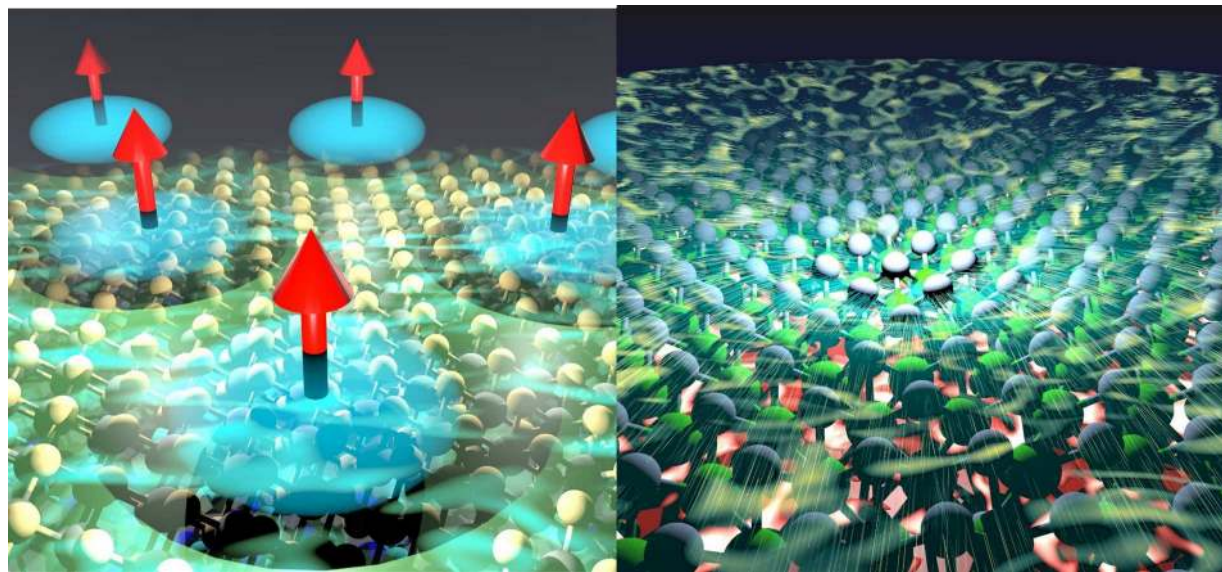


Deconstructing 4Hb-TaS₂: heavy-fermion Kondo insulators and nodal superconductivity



Jose Lado

Department of Applied Physics, Aalto University, Finland



Nature 599, 582–586 (2021)

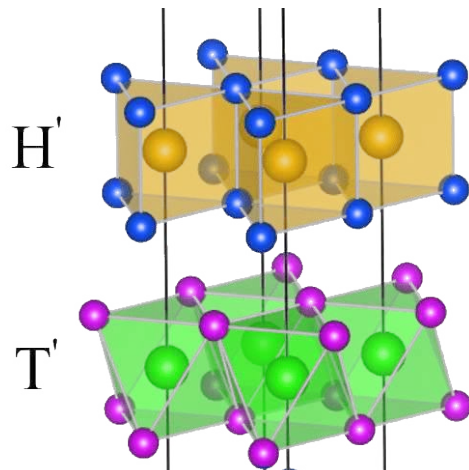
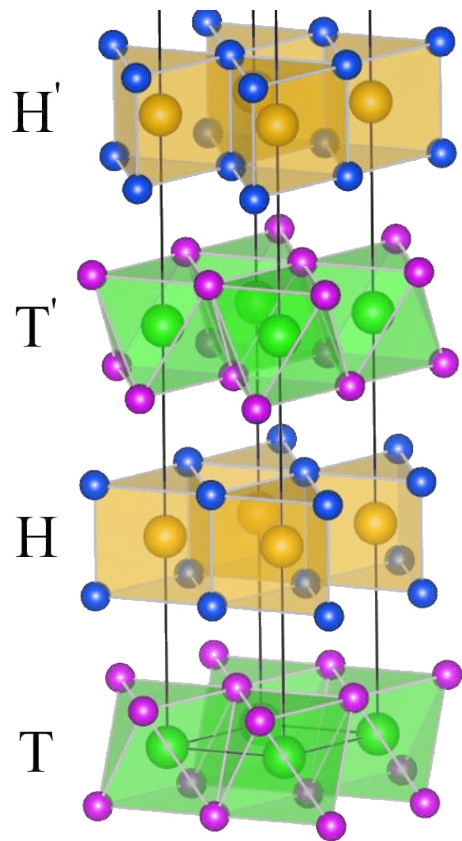
arXiv:2112.07316 (2021)

Hacking 4Hb

September 5th 2022, Weizmann Institute, Israel

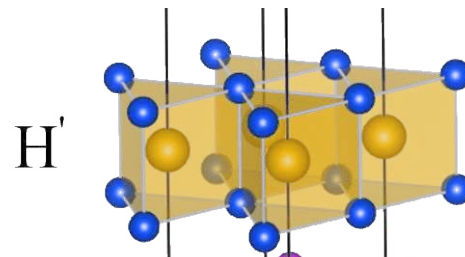
Deconstructing 4H-TaS₂

We will explore the physics of a deconstructed version



**Heavy fermion
Kondo insulators**

1H-1T bilayer



Nodal superconductivity

Ultraclean 1H monolayer

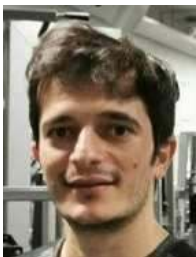
Behind the scenes

Experiment

S. Ganguli



M. Amini



M. Aapro



L. Yan



V. Vaňo



S. Kezilebieke



Md. Huda



P. Liljeroth



Theory

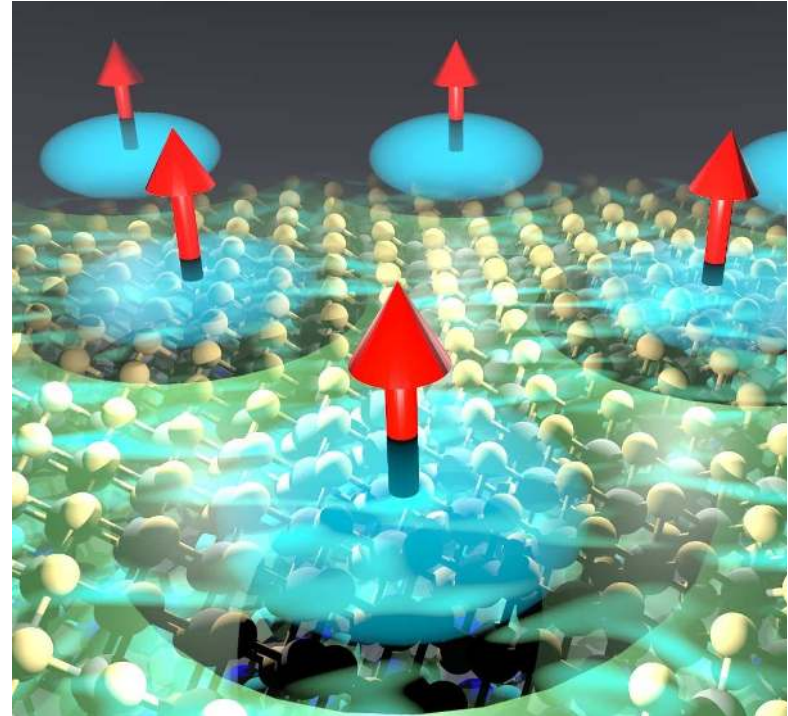
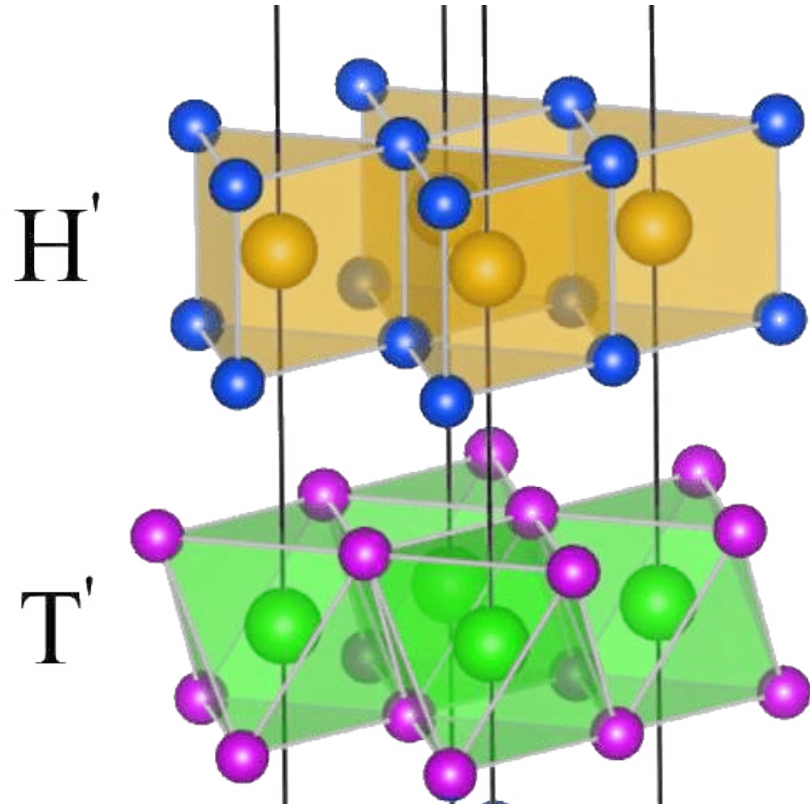
G. Chen



M. Khosravian



Heavy-fermion Kondo insulators in 1H-1T bilayer



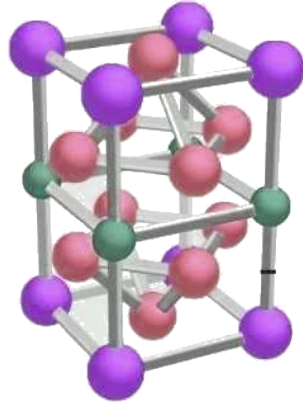
Nature 599, 582–586 (2021)

Searching for new heavy fermion compounds

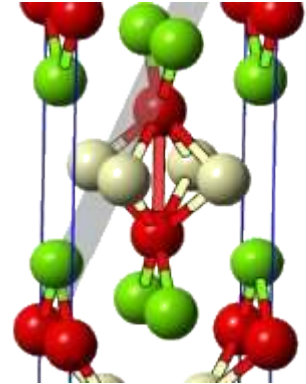
We have a rich family of bulk heavy-fermion compounds



Science 375, 6576, 76-81 (2021)



Nature 579, 523–527 (2020)



- Strongly correlated matter, dominated by Kondo lattice physics
- Unique system to realize quantum criticality and unconventional superconductivity
- Found in rare-earth compounds, such as UTe_2 or UPt_3

Building an artificial heavy fermion state



Triangular lattice of
local magnetic moments



1T-TaS₂

Two-dimensional electron gas

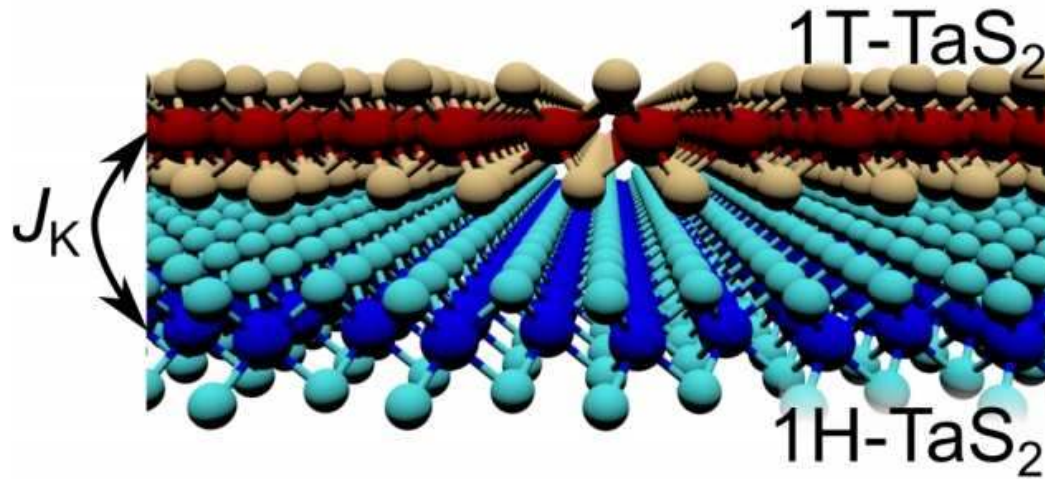


1H-TaS₂

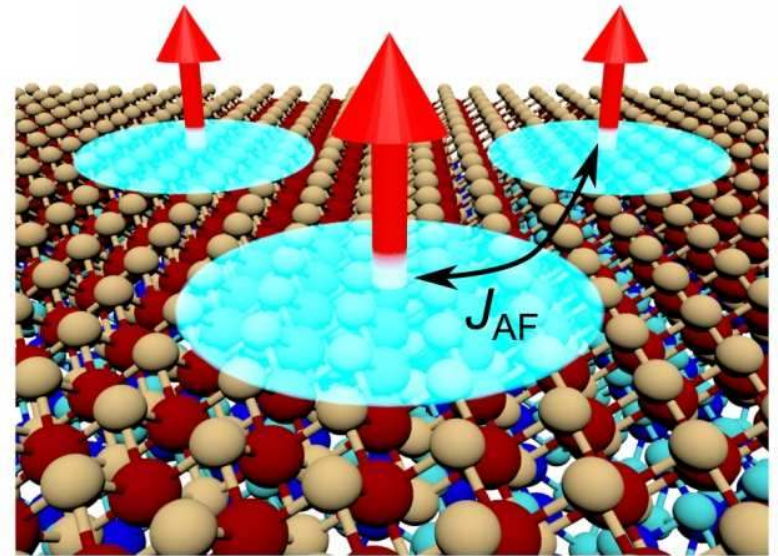


Heavy-fermions in dichalcogenide bilayers

Interlayer coupling create
the Kondo coupling



Intralayer exchange between
local moments



The bilayer realizes all the ingredients for heavy-fermion physics

The Kondo problem

Conduction electrons

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right)$$

Kondo coupling

$$H_K = \sum_{\alpha\beta} \left(c_{0\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{0\alpha} \right) \cdot \vec{S}$$

We now take a quantum spin $S=1/2$

$$|GS\rangle \sim \frac{1}{\sqrt{2}} [|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle]$$

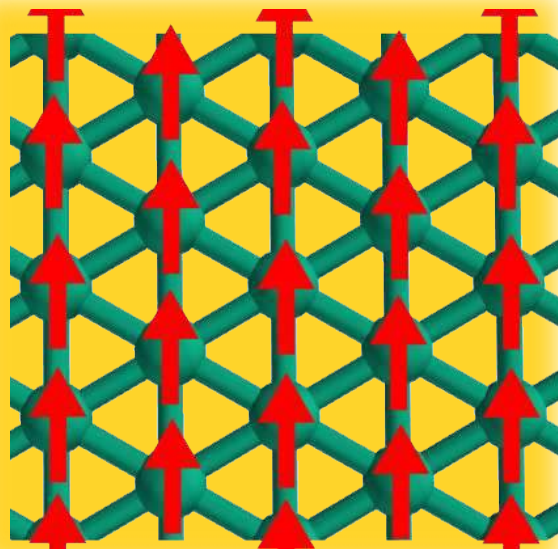


The Kondo lattice problem

The Kondo lattice problem

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right) + J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{j\alpha} \right) \cdot \vec{S}_j$$

Conduction electrons

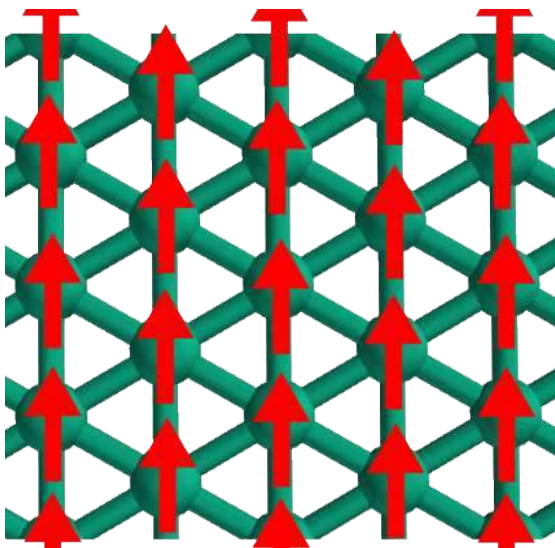


Kondo coupling

Kondo sites

Building an artificial heavy fermion state

Lattice of Kondo impurities



Dispersive electron gas

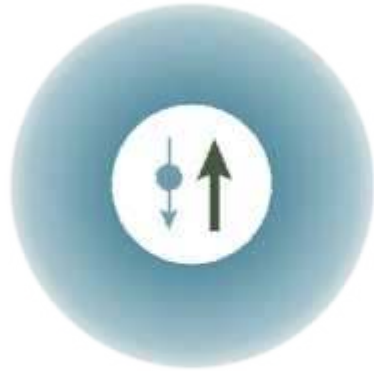
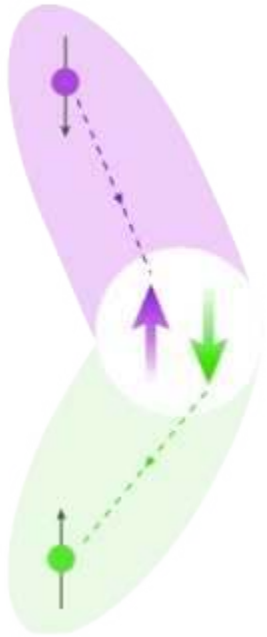


$$\longleftrightarrow J_K$$

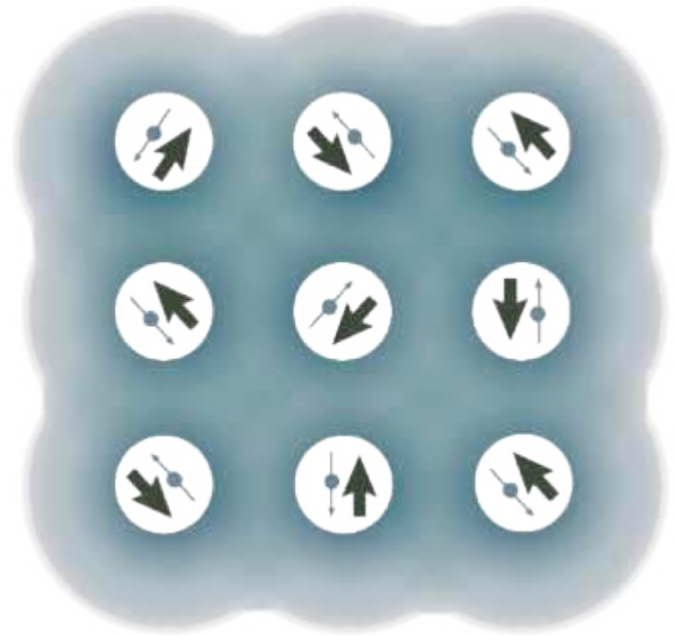
Both ingredients coupled through Kondo coupling

Building an artificial heavy fermion state

**Conduction electrons form
Kondo singlets with the impurities**



Kondo-lattice model



Associated with Kondo lattice physics:

- Colossal mass enhancement of electrons
- Quantum criticality
- Unconventional (topological) superconductivity

Solving the Kondo lattice problem

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right) + J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{j\alpha} \right) \cdot \vec{S}_j$$

Replace the spin sites by auxiliary fermions $S_{\alpha\beta}(j) = f_{j\alpha}^\dagger f_{j\beta} - \frac{n_f(j)}{N} \delta_{\alpha\beta}$

This makes the effective Hamiltonian an “interacting” fermionic Hamiltonian

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger f_{j\beta} \right) \left(f_{j\alpha}^\dagger c_{j\alpha} \right)$$

Solving the Kondo lattice problem

Now we decouple the fermions with a mean-field approximation

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - J \sum_{j,\alpha\beta} \left(c_{j\beta}^{\dagger} f_{j\beta} \right) \left(f_{j\alpha}^{\dagger} c_{j\alpha} \right)$$

Obtaining a quadratic Hamiltonian

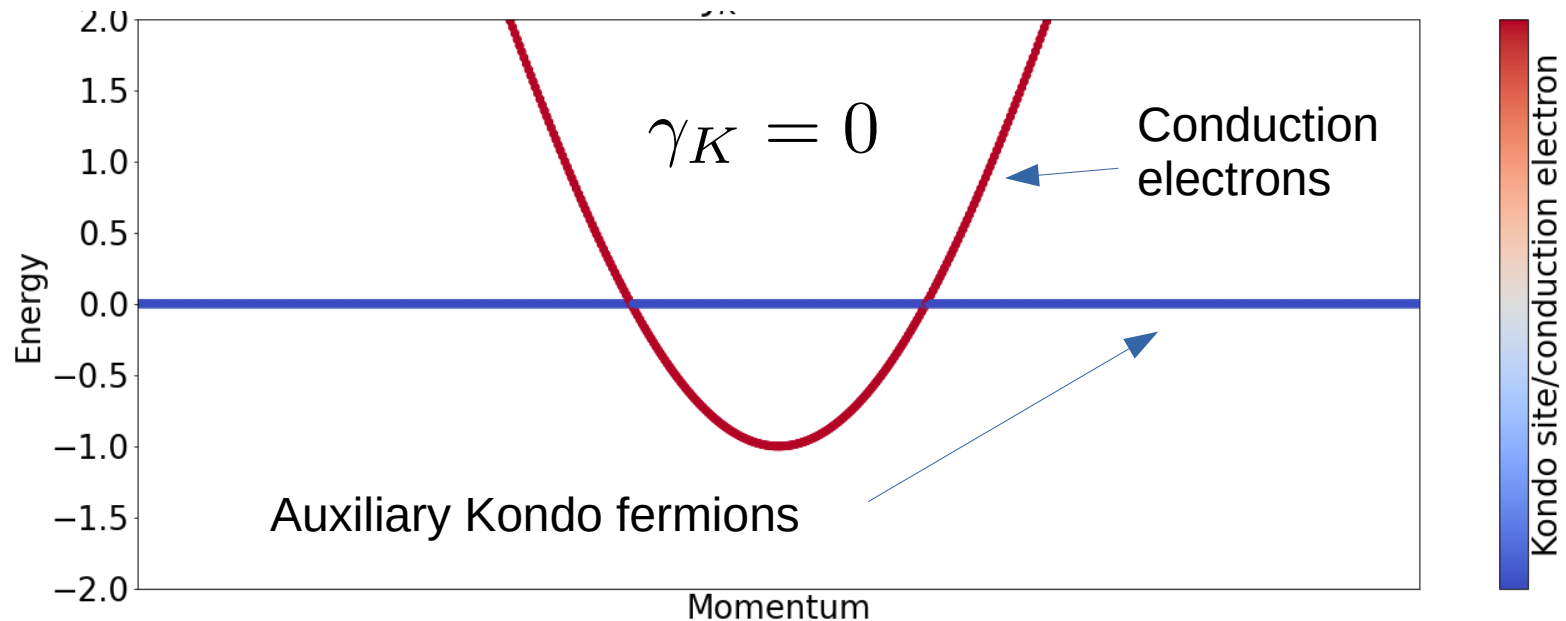
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha\beta} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$

Conduction band dispersion

Kondo hybridization

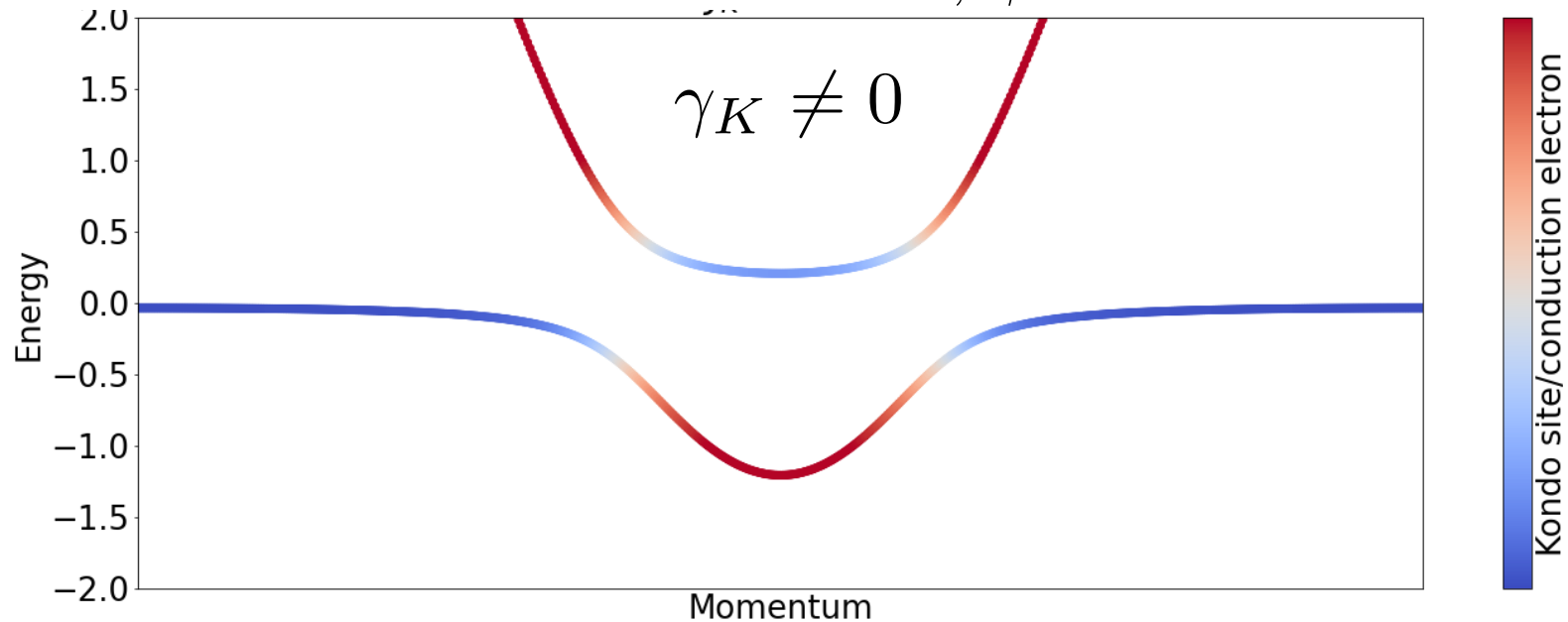
Electronic structure of the Kondo lattice problem

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k}, \alpha\beta} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$



Electronic structure of the Kondo lattice problem

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k}, \alpha\beta} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$

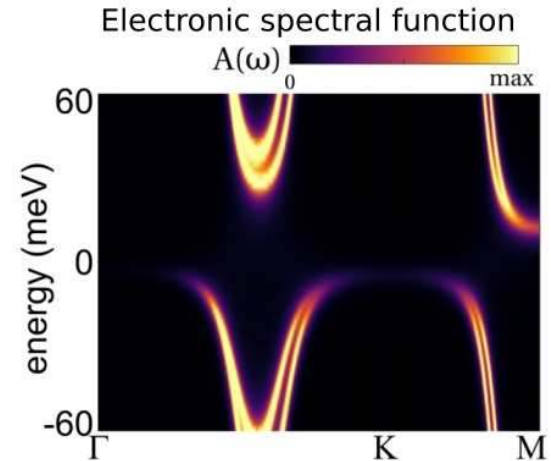
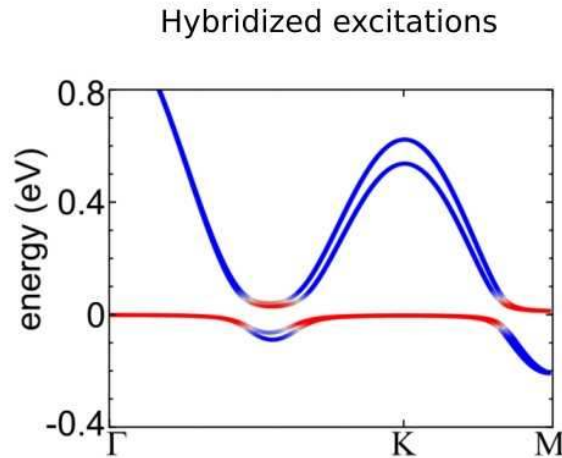
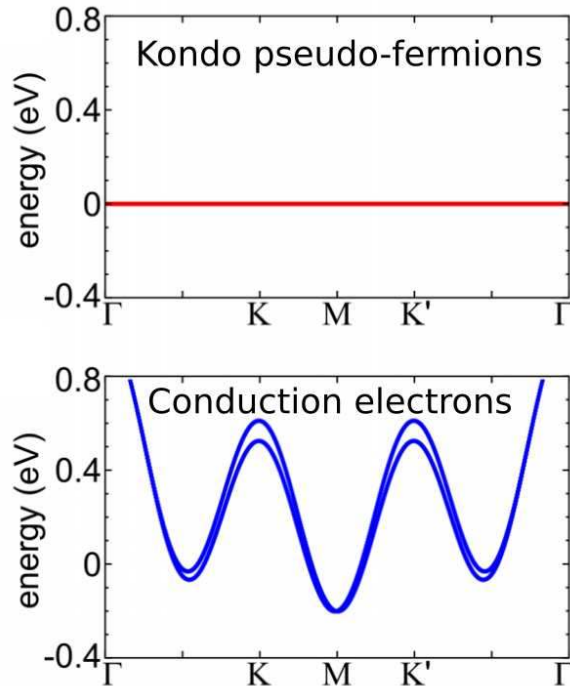


The Kondo coupling opens up a gap in the electronic structure

Brief theory of heavy-fermions

Kondo physics introduces resonant pseudo-fermions at the chemical potential

Leading to the opening of a heavy-fermion gap

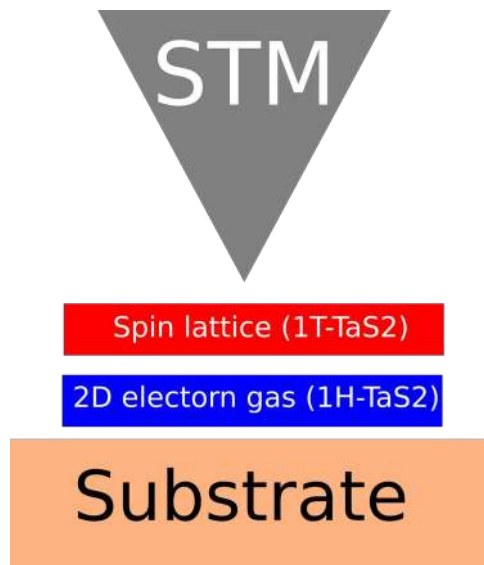


Experimental signatures of heavy-fermion physics

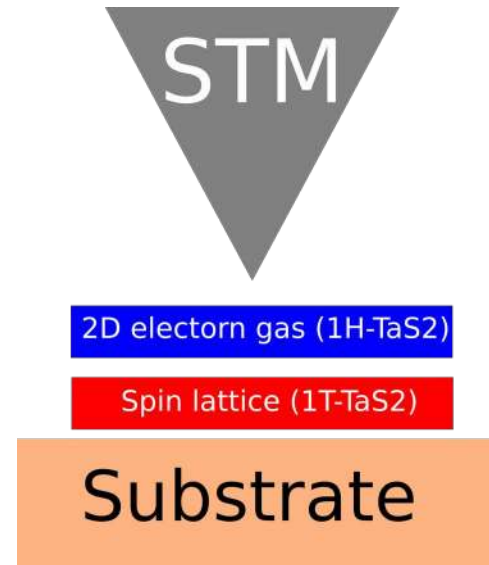
- Kondo physics in the magnetic lattice
- Gap opening in the metallic layer

Both signatures can be probed with scanning tunnel microscopy by growing two heterostructures

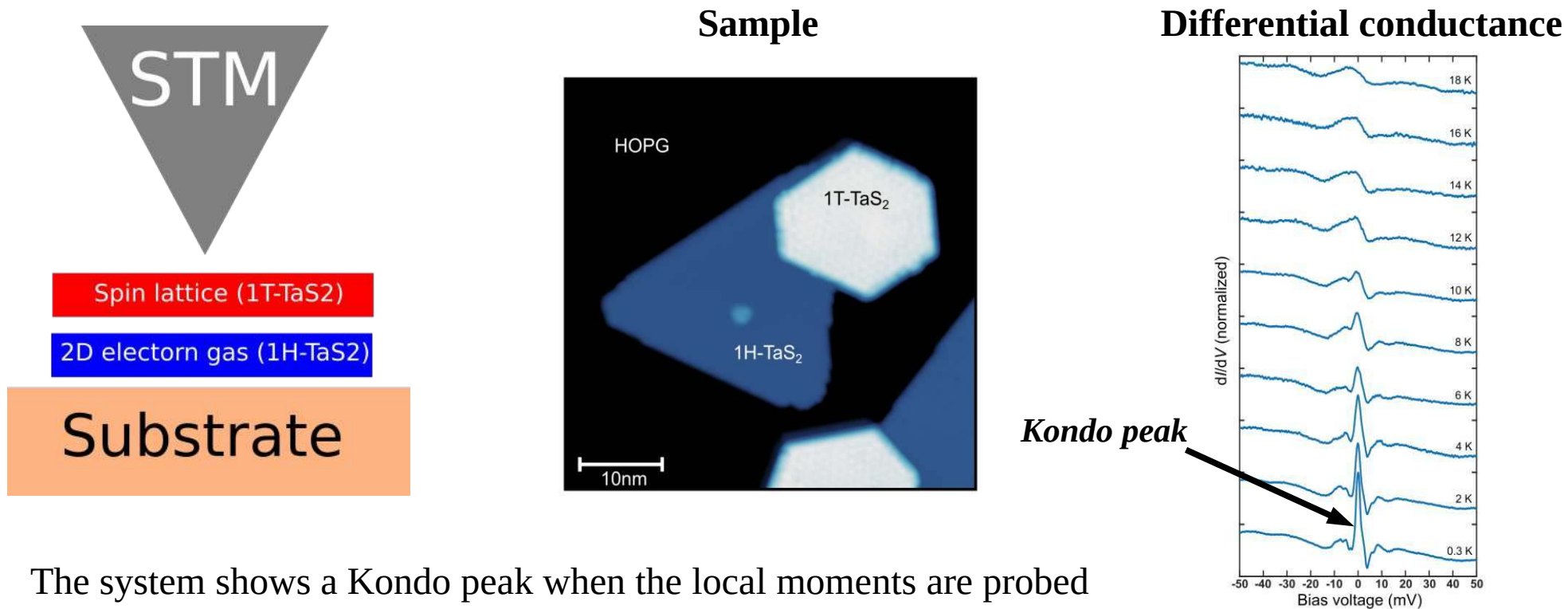
Probing the Kondo peak



Probing the heavy-fermion gap



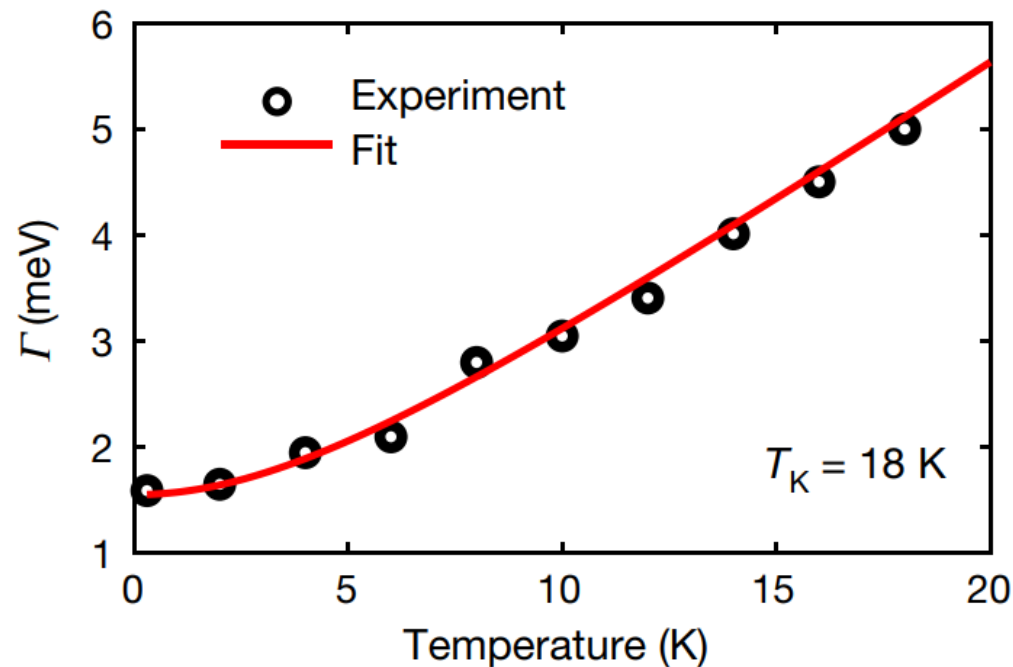
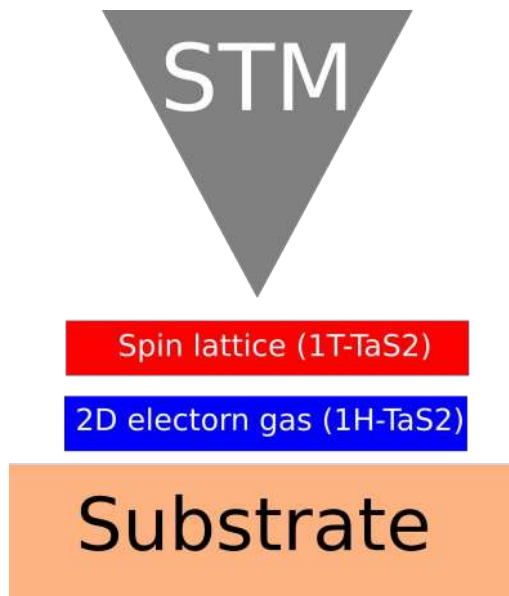
Kondo physics in the magnetic lattice



The system shows a Kondo peak when the local moments are probed

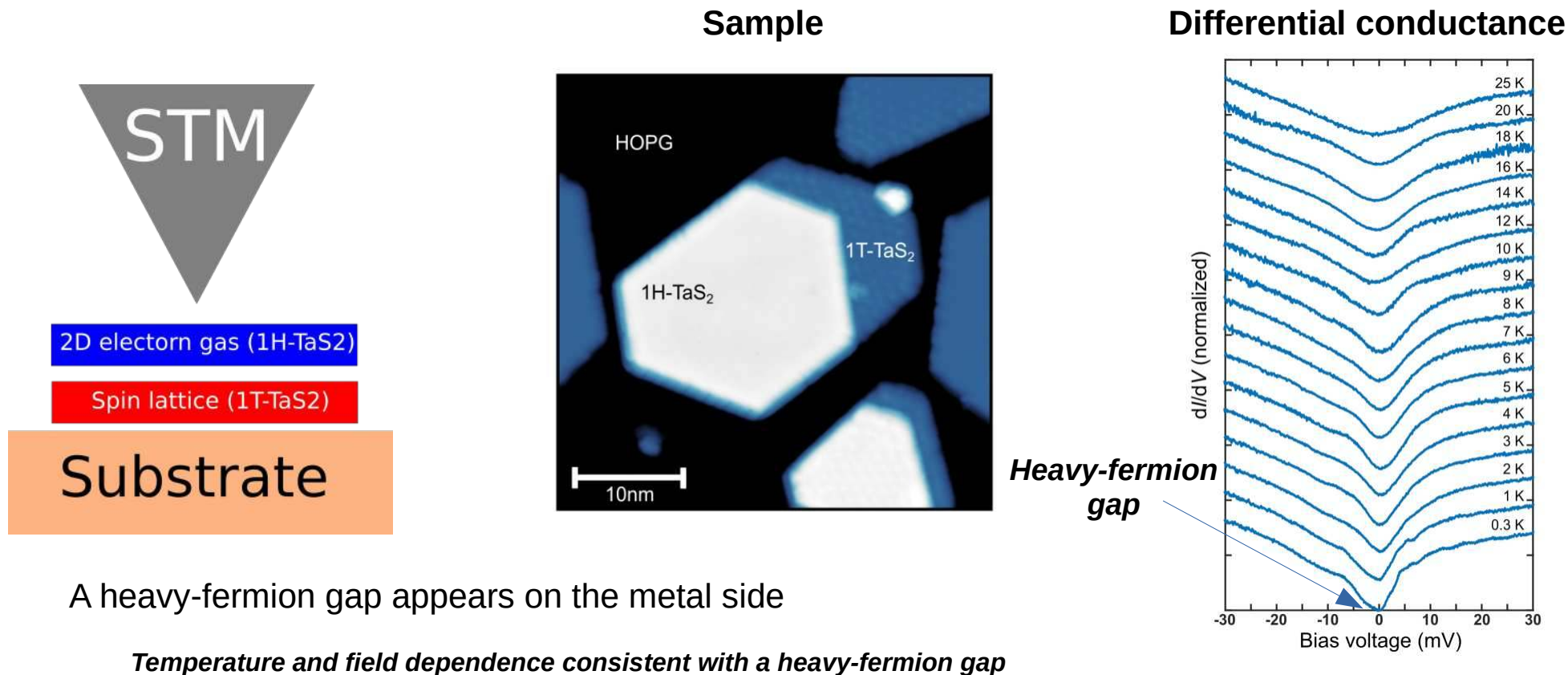
Temperature and field dependence consistent with Kondo

Kondo physics in the magnetic lattice



The Kondo peak follows the expected temperature dependence

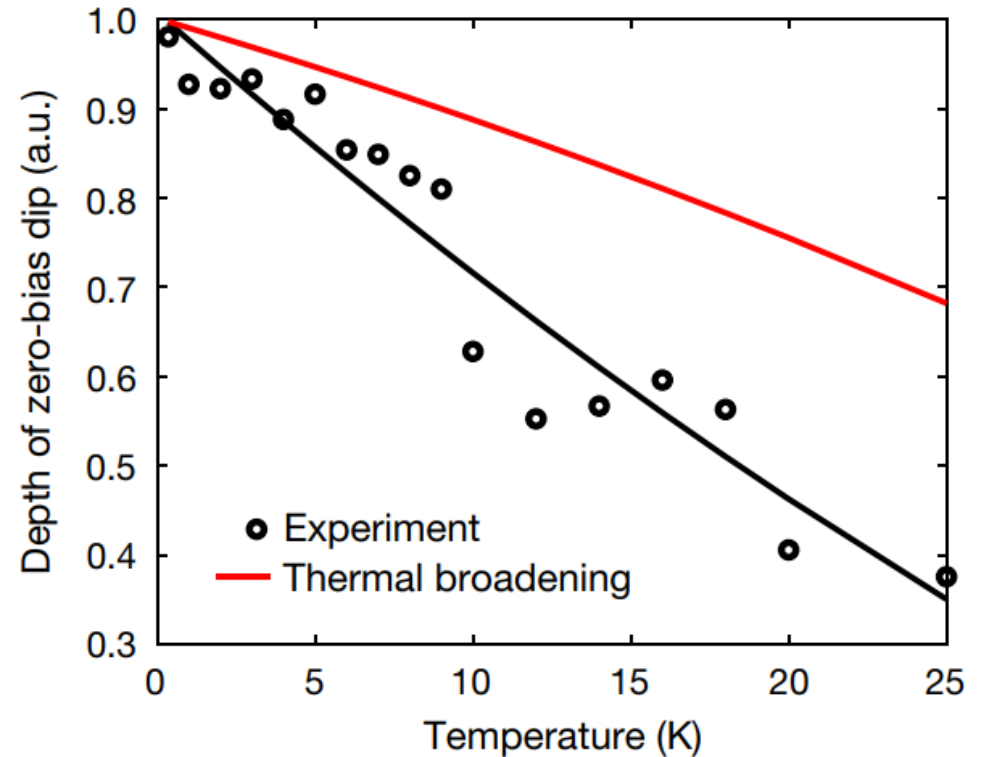
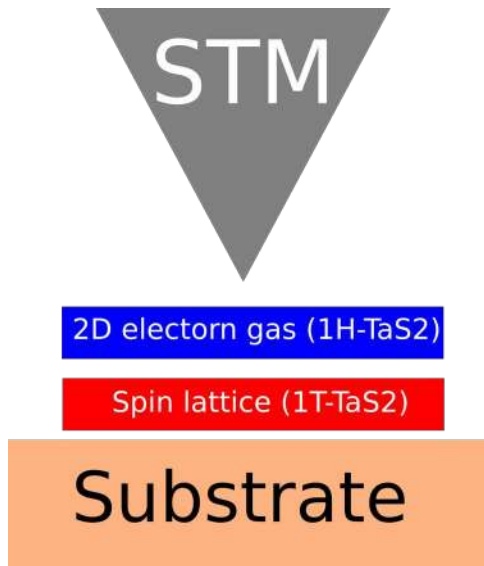
Heavy-fermion gap in the 2D electron gas



A heavy-fermion gap appears on the metal side

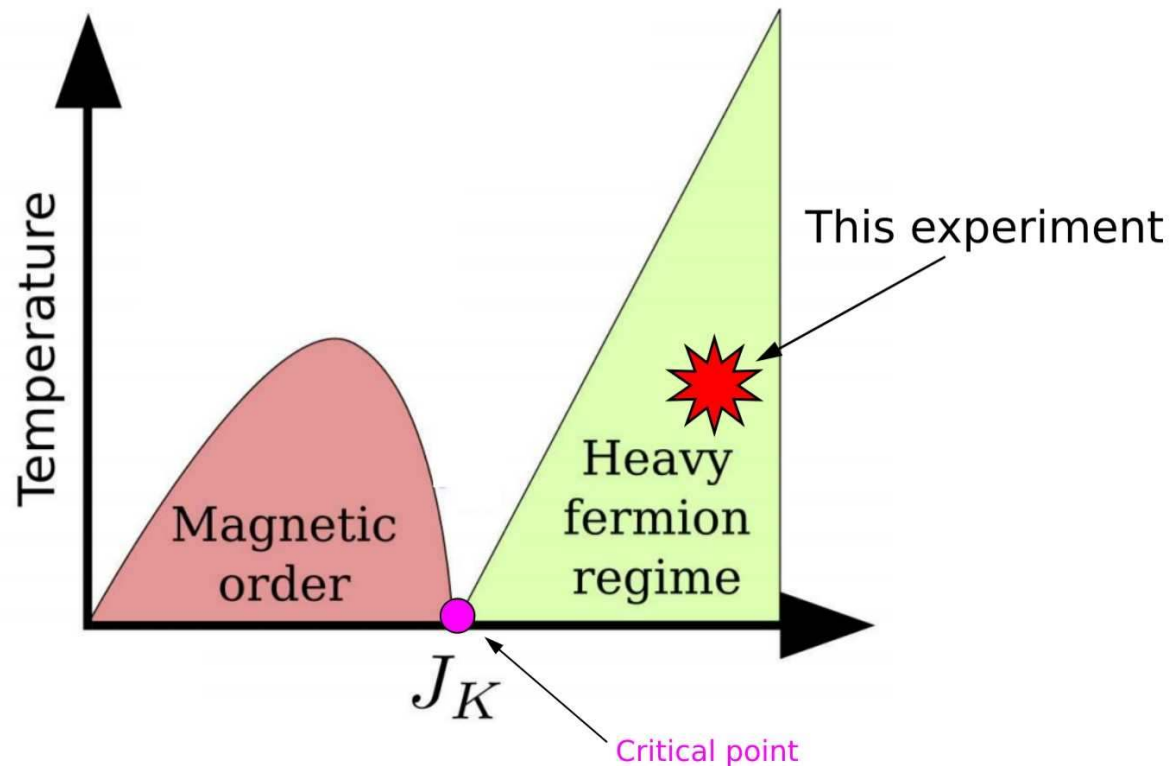
Temperature and field dependence consistent with a heavy-fermion gap

Heavy-fermion gap in the 2D electron gas



The gap shows a temperature dependence beyond thermal broadening, expected from a heavy-fermion gap

Next steps in van der Waals heavy-fermions



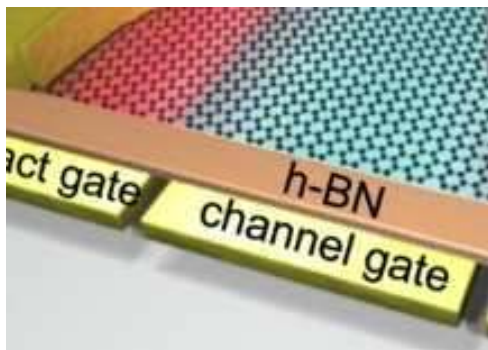
Can we push this system to critical and different unconventional superconducting regime

Controlling the heavy-fermion state

*Of course, we can use the typical knobs of bulk compounds
(pressure, chemical doping, etc)*

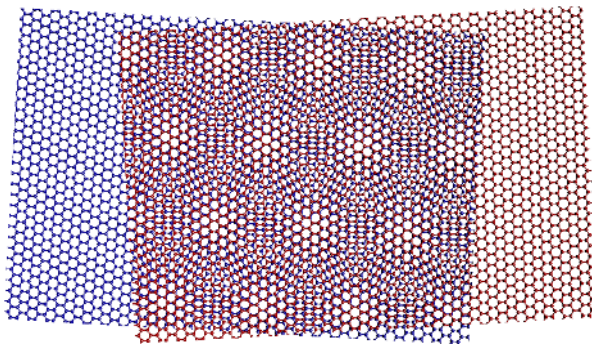
Most importantly, we can exploit the full tunability of van der Waals materials

Gating



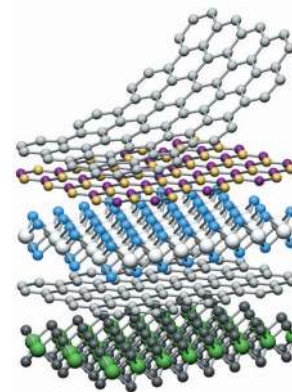
Science 306, 5696, 666-669 (2004)

Twist engineering



Nature Reviews Materials 6, 201–206 (2021)

Materials engineering

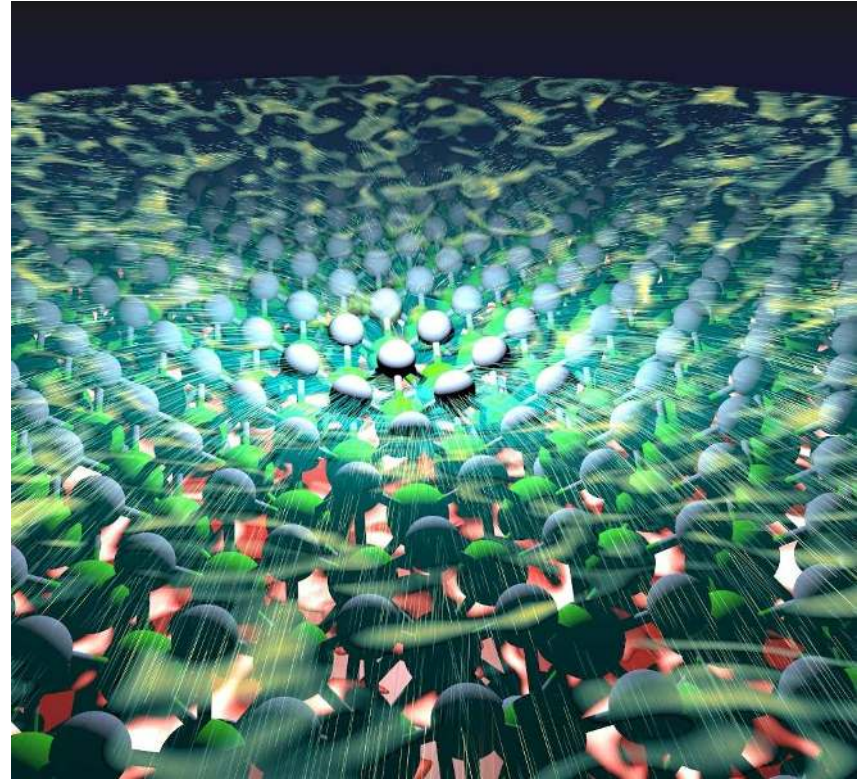
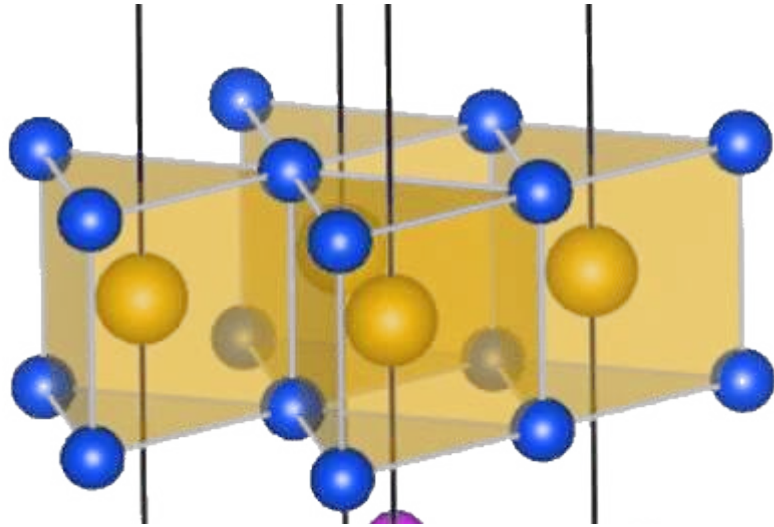


Nature 499, 419–425 (2013)

Allowing to independently control Kondo coupling, exchange coupling, doping and electronic dispersion

Nodal superconductivity in ultraclean TaS_2

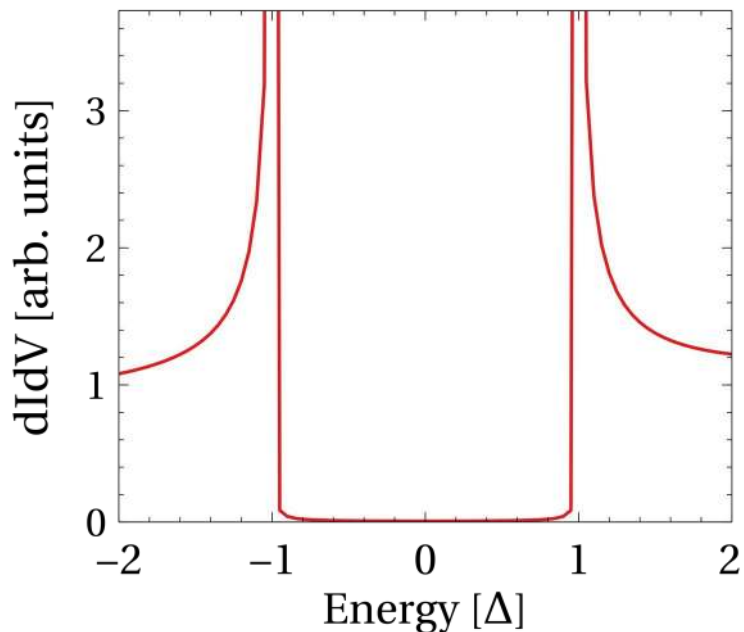
H'



arXiv:2112.07316 (2021)

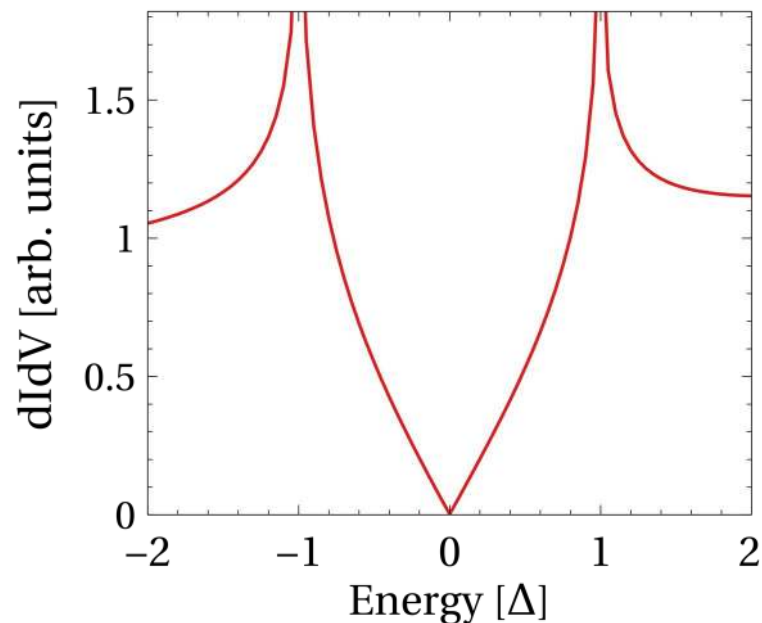
Gapped and nodal superconductivity

Conventional s-wave



Driven by electron-phonon

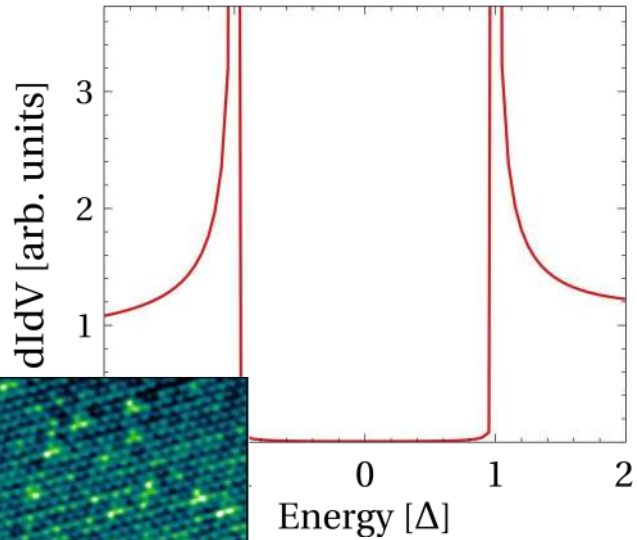
Unconventional nodal f-wave



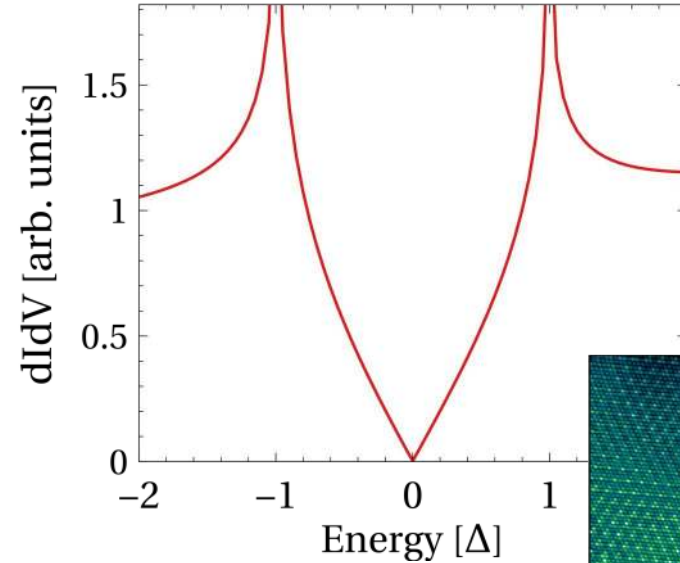
Driven by unconventional mechanism
(magnons, valence fluctuations)

Two superconducting states in TaS₂ monolayer

Strong disorder
Fully-gaped s-wave

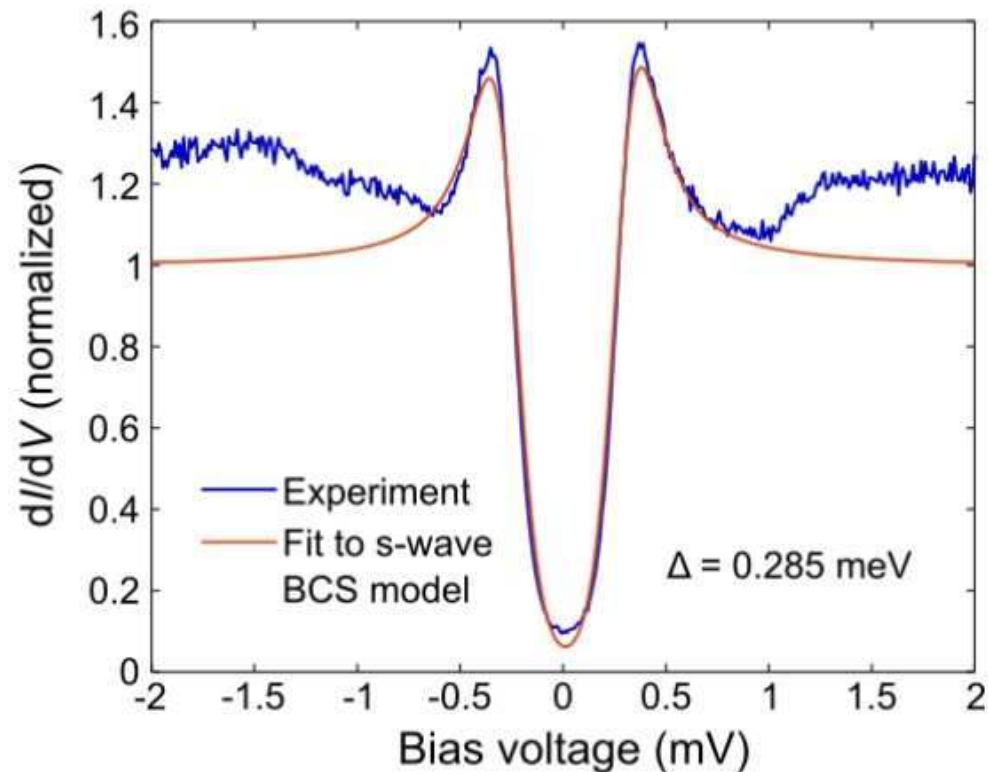
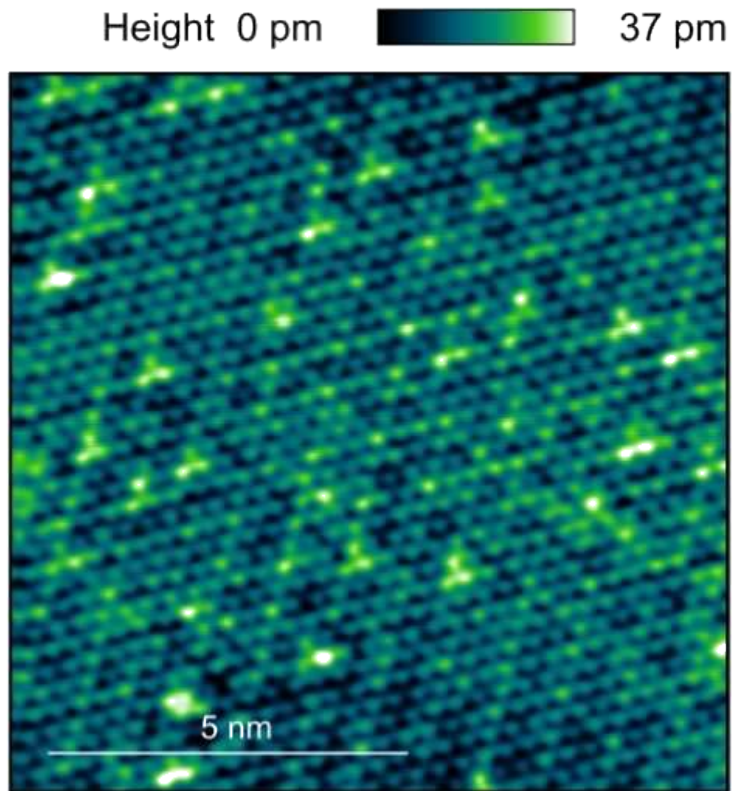


Weak disorder
Gapless nodal f-wave

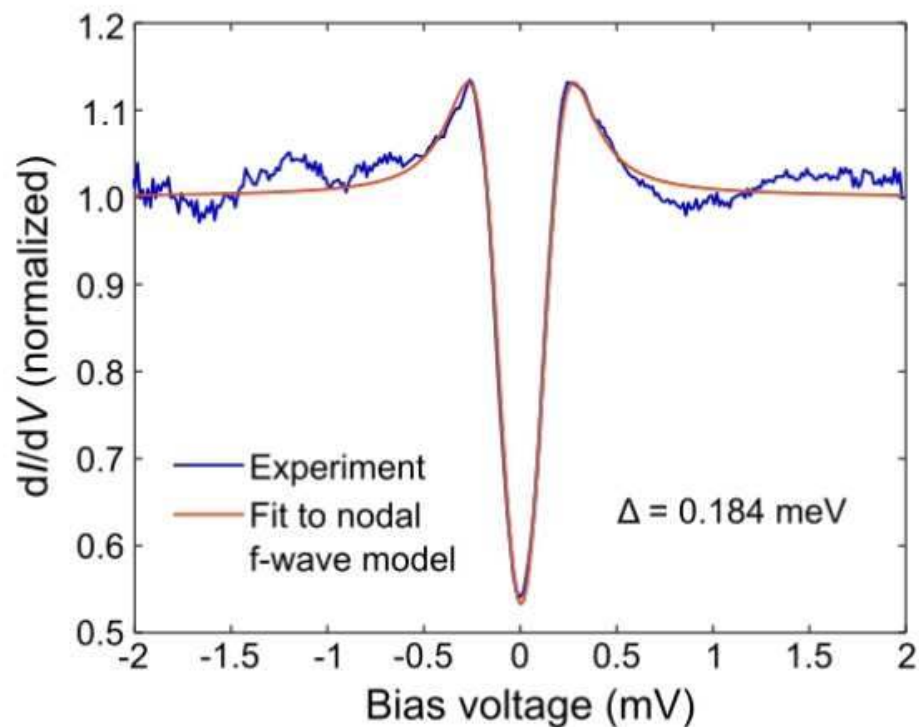
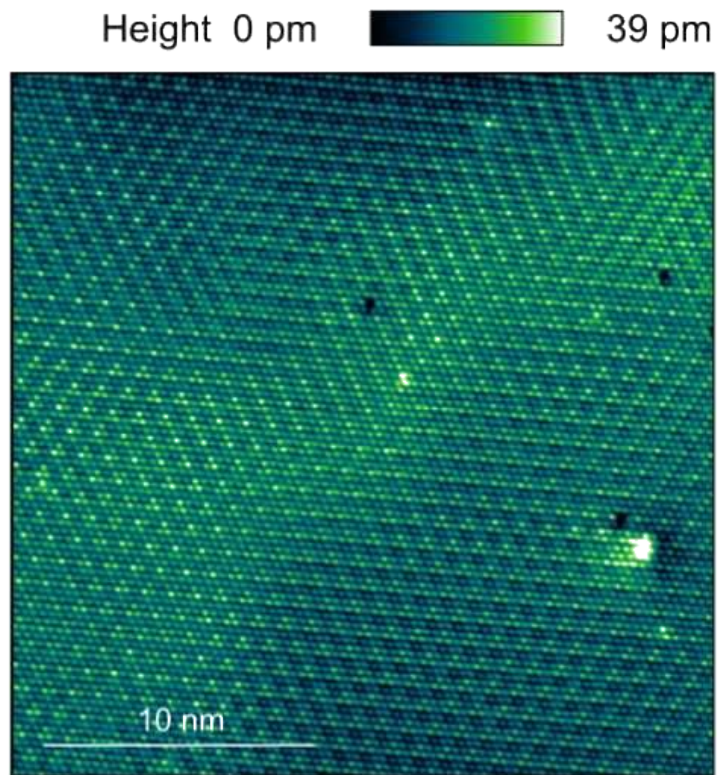


Superconducting phase transition associated with disorder

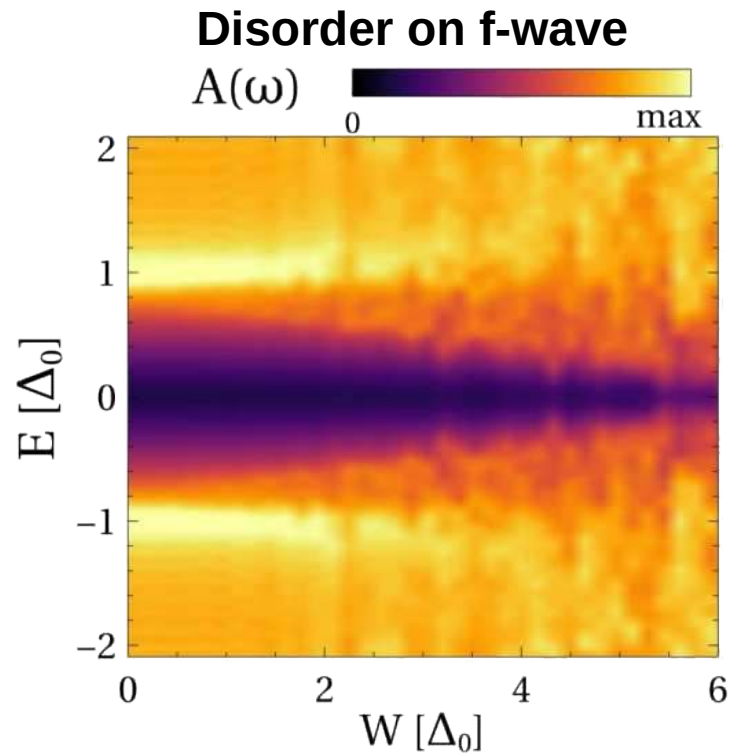
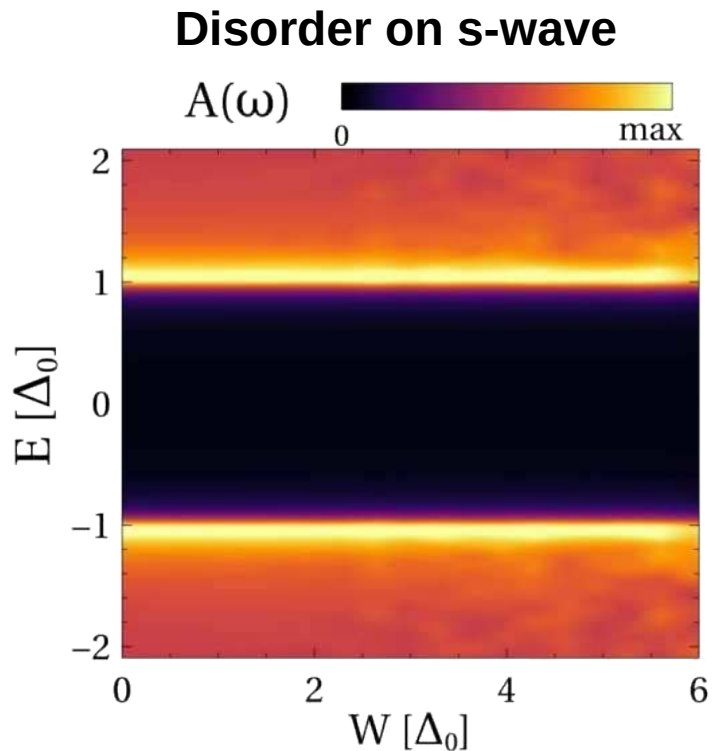
Dirty TaS₂ monolayer sample



Ultra-clean TaS₂ monolayer sample

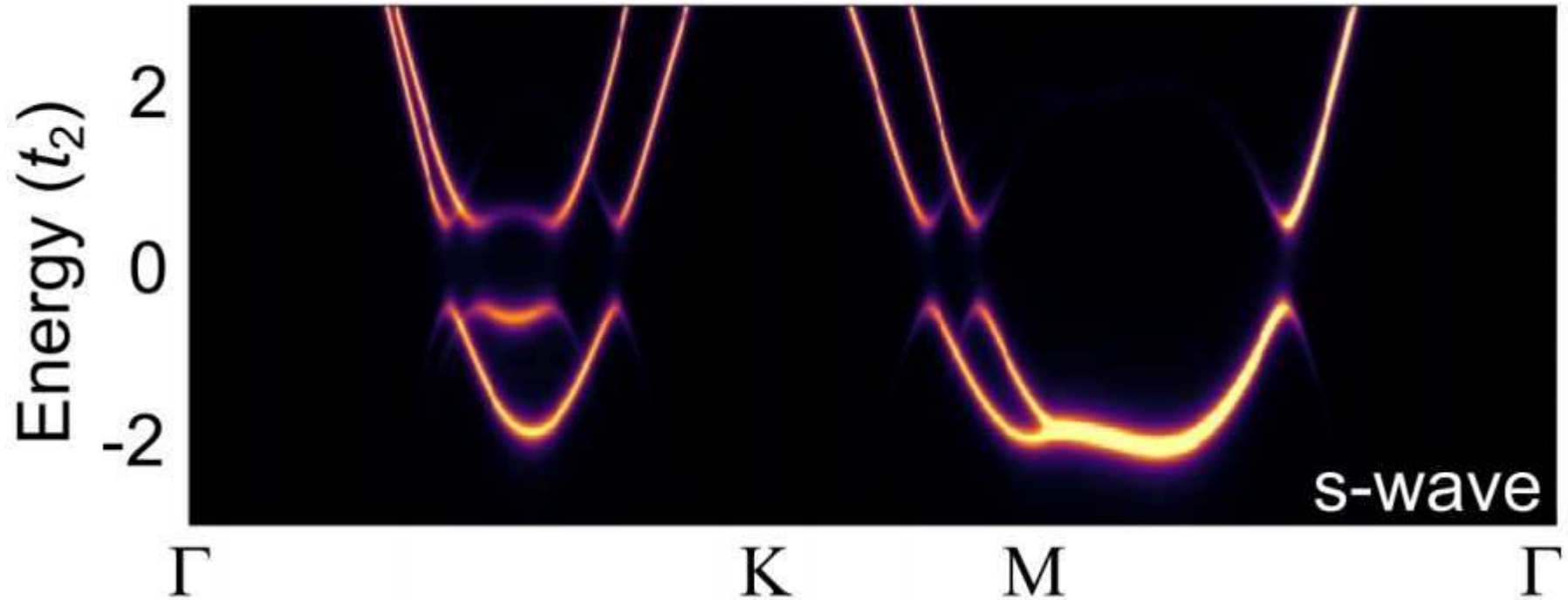


Impact of disorder for f-wave and s-wave



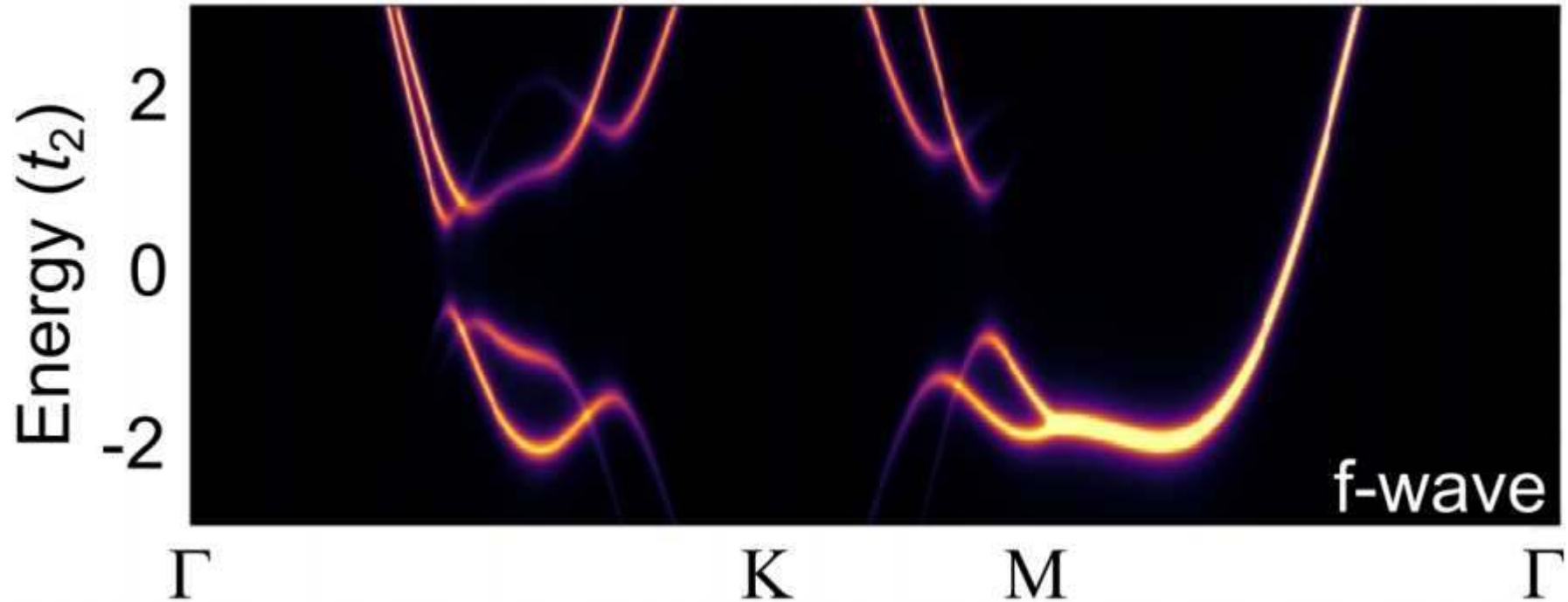
$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{SC} + \sum_{n,s} W_n c_{n,s}^\dagger c_{n,s}$$

Electronic structure with an s-wave gap



A full gap appear in the band structure in the presence of superconductivity

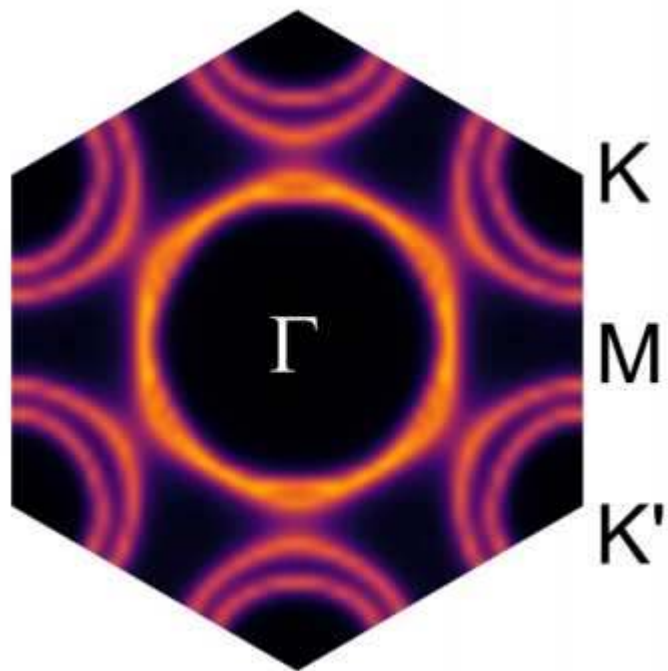
Electronic structure with an f-wave gap



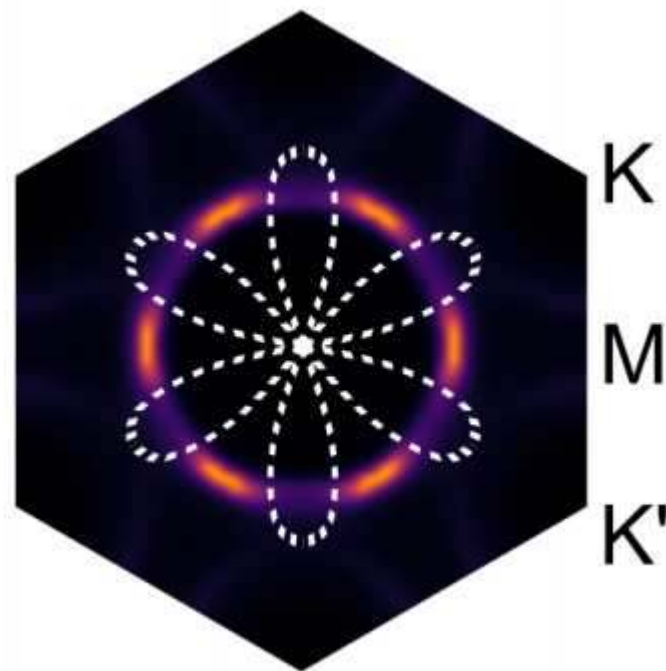
Part of the band structure remains gapless in the presence of superconductivity

Fermi surface of 1H-TaS₂

Normal State

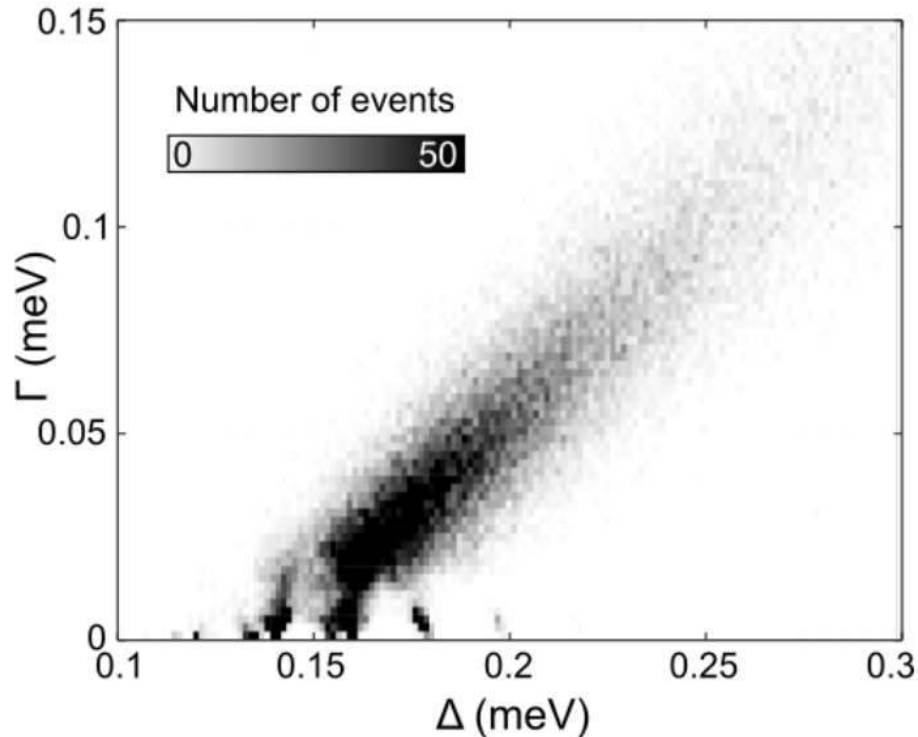


Nodal superconducting state



Nodal f-wave order

Gap versus pair breaking in ultraclean TaS₂



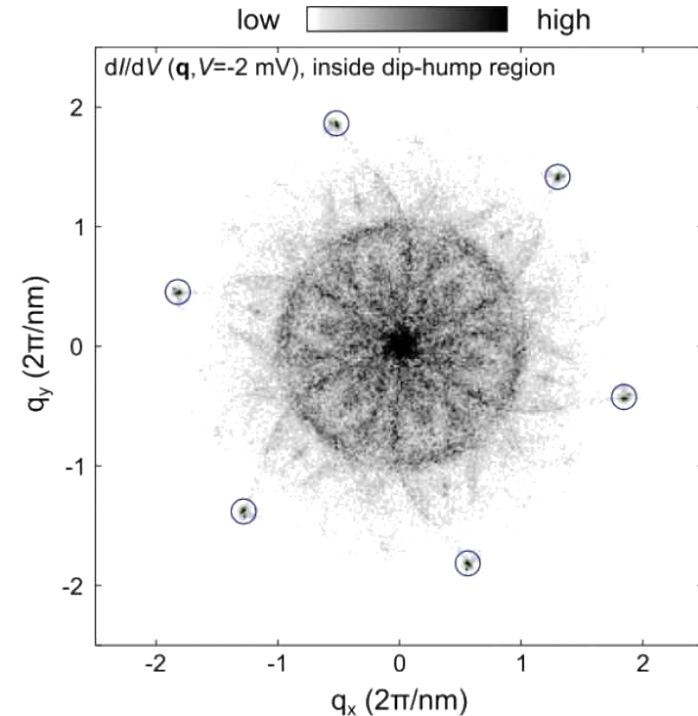
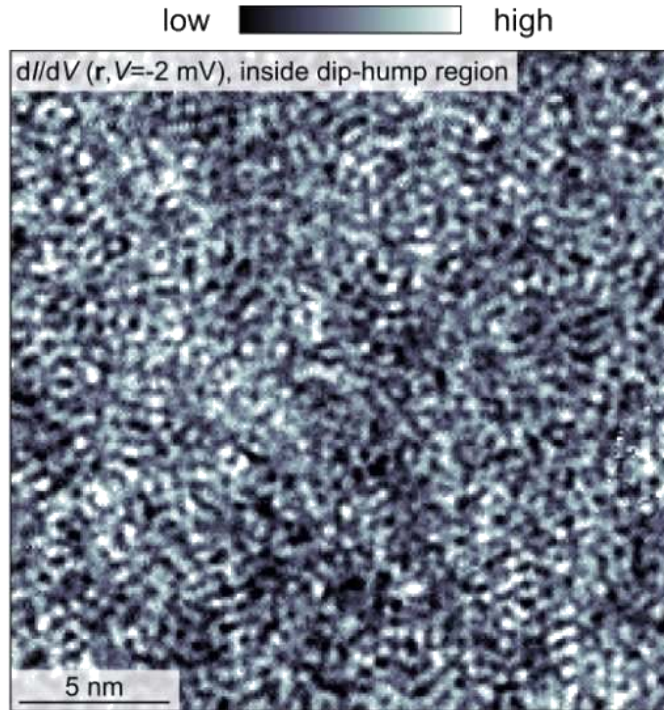
Let assume that the ultraclean sample is actually s-wave with pair breaking (Dynes)

$$\rho(E) = \left| \Re \left[\frac{E - i\Gamma}{\sqrt{(E - i\Gamma)^2 - \Delta^2}} \right] \right|$$

Physically, the gap must be anticorrelated with the pair-breaking

The ultraclean sample is incompatible with s-wave gap with pair breaking

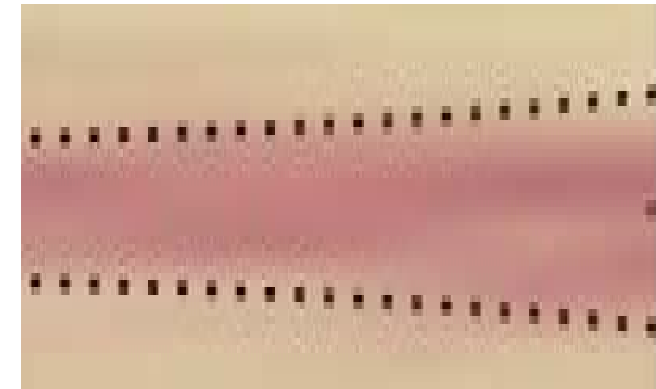
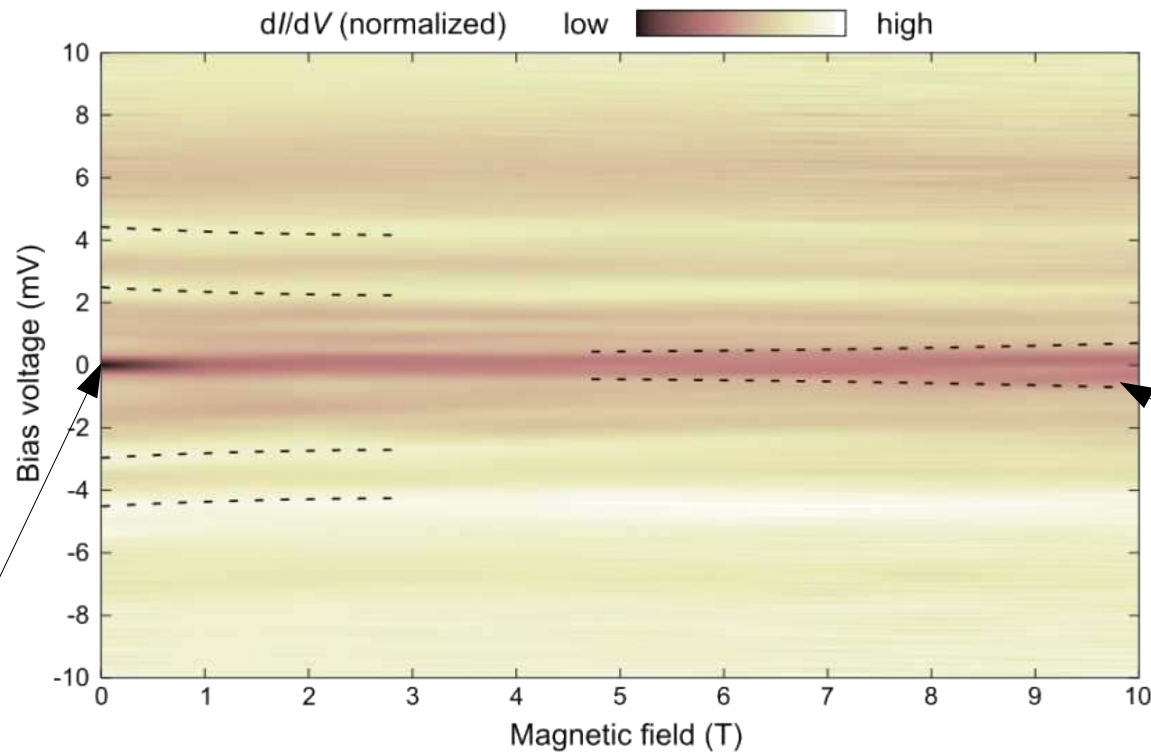
Hidden order many-body fluctuations



Inelastic excitations with a characteristic length scale emerge above the coherence peaks

Existence of collective many-body modes, potentially related with the superconducting state

Ultraclean sample versus magnetic field



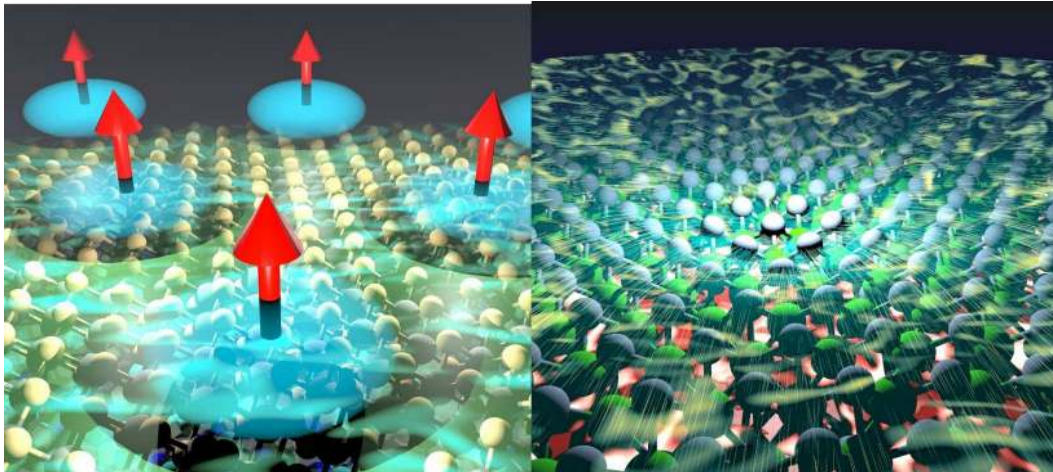
Pseudo-gap

Superconducting gap

A magnetic field increases a pseudo gap at the chemical potential

Take home

- 1H-1T TaS₂ bilayer shows heavy-fermion Kondo insulator behavior
- Ultraclean 1H TaS₂ monolayer shows nodal superconductivity



Nature 599, 582–586 (2021)

arXiv:2112.07316 (2021)

Thank you

Funding from

