



# Cuprate twistrionics towards a new generation of quantum hardware



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2D Quantum Materials

Engineered Materials

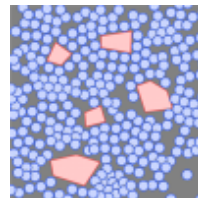
July 15-28, 2024

Ramanujan Lecture Hall

Speakers |  
Abhay Pasupathy  
Aditya Lakshminarayana

Disakar Valsak  
Peter B Littlewood  
Priya Mahalingam  
Romy Thomale\*  
Kooda Perichova  
Sergey Kumar

This meeting will focus on recent developments exploring emergent electronic, magnetic, and topological phenomena in two-dimensional synthetic structures of quantum materials. The strong interplay between electronic correlations, spin-orbit coupling, crystal structure, symmetry, and topology often leads to various intriguing phenomena in bulk crystals, such as...



Superpuddles Lab

# Acknowledgments: the members and funding sources

## Current Members:

Tommaso Confalone, Mickey Martini, Sanaz Shokri, Giuseppe Serpico, Yejin Lee, Sushmita Chandra, Flavia Lo Sardo, Haolin Jin, Kristiane Kranz, Rongxin Li, Narayan Kunchur, **Golam Haider**

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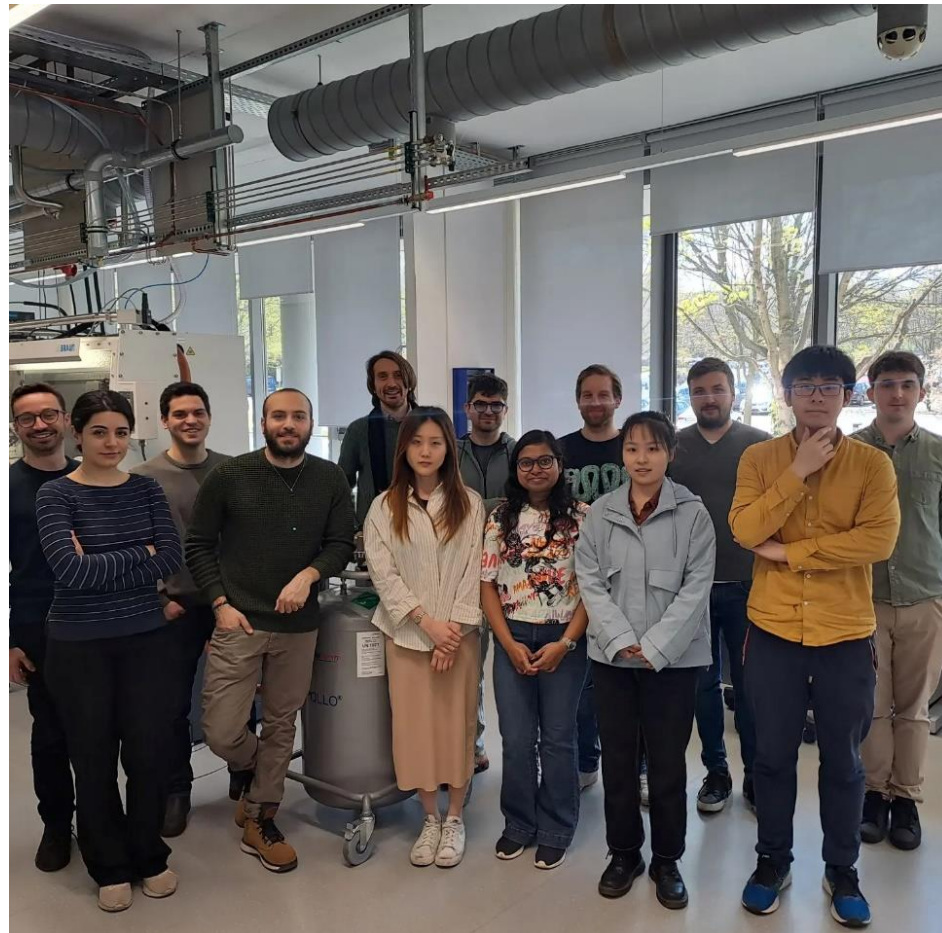


photo in 2023

# Collaborators

Thank you to my main current collaborators in this journey that I will present you today



Kornelius Nielsch



Uri Vool



Stefan Kaiser



Claudia Felser



Daniel Wolf



Francesco Tafuri



Domenico Montemurro



Davide Massarotti



Valentina Brosco



Federico Cagliaris



Valerii Vinokur



Claudio Mazzoli



Genda Gu



Luca Chirolli



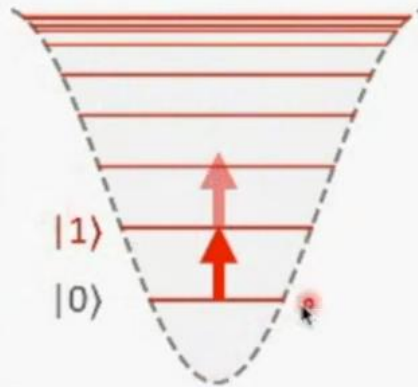
Hiroshi Eisaki

# From idealization to physical reality

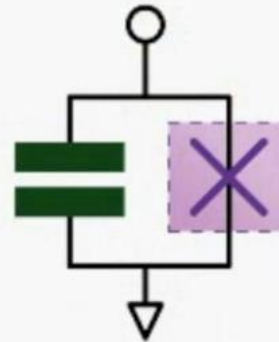
Idealization of qubit



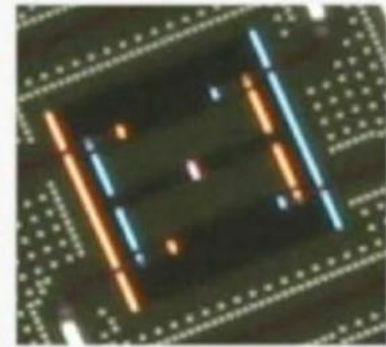
Anharmonic oscillator



Physical circuit model



Physical layout



Idealization



Physical reality

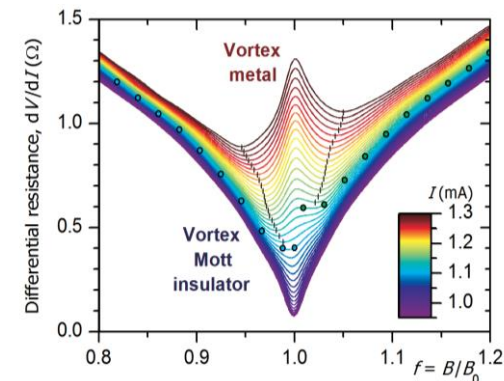
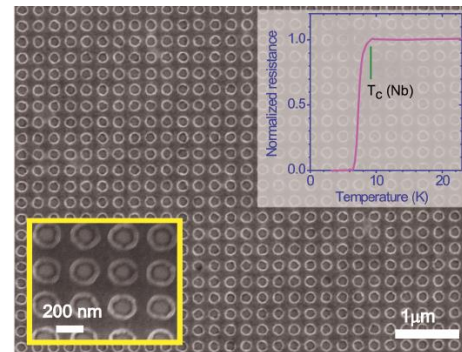
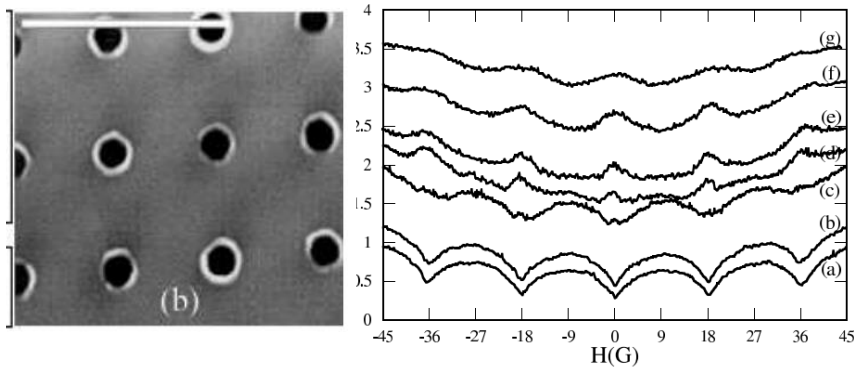
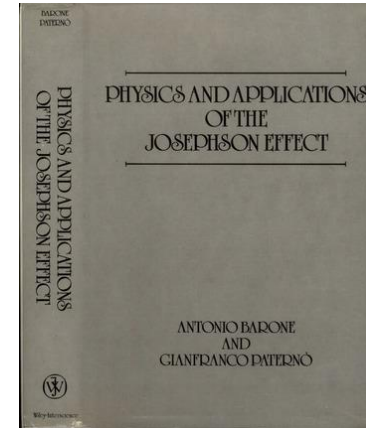
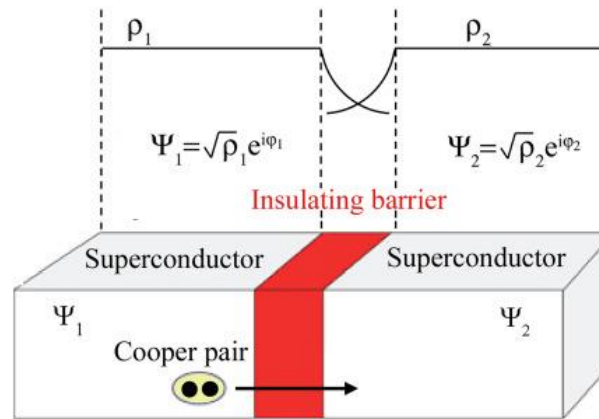
# The grand-challenge in the Josephson-effect: the detrimental disorder

$$I(t) = I_c \sin(\phi(t))$$

$$\frac{\partial \phi}{\partial t} = \frac{2eV(t)}{\hbar}$$

B. D. Josephson, *Physical Letters*. **1**, 251-253 (1962).

A. Barone, G. Paterno. *Physics and applications of the Josephson effect*. 1982.

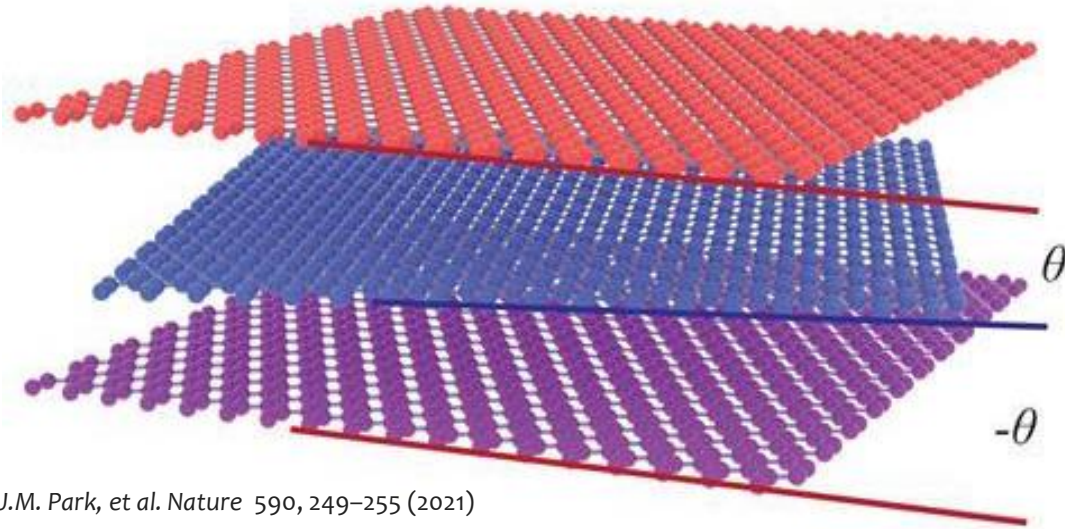


Z. Jiang, et al. *Applied physics letters* 84 5371-5373 (2004).

NP et al., *Science* 349, 202-1205 (2015)

# A revolution in materials science

Twistronics = 2D materials + twist angle



J.M. Park, et al. Nature 590, 249–255 (2021)

## Unprecedented tunability

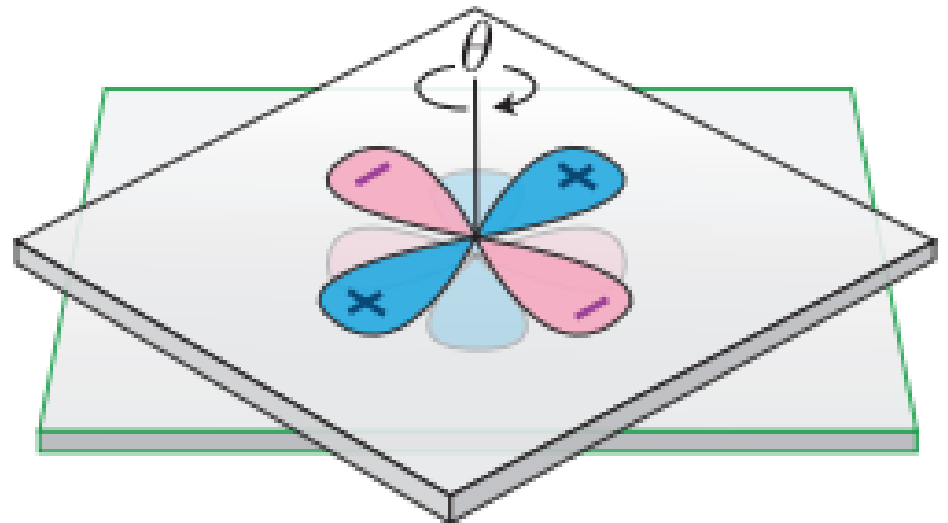
- Correlated physics
- Superconductivity
- Topological matter

- Cuprates materials show an inexplicable macroscopic quantum state.

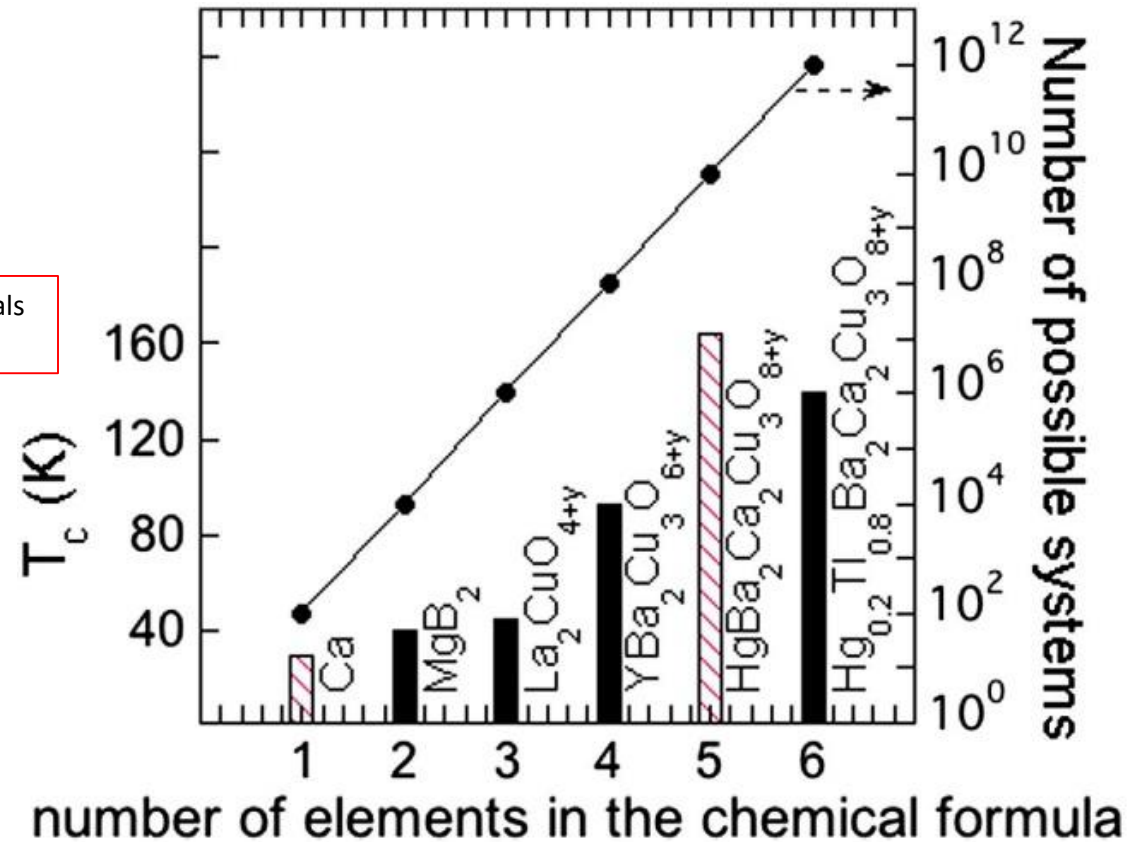
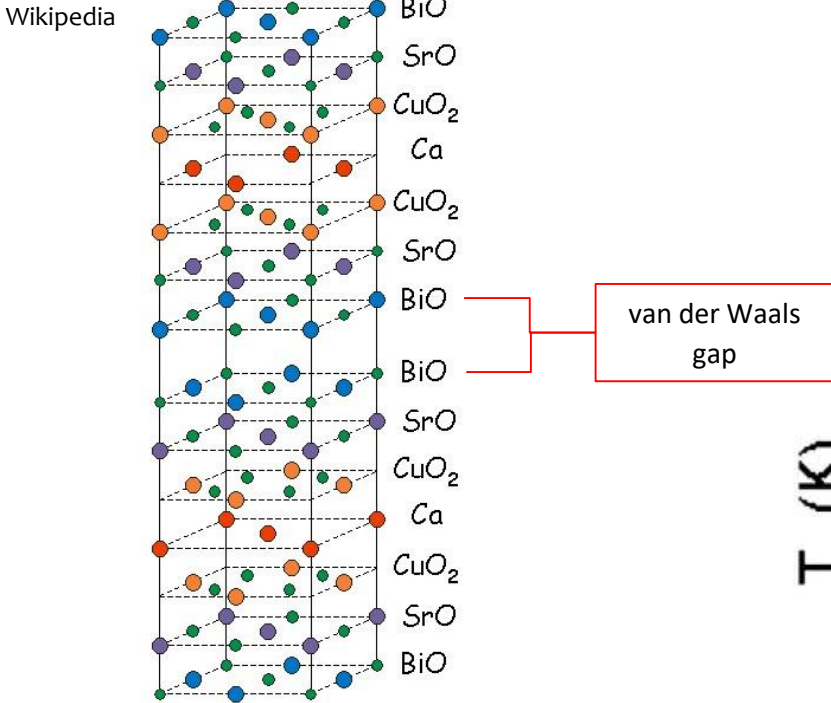
J. Bednorz and K.A. Müller *Zeitschrift für Physik B Condensed Matter* 64, 189-193 (1986).

- Twisted cuprate structures proposed for exploring new physics.

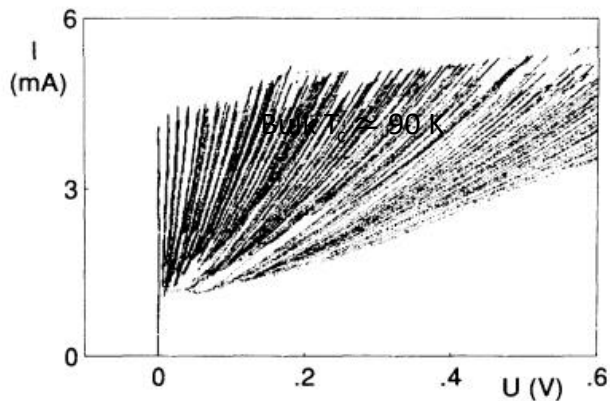
M. Sigrist, *Progress of theoretical physics* 99, 899-929 (1998)



# Physics of complexity meet macroscopic quantum states



NP et al. *Proceedings of the National Academy of Sciences* 109, 15685-15690 (2012).

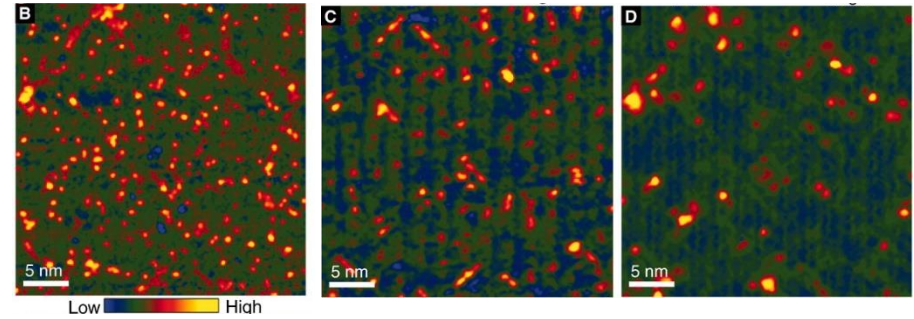
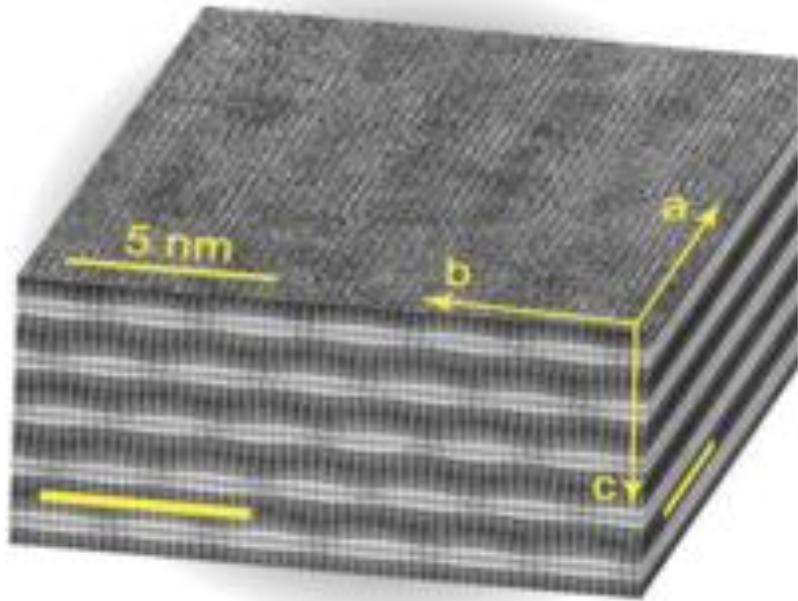


- Intrinsic Josephson junctions in the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  crystal structure

R. Kleiner, et al. *Physical review letters* 68 2394 (1992).

# The complexity created by the oxygen interstitials

the most vagabond among the elements



Apical oxygen,

O-i near SrO,

O-i near BiO,

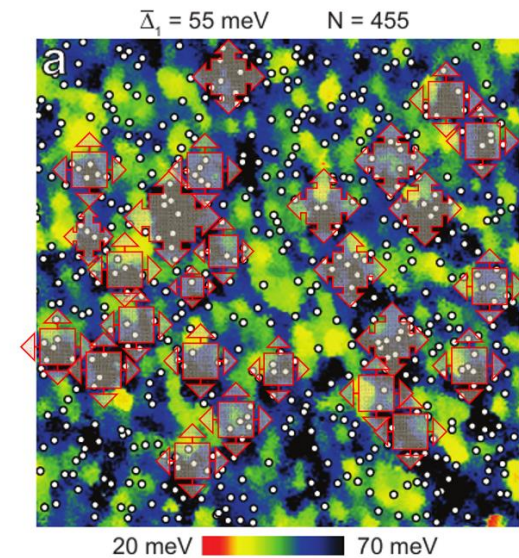
I. Zeljkovic, et al. *Science* 337, 320-323 (2012).

- Incommensurate superlattice modulation

NP et al, *Physical Review Materials* 4, 114007 (2020).

- Oxygen interstitials nanopuddles in Bi2212

V. Velasco, et al. *Journal of Condensed Matter Physics* 35, 415602 (2023)



- Superconducting gaps inhomogeneity in Bi2212

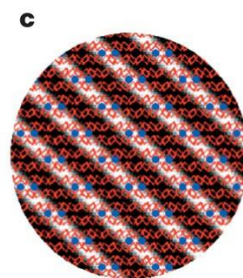
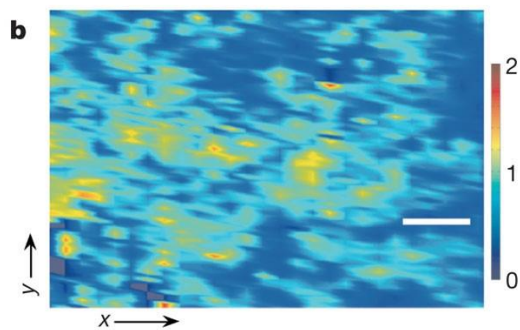
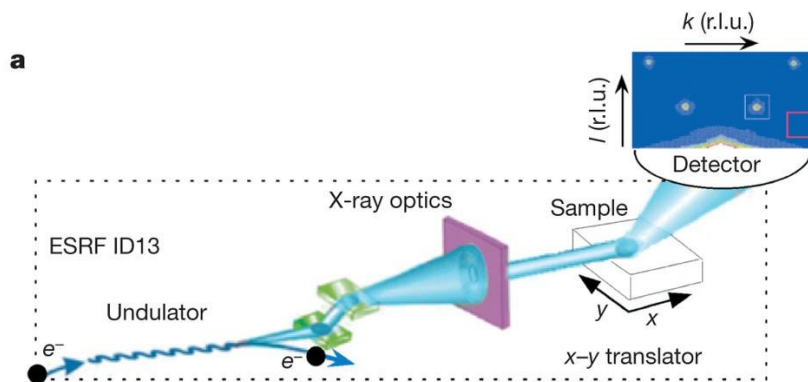
Fischer, Øystein, et al. *Reviews of Modern Physics* 79 353 (2007)



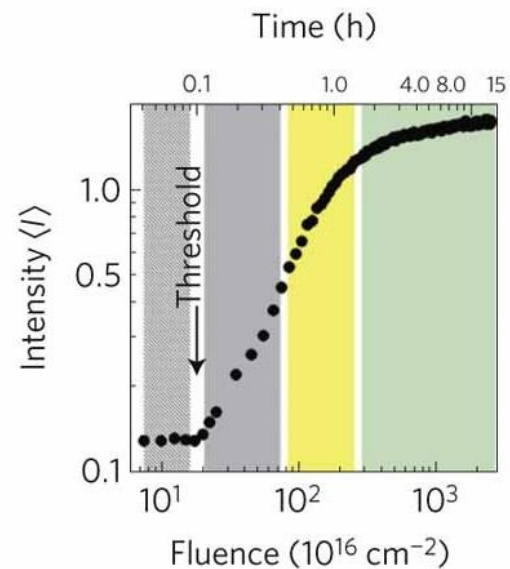
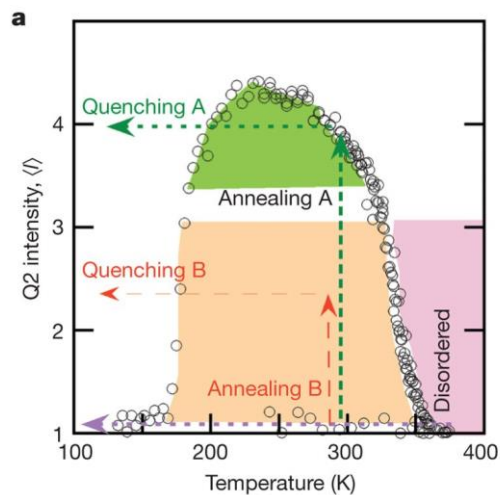
# Evolution of oxygen order in cuprate superconductors

How do we know that oxygen interstitials are frozen below 200K?

- Oxygen interstitials order form fractal structures that seem to promote high-temperature superconductivity



M. Fratini, NP, et al. *Nature* 466, 841-844 (2010).



- Oxygen interstitials order can be tuned with continuous exposition to synchrotron X-ray light in between 200-300 K.

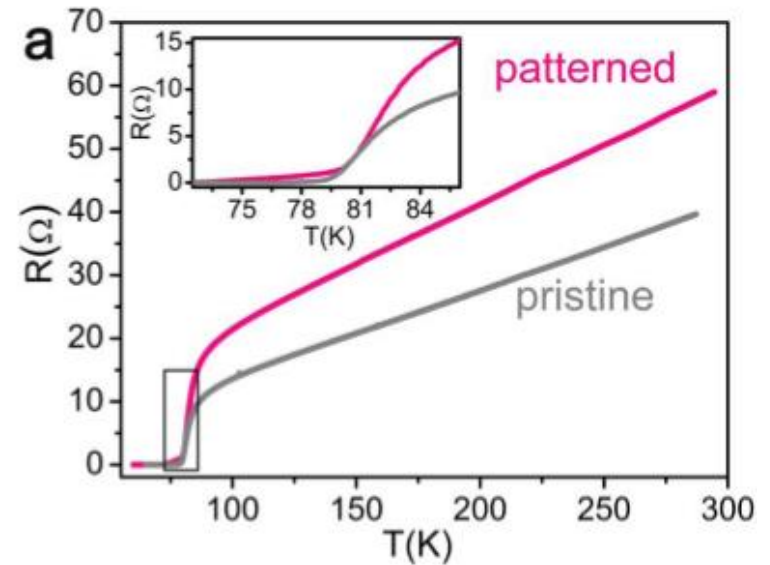
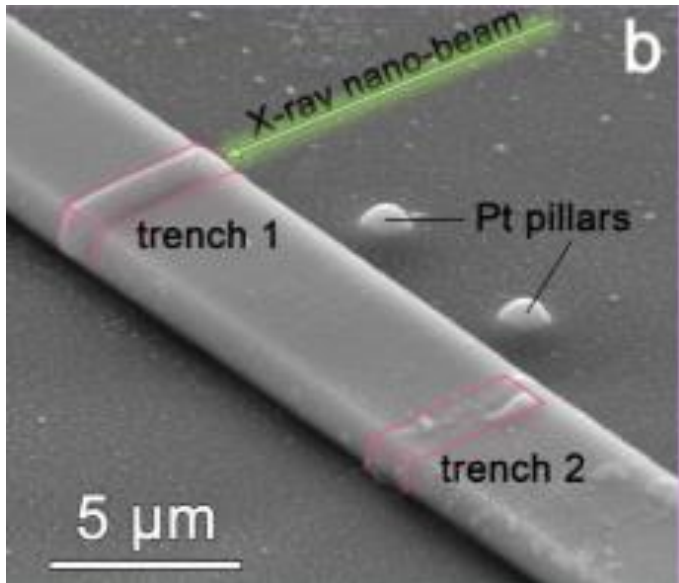
NP et al. *Nature Materials* 10, 733-736.(2011)

- A solvent-polymer free lithographic process is proposed using continuous X-ray light illumination for creating **Josephson junctions** with a cuprate superconductor.

NP, et al. *Superconductor Science and Technology* 25 124004 (2012)

# Solvent-polymer free X-ray lithographic process

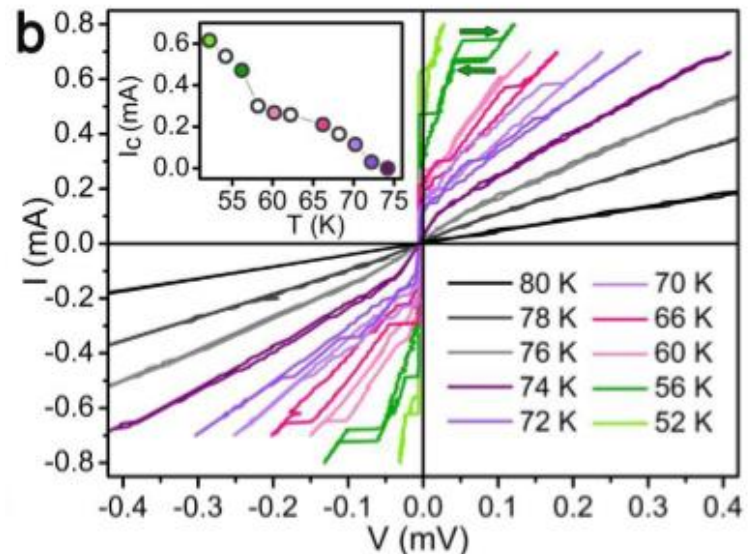
Detrimental disorder created by the depletion and diffusion of oxygen



- Fully functional Josephson junctions devices have been obtained by locally turning the material into a nonsuperconducting state by means of hard nano X-ray exposure.

M. Truccato, et al. *Nano Letters* 16 1669-1674 (2016)

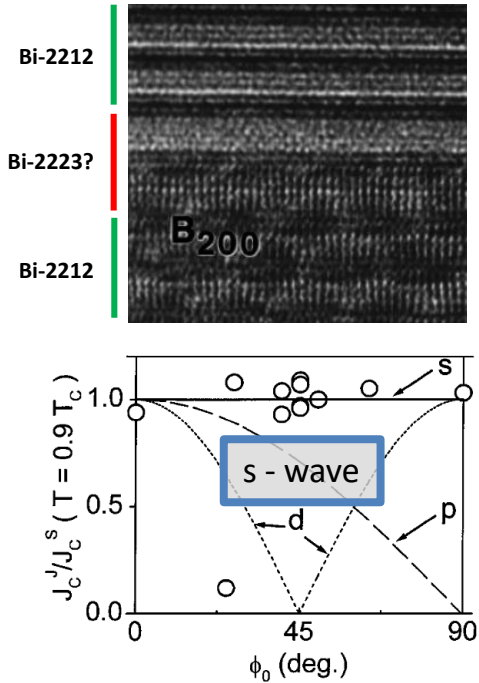
However,  $I_c$  and  $T_c$  control in the Josephson junction is challenging



# Stacked and twisted $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ crystals.

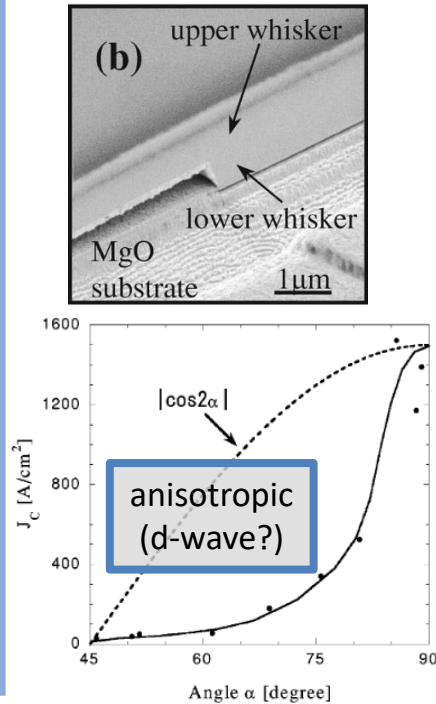
The detrimental disorder is introduced by the high temperature (up to 850 °C) oxygen annealing

## Bulk Crystals



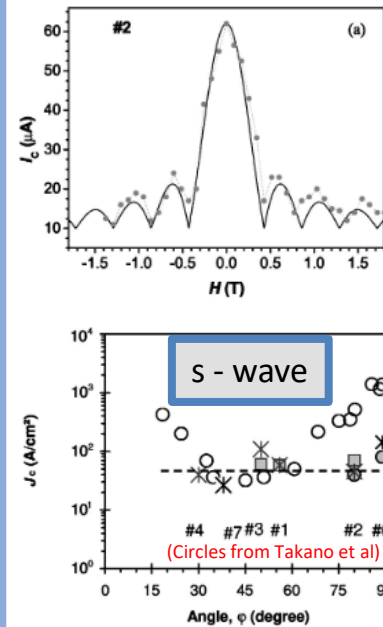
Q. Li, et al. *Physical review letters* 83, 4160 (1999).

## Artificial Whisker Junctions



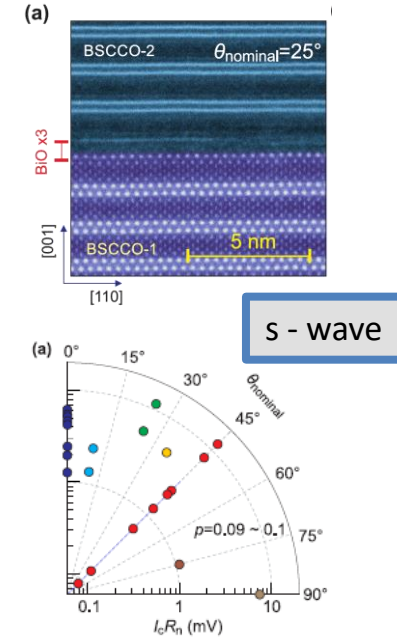
Y. Takano, et al. *Physical Review B* 65, 140513 (2002).

## Natural Whisker Junctions



Y. I. Latyshev, et al. *Physical Review B* 70 094517 (2004).

## Exfoliated Flakes

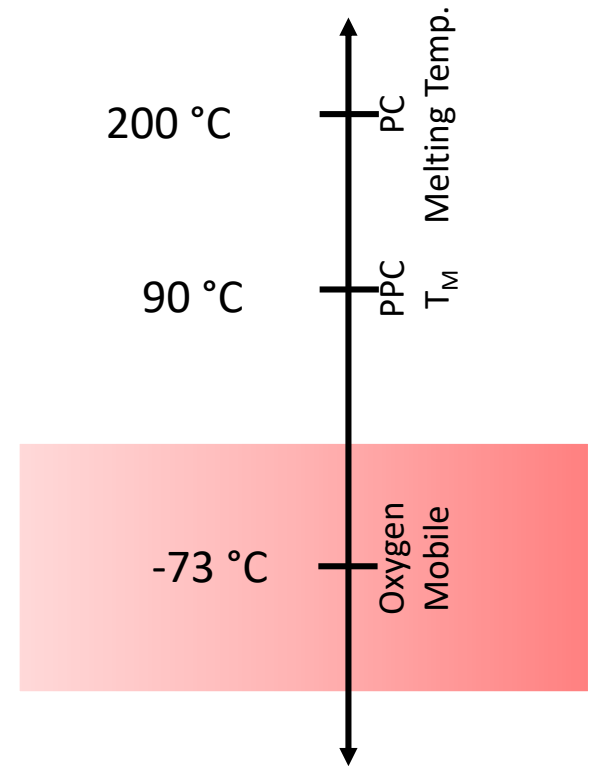
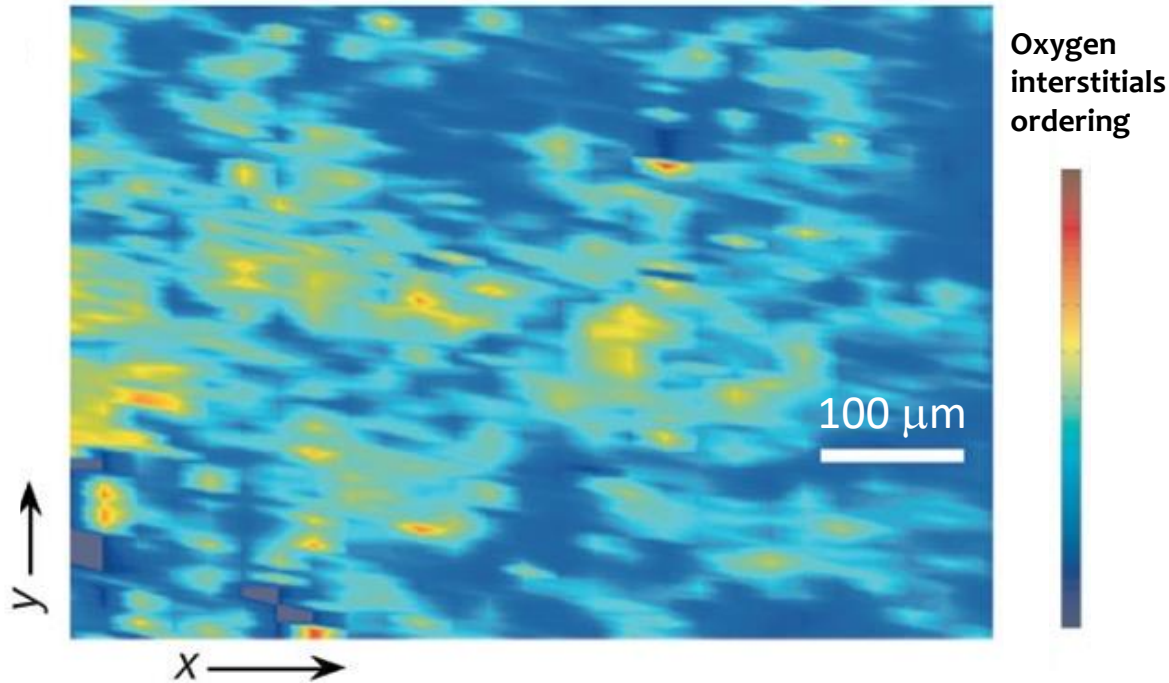


Y. Zhu, et al. *Physical Review X* 11, 031011 (2021).

Oxygen diffusion mechanisms are different in the ab-plane (activation 0.9eV) and along the c-axis (activation 2eV).  
The c-axis diffusion coefficient increases from  $10^{-17}$  to  $10^{-12} \frac{\text{cm}^2}{\text{s}}$  respectively for 400 °C and 700 °C.

M. Runde, et al. *Physical Review B* 45, 7375 (1992)

# Minimizing detrimental disorder in twisted cuprate interfaces is a 25 years long challenge.

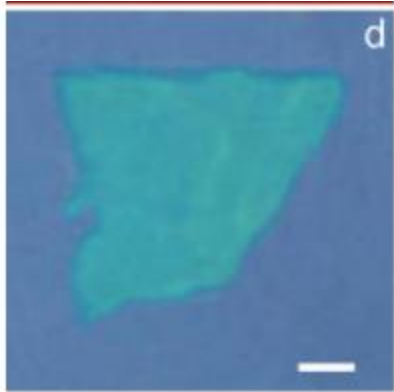


**Essential heterogeneities** are determined in cuprates by the oxygen interstitials self-organization which **can be erased at high temperatures**.

M. Fratini, NP, et al. *Nature* 466, 841-844 (2010).  
NP et al. *Nature Materials* 10, 733-736.(2011)  
NP, et al. *PNAS* 109 15685-15690 (2012).  
G. Campi, NP, et al *Nature* 525, 359-362 (2015)

**Challenge: Finding a stacking methodology which works below -73 °C .**

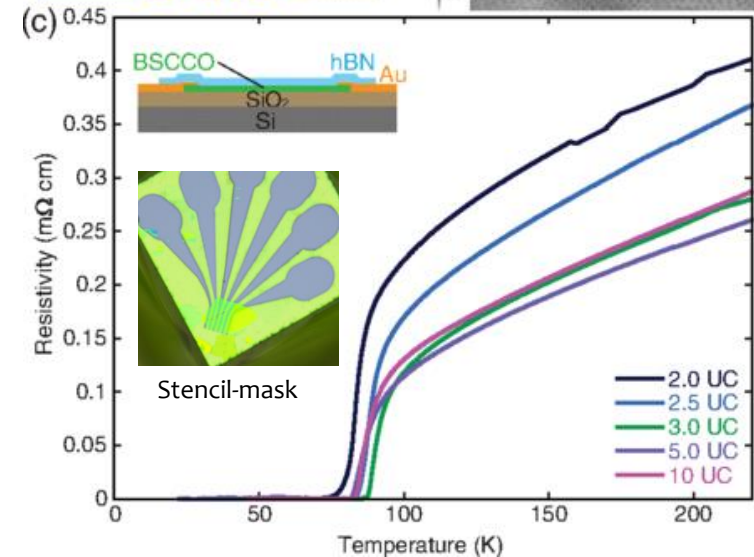
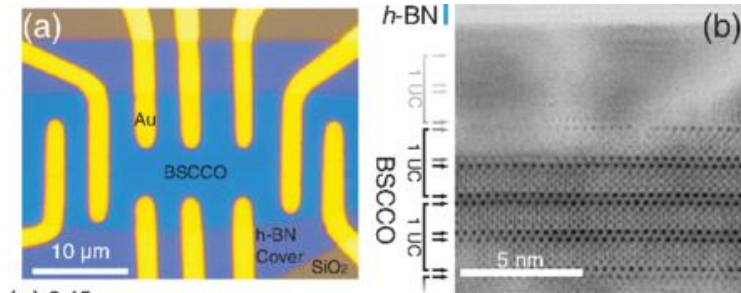
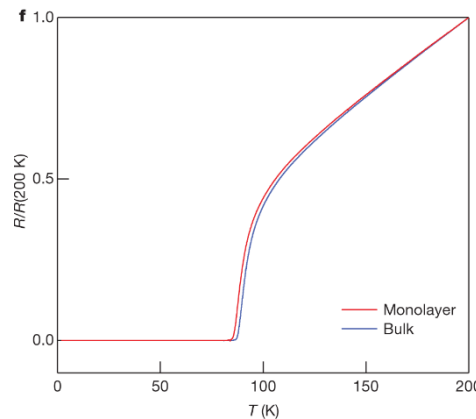
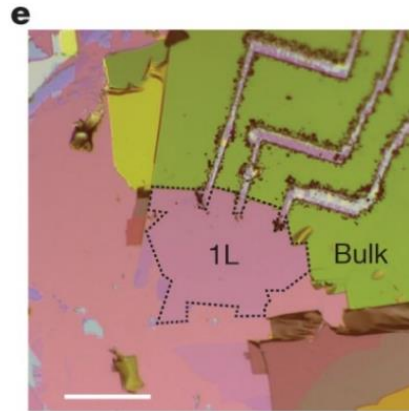
# Starting with the isolation of optimally superconducting $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ atomically thin crystals.



$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  crystals are exfoliated on a  $\text{SiO}_2$  substrate with the scotch-tape method.

Chemical reactivity turns the crystals insulating.

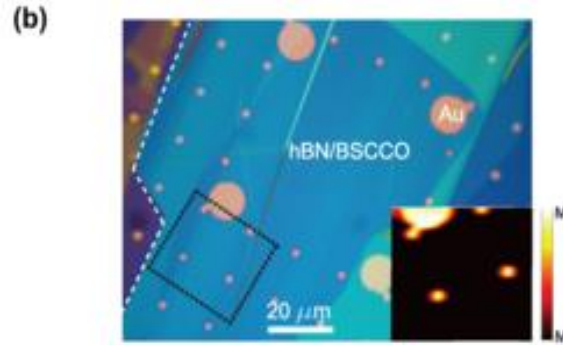
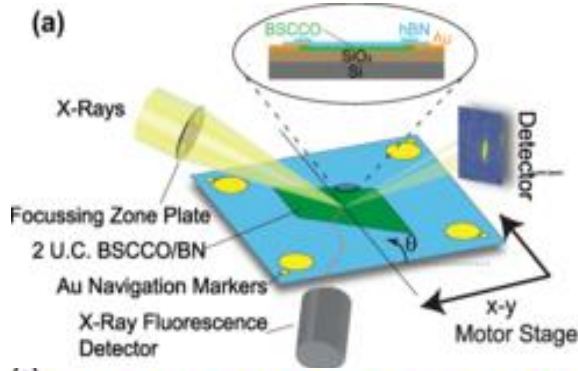
K.S. Novoselov, et al. *Proceedings of the National Academy of Sciences* 102, 10451-10453 (2005)



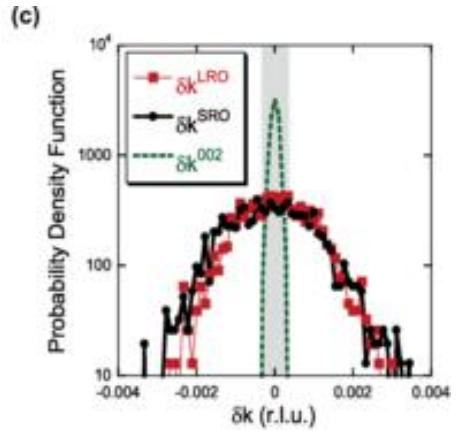
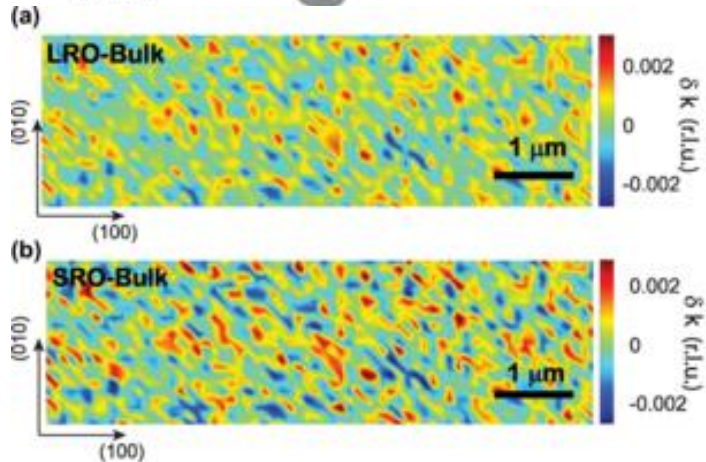
Isolation of optimally superconducting  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  atomically thin crystals achieved in 2019.

- **Heat sensitivity** during the exfoliation is overcome by cooling the  $\text{SiO}_2$  substrate at  $-40^\circ\text{C}$ .  
Y. Yu, et al. *Nature* 575 156-163 (2019)
- **Chemical reactivity** for the realization of the electrical contacts in a nanodevice is overcome by the stencil mask method  
S.Y. F. Zhao, NP, et al. *Physical review letters* 122 247001 (2019)

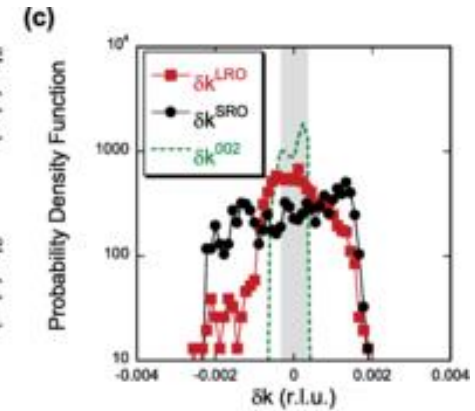
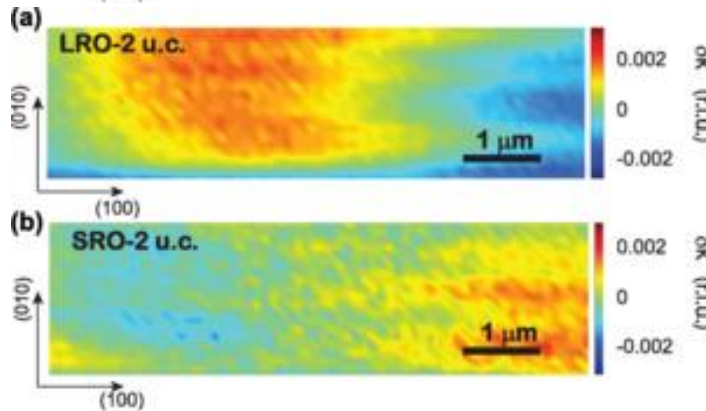
# Detrimental dislocations in optimally superconducting $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ atomically thin crystals.



- Lattice must be as frozen as possible during any fabrication involving  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  surfaces.



r-k gaussian-like disorder in the incommensurate superlattice

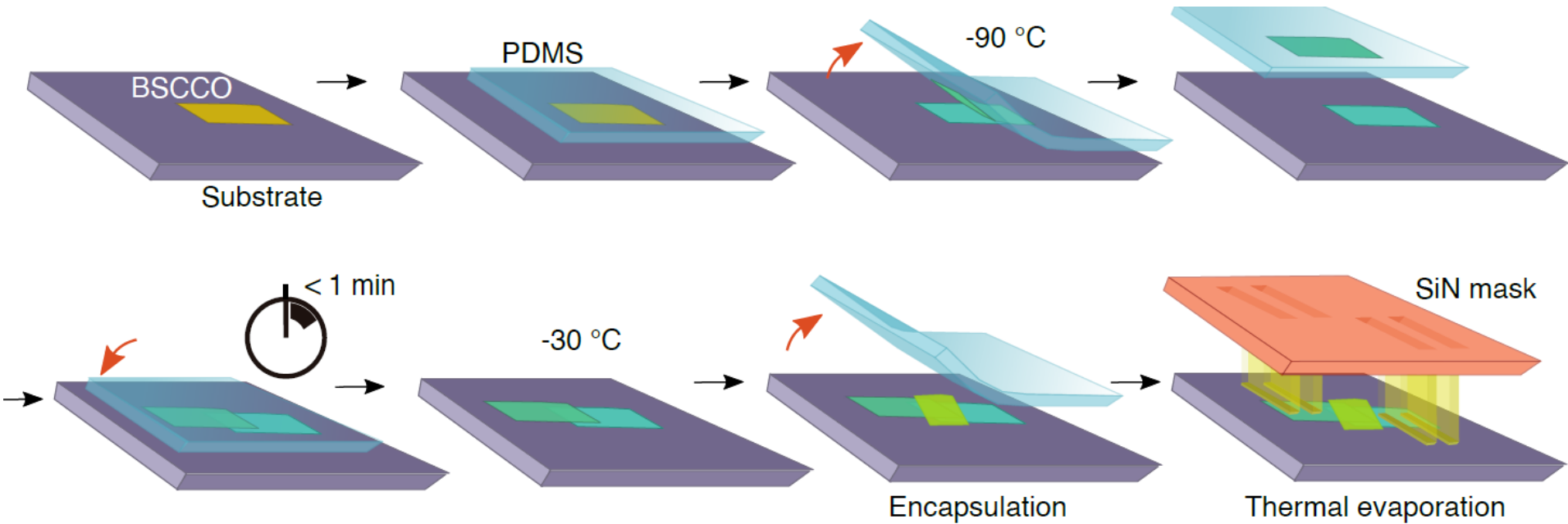


r-k non-gaussian disorder in the incommensurate superlattice





# The silicon polymer PDMS becomes stickier close to its glass transition temperature $T_g \sim -120^\circ\text{C}$



Low  $T$        $-120^\circ\text{C}$        $-73^\circ\text{C}$        $90^\circ\text{C}$        $200^\circ\text{C}$       High  $T$

$T_g$   
PDMS

Oxygen  
Mobile

$T_M$   
PPC

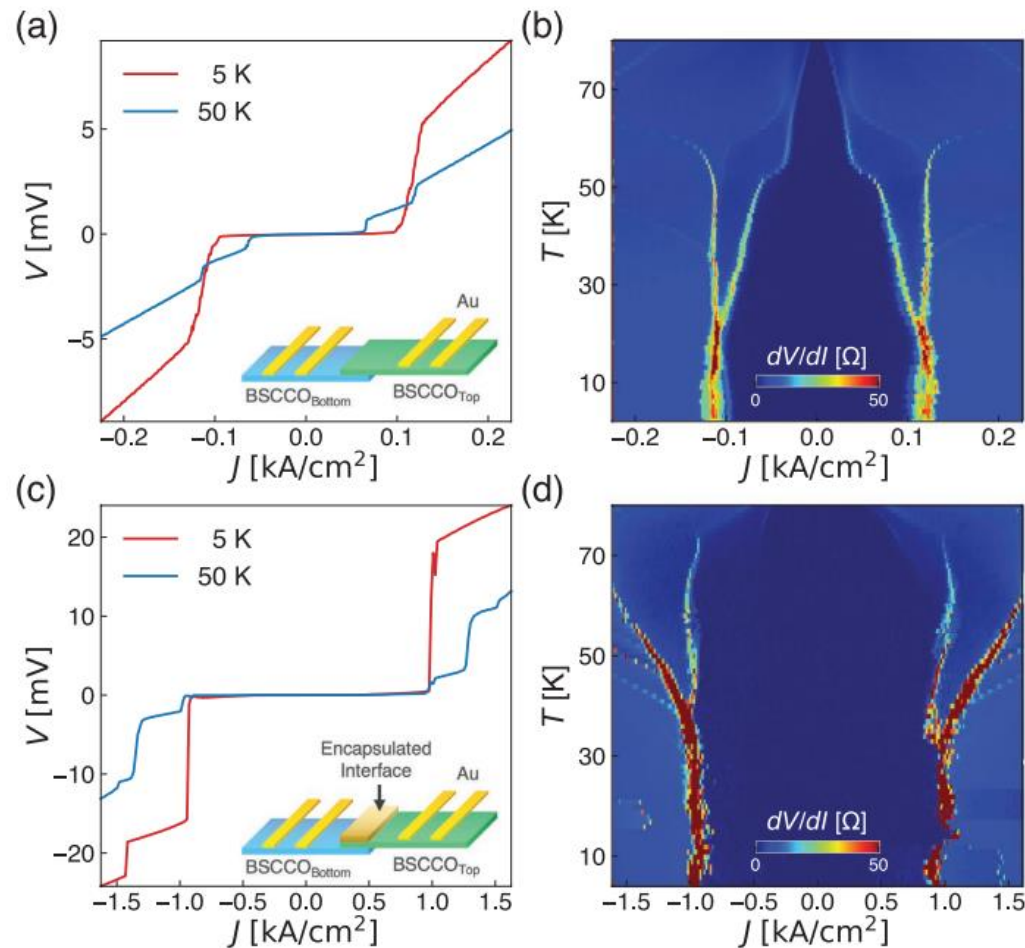
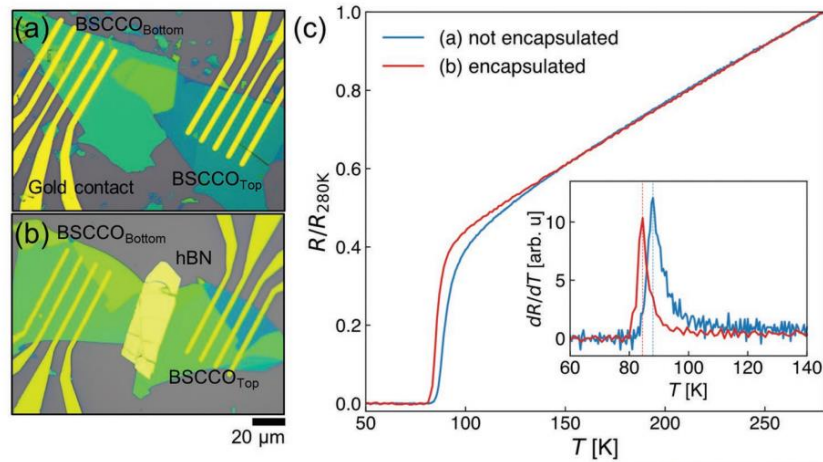
$T_M$   
PC

Interfaces are realized with a  $T_d \sim -100^\circ\text{C}$   
Measurements of  $T_d$  are done with an optical microscope.

S.Y. F. Zhao, NP et al. *Science* 382, 1422-1427 (2023)  
Y. Lee, NP et al. *Advanced Materials* 35, 2209135 (2023)  
M. Martini, NP et al. *Materials Today*, 67, 106-112 (2023)



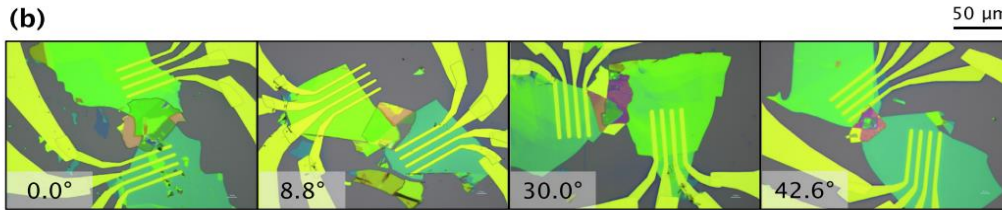
# Encapsulation protects from detrimental disorder if the electrical contacts are evaporated close to $10^{-6}$ Torr



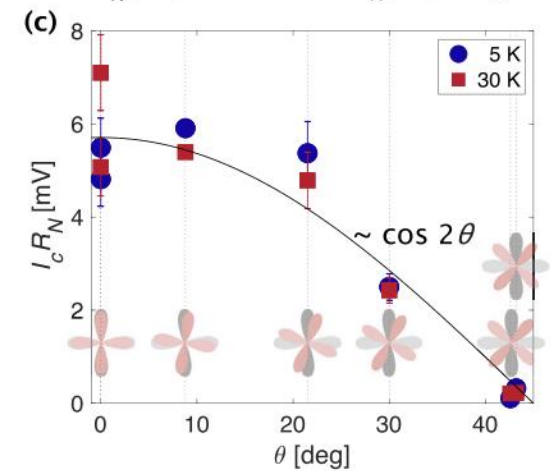
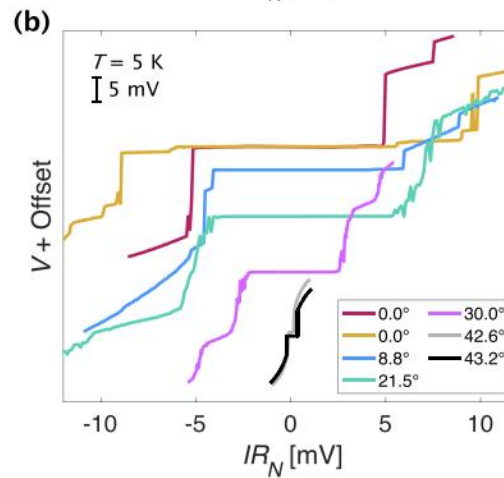
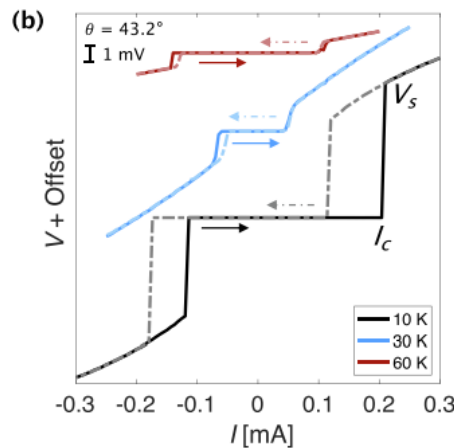
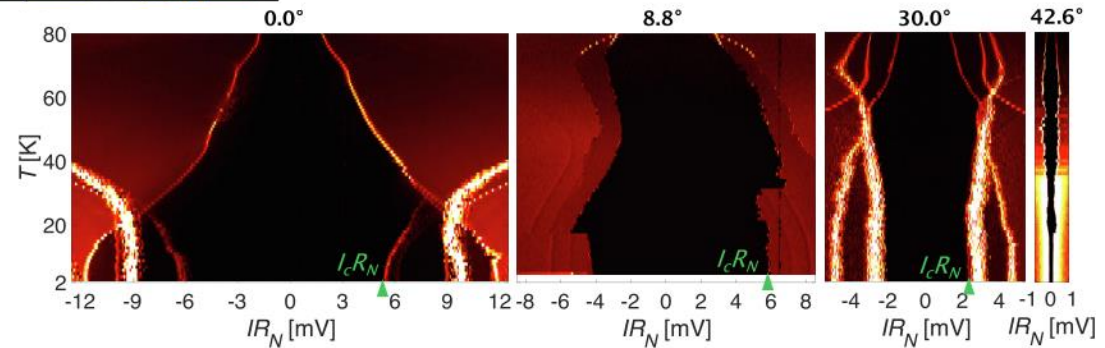
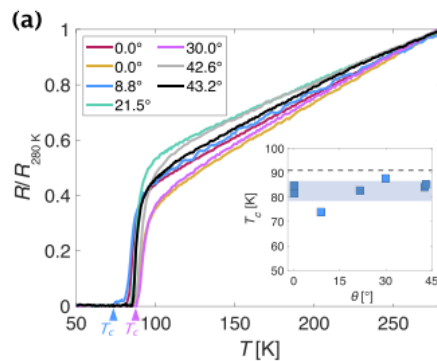
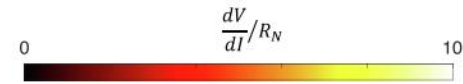
- Optimal superconducting critical transition ( $T_c \sim 90$  K) is a **requirement** for coherence in the twisted cuprate Josephson junctions, **but is not sufficient**.
- An encapsulated and not encapsulated twist junction may share the same  $T_c$ , but have very different transport properties for the same angle.

# Josephson coupling dependence with the twist in hBN encapsulated junctions

Au contacts are thermally evaporated close to  $10^{-6}$  Torr



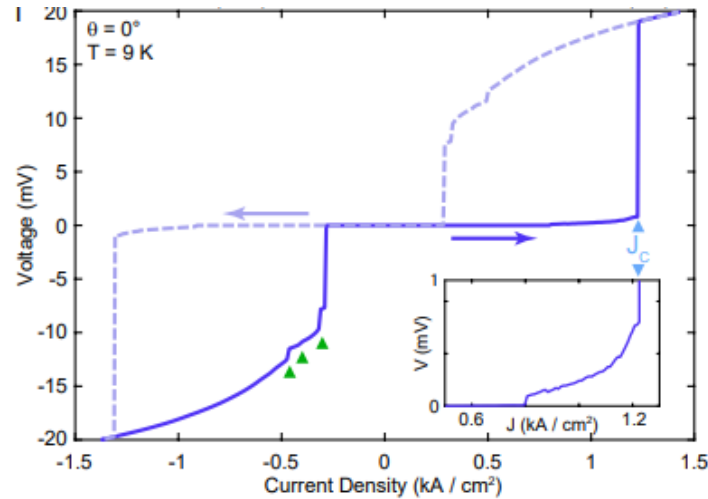
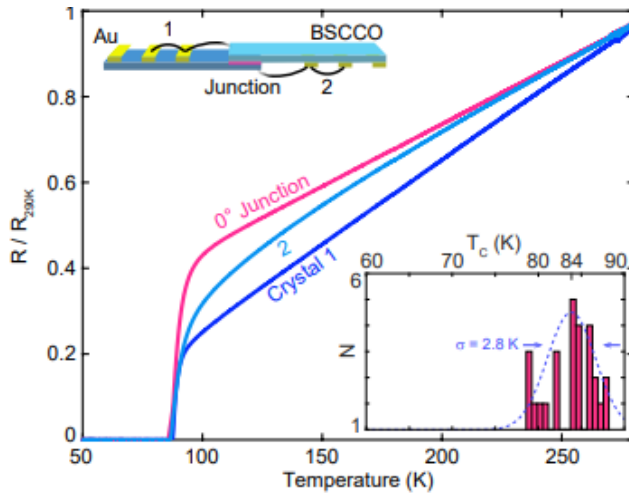
- Intrinsic Josephson junction characteristics at  $\sim 0^\circ$  twist angle  $\sim 1.2$  kA/cm<sup>2</sup>



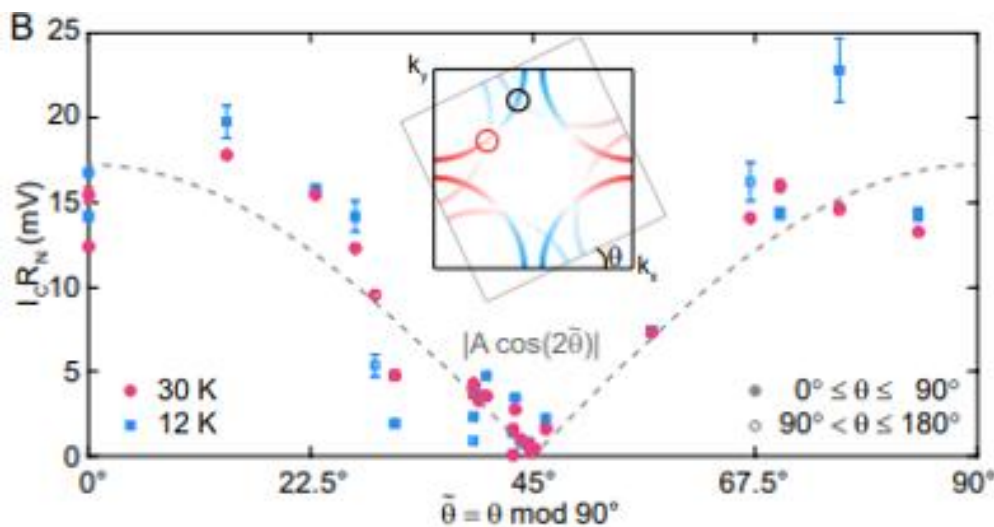
- Two order of magnitude Josephson energy change near  $45^\circ$  twist angle consistent with a d-wave angular dependence  $\sim \cos(2\theta)$ .

# Josephson coupling dependence with the twist

Au contacts are thermally evaporated close to  $10^{-8}$  Torr therefore there is no need of encapsulation.



- Intrinsic Josephson junction characteristics at  $\sim 0^\circ$  twist angle  $\sim 1.2$  kA/cm<sup>2</sup>
- Two order of magnitude Josephson energy change near  $45^\circ$  twist angle consistent with a d-wave angular dependence  $\sim \cos(2\theta)$ .



The d-wave angular dependence can be obtained with same cryogenic transfer technology and alternative methods for electrical contacts fabrication.

$I_c(\theta) \sim \cos(2\theta)$  provides evidence for incoherent tunnelling.

Theoretical details in: T. Tummuru et al., *Phys. Rev. B* **105**, 064501 (2022)

# Coherent tunnelling in d-wave in twisted Josephson junctions from the anomalous $I_c(T)$

Theoretical details in: T. Tummuru et al., *Phys. Rev. B* **105**, 064501 (2022)

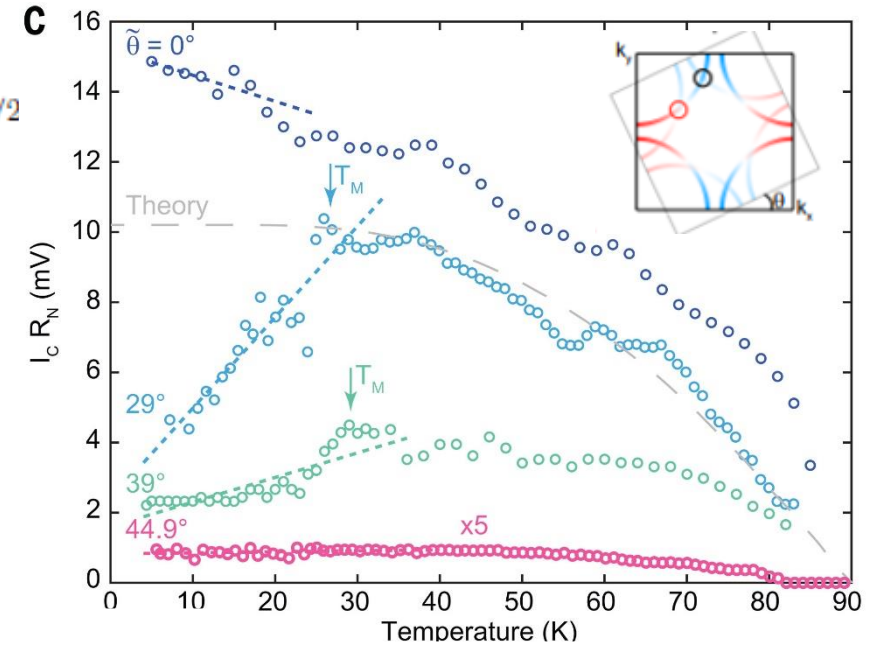
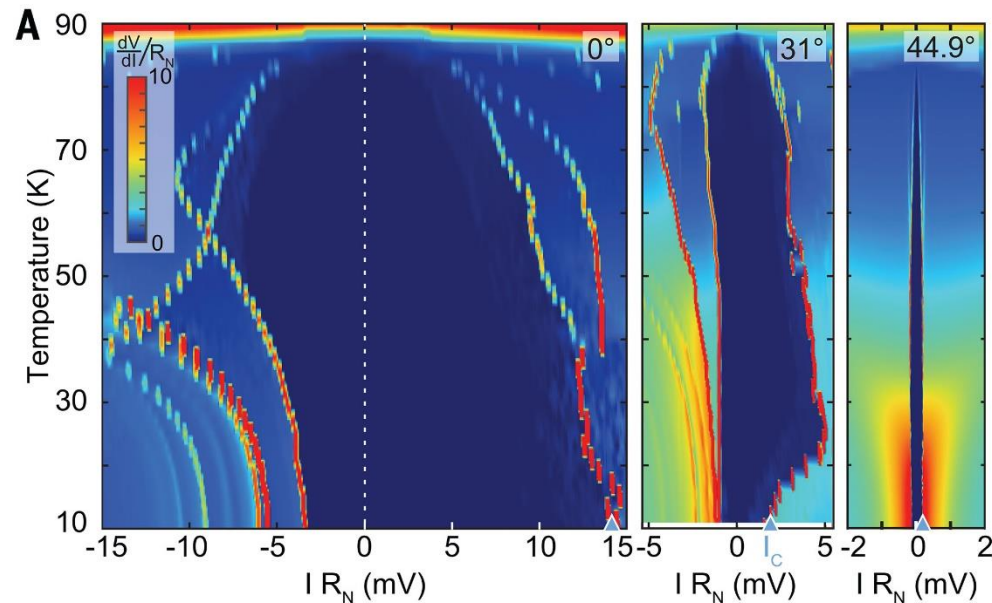
$$I_c(T) \simeq \frac{et^2}{2\hbar} \sum_k \frac{\Delta_{k1}\Delta_{k2}}{D_k(\pi/2)} \sum_{a=\pm} \left[ \frac{-a}{E_{ka}} \tanh \frac{1}{2}\beta E_{ka} \right]_{\varphi \rightarrow \pi/2}$$

$$E_{k+} > E_{k-}$$

- It is always positive by definition.

$$\Delta_{k1}\Delta_{k2} = \Delta^2 \cos(2\alpha_k + \theta) \cos(2\alpha_k - \theta)$$

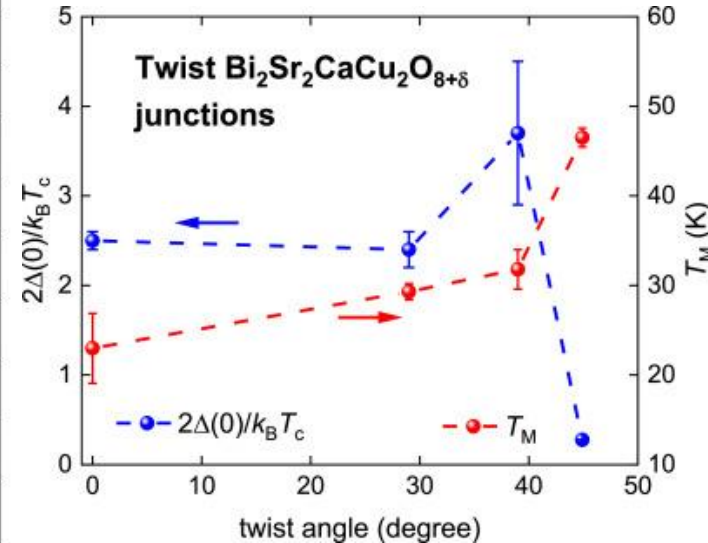
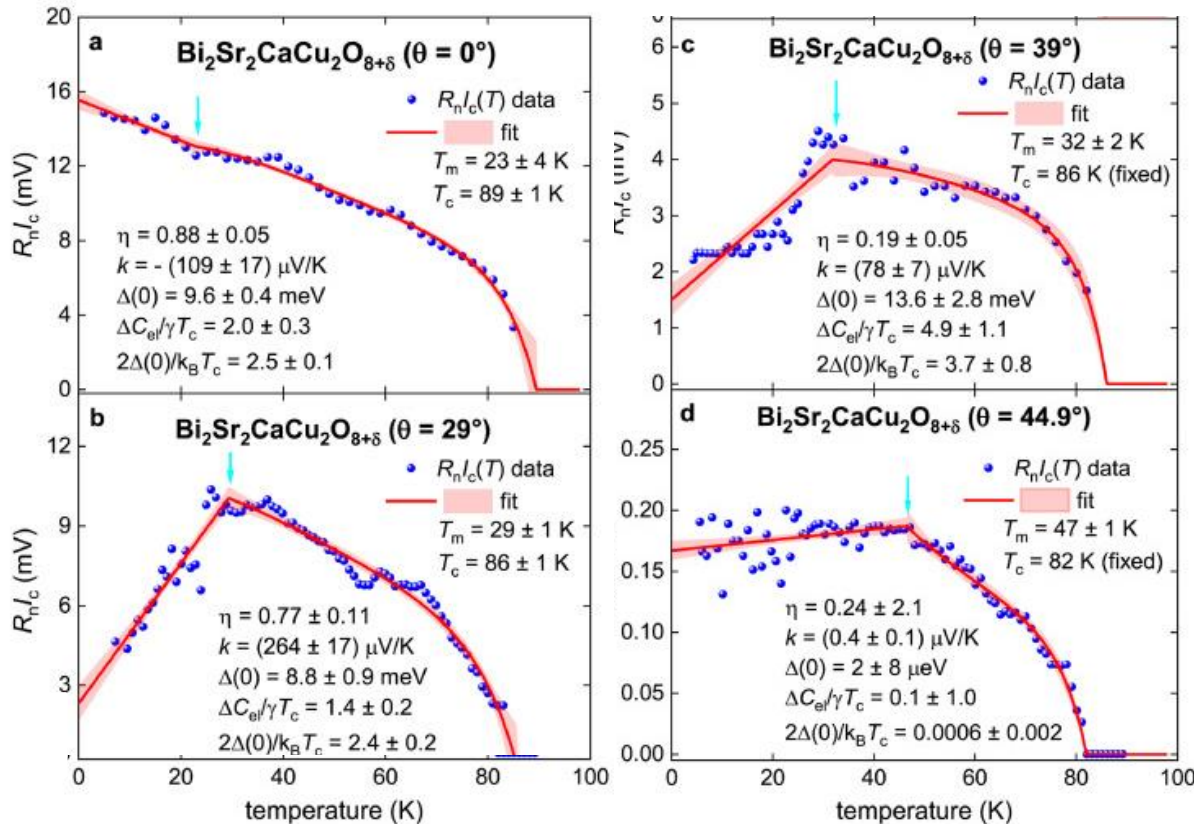
- For non-zero twist this product is negative in the vicinity of the Brillouin zone (Bz) diagonals, i.e. the nodal region of the original untwisted d-wave superconductor, and is positive in the rest of the Bz.



- In a twisted configuration nodal regions give a negative contribution to  $I_c(0)$ . Reducing this negative contribution by thermal excitations therefore produces a net increase in the total supercurrent.

D-wave coherent tunnelling is consistent with the experimental observations

# d-wave symmetry of c-axis superconducting gap



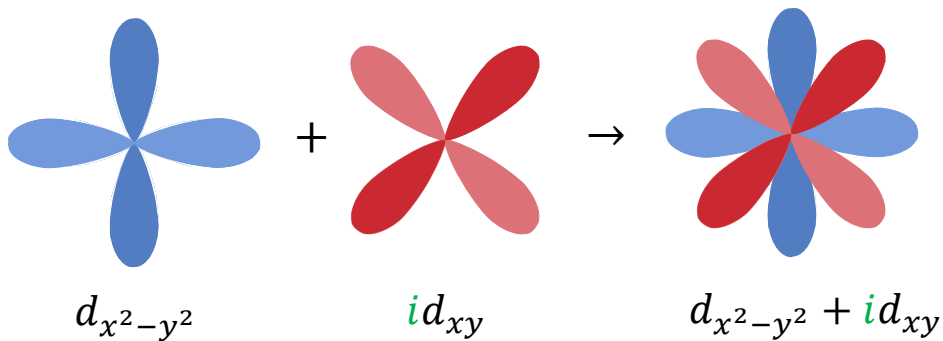
- The implementation of the more accurate gap equation (which allows variable coupling strength and gap symmetry) in the Ambegaokar-Baratoff equation provides further evidence of the d-wave symmetry for the c-axis superconducting gap in twisted Bi-2212 junctions.

# Supercurrent flowing through second order-mechanisms

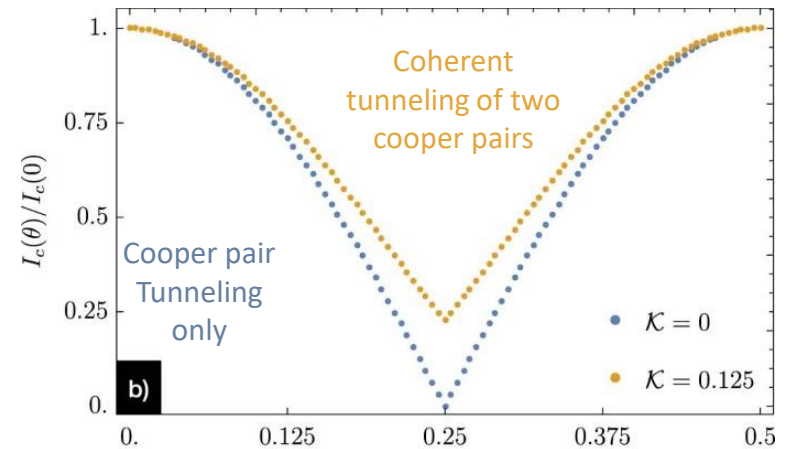
Theory predicts close to  $\theta = 45^\circ$ .

$$j(\varphi) = j_1 \sin(\varphi) + j_2 \sin(2\varphi)$$

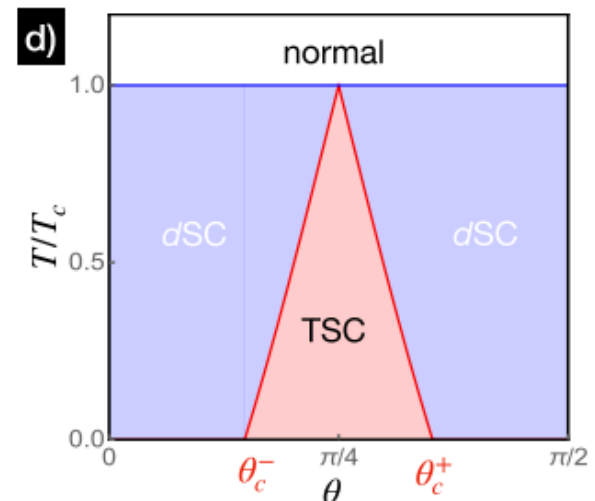
- The maximally mismatched superconducting order parameter phase eliminates the conventional direct Cooper pair tunneling term  $j_1$



- Second-order mechanisms are predicted to support an interfacial superconductivity with **doubly degenerate Josephson energy** owing to the inherent  $d_{x^2-y^2} + id_{xy}$  symmetry of the superconducting order parameter within each flake.

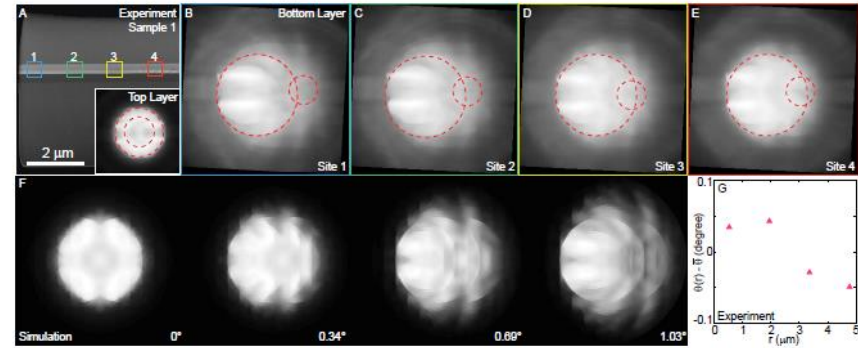
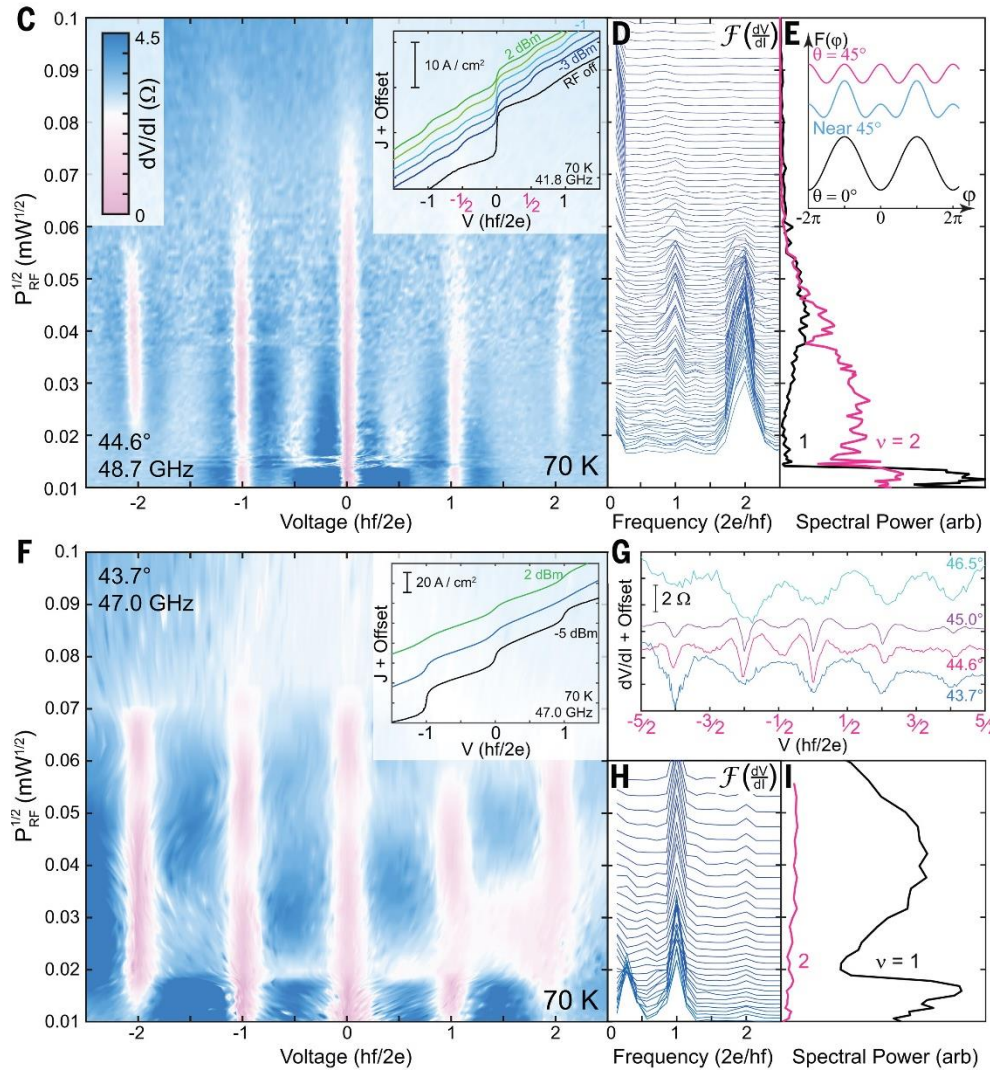


P.A. Volkov., NP et al. *arXiv preprint arXiv:2108.13456* (2021).



O. Can, et al. *Nature Physics* 17 519-524.(2021)

# Half-integer Shapiro steps emerge close to $\theta = 45^\circ$ .



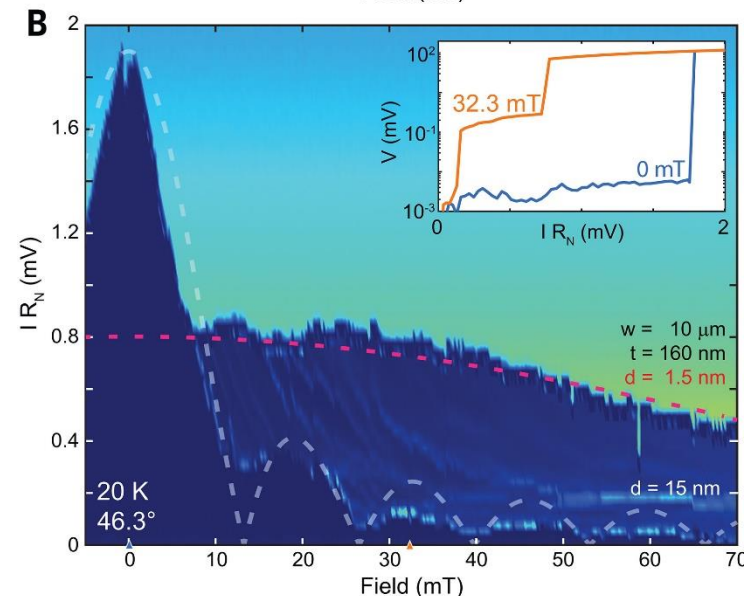
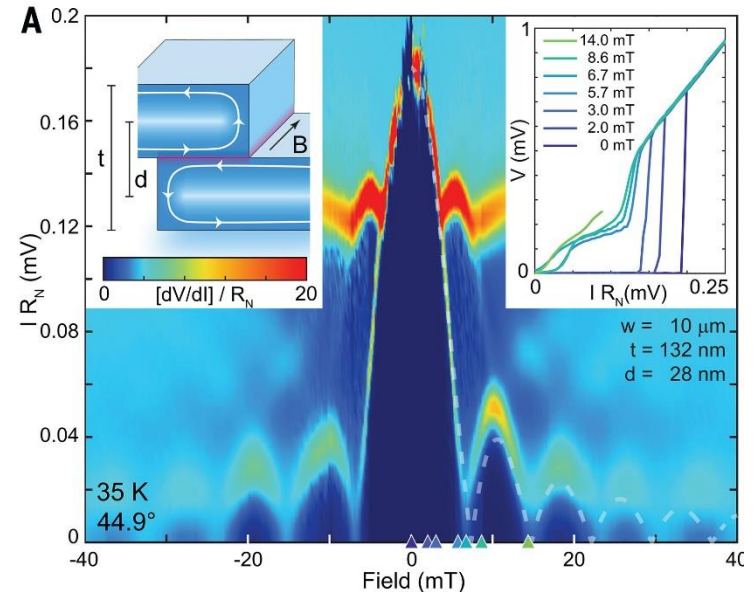
- Convergent beam electron diffraction measurement of the rotation angle  $\theta$  provides an upper bound to angular disorder at  $0.2^\circ$

Half-integer Shapiro step signatures observed in our experiments are fully consistent with a dominant second-harmonic Josephson current-phase relation

# Fraunhofer patterns near $\theta = 45^\circ$ .

- The Meissner currents (white lines) in the flakes affect the phase difference at the twist junction, enhancing its effective thickness  $d$  for the magnetic flux.
- $\theta = 44.9^\circ$  JJ with IC(B||) oscillation with a period about 20 times shorter than that expected for intrinsic junctions.
- At a nearby angle  $\theta = 46.3^\circ$ , the short-period oscillations appear to coexist with a long-period oscillation characteristic of IJJs.

$d/t \approx 0.1$  for both devices only if we assume that the  $44.9^\circ$  junction is coupled purely through the **second-order process with a doubled Fraunhofer interference pattern period.**

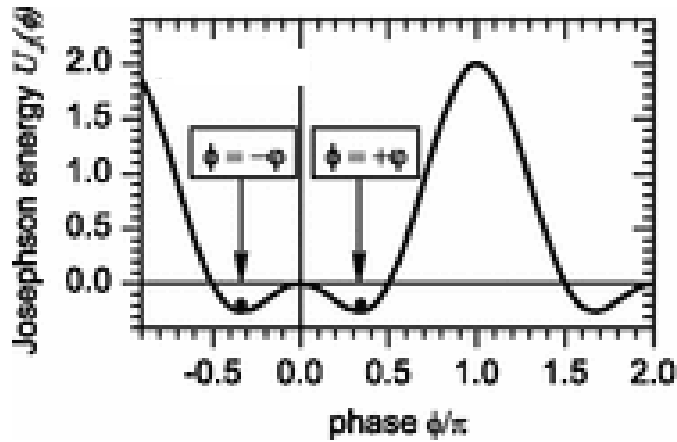




# Prepare the Time-Reversal-Symmetry broken initial $\pm\varphi_0$ states by the “training current sequence”

$$j(\varphi) = j_1 \sin(\varphi) + j_2 \sin(2\varphi); \quad J_2 > J_1$$

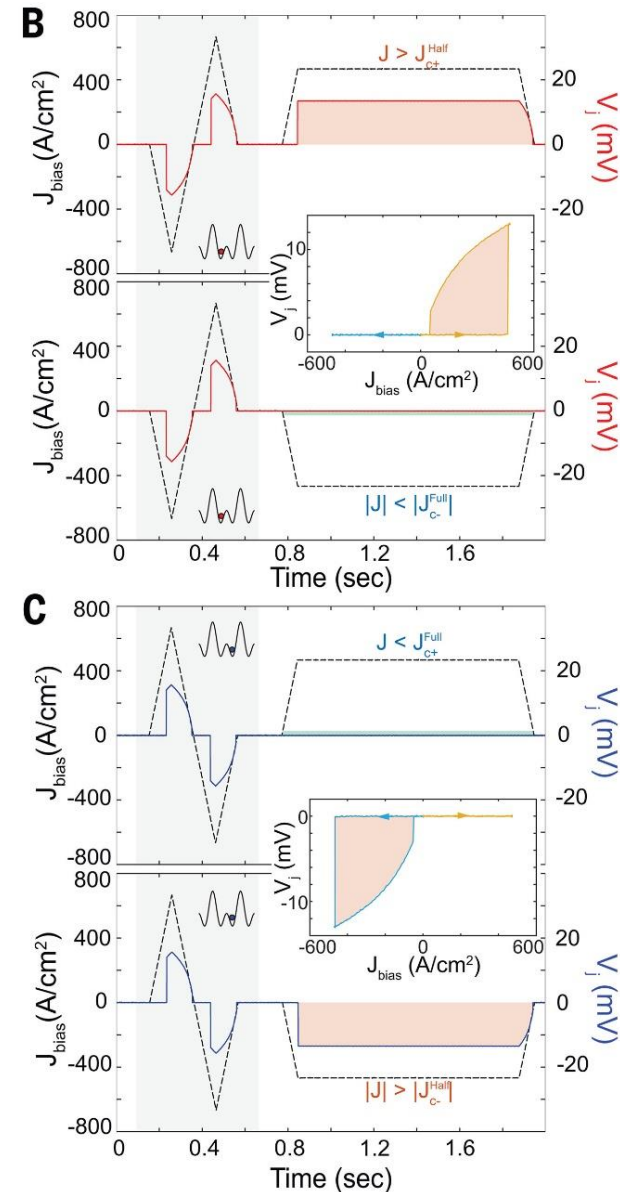
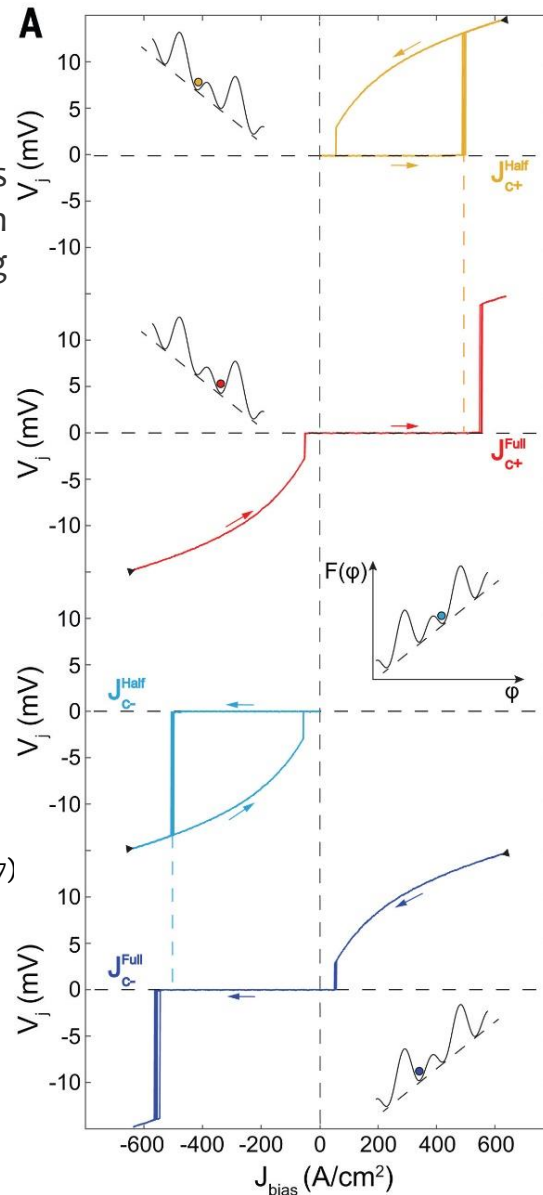
the Josephson junction ground state splits into a pair of degenerate states with complex phase angle  $\varphi = \pm\varphi_0$  lying between 0 and  $\pi$



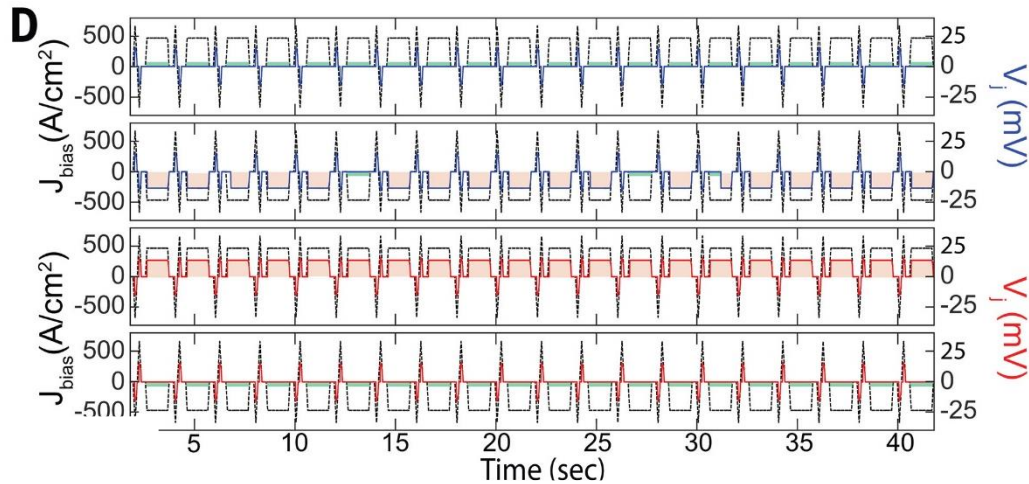
Theory details in: E. Goldobin, et al. *Phys. Rev. B* 76, 224523 (2007)

Junction I-V characteristics obtained from a JJ with  $\theta = 39.5^\circ$  at 12 K revealing four distinct  $J_C$  accessible via different current sweep directions and histories

S.Y. F. Zhao, NP et al. *Science* 382, 1422-1427 (2023)

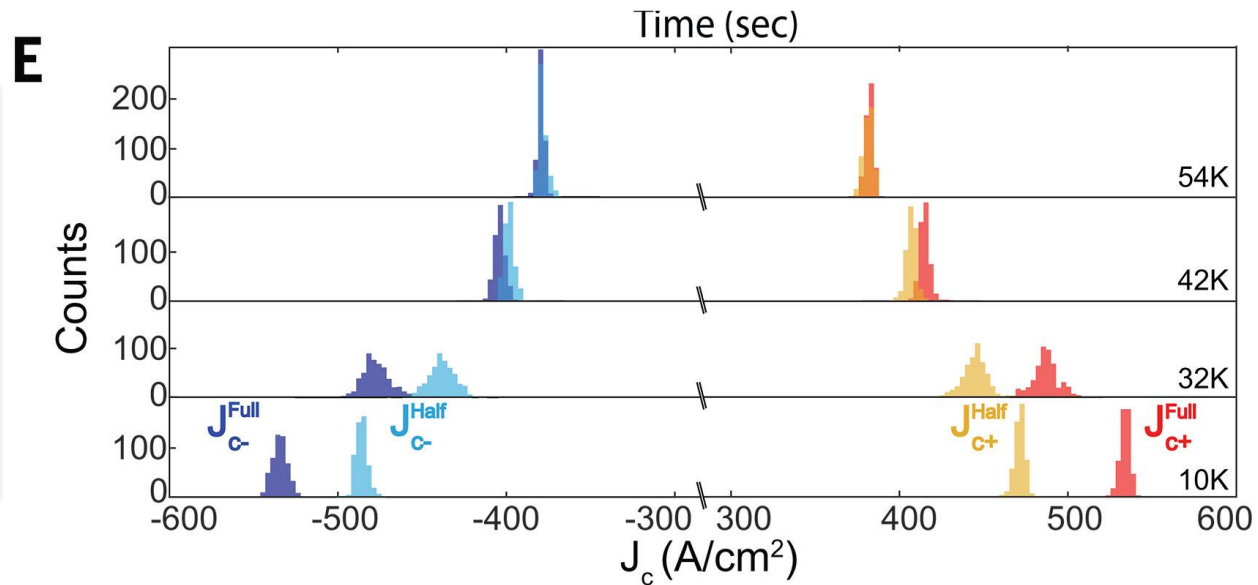


# Current-trainable Josephson diode effect



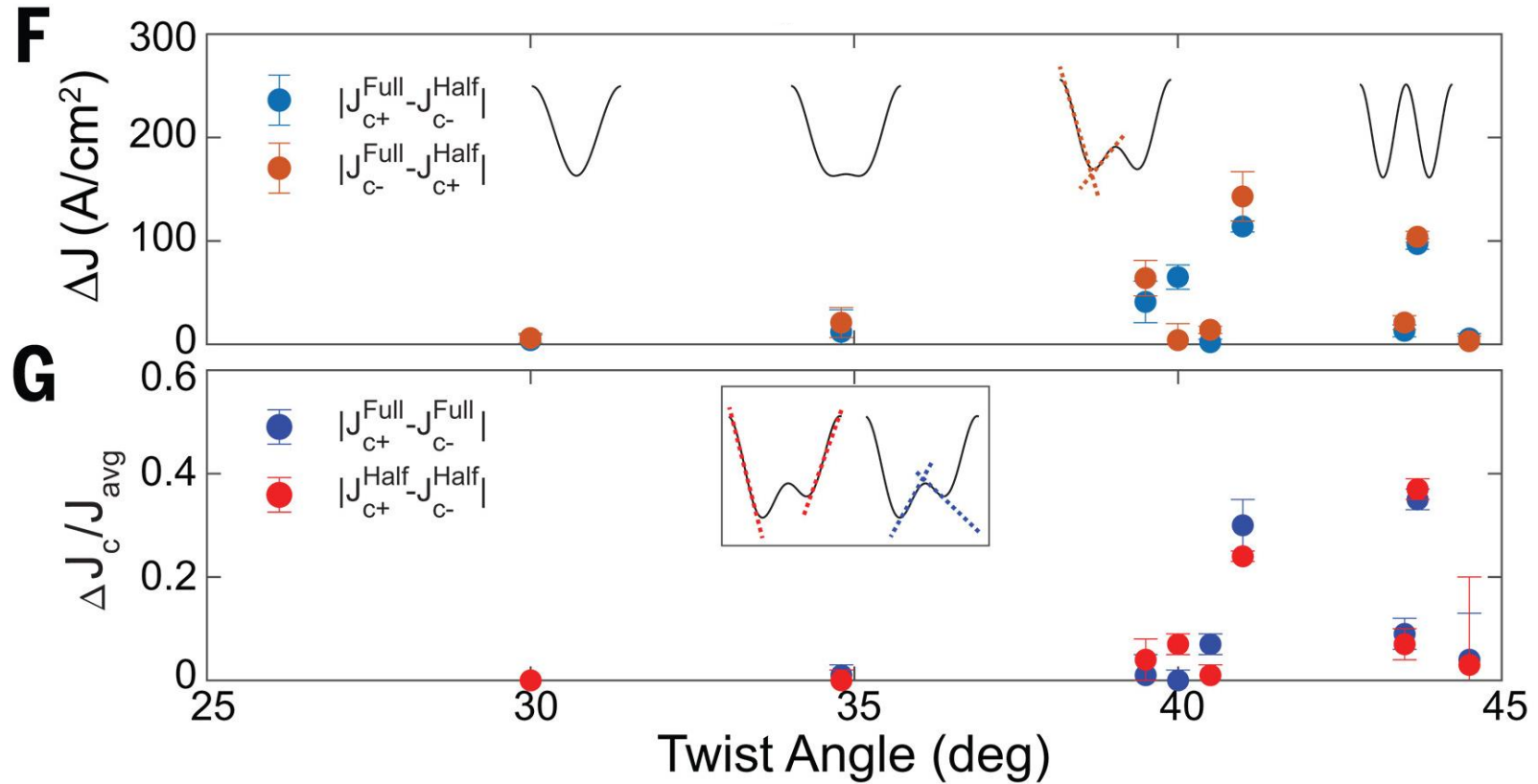
- Junction response to consecutive training and test pulses, showing controllable behavior
- Temperature-dependent switching current distributions of the four different critical currents

At higher temperature, the phase dynamics become damped, and the  $J_c$  asymmetry becomes less prominent upon heating and vanishes at about 50 K



# Time reversal symmetry breaking superconductivity between twisted cuprate superconductors

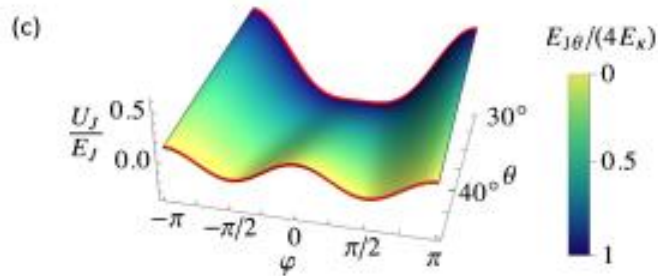
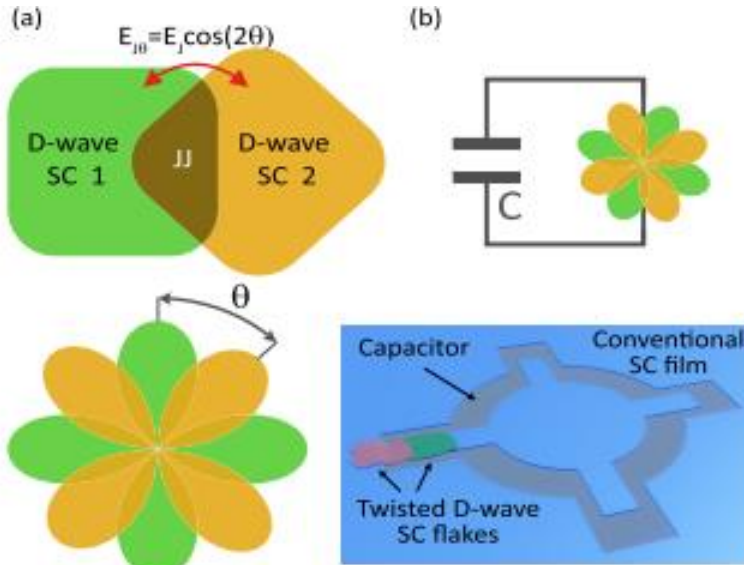
Observation of the valley asymmetry in the pair of degenerate states



At exactly  $45^\circ$ , on the other hand, the purely second-harmonic Josephson potential has an additional symmetry around  $\pm\varphi_0$ :  $\pm\varphi_0 \pm \varphi \leftrightarrow \pm\varphi_0 \mp \varphi$ , which forbids the diode effect

The observed diode effect is closely related to the free energy landscape of coexisting first- and second-harmonic Current-Phase relation terms

# Towards a new generation of macroscopic quantum bit based on twisted cuprate van der Waals heterostructures



- Hamiltonian for a s-wave junction circuit

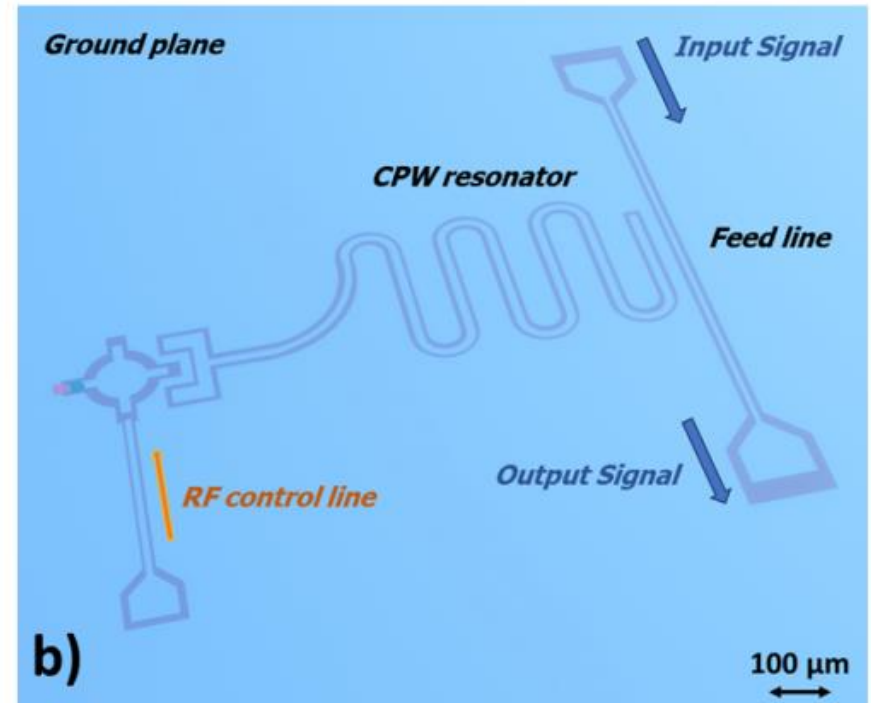
$$H_J = -E_J \cos(\hat{\varphi}),$$

- Hamiltonian of the **flowermon** circuit

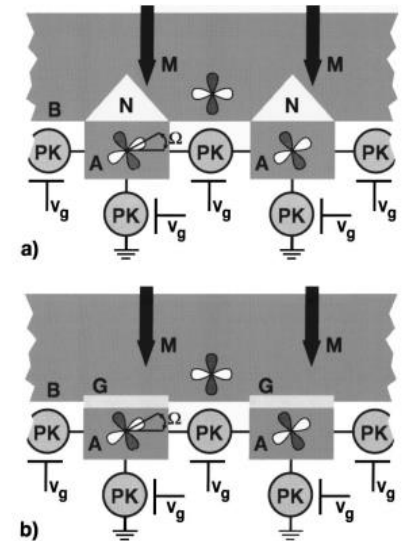
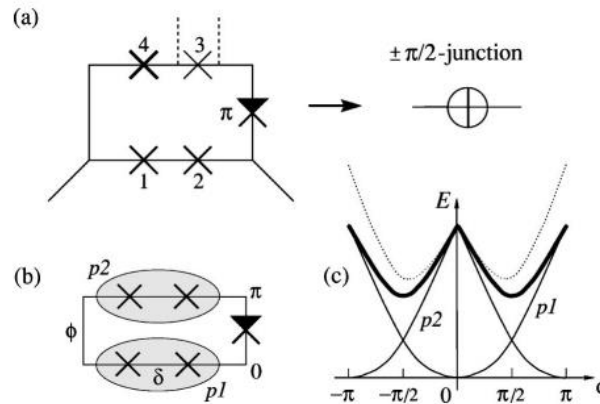
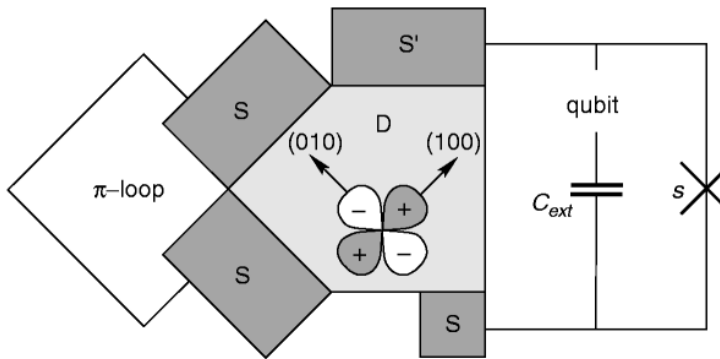
$$H = 4E_C(\hat{n} - n_g)^2 - E_{J\theta} \cos(\hat{\varphi}) + E_K \cos(2\hat{\varphi}),$$

Cuprate twisted junctions to realize a capacitively shunted qubit that we call **flowermon**. The d-wave nature of the order parameter endows the flowermon with inherent protection against charge-noise-induced relaxation and quasiparticle-induced dissipation.

V. Brosco, NP, et al. Physical Review Letters 132, 017003 (2024)



# Pioneering theoretical and experimental works on cuprates qubits

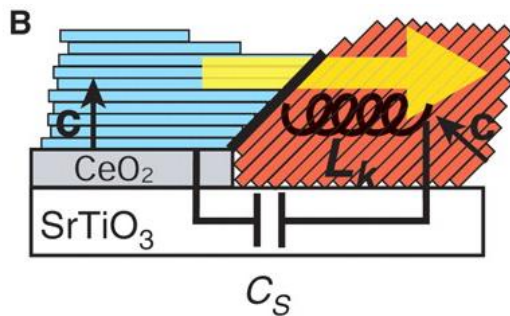


- Proposals for the suppression of tunneling in d-wave based Josephson junctions, based on momentum mismatch, to realize superconducting qubits with an enhanced coherence

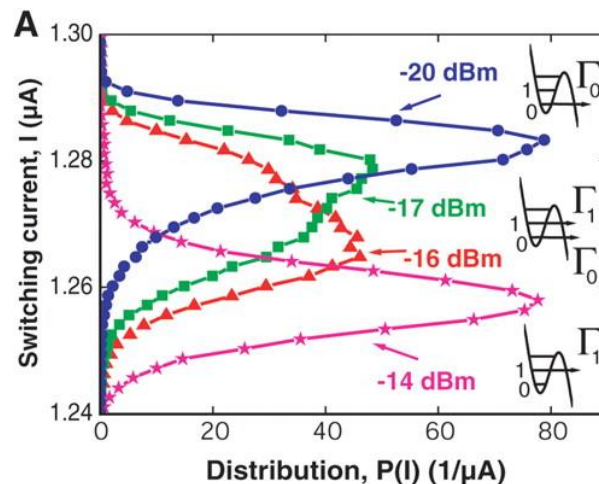
L. B., Ioffe, et al. *Nature* 398 679-681 (1999)

G. Blatter, et al. *Phys. Rev. B* 63, 174511 (2001)

- Two d-wave qubits coupled are shown  
A. Blais, et al. *Phys. Rev. A* 61, 042308 (2000)



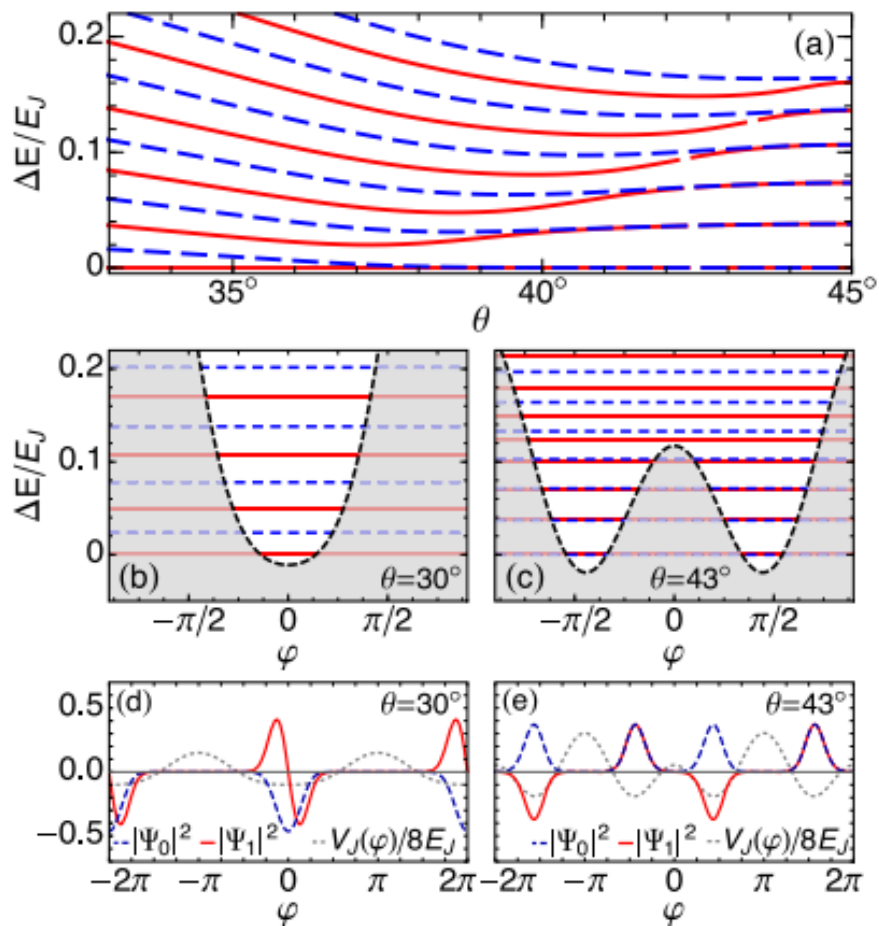
T. Bauch, et al., *Science* 311, 57 (2006)



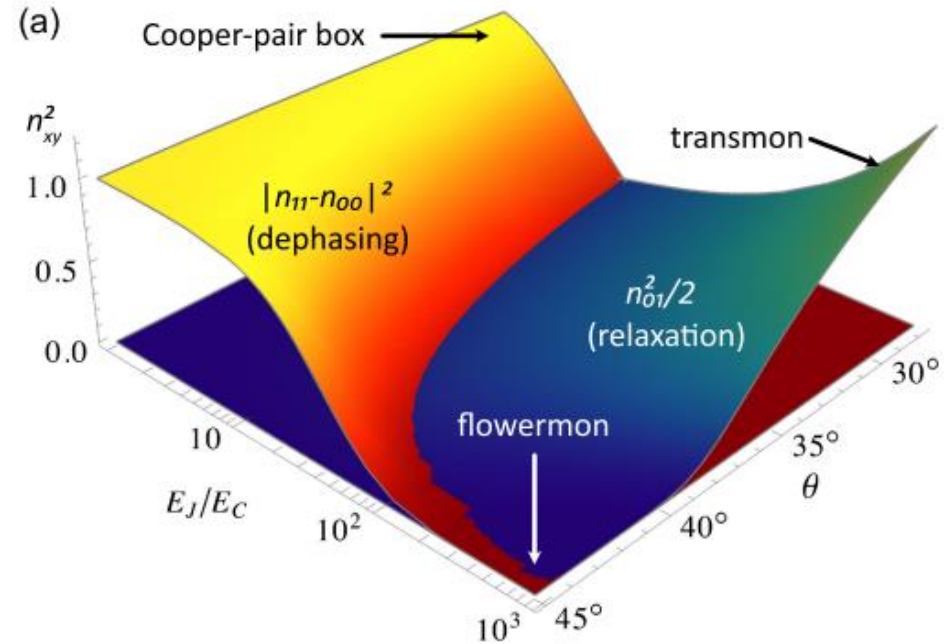
- Experimental early realization of momentum mismatch with grain boundary  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  indicated the degeneracy of the ground state.

**Detrimental disorder and fabrication complexity** hindered the realization of the qubit based on grain boundary junctions.

# New class of hybrid devices which combine the benefits of quantum materials and coherent quantum circuits



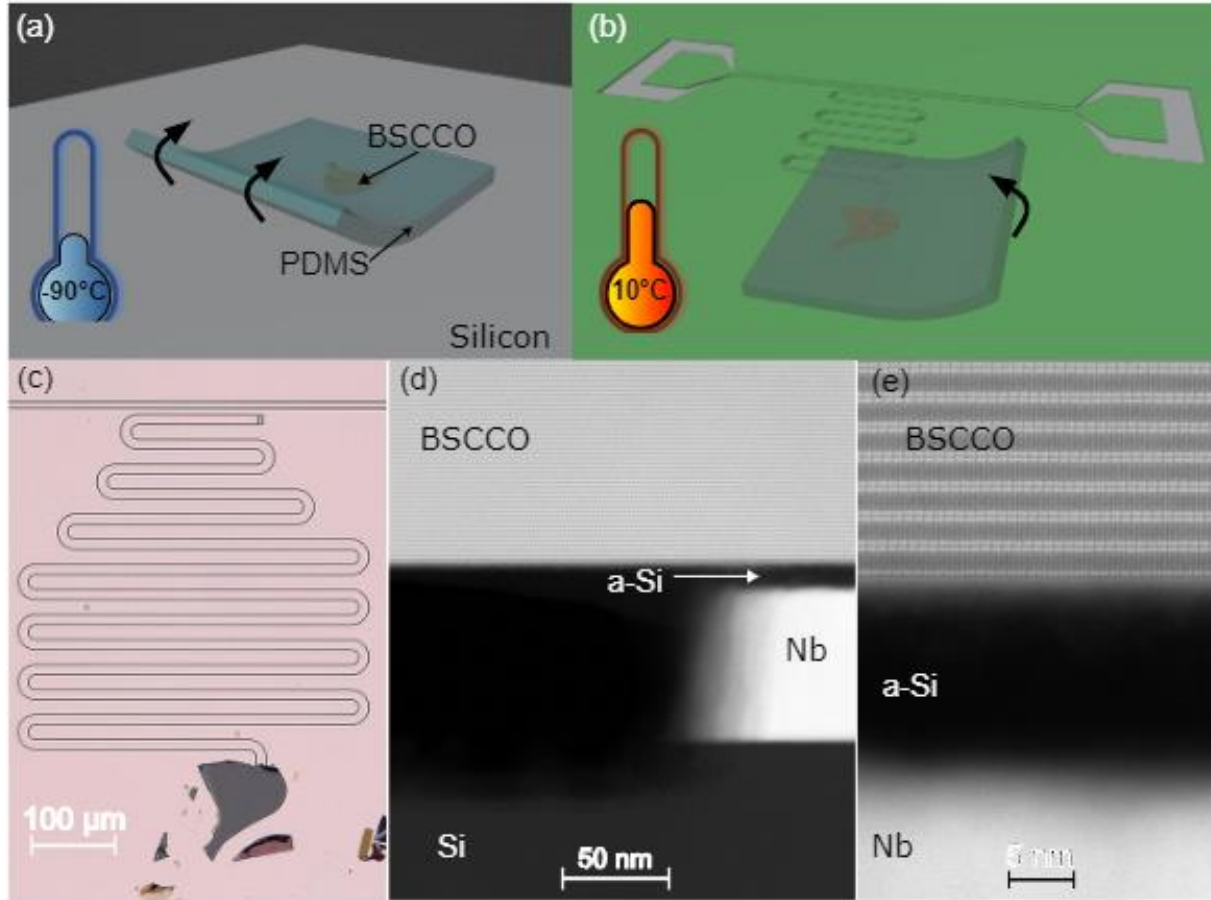
Flowermon low-energy spectrum. (a) Energy levels of the flowermon qubit as a function of the angle  $\theta$ . Even and odd levels are shown in red and blue lines respectively



$$\Gamma_{\downarrow,qp} = t^2 |\langle \psi_0 | \sin(\varphi/2) | \psi_1 \rangle|^2 S_{qp}^\theta(\omega_{01}),$$

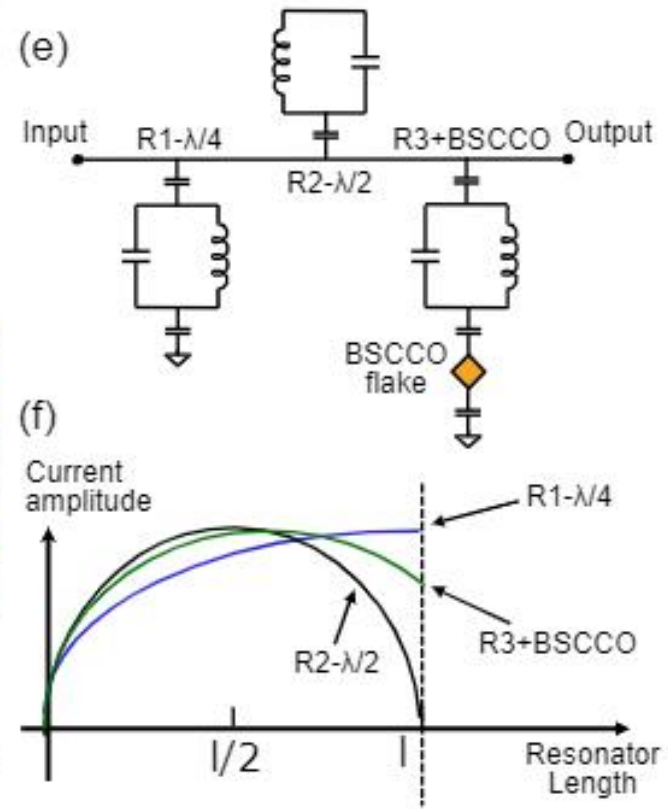
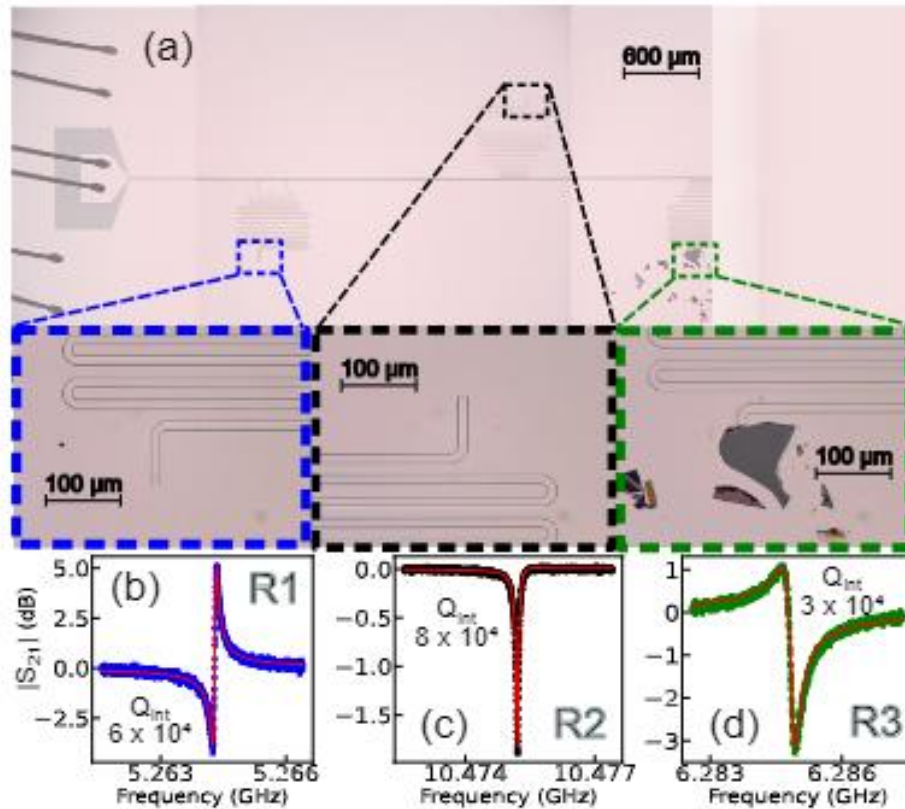
At twisting angles close to  $45^\circ$ , the flowermon shows exponential suppression of the charge-induced noise, as well as protection from nodal quasiparticle tunneling.

# Exploring van der Waals cuprate superconductor using hybrid microwave resonator



Advanced cryogenic stacking technology can be used to integrate fragile complex van der Waals cuprate architectures in superconducting circuits and explore their coupling!

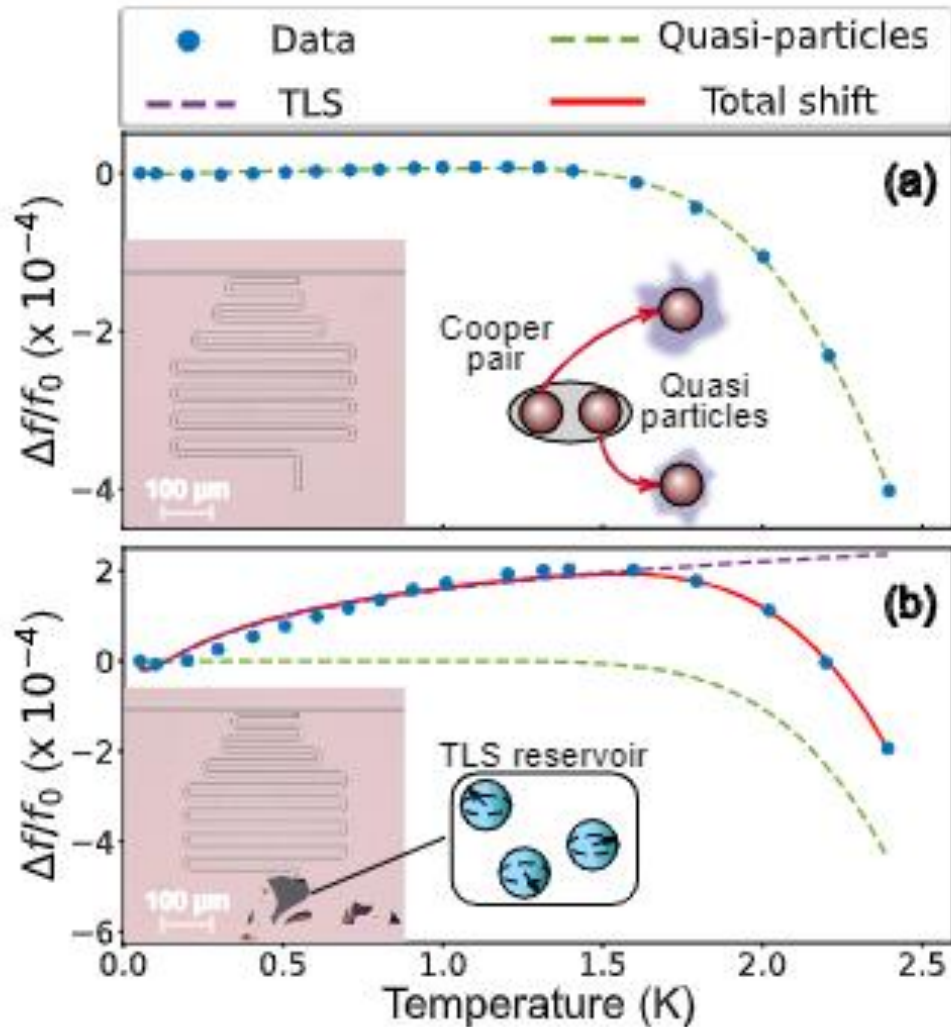
# Measuring the coupling



Low-power transmission spectrum of the resonator R1 (left, blue), R2 (middle, black) and R3 (with BSCCO flake, right, green) and their internal quality factors at 50 mK.



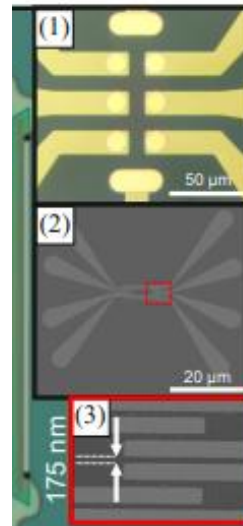
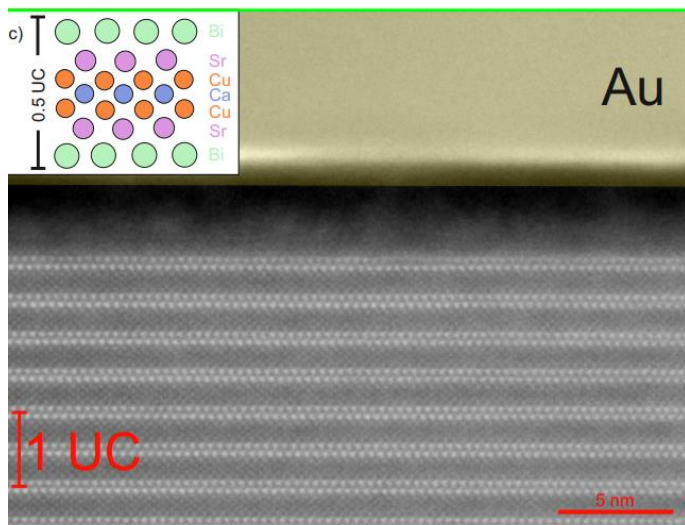
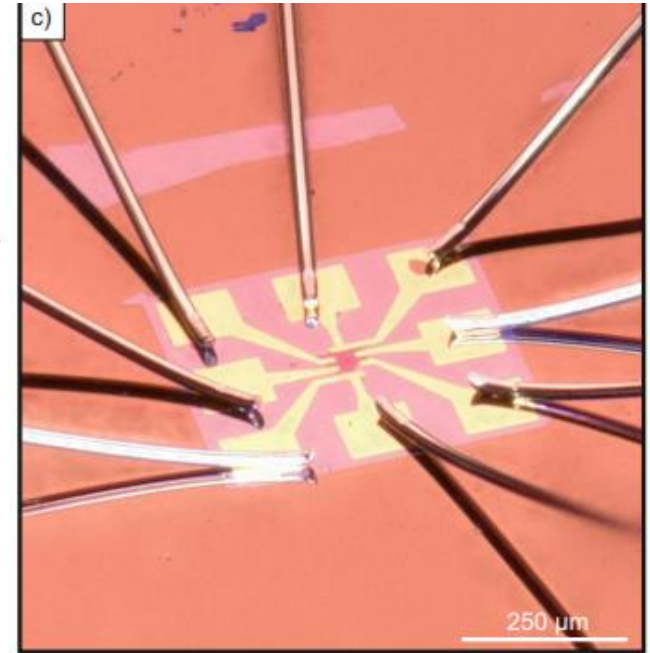
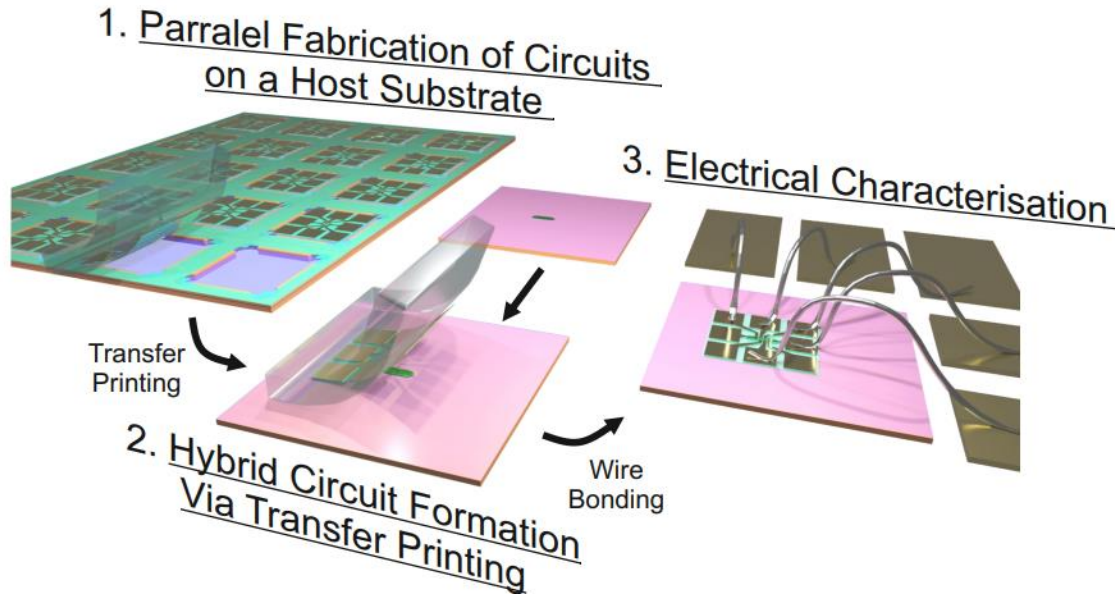
# Temperature dependence of the resonance frequency.



The green dashed line is a fit to a shift due to the thermal generation of quasiparticles, while the purple dashed line is a fit to coupling with a TLS reservoir.

# Next generation: Silicon nitride technology for micro-printed circuits

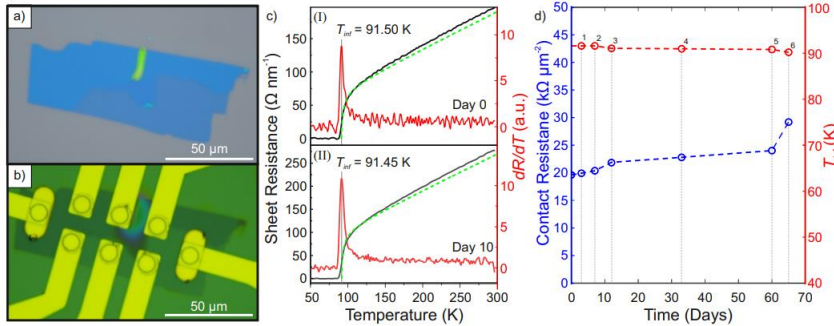
Leading the integration of complex circuits into 2d twisted cuprate junctions



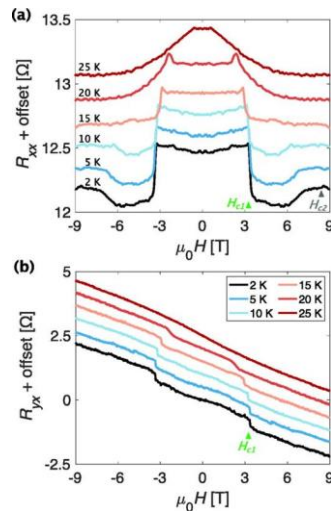
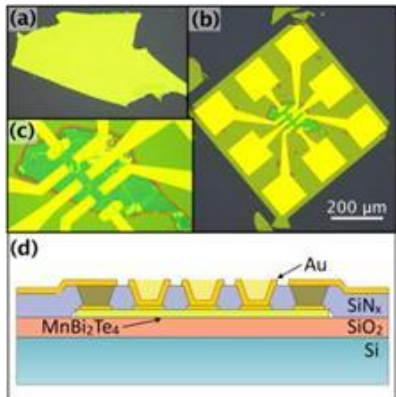
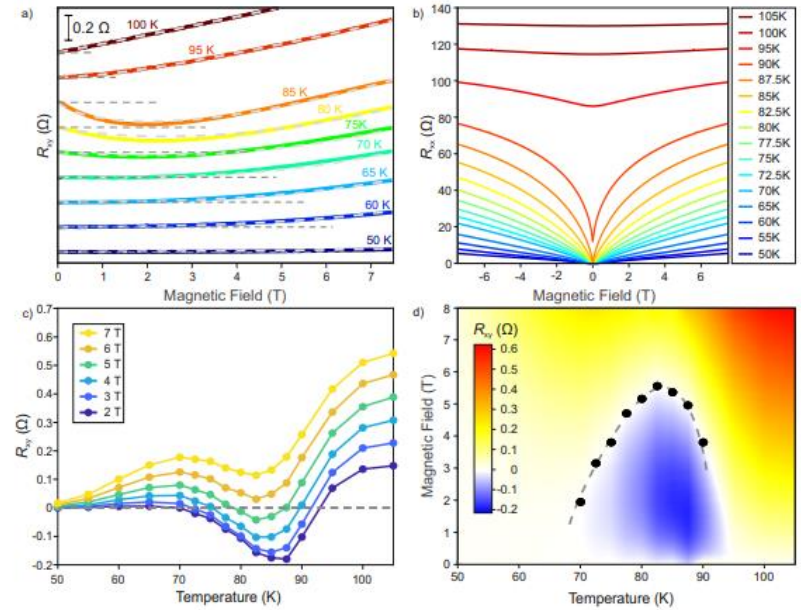
Cryogenically transferred circuit boards **entirely separates the circuit fabrication** stage requiring chemically reactive substances and ionizing physical processes from the creation of the thin superconducting structures.

# Testing the silicon nitride micro printed circuits boards

The results show optimal contact resistance, versatility in the choice of materials, nanometerprecision and long-stability



- The technology preserves the superconducting properties of the BSCCO crystal and that the membrane acts as an encapsulation layer, blocking the detrimental effects of disorder over a long time.
- The robustness of the membrane is demonstrated by the numerous thermal cycles performed for our experiments



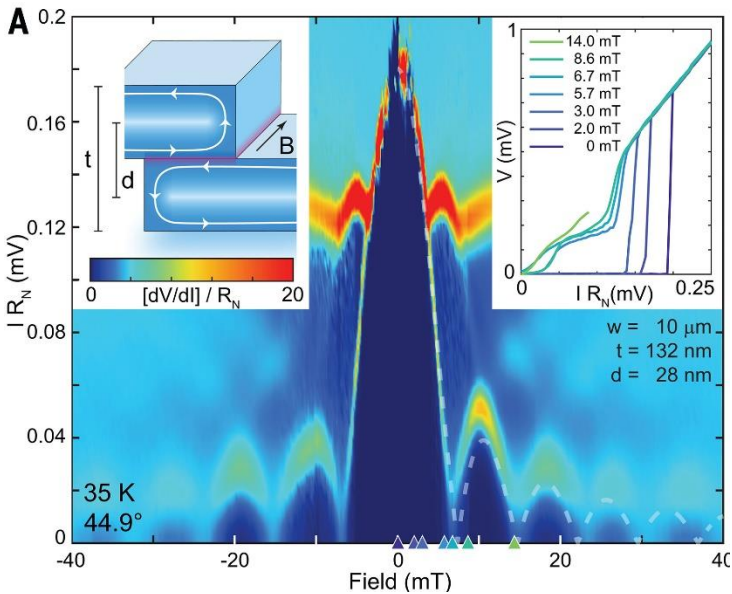
The technological solution will be implemented for integrating quantum electrodynamical circuits for the next generation of nanodevices with the **goal to reduce maximally the detrimental disorder in quantum materials.**

C.N.Saggau, NP., et al. *ACS Applied Materials & Interfaces* 15 51558-51564 (2023).

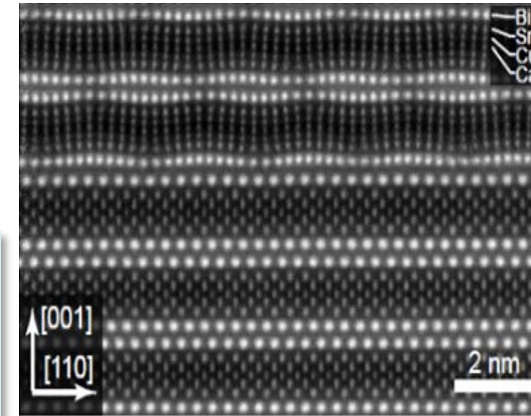
M. Martini, NP et al. *Applied Physics Letters* 123 (2023).

# Conclusion. Artificial 2D cuprate heterostructures for:

Engineering **complexity**, **topological** and **strongly correlated physics** in **one system** and in a **wide temperature range**



Advancing methodologies for the fabrication of **artificial 2D complex quantum materials**



Producing a **new generation of quantum circuits** harnessing the unique quantum complexity created by the geometry.

