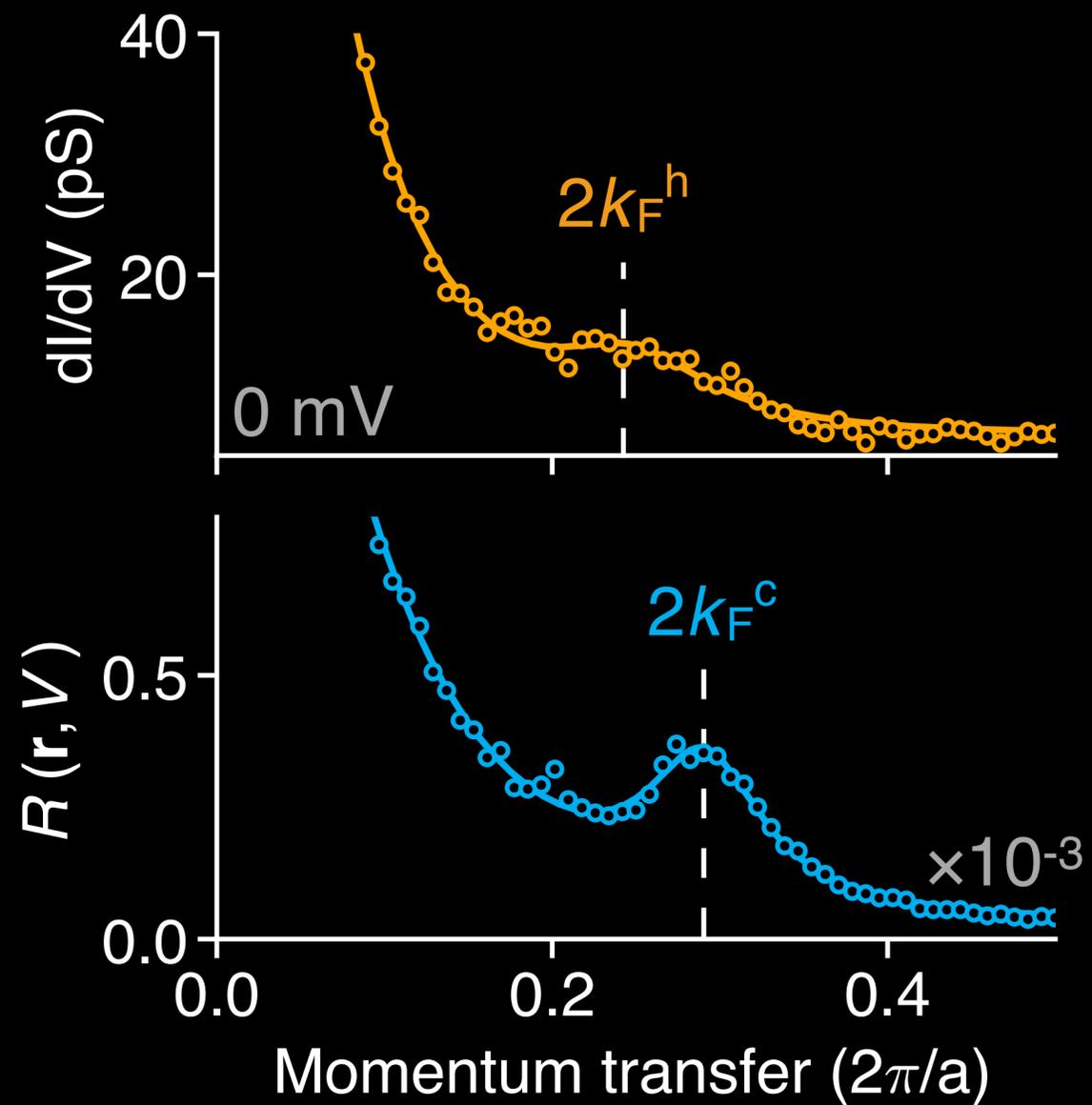
$$R(\mathbf{r}, V) = \frac{I(\mathbf{r}, +V)}{I(\mathbf{r}, -V)}$$

# $R(\mathbf{r}, V)$ reveals itinerant charge

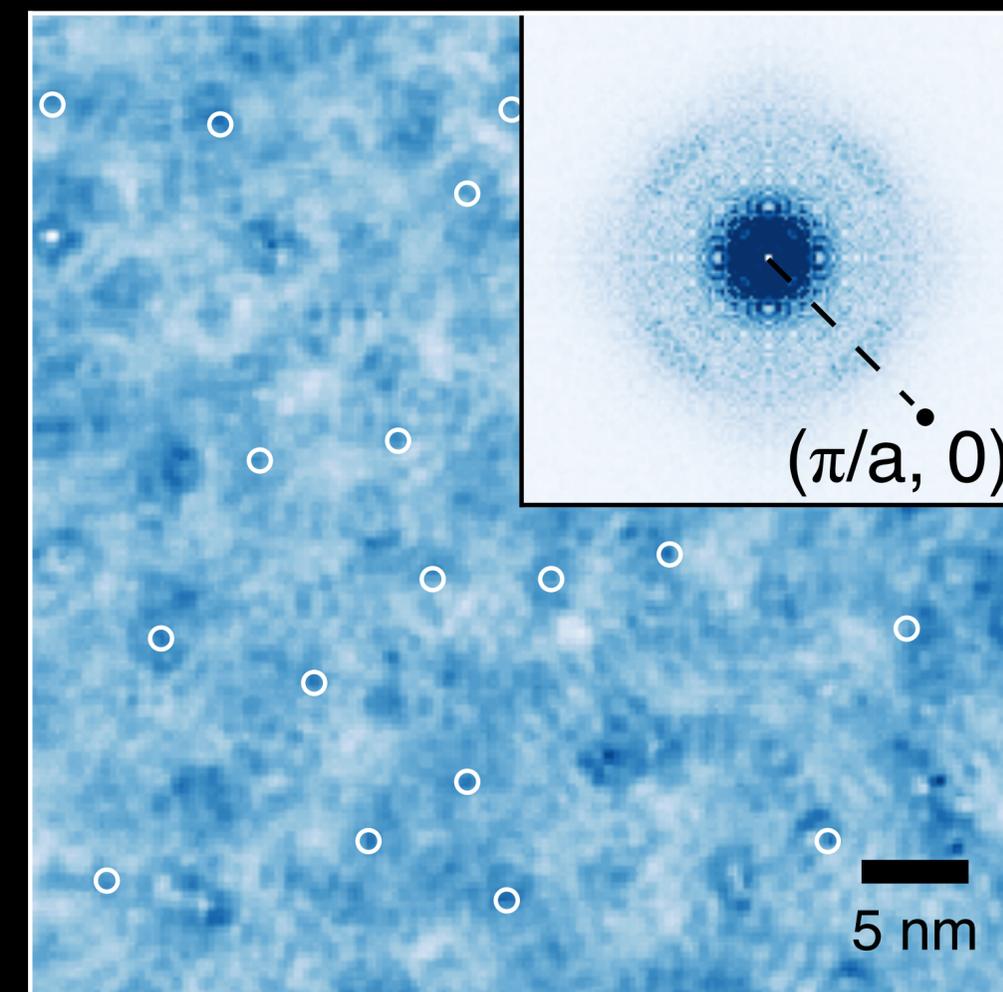
Heavy fermions at  $T < 16\text{K}$



Measured at  $T = 5.9\text{K}$



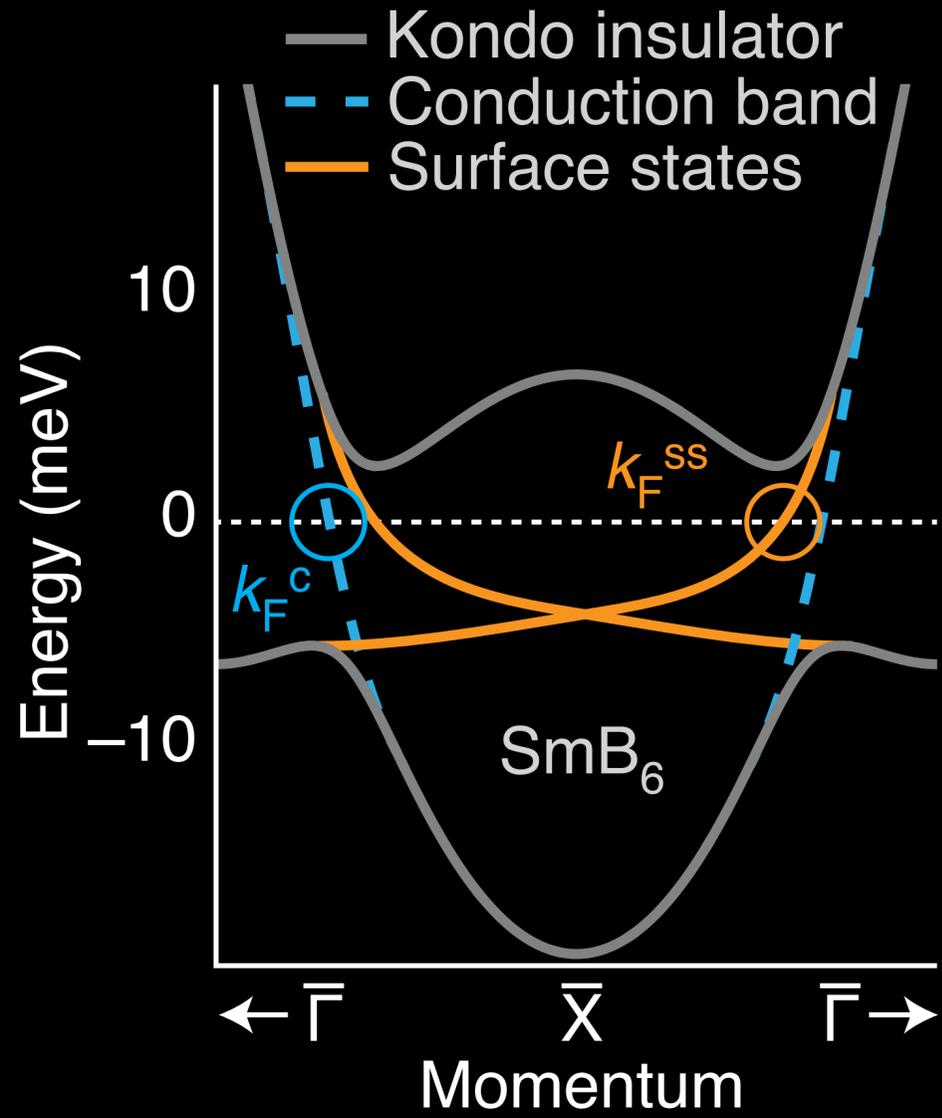
$R(\mathbf{r}, V = -3\text{ meV})$   
0.8  1.3



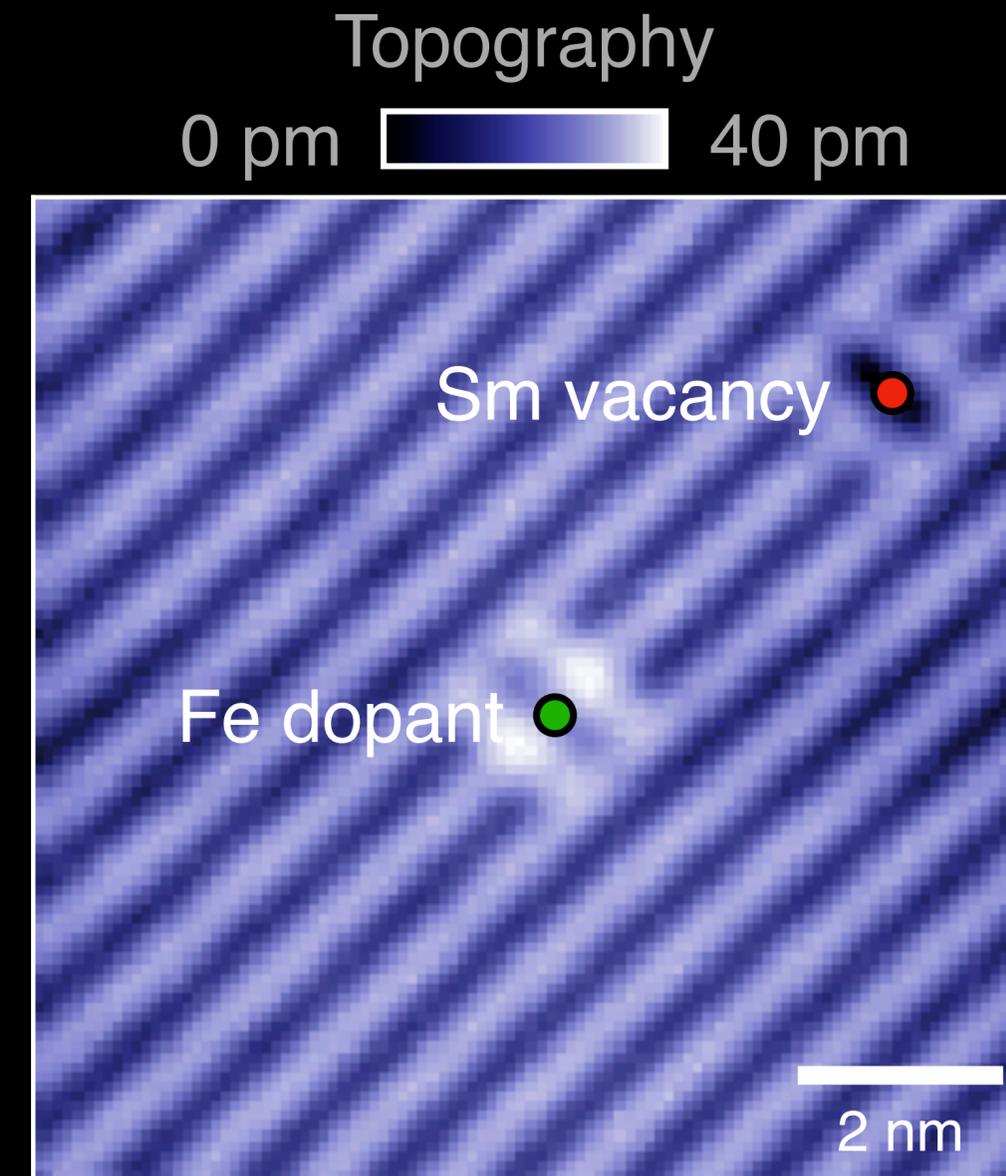
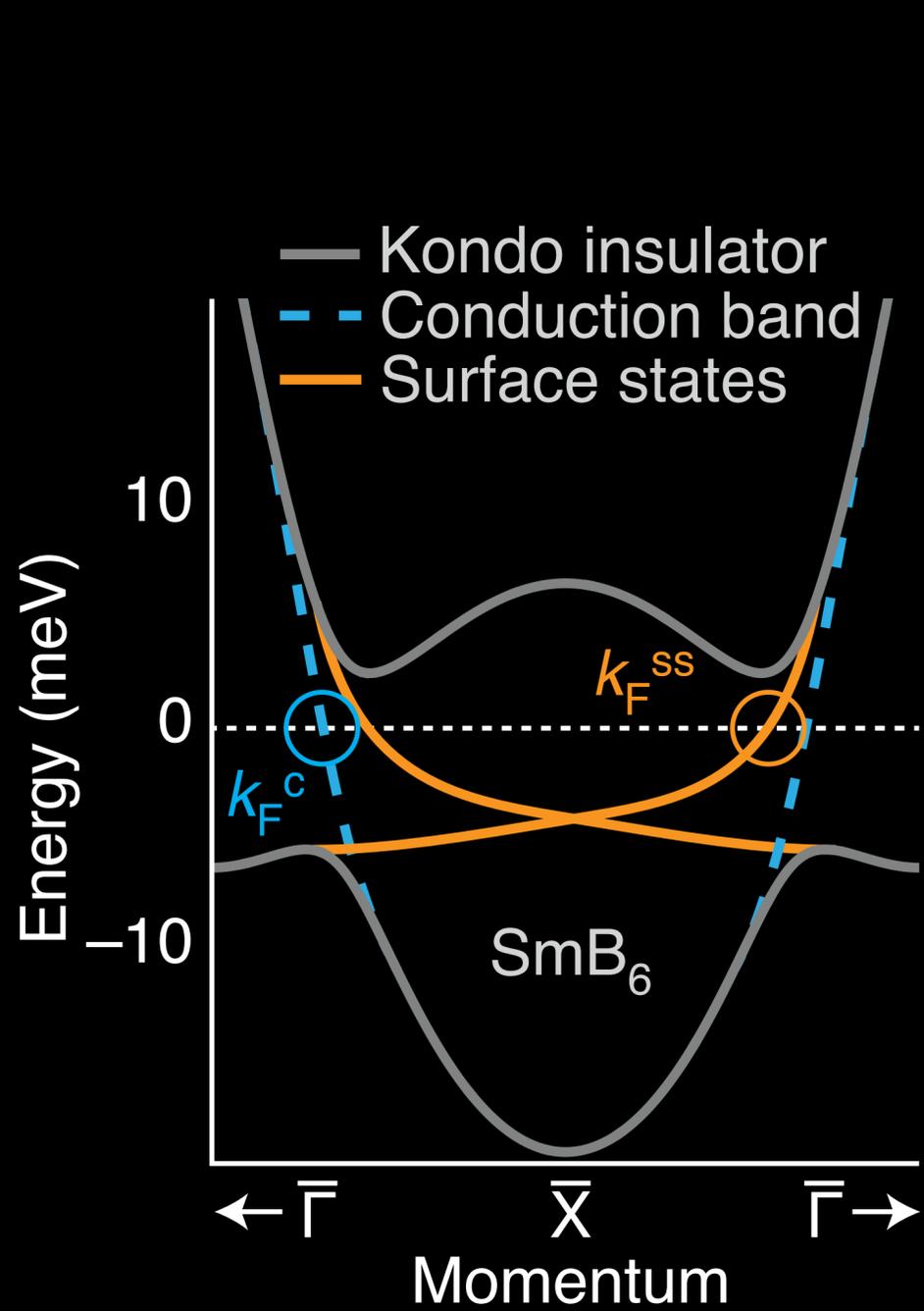
$$R(\mathbf{r}, V) = \frac{I(\mathbf{r}, +V)}{I(\mathbf{r}, -V)}$$

Does this work in  $\text{SmB}_6$ ?

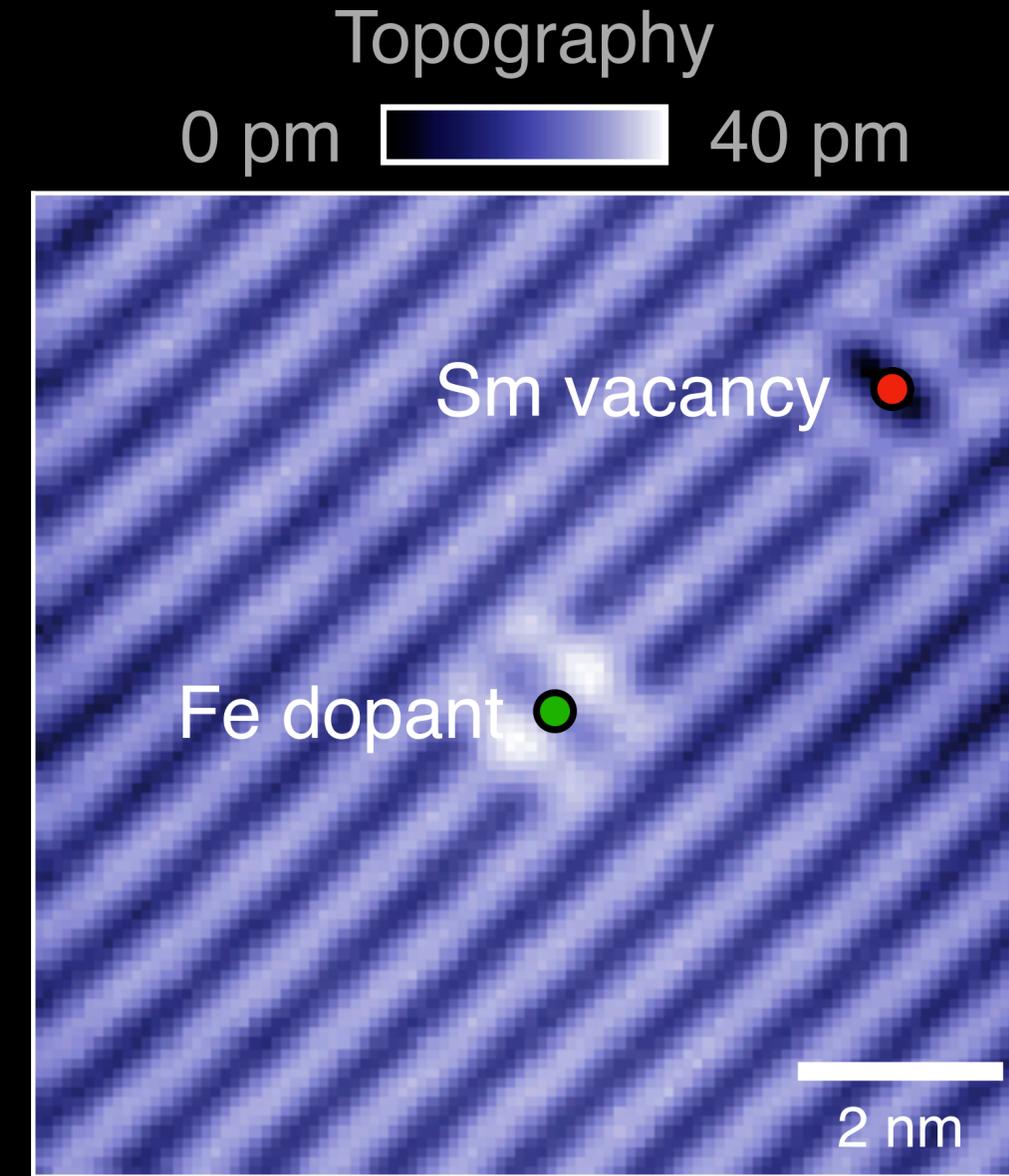
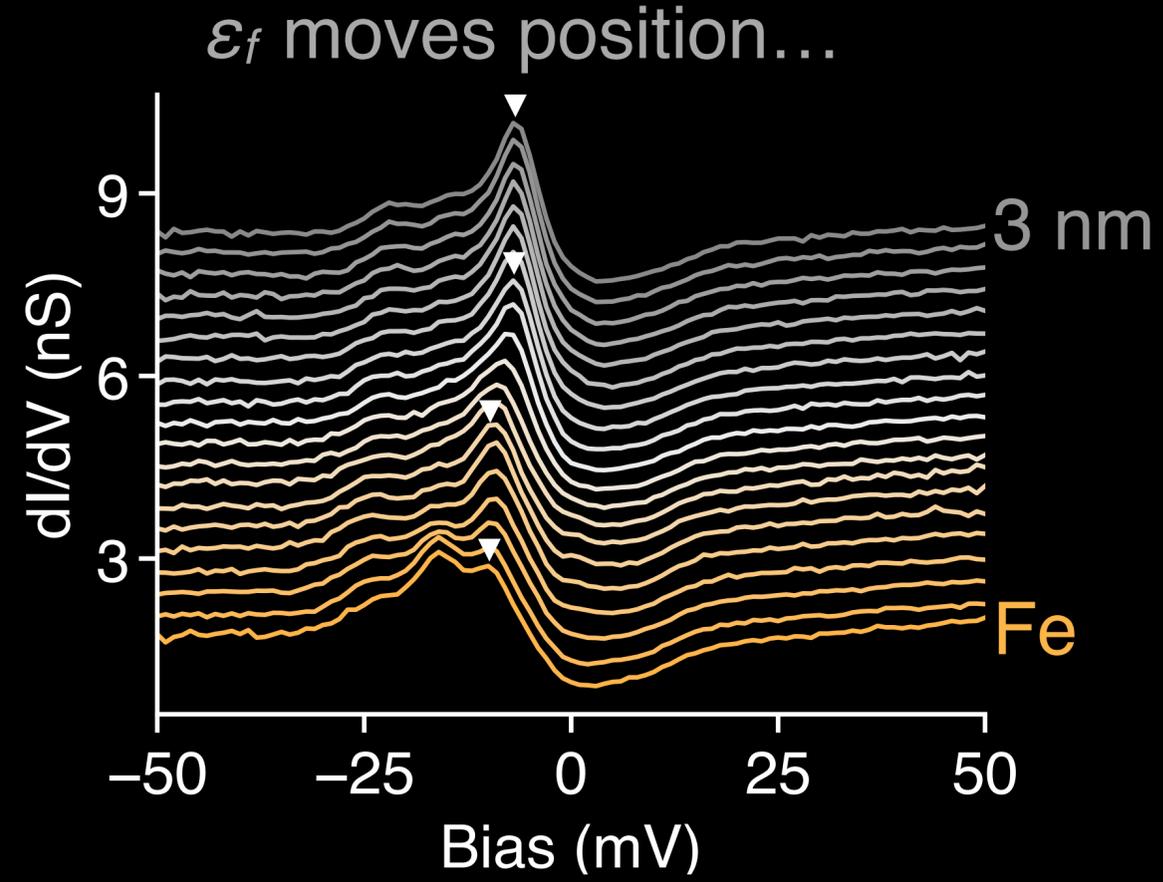
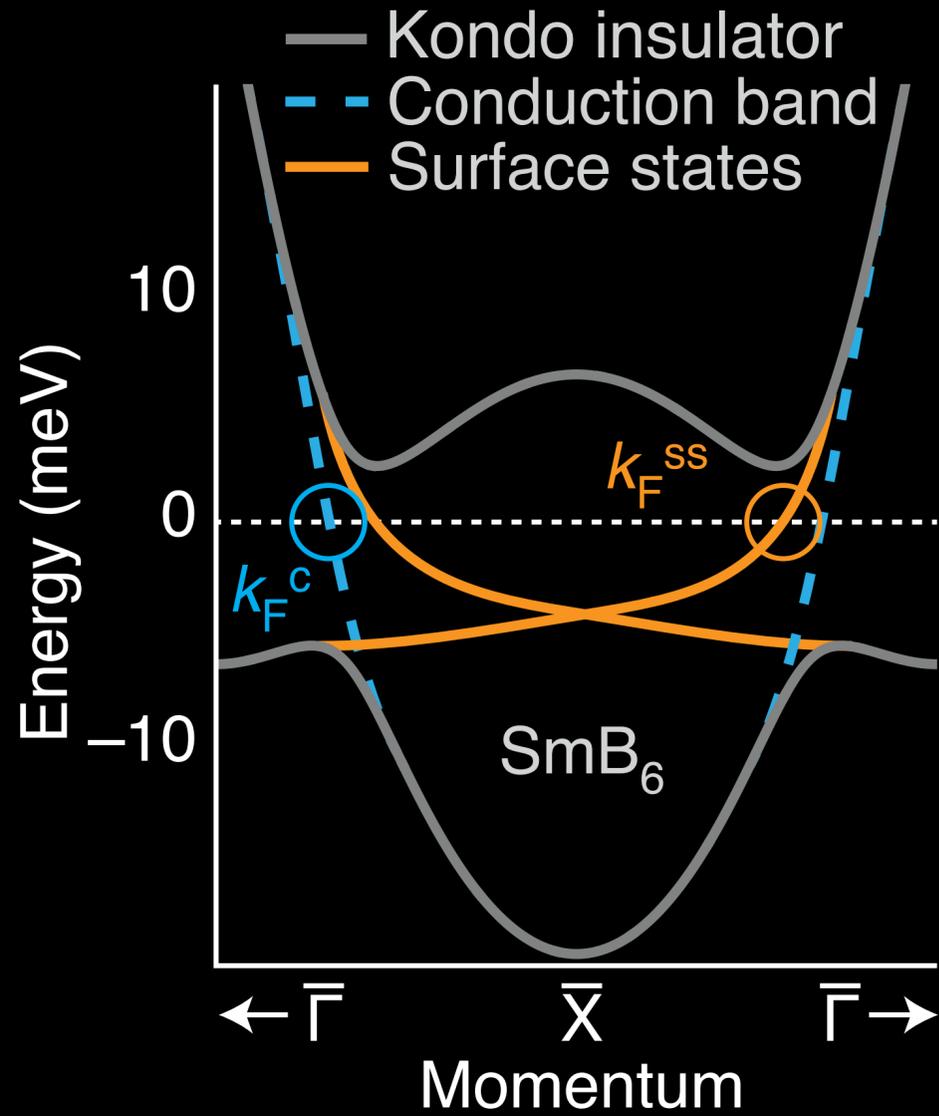
# SmB<sub>6</sub> has heavy Dirac fermions on surface



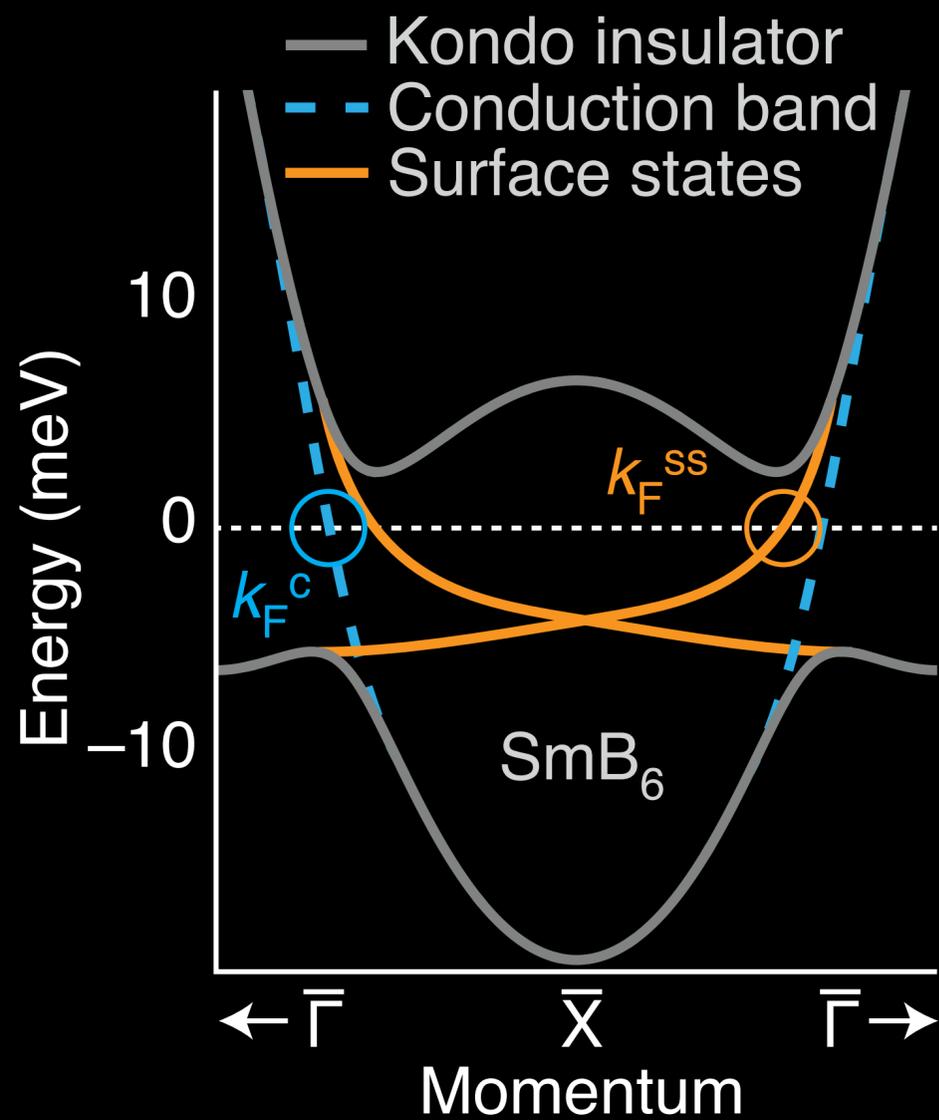
# Fe dopants and Sm vacancies replace $f$ moments



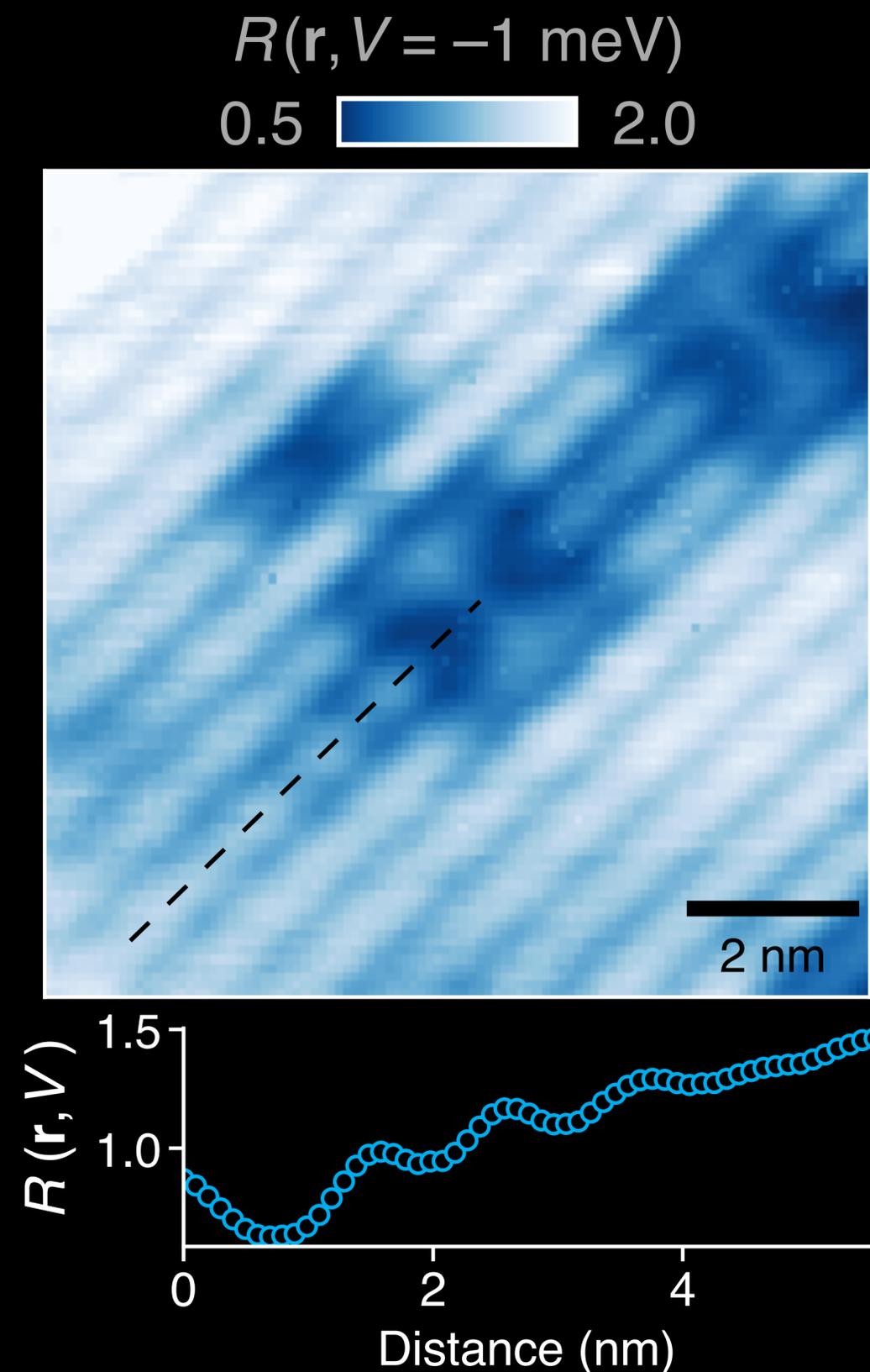
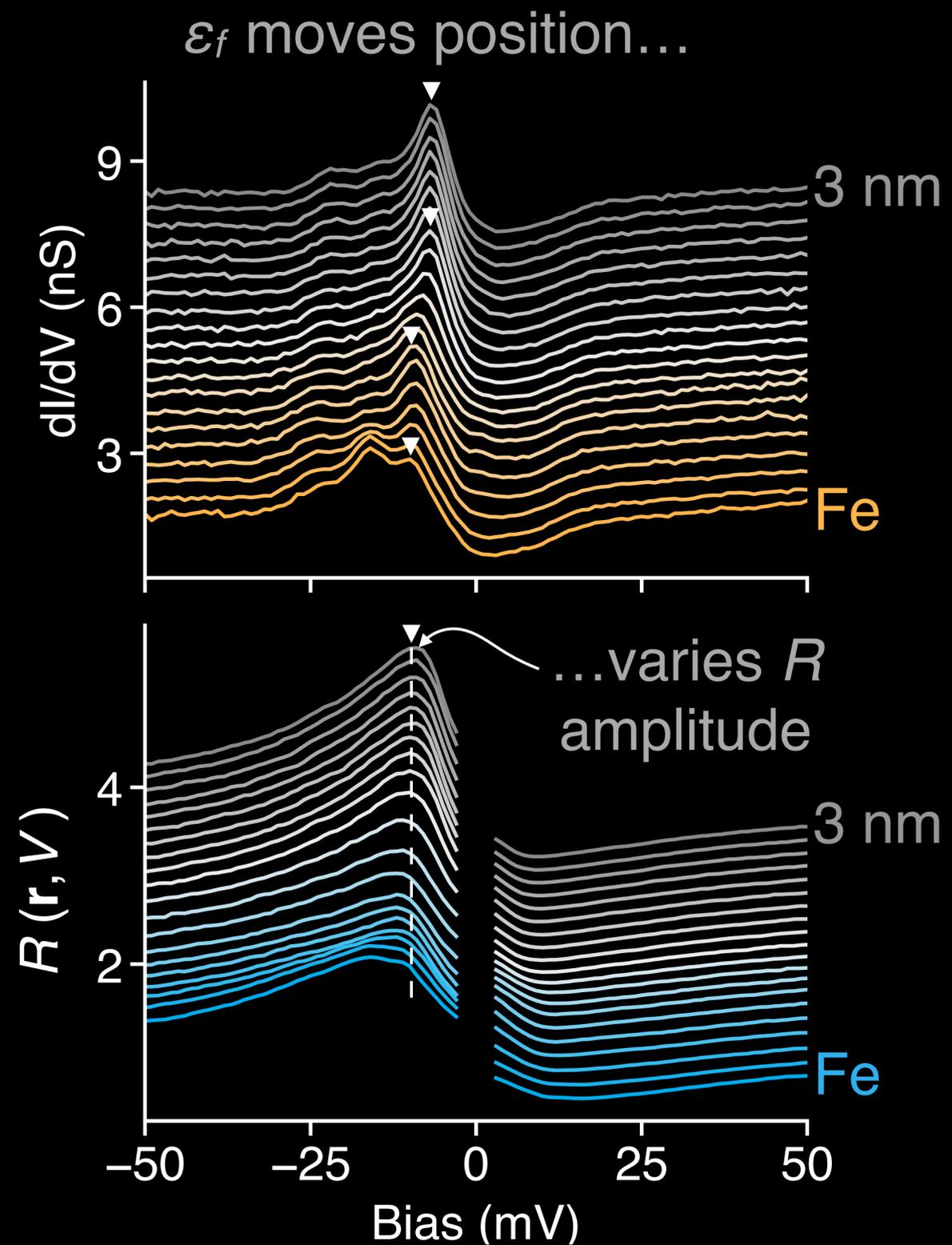
# Measured $\epsilon_f$ around dopants



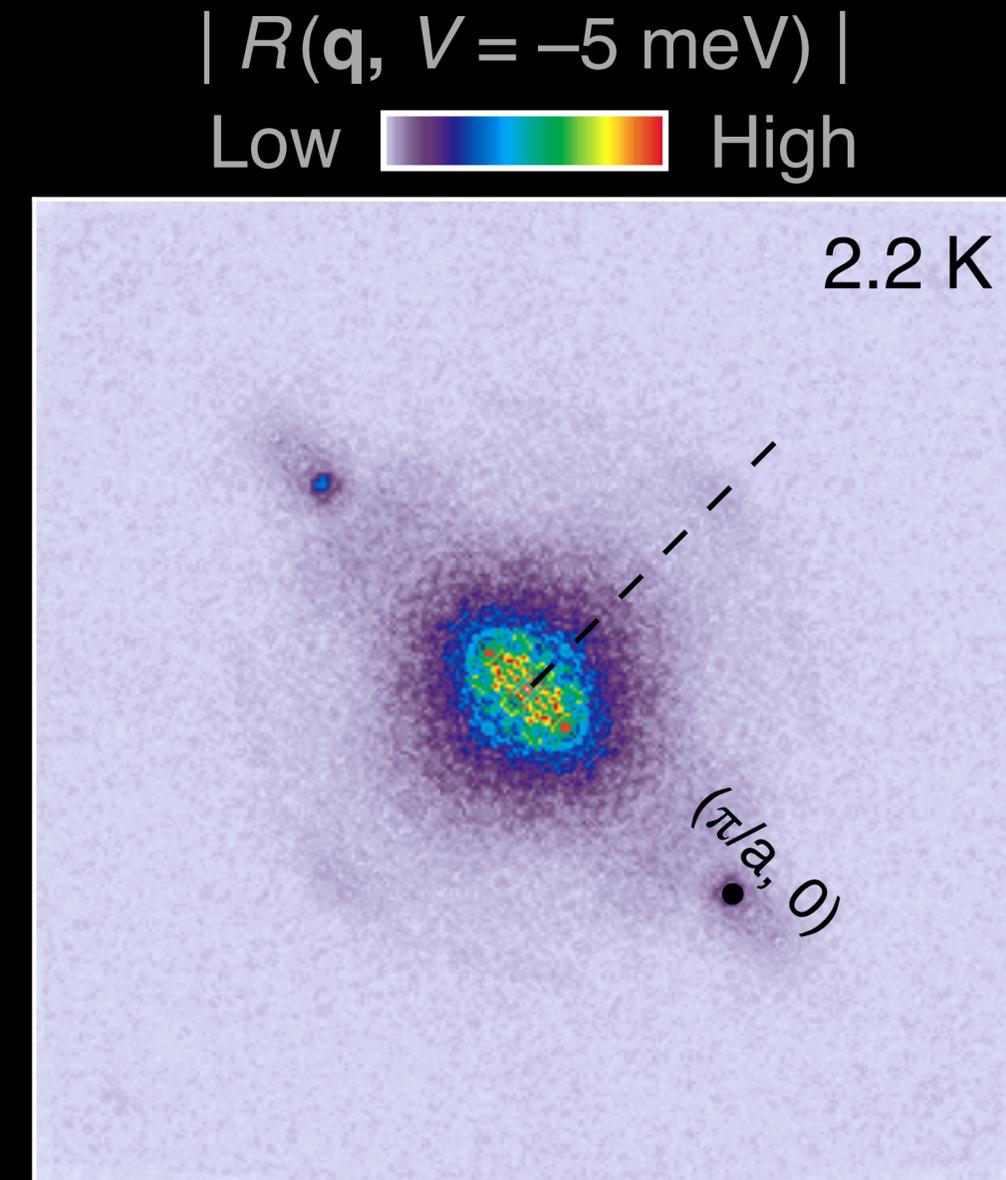
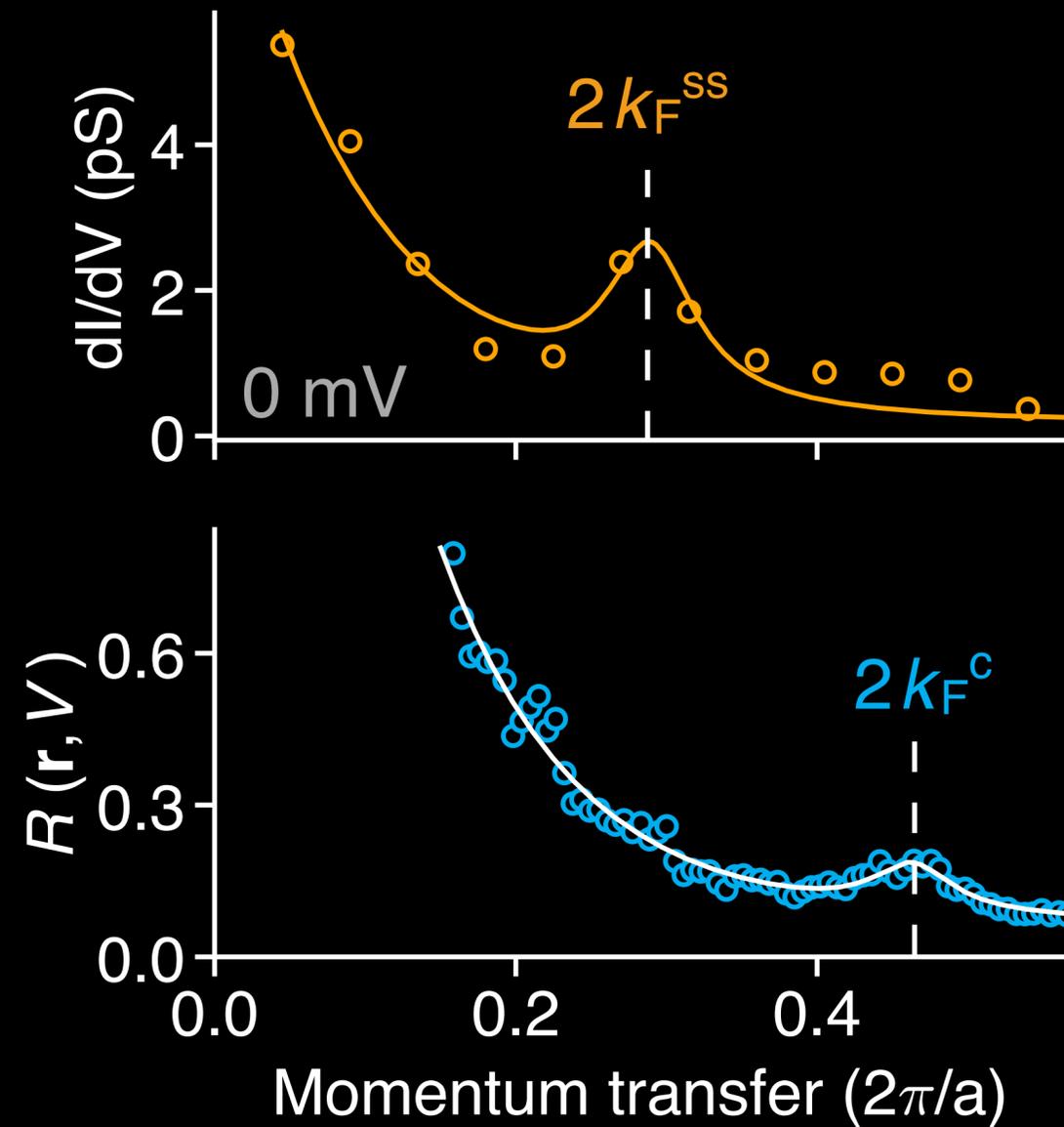
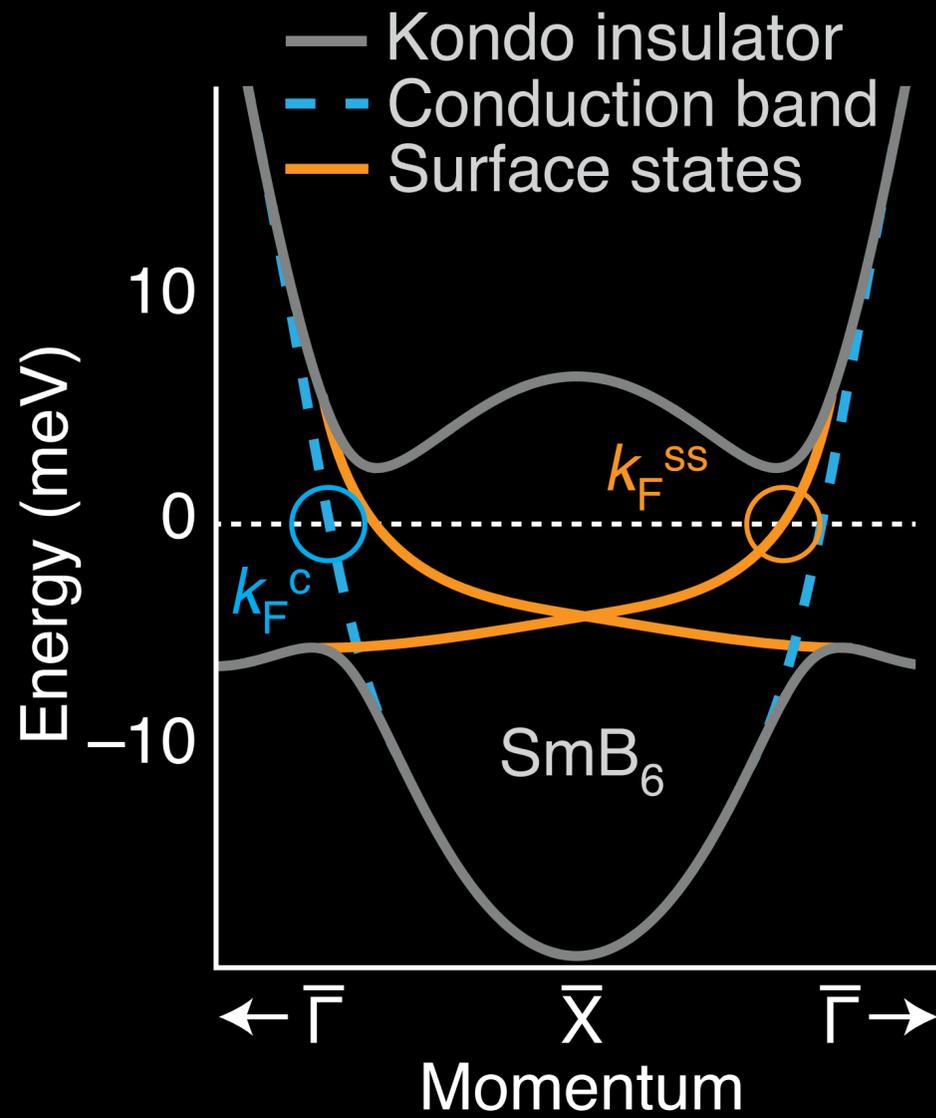
# The $\epsilon_f$ shifts are amplified in $R(\mathbf{r}, V)$



$$R(\mathbf{r}, V) = \frac{I(\mathbf{r}, +V)}{I(\mathbf{r}, -V)}$$

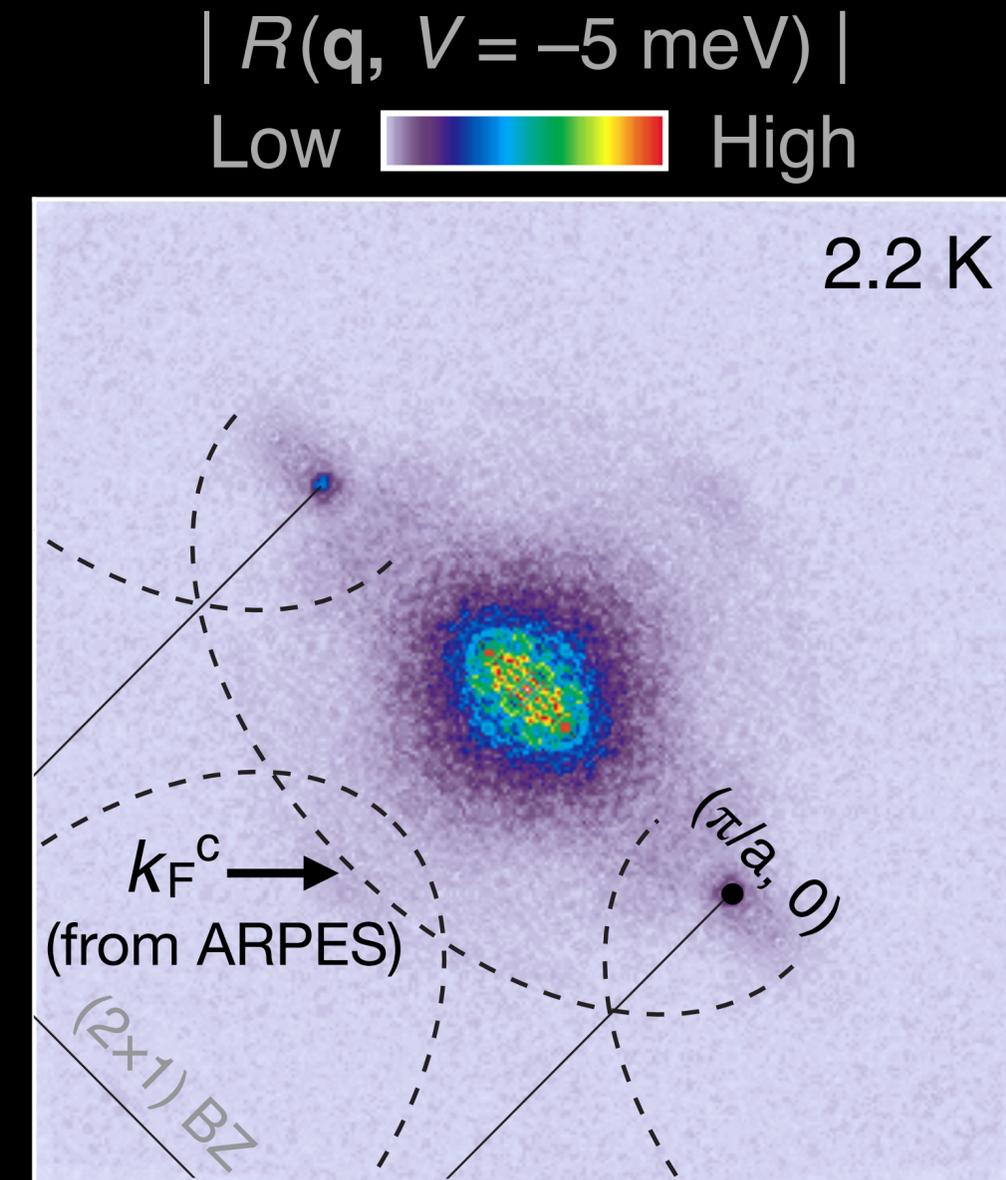
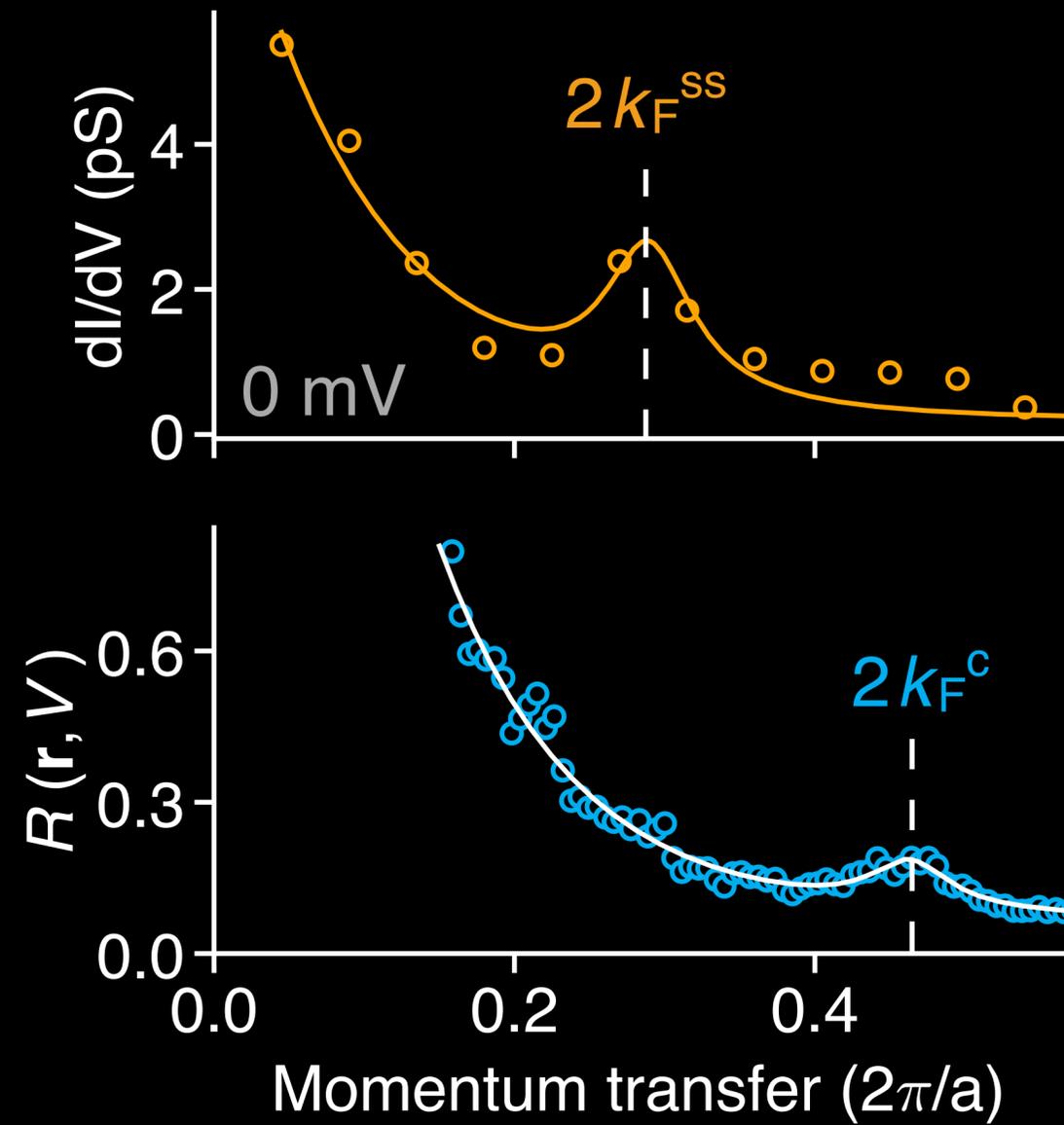
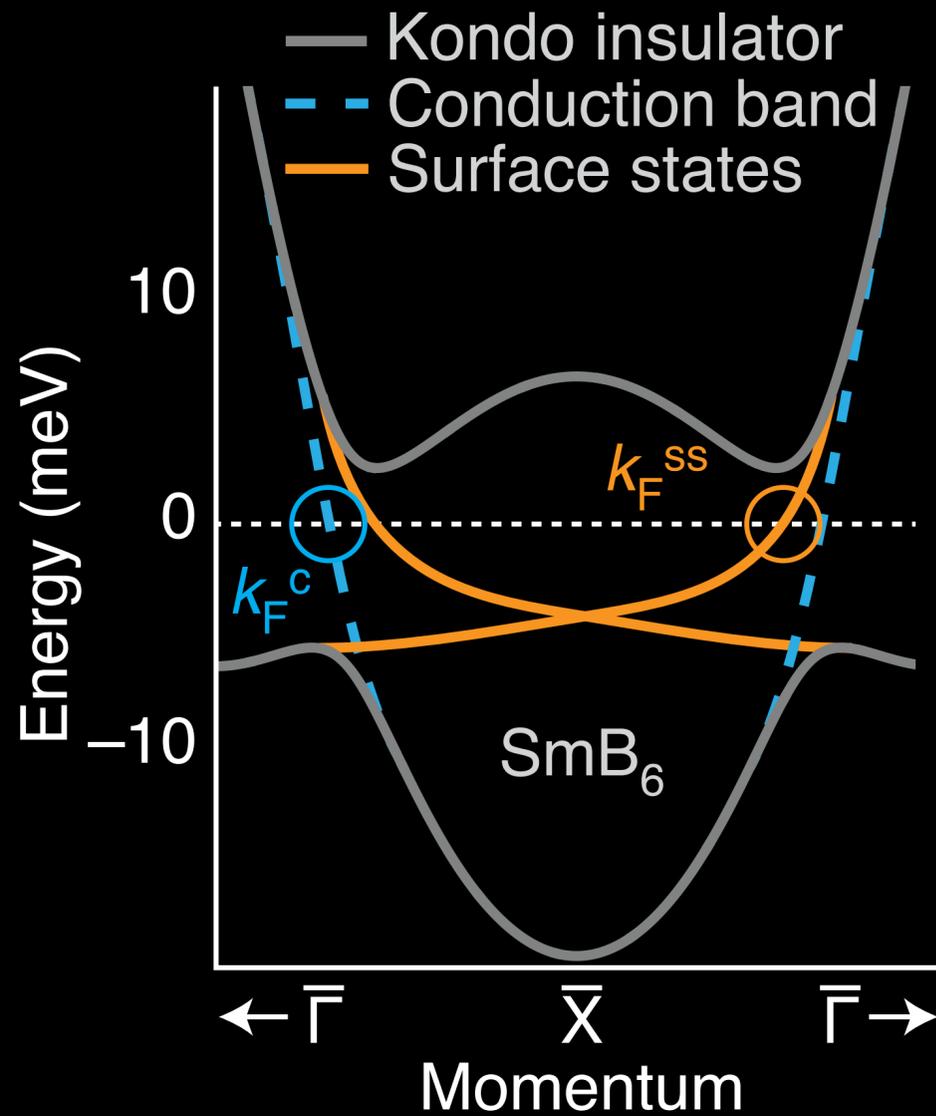


# Charge oscillations come from light electrons



$$R(\mathbf{r}, V) = \frac{I(\mathbf{r}, +V)}{I(\mathbf{r}, -V)}$$

# Charge oscillations come from light electrons



$$R(\mathbf{r}, V) = \frac{I(\mathbf{r}, +V)}{I(\mathbf{r}, -V)}$$

# Is the origin **intrinsic** or **extrinsic**?

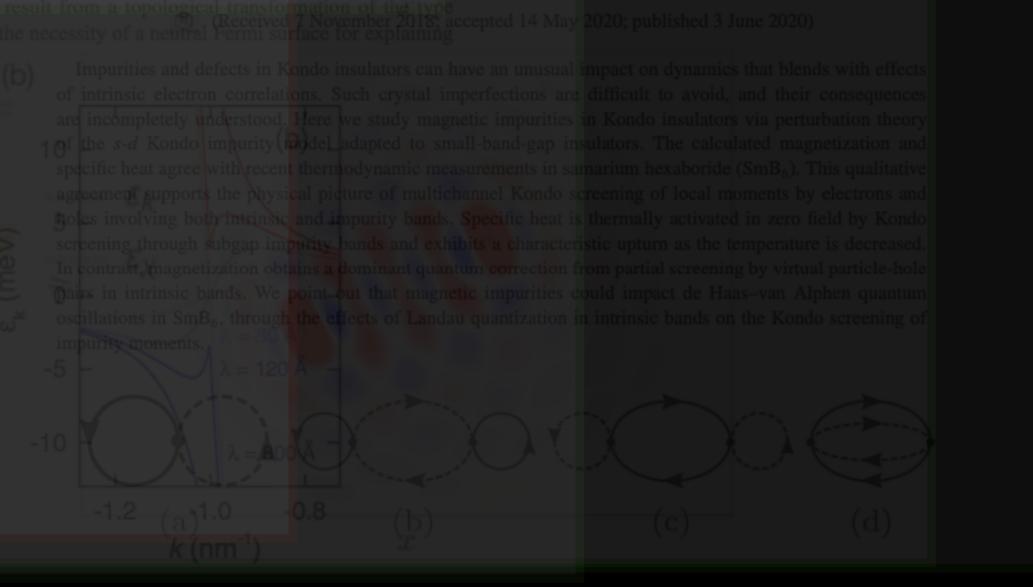
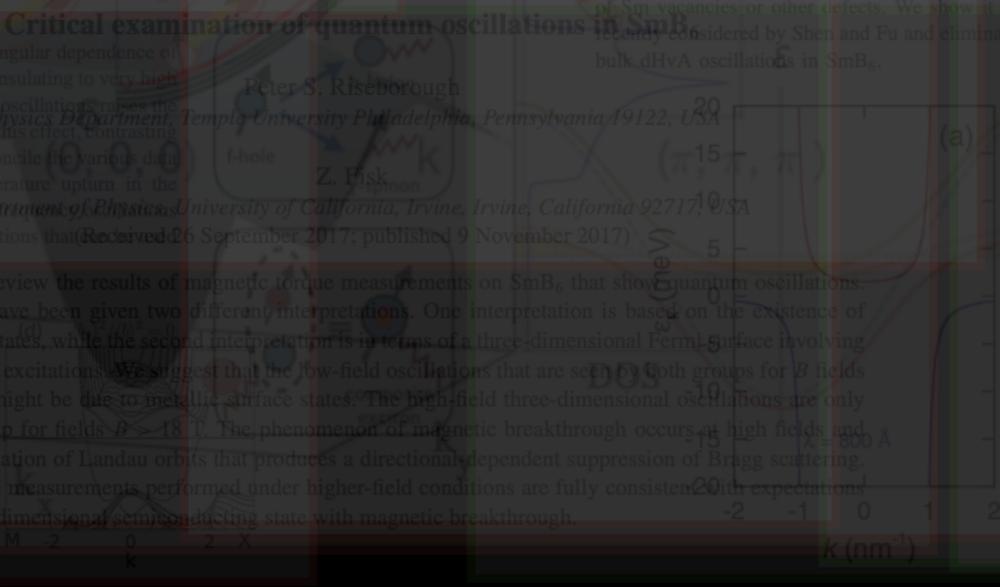
We will answer this question experimentally:

*Question:* Is there microscopic evidence of light electrons?

*Answer:* Yes, light electrons are released by Kondo holes

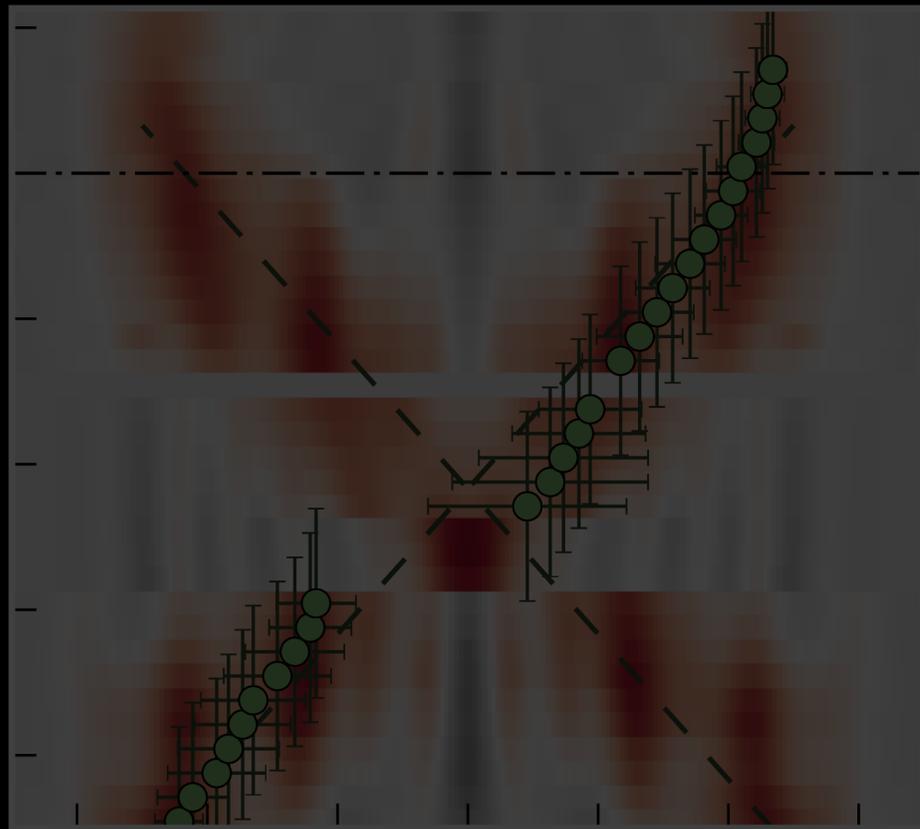
**Majorana Fermi Sea in Insulating  $\text{SmB}_6$ :**  
A proposal and a Theory of Quantum Oscillations in Kondo Insulators  
D. S. S. Kumar, P. S. Rieborgh, and Z. Fisk  
The Institute of Mathematics and Perimeter Institute for Theoretical Physics

In an exciting development,  $\text{SmB}_6$  has been shown to exhibit bulk quantum oscillations. We propose that  $\text{SmB}_6$  is a bulk scalar Majorana Fermi Liquid (MFL) with a finite Fermi surface for charge and spin excitations. In their study of Kondo insulators in 1993, Coleman, Mielke, and Tsvetkiv (CMT) envisaged such a remarkable possibility, being a mean field ansatz. We generalize CMT theory to non-zero magnetic fields and show a counter intuitive result, that the scalar Majorana fermi liquid, while remaining electrically insulating, responds to external magnetic field and Landau diamagnetism and quantum oscillations. Physics of an emergent compactified Kondo lattice physics that is behind formation of the novel scalar Majorana fermi liquid is discussed. It is also suggested that a known resistivity saturation in  $\text{SmB}_6$  as well as strong deviation of quantum oscillation amplitude from Lifshitz-Kosevich formula in  $\text{SmB}_6$  at low temperatures are due to a coherent fluctuation of charge of a neutral scalar Majorana fermion. Possible presence of 2-dimensional Majorana fermion excitations in surfaces of  $\text{SmB}_6$ , and other Topological Kondo Insulators (TKI) is pointed out.



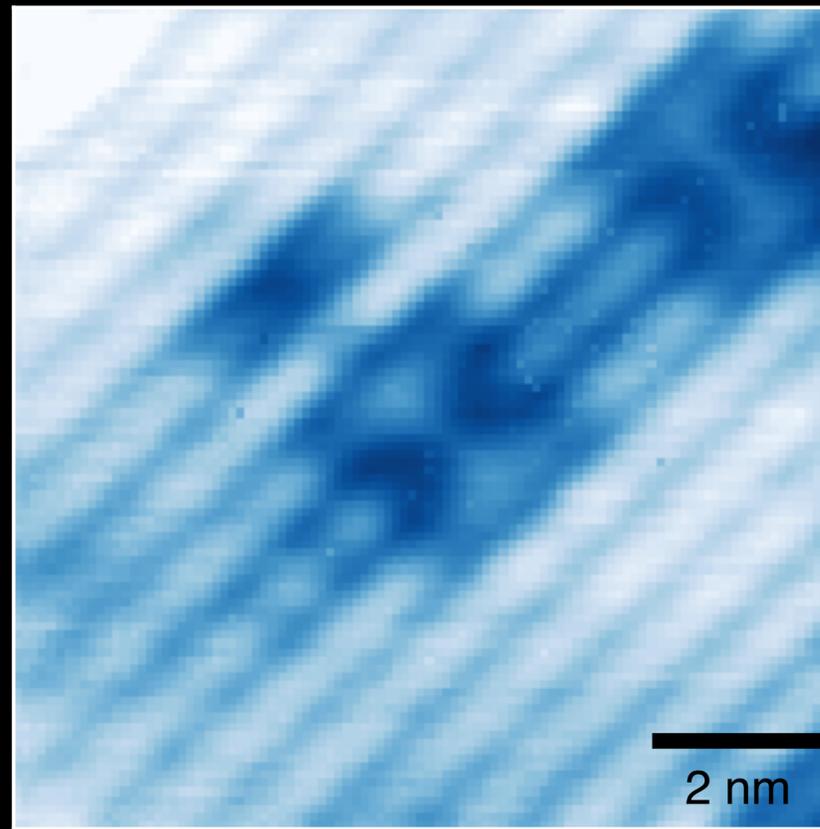
# Outline

(1) Dirac fermions **410 times** heavier than a bare electron!



Pirie et al., *Nat. Phys.* **16**, 52 (2020)  
Matt et al., *Phys. Rev. B* **101**, 085142 (2020)

(2) Metallicity in an **insulator!**



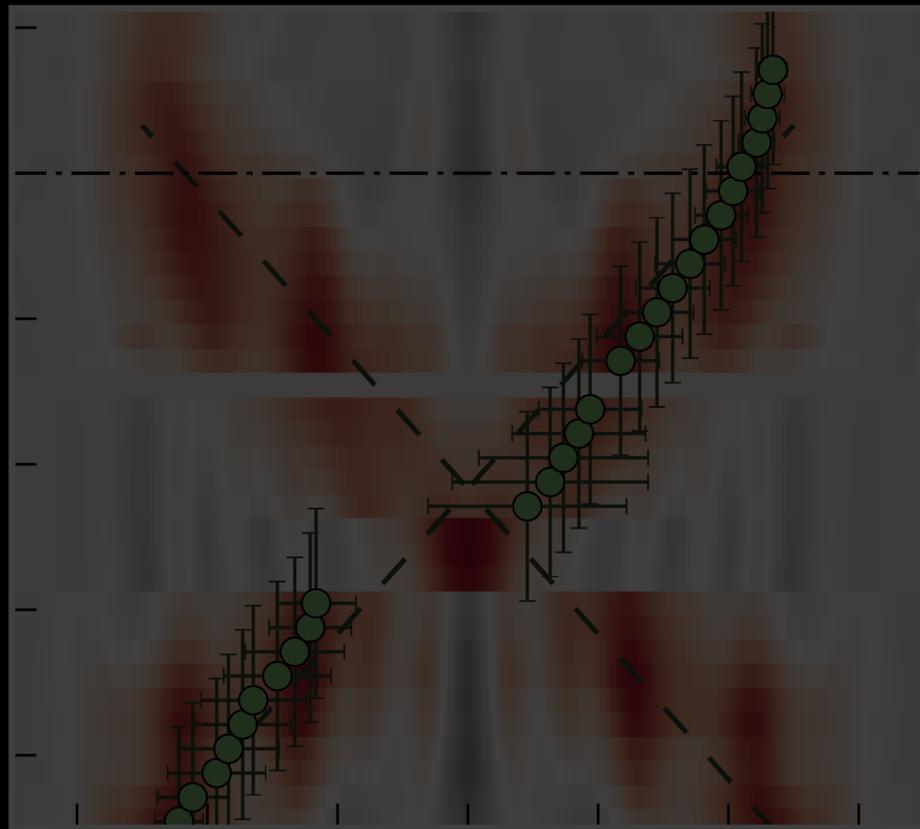
Pirie et al., *Science* **379**, 1214 (2023)

(3) Breakdown of **topological protection!**



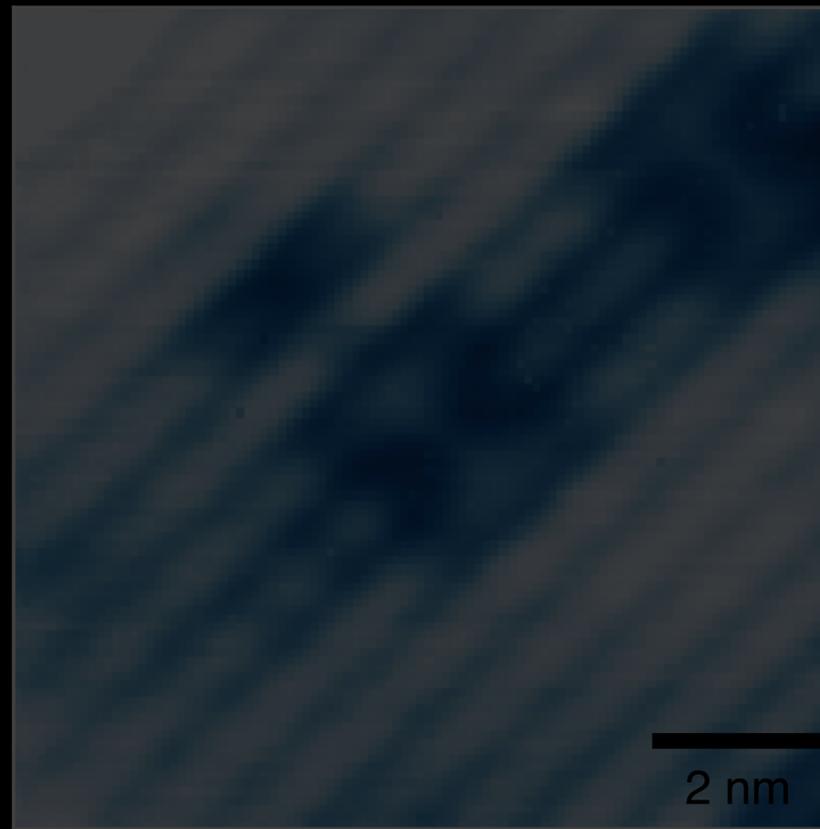
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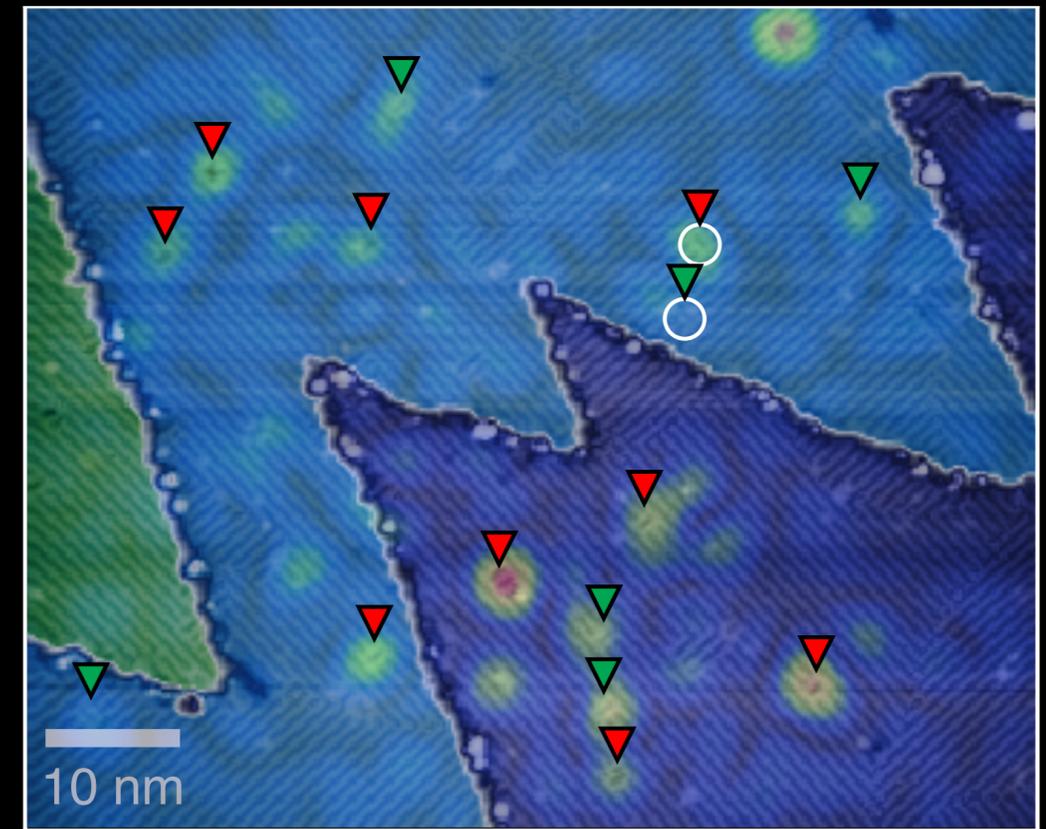
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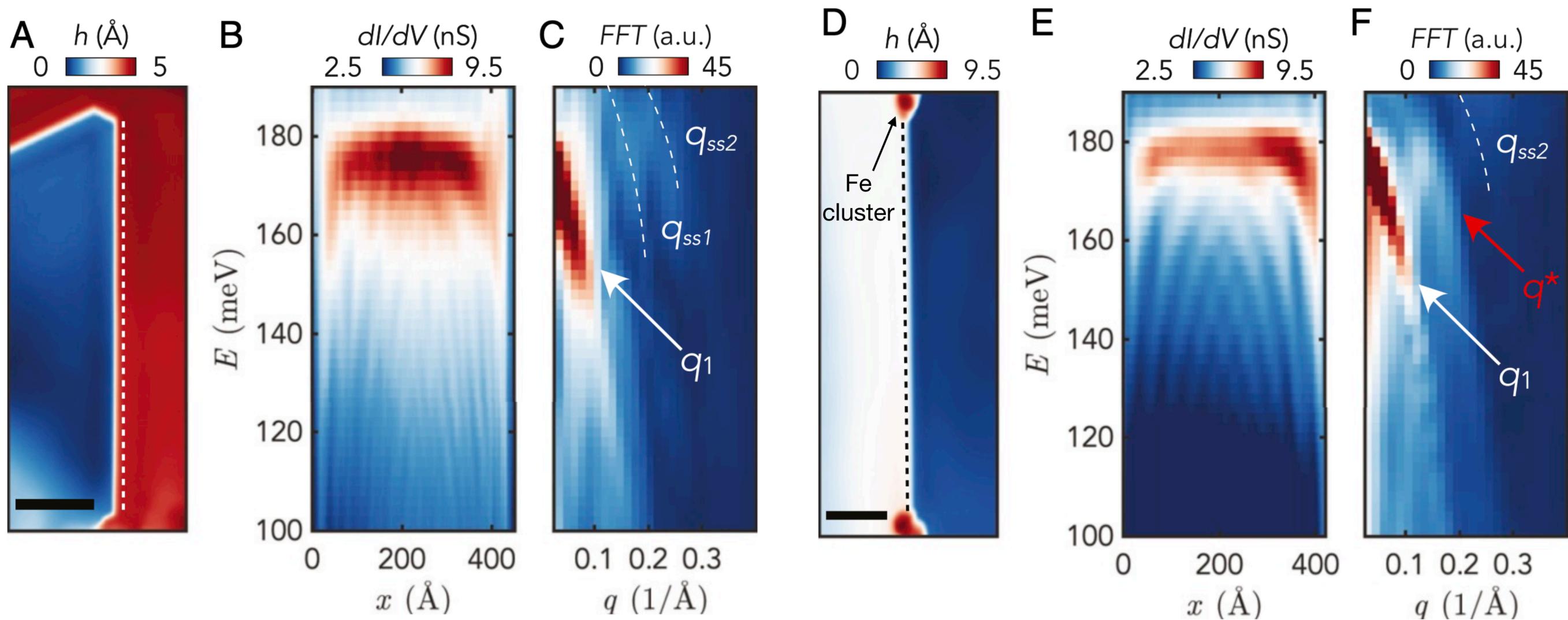
*Answer:* **Yes**, light electrons are released by Kondo holes

*Question:* Do the metallic puddles have a finite magnetic susceptibility?

# Topological states are a magnetic probe

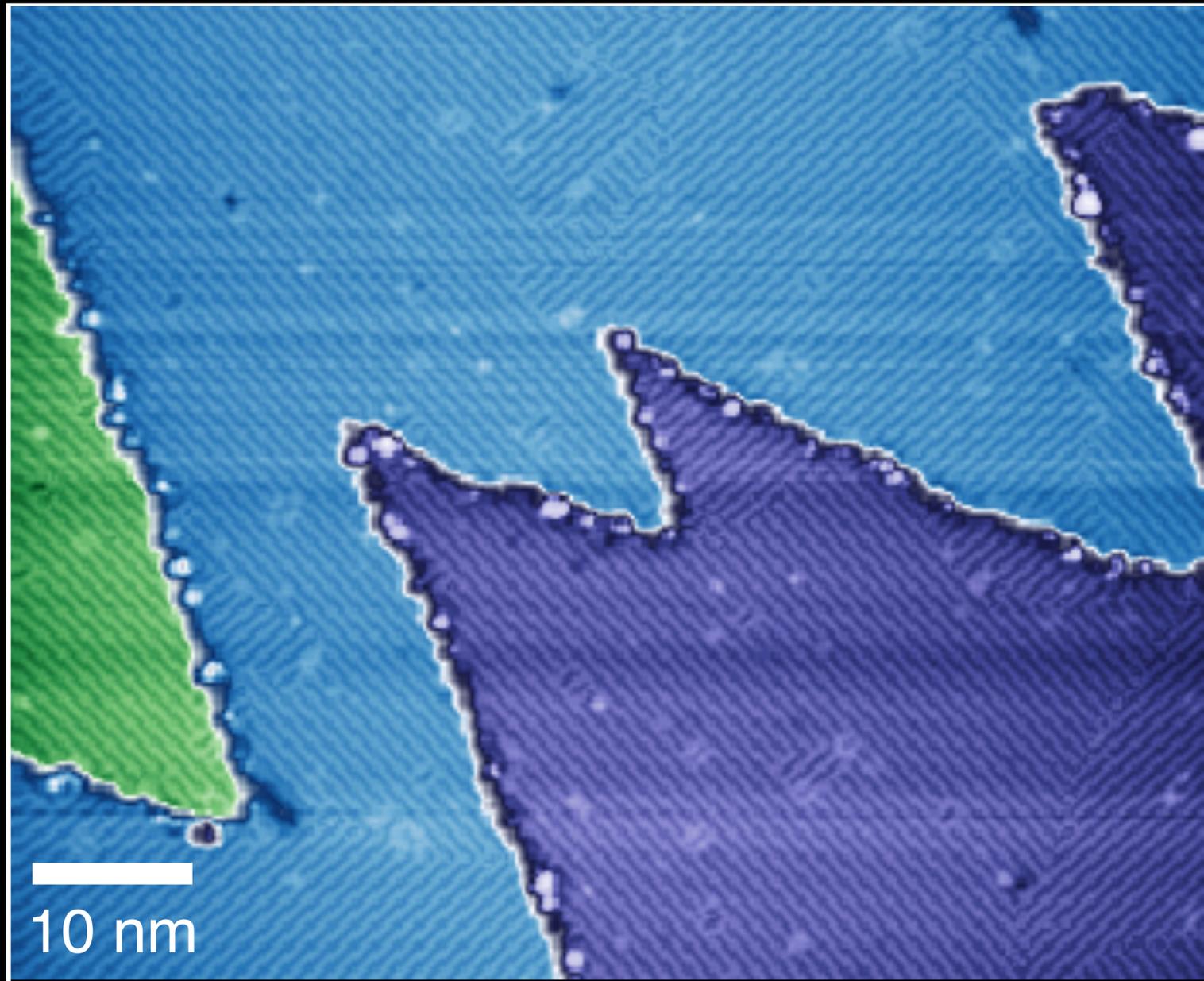
## Observation of backscattering induced by magnetism in a topological edge state

Berthold Jäck<sup>a,b</sup>, Yonglong Xie<sup>a,b</sup>, B. Andrei Bernevig<sup>a,b</sup>, and Ali Yazdani<sup>a,b,1</sup>

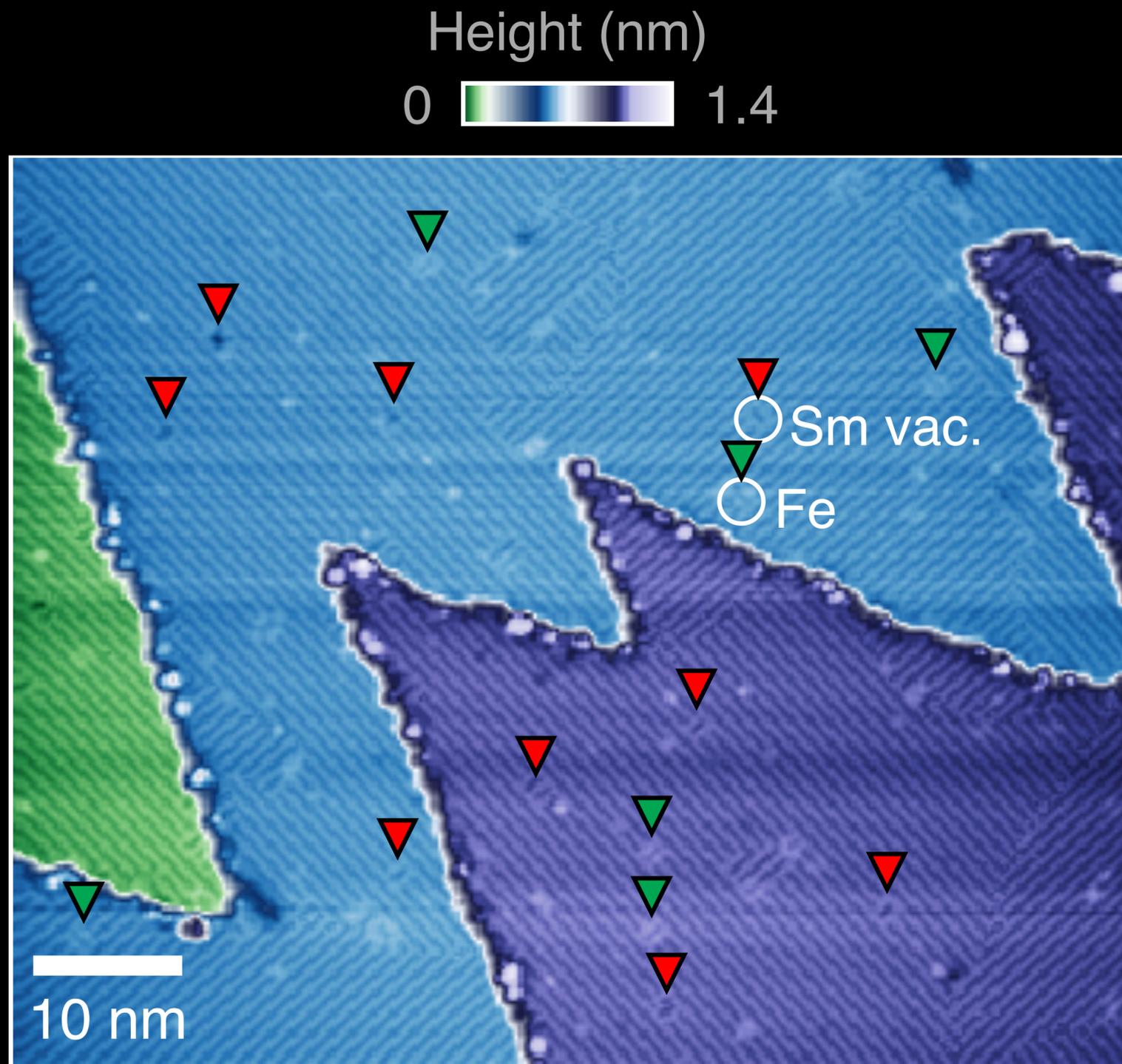


# Which defects backscatter Dirac fermions?

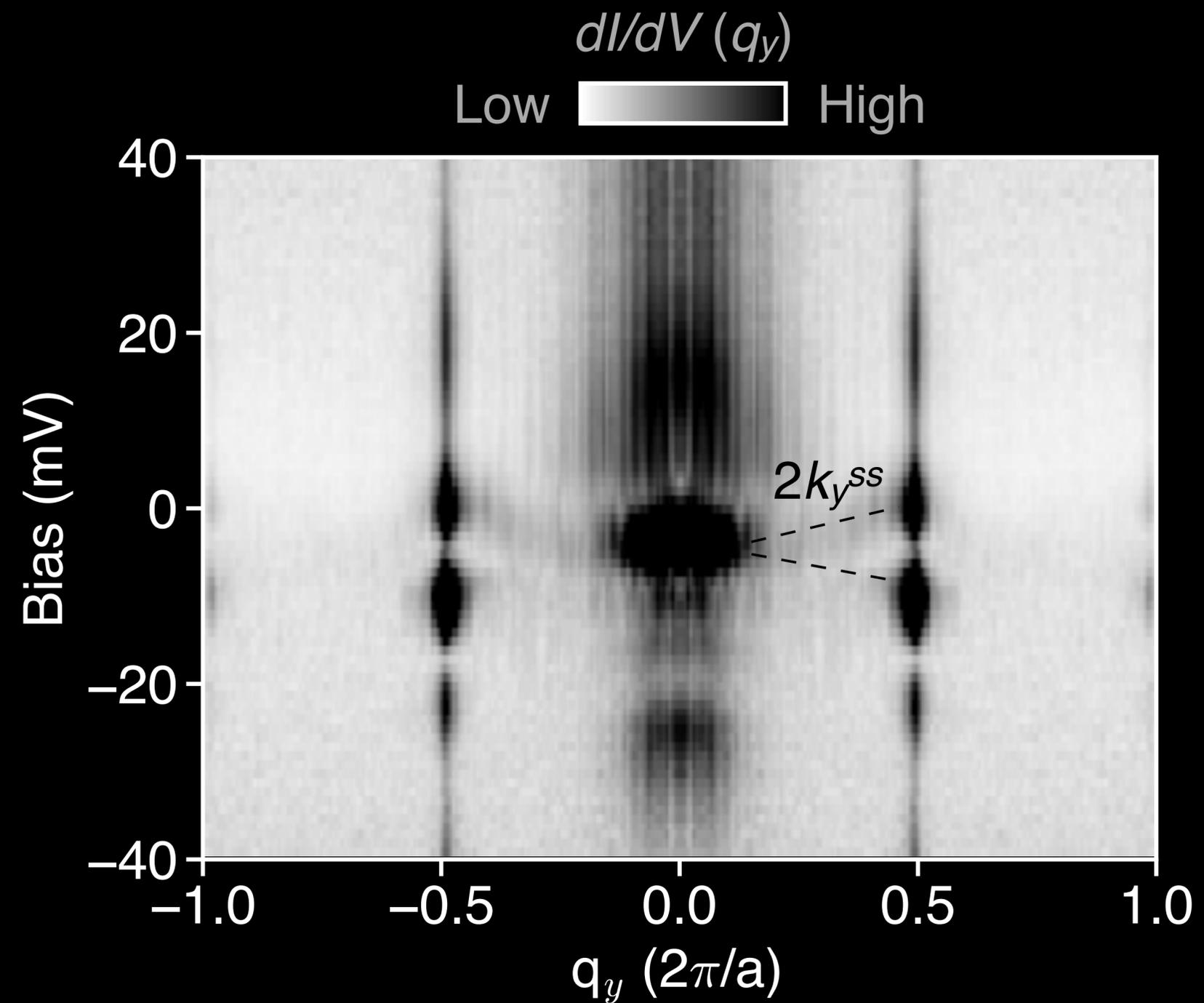
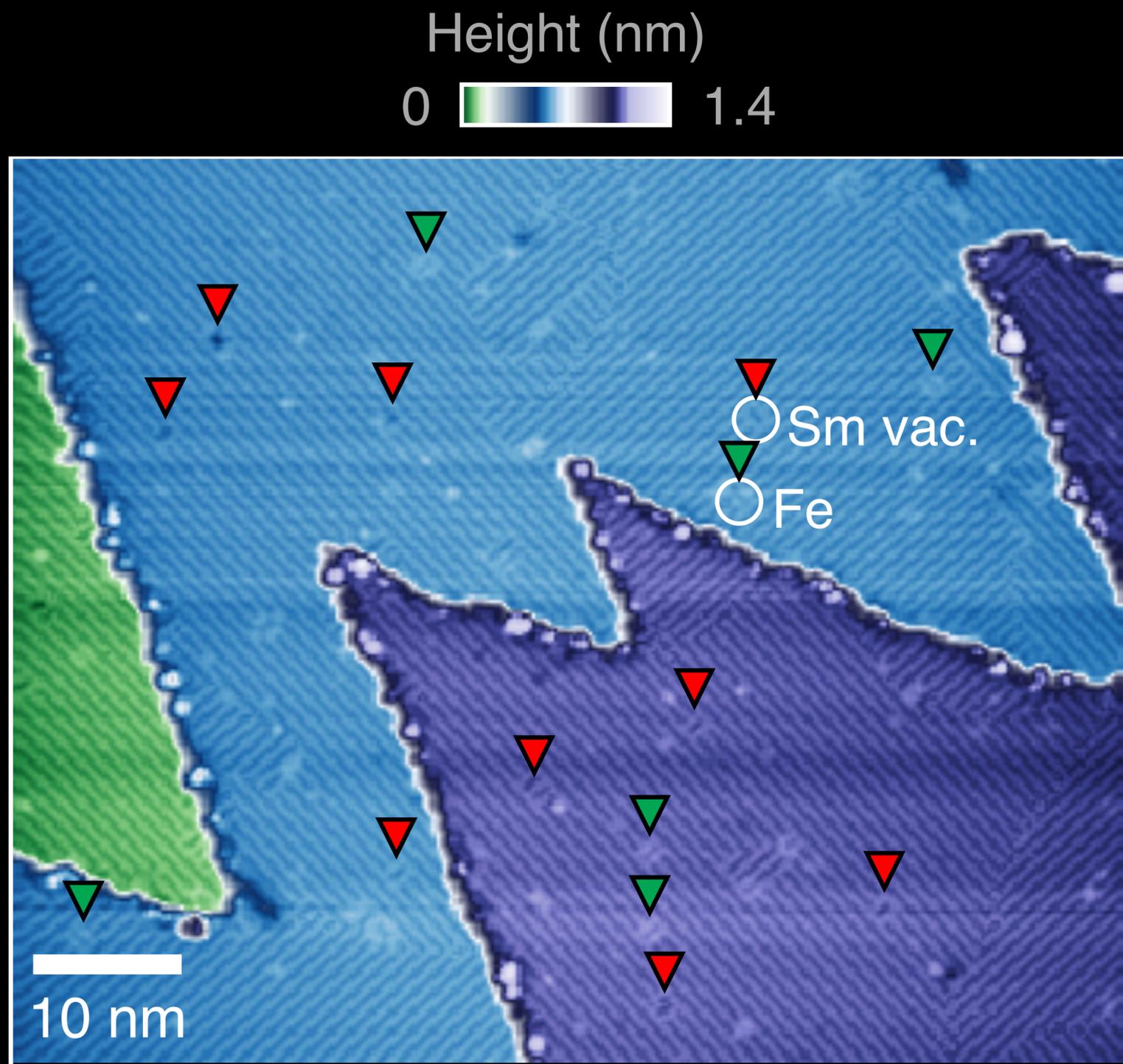
Height (nm)  
0 1.4



# Which defects backscatter Dirac fermions?

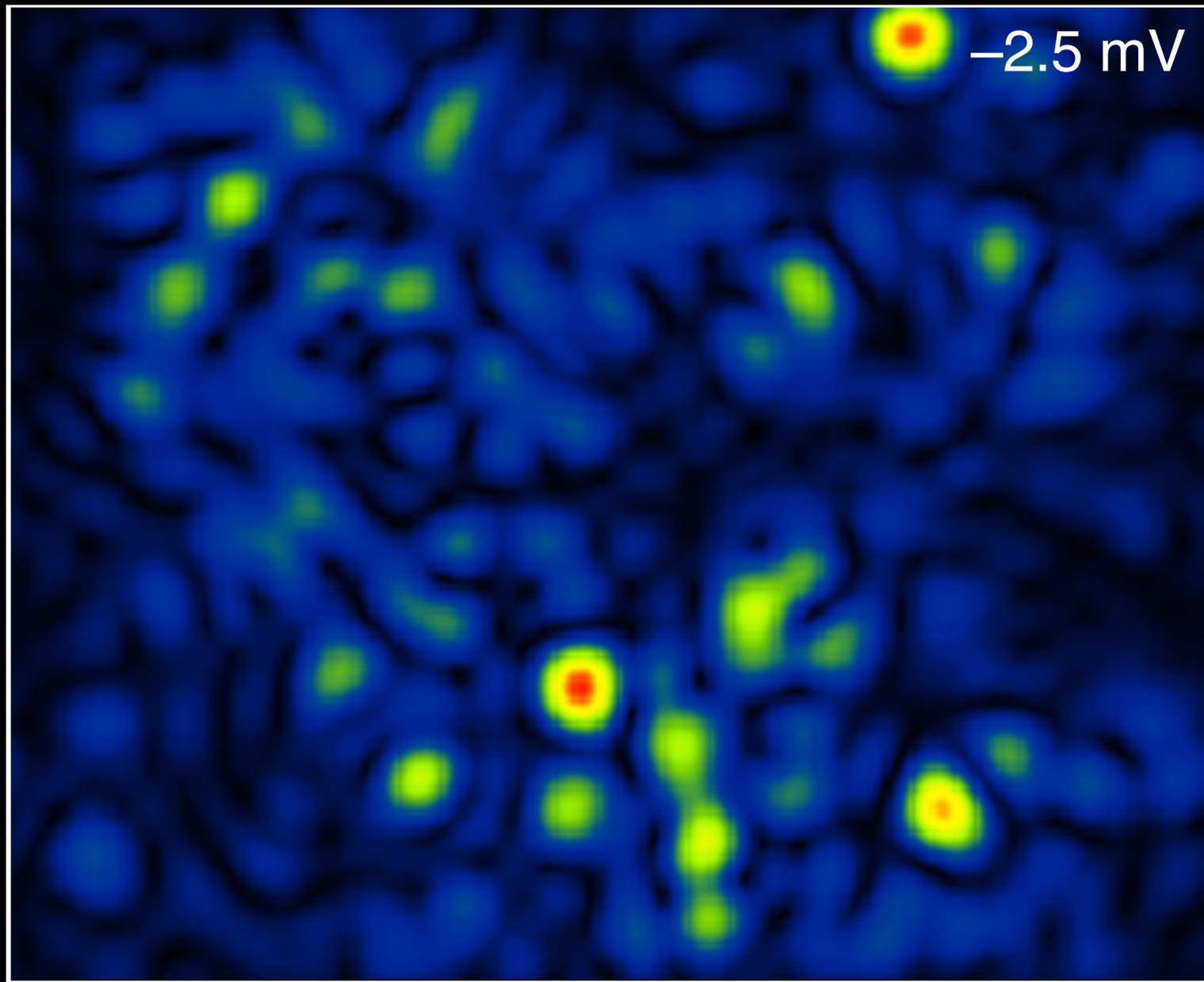


# Heavy Dirac fermions are visible in raw $dI/dV$

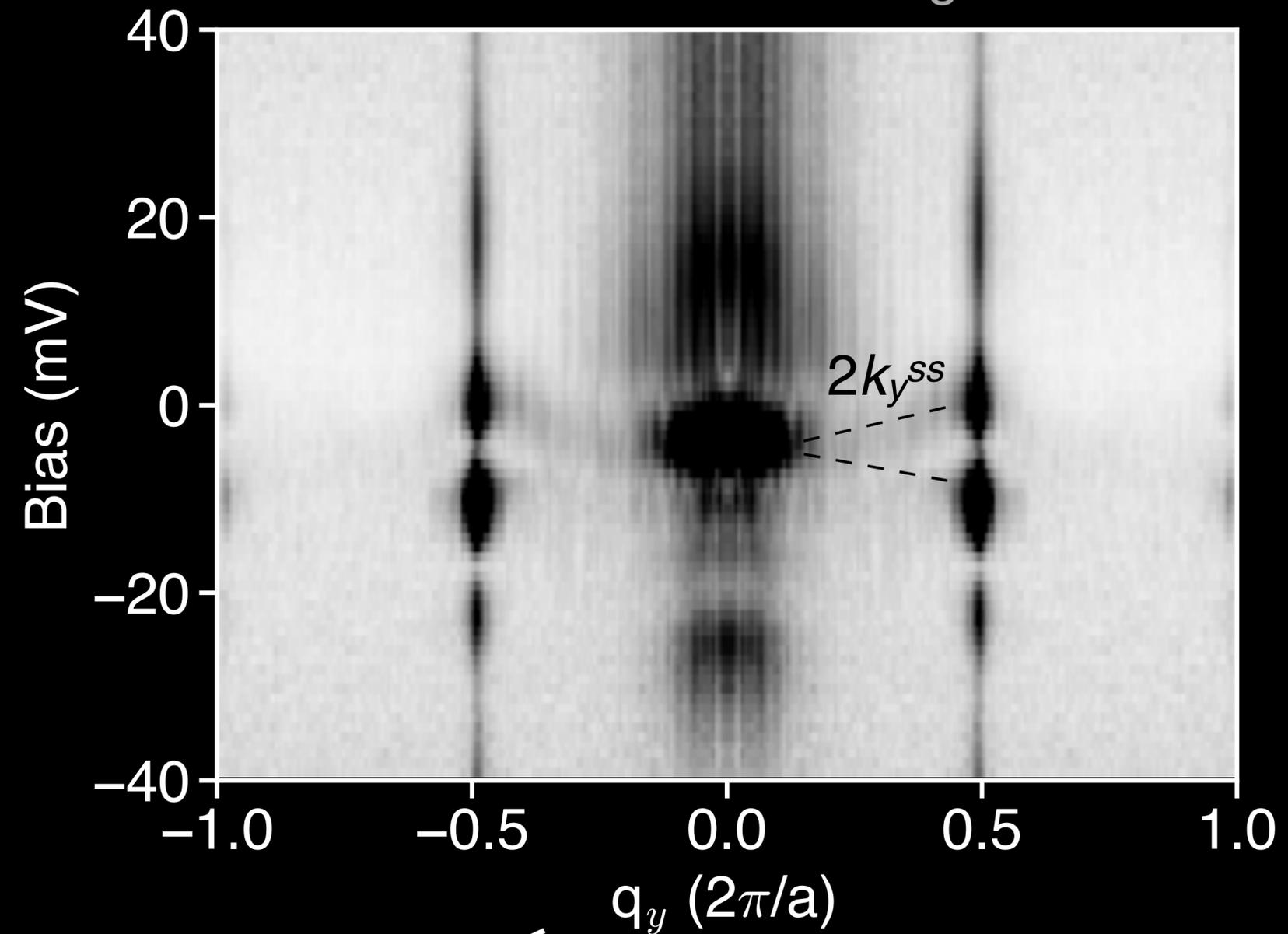


# Imaging backscattering centres

Backscattering  
Low  High

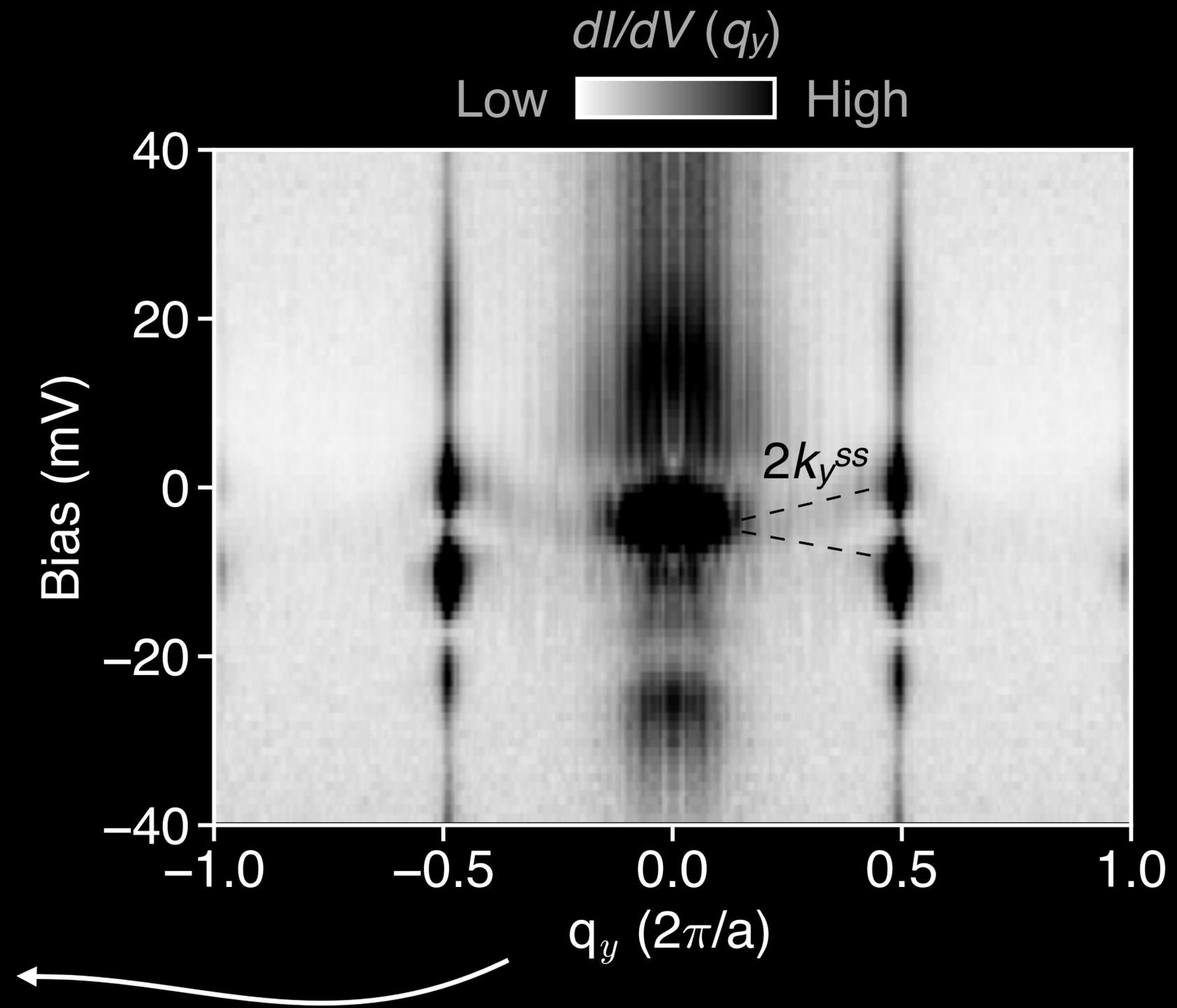
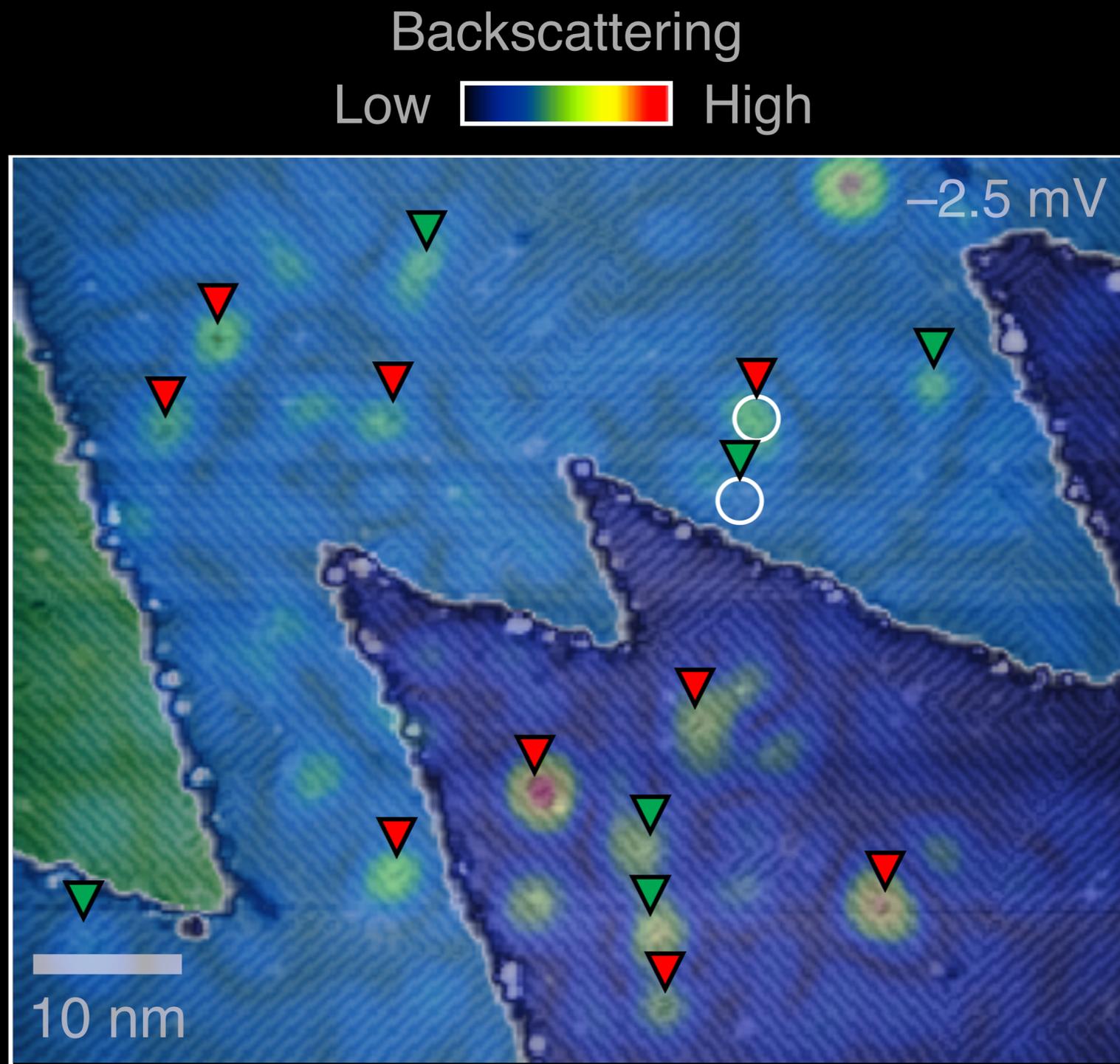


$dI/dV(q_y)$   
Low  High



Inverse Fourier transform at  $q = 2k_y^{ss}$

# Kondo holes backscatter heavy Dirac fermions



Inverse Fourier transform at  $q = 2k_y^{ss}$

# Is the origin **intrinsic** or **extrinsic**?

We will answer this question **experimentally**:

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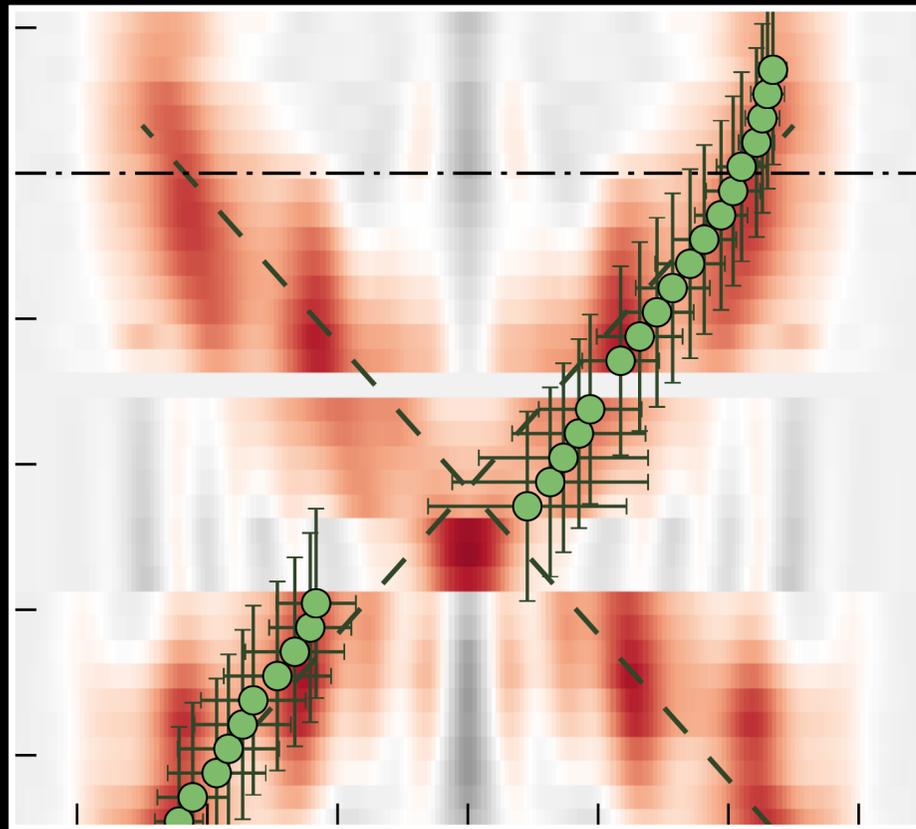
*Answer:* **Yes**, light electrons are released by Kondo holes

*Question:* Do the metallic puddles have a finite magnetic susceptibility?

*Answer:* **Yes**, Kondo holes magnetically scatter Dirac fermions

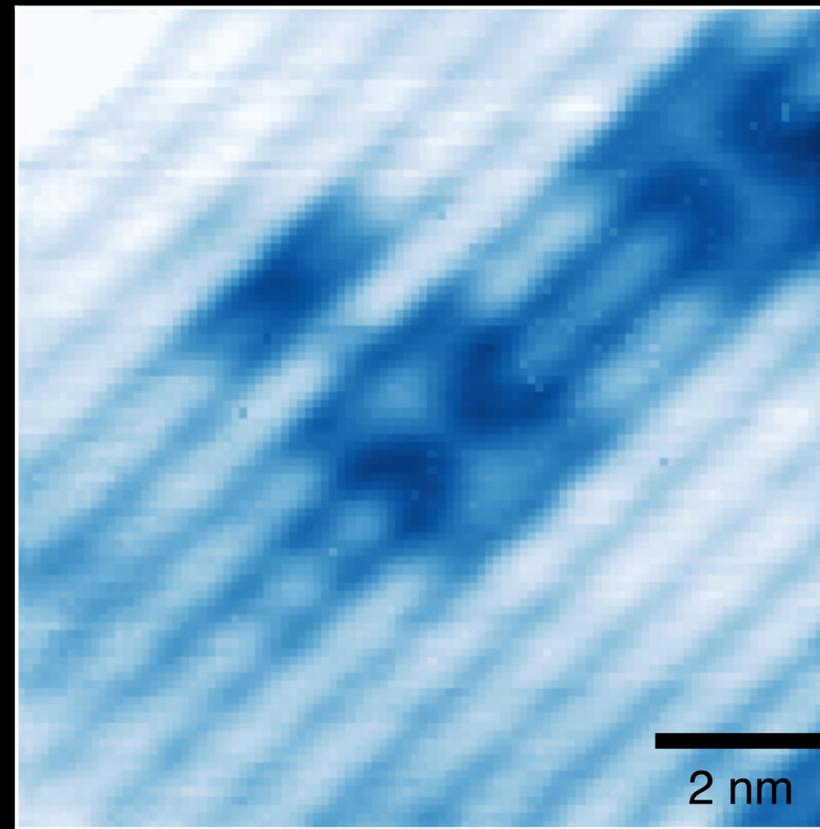
# Conclusion

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