Superconductivity in Two Dimensional atomic quasicrystal

Philip Kim
Department of Physics, Harvard University

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Sangmin Lee

Lucy Nathwani

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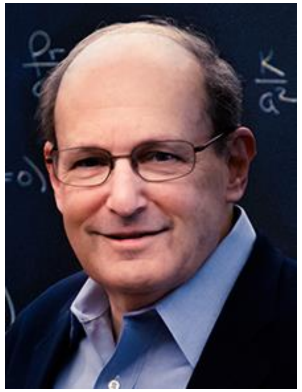
(University of Marburg)



Paul Steinhardt
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Alex Cui

Talieh Ghiasi
(now at Delft)

Jonathan Zauberman

Nadine Leisgang
(now at University Marburg)

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National Research Foundation of Korea

Quasi-periodic & Quasicrystals

VOLUME 53, NUMBER 20 PHYSICAL REVIEW LETTERS 12 NOVEMBER 1984

Metallic Phase with Long-Range Orientational Order and No Translational Symmetry

D. Shechtman and I. Blech
Department of Materials Engineering, Israel Institute of Technology–Technion, 3200 Haifa, Israel

and

D. Gratias
Centre d'Etudes de Chimie Métallurgique, Centre National de la Recherche Scientifique, F-94400 Vitry, France

and

J. W. Cahn
Center for Materials Science, National Bureau of Standards, Gaithersburg, Maryland 20760
(Received 9 October 1984)

VOLUME 53, NUMBER 26 PHYSICAL REVIEW LETTERS 24 DECEMBER 1984

Quasicrystals: A New Class of Ordered Structures

Dov Levine
Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania 19104

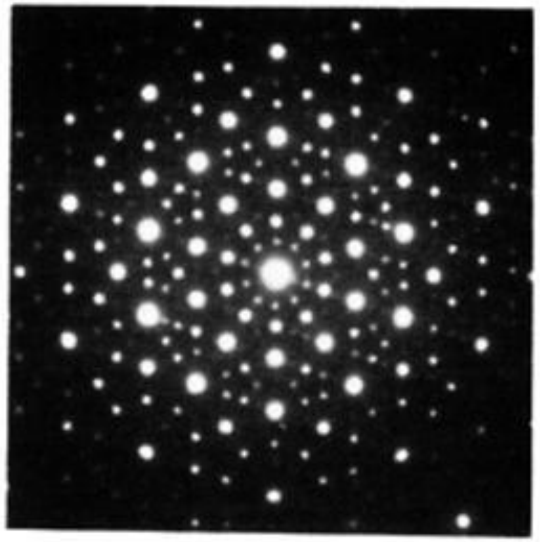
and

Paul Joseph Steinhardt
Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania 19104, and IBM Thomas J. Watson Research Laboratory, Yorktown Heights, New York 10598
(Received 2 November 1984)

A quasicrystal is the natural extension of the notion of a crystal to structures with *quasi-periodic*, rather than periodic, translational order. We classify two- and three-dimensional quasicrystals by their symmetry under rotation and show that many disallowed crystal symmetries are allowed quasicrystal symmetries. We analytically compute the diffraction pattern of an ideal quasicrystal and show that the recently observed electron-diffraction pattern of an Al-Mn alloy is closely related to that of an icosahedral quasicrystal.

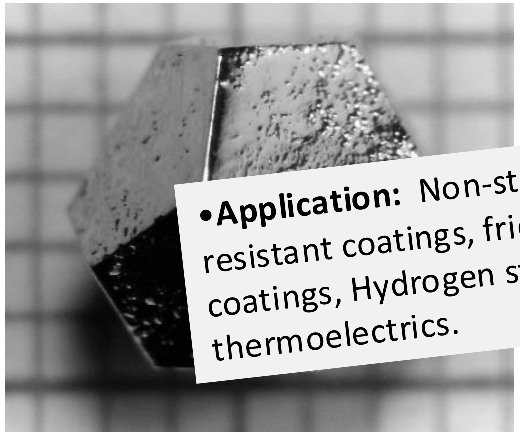
PACS numbers: 61.50.Em, 61.55.Hg, 64.70.Ew

Al –Mn (at 14%) alloy



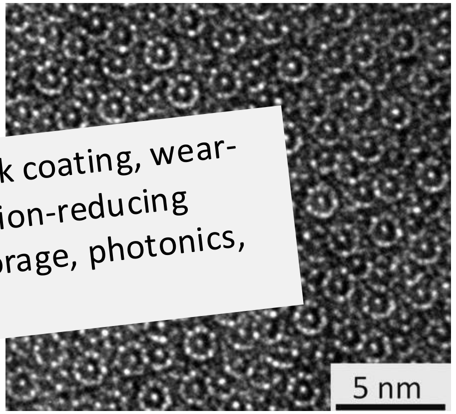
10-fold symmetry!!

Single grain of icosahedral Ho-Mg-Zn quasicrystal



Fisher et al., Phys. Rev. B59, 308 (1999)

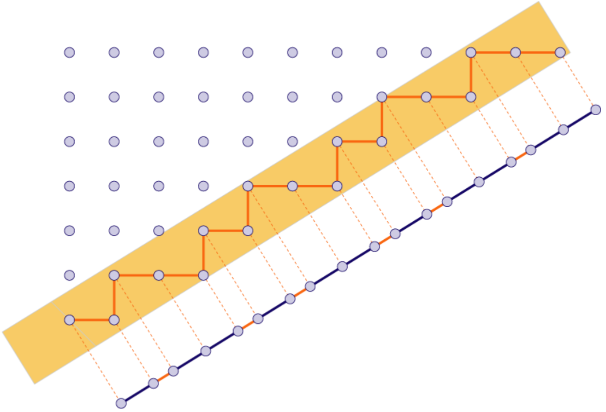
Al71Ni24Fe5 quasicrystal from meteorite



Bindi, et al., Science Report (2015)

- **Application:** Non-stick coating, wear-resistant coatings, friction-reducing coatings, Hydrogen storage, photonics, thermoelectrics.

Projection from Higher Dimensional Periodic Crystals to Lower Dimensional Quasicrystals



Superconducting Quasicrystals

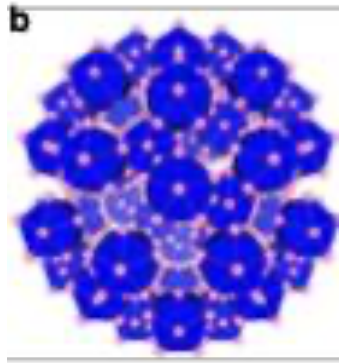
ARTICLE

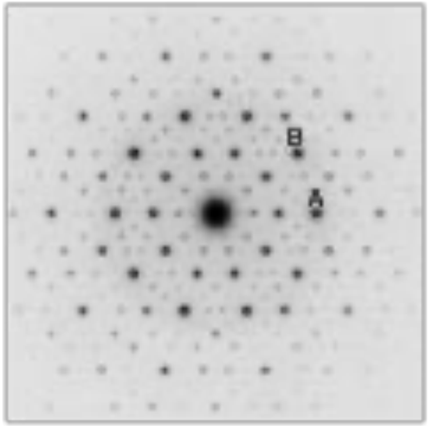
DOI: 10.1038/s41467-017-02667-x OPEN NATURE COMMUNICATIONS | (2018)9:154

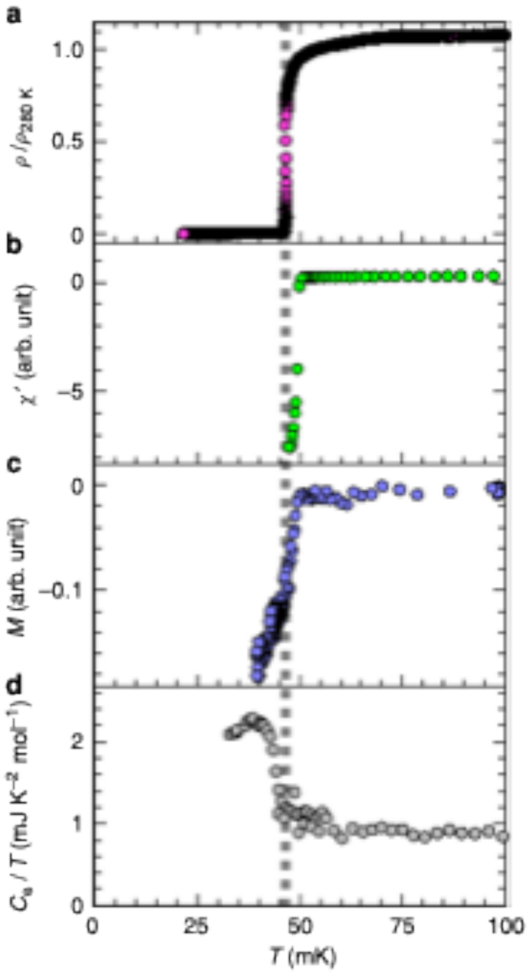
Discovery of superconductivity in quasicrystal

K. Kamiya[1,5], T. Takeuchi[2], N. Kabeya[3], N. Wada[1], T. Ishimasa[4], A. Ochiai[3], K. Deguchi[1], K. Imura[1] & N.K. Sato[1]

Al-Zn-Mg QC,







PHYSICAL REVIEW B 95, 024509 (2017)

Superconductivity on a quasiperiodic lattice: Extended-to-localized crossover of Cooper pairs

Shiro Sakai,[1] Nayuta Takemori,[1] Akihisa Koga,[2] and Ryotaro Arita[1]
[1]Center For Emergent Matter Science, RIKEN, Wako, Saitama 351-0198, Japan
[2]Department of Physics, Tokyo Institute of Technology, Meguro, Tokyo 152-8551, Japan

(Received 12 August 2016; revised manuscript received 20 November 2016; published 18 January 2017)

PHYSICAL REVIEW B 100, 014510 (2019)

Conventional superconductivity in quasicrystals

Ronaldo N. Araújo and Eric C. Andrade
Instituto de Física de São Carlos, Universidade de São Paulo, C.P. 369, São Carlos, SP, 13560-970, Brazil

PHYSICAL REVIEW RESEARCH 3, 023195 (2021)

- Quasiperiodic/fractal gap structures
- Modified critical fluctuation
- Unusual collective modes

Letter

Gapless superconductivity and its real-space topology in quasicrystals

Kazuma Saito,[1] Masahiro Hori,[1,2] Ryo Okugawa,[1] K. Tanaka,[2] and Takami Tohyama[1]
[1]Department of Applied Physics, Tokyo University of Science, Katsushika, Tokyo 125-8585, Japan
[2]Department of Physics and Engineering Physics, and Centre for Quantum Topology and Its Applications (quanTA), University of Saskatchewan, 116 Science Place, Saskatoon, Saskatchewan, Canada S7N 5E2

(Received 1 October 2024; revised 19 February 2025; accepted 13 June 2025; published 25 June 2025)

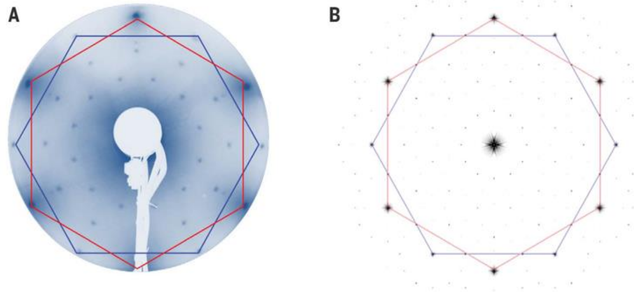
……

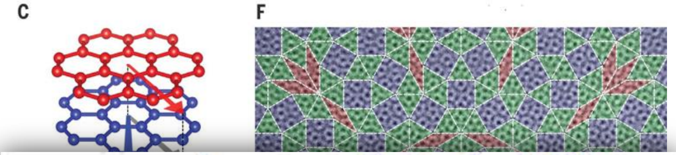
Van der Waals Interlayer Quasi Crystals

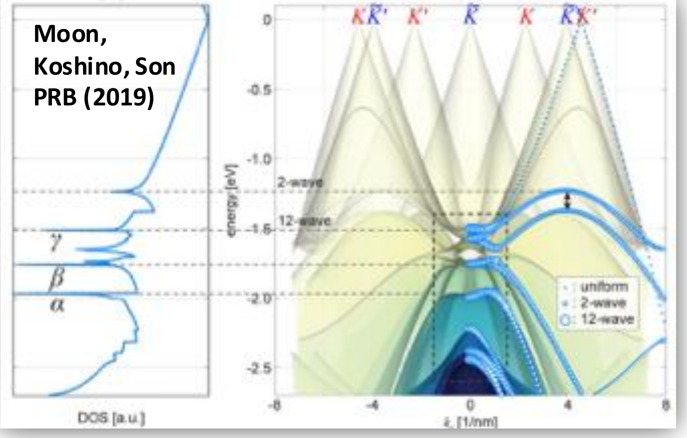
QUASICRYSTALS Ahn *et al.*, *Science* **361**, 782–786 (2018) 24 August 2018

Dirac electrons in a dodecagonal graphene quasicrystal

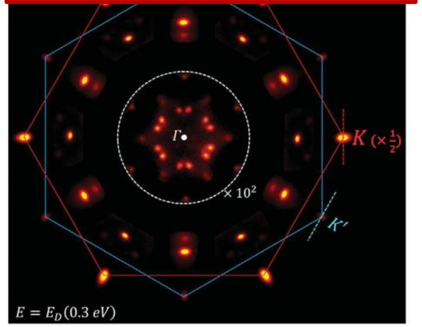
Sung Joon Ahn[1]*, Pilkyung Moon[2,3]*, Tae-Hoon Kim[4]*, Hyun-Woo Kim[1], Ha-Chul Shin[1], Eun Hye Kim[1], Hyun Woo Cha[4], Se-Jong Kahng[5], Philip Kim[6], Mikito Koshino[7], Young-Woo Son[8]†, Cheol-Woong Yang[4]†, Joung Real Ahn[1,9]†

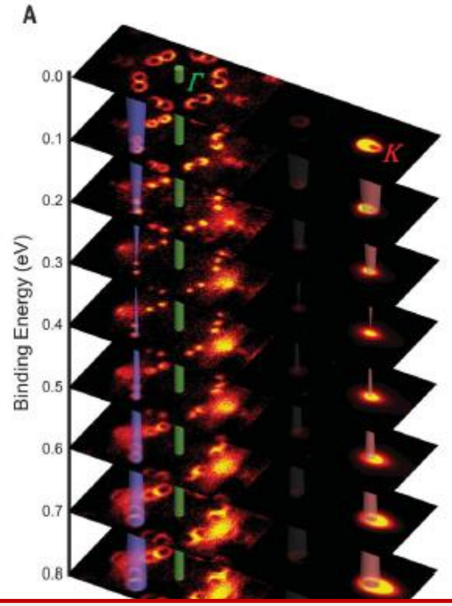






Appearance of higher order Umklapp features



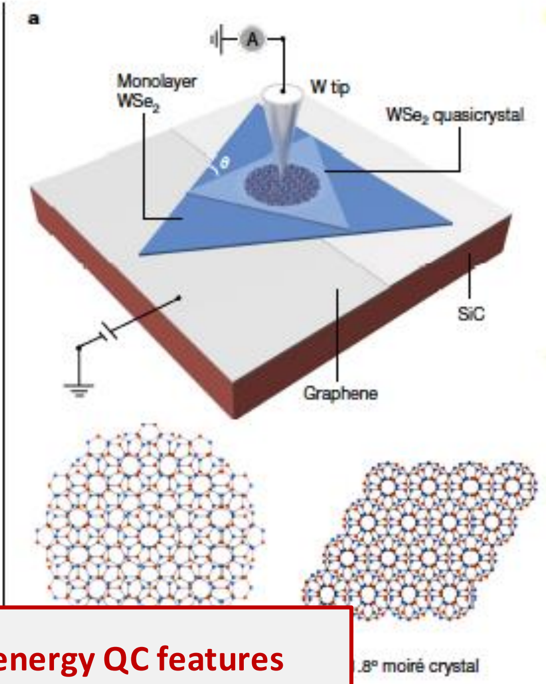


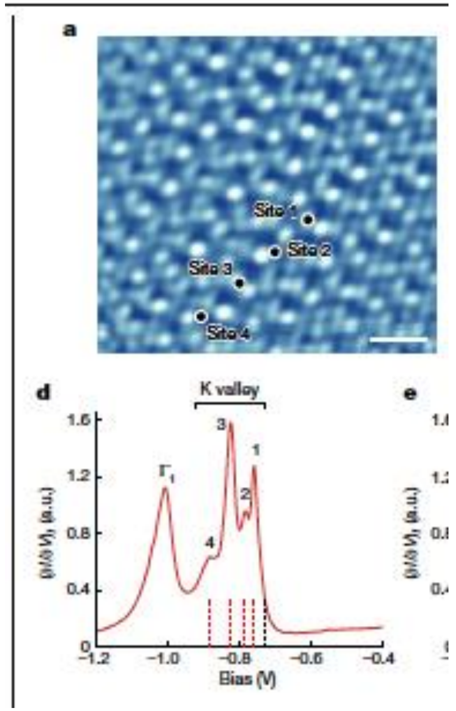
Article 494 | Nature | Vol 625 | 18 January 2024

Tuning commensurability in twisted van der Waals bilayers

https://doi.org/10.1038/s41586-023-06904-w
Received: 26 March 2023
Accepted: 28 November 2023
Published online: 17 January 2024

Yanxing Li[1,7], Fan Zhang[1,7], Viet-Anh Ha[1,2], Yu-Chuan Lin[3,4], Chengye Dong[3], Qiang Gao[1], Zhida Liu[1], Xiaohui Liu[1], Sae Hee Ryu[5], Hyunsue Kim[1], Chris Jozwiak[5], Aaron Bostwick[5], Kenji Watanabe[6], Takashi Taniguchi[6], Bishoy Kousa[1], Xiaoqin Li[1], Eli Rotenberg[5], Eslam Khalaf[1], Joshua A. Robinson[3], Feliciano Giustino[1,2] & Chih-Kang Shih[1]





- Weak interlayer interaction/ High energy QC features
- Long wavelength moire approximation

2D Atomic Quasi Crystal

$$\frac{3+2\sqrt{3}}{4} = 1.616025404\cdots$$

Angew. Chem. Int. Ed. **1998**, *37*, No. 10

A Dodecagonal Quasicrystalline Chalcogenide**

Matthias Conrad, Frank Krumeich, and Bernd Harbrecht*

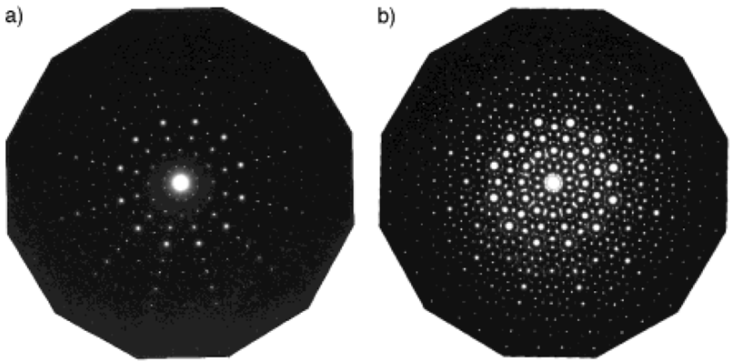
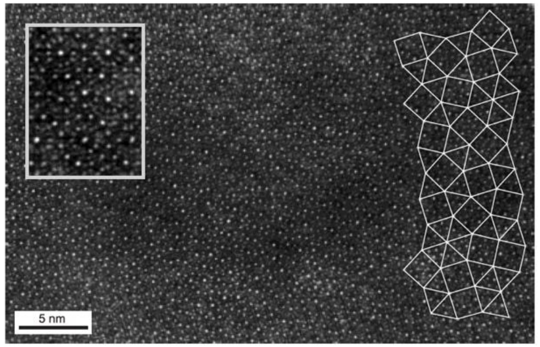
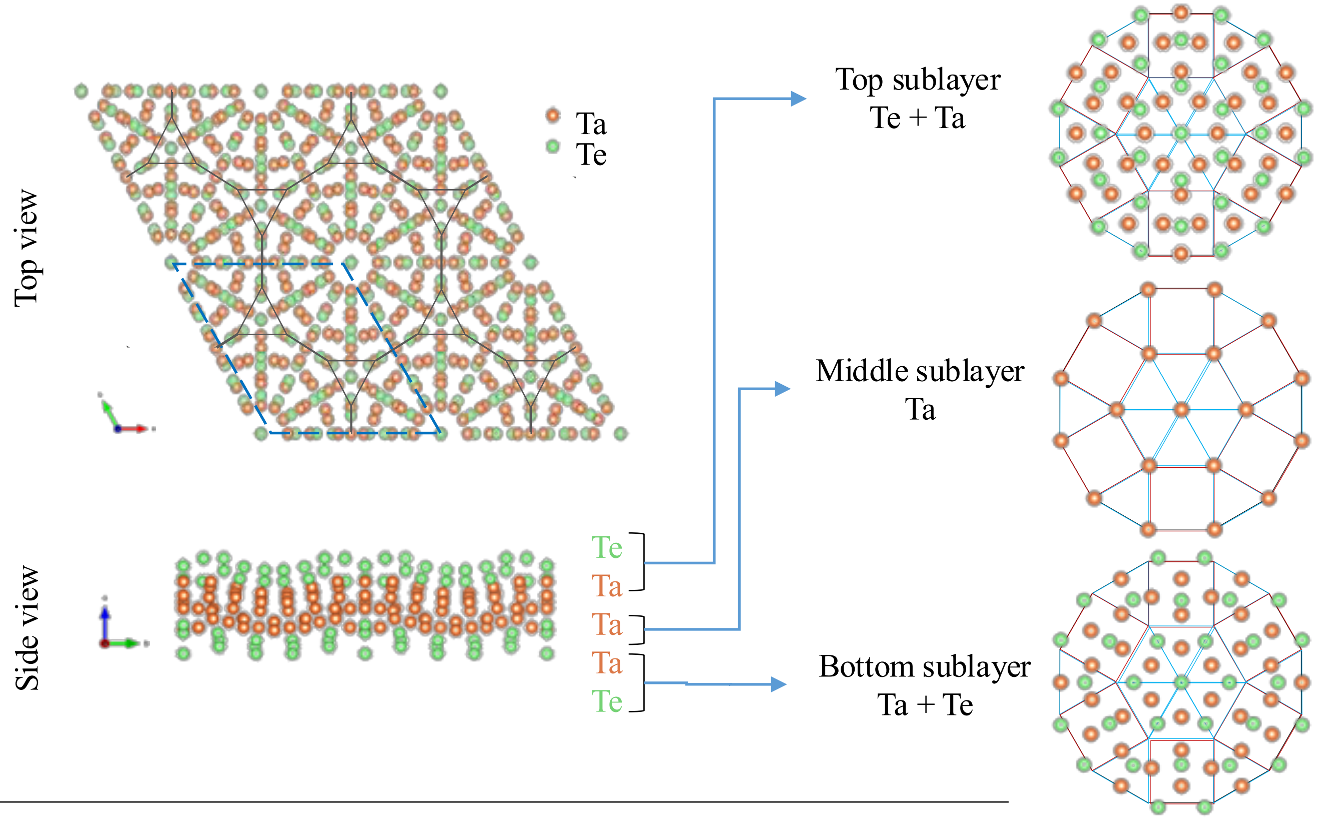
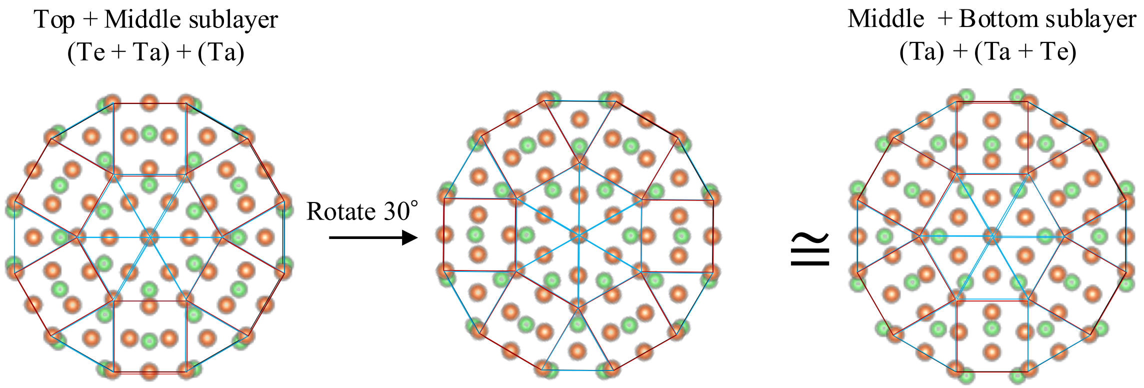


Figure 2. Reflection-poor (a) and reflection-rich (b) electron diffractograms of dd-$Ta_{1.6}Te$.







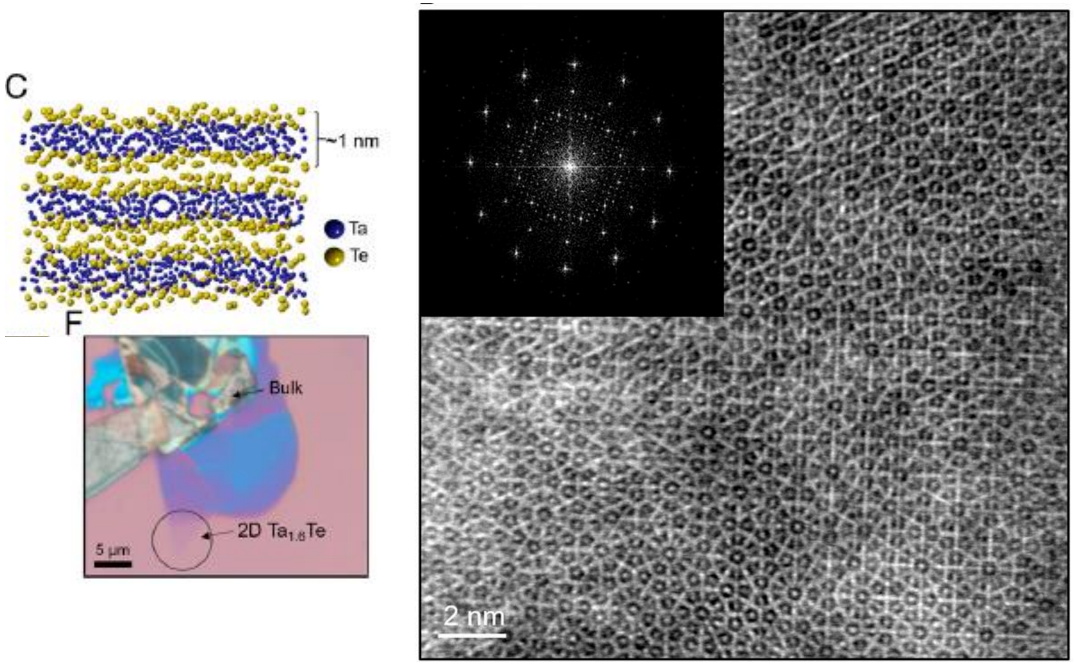
van der Waals 2D Atomic Quasi Crystals

Layer-dependent topological phase in a two-dimensional quasicrystal and approximant

Jeffrey D. Cain[a,b,c,1], Amin Azizi[a,c,1], Matthias Conrad[d], Sinéad M. Griffin[b,e], and Alex Zettl[a,b,c,2]

[a]Department of Physics, University of California, Berkeley, CA 94720; [b]Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720; [c]Kavli Energy NanoSciences Institute, University of California, Berkeley and Lawrence Berkeley National Laboratory, Berkeley, CA 94720; [d]Fachbereich Chemie, Philipps-Universität Marburg, 35032 Marburg, Germany; and [e]The Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

Edited by Angel Rubio, Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany, and approved August 24, 2020 (received for review July 17, 2020)



- Can be cleaved down to monolayer!
- Quasicrystals & Approximants

Nature Communications | (2024)15:1529

Article

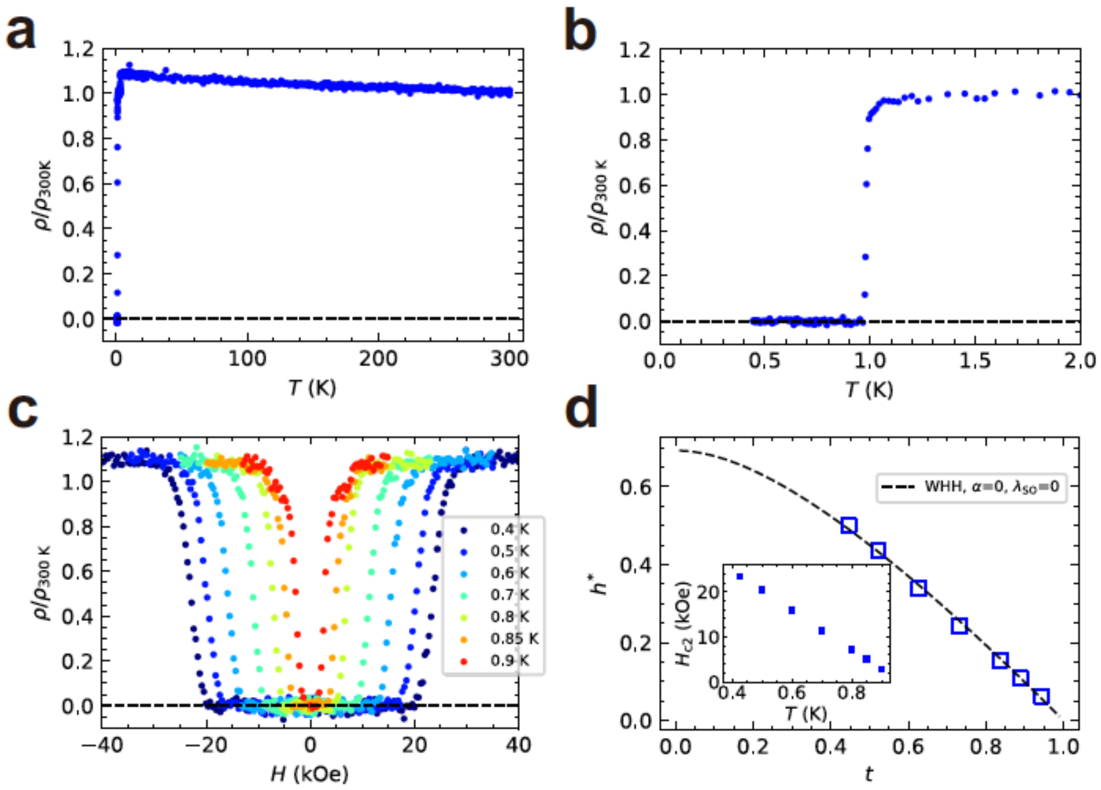
Superconductivity in a van der Waals layered quasicrystal

Measurement was done on polycrystals

Received: 29 August 2023

Accepted: 8 February 2024

Yuki Tokumoto[1], Kotaro Hamano[1], Sunao Nakagawa[1], Yasushi Kamimura[1], Shintaro Suzuki[2], Ryuji Tamura[3] & Keiichi Edagawa[1]

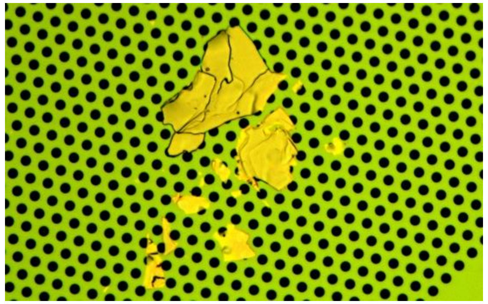


Opening Up 25-Year-Old Samples

Flux grown single crystals of
Ta-Te Quasicrystals and Approximants

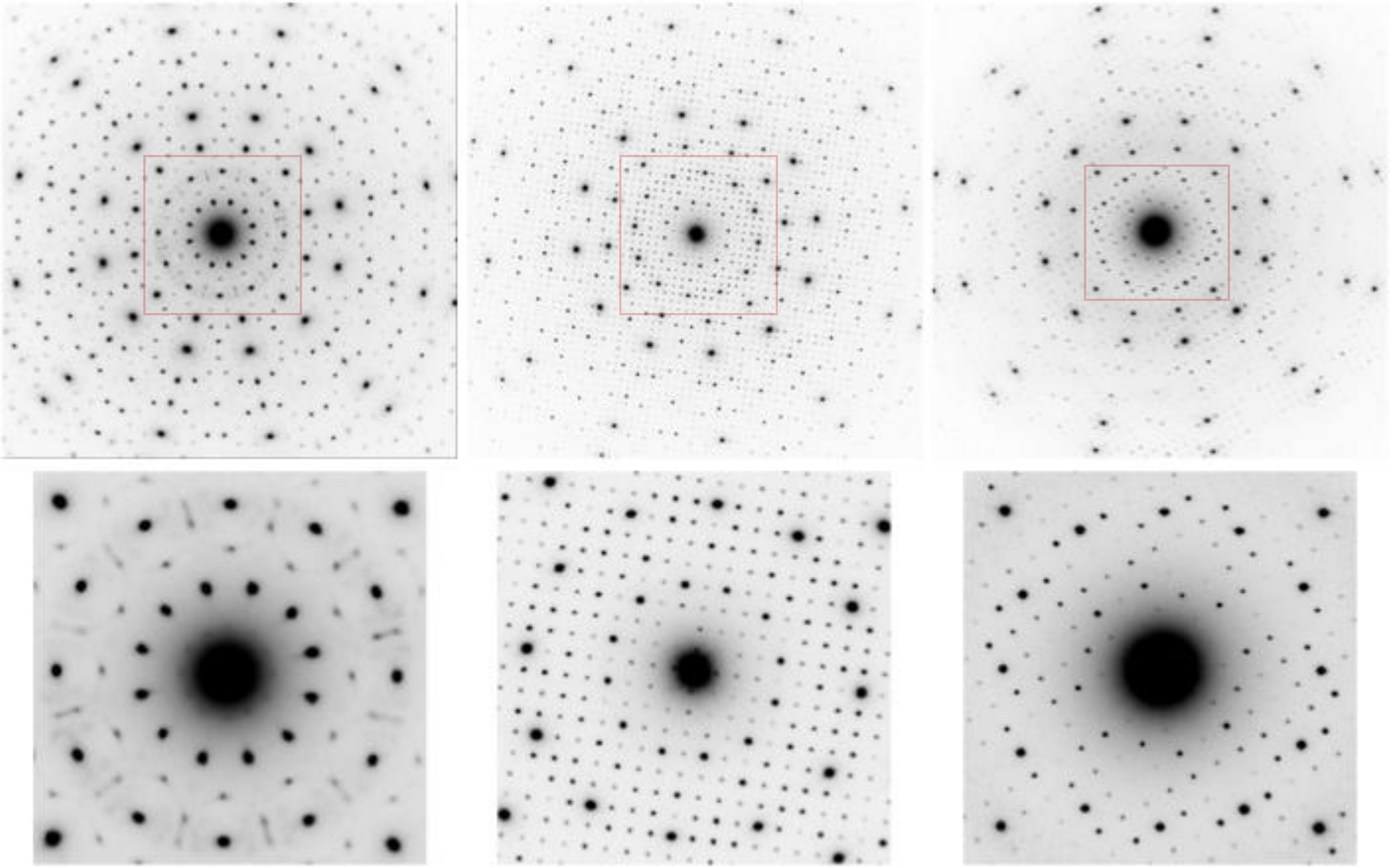
Samples are air-sensitive!

TEM sample preparation

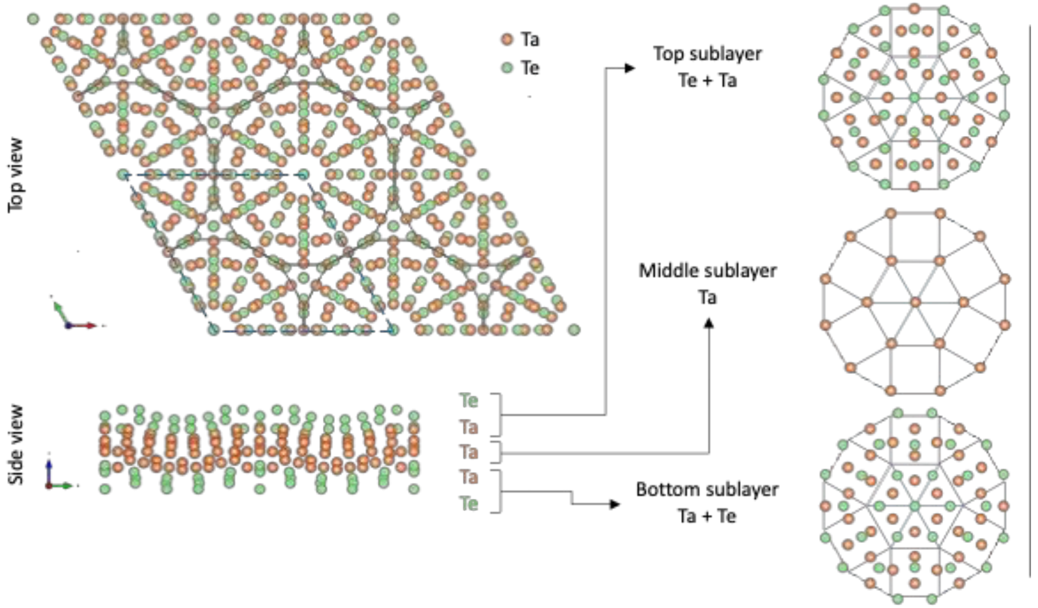


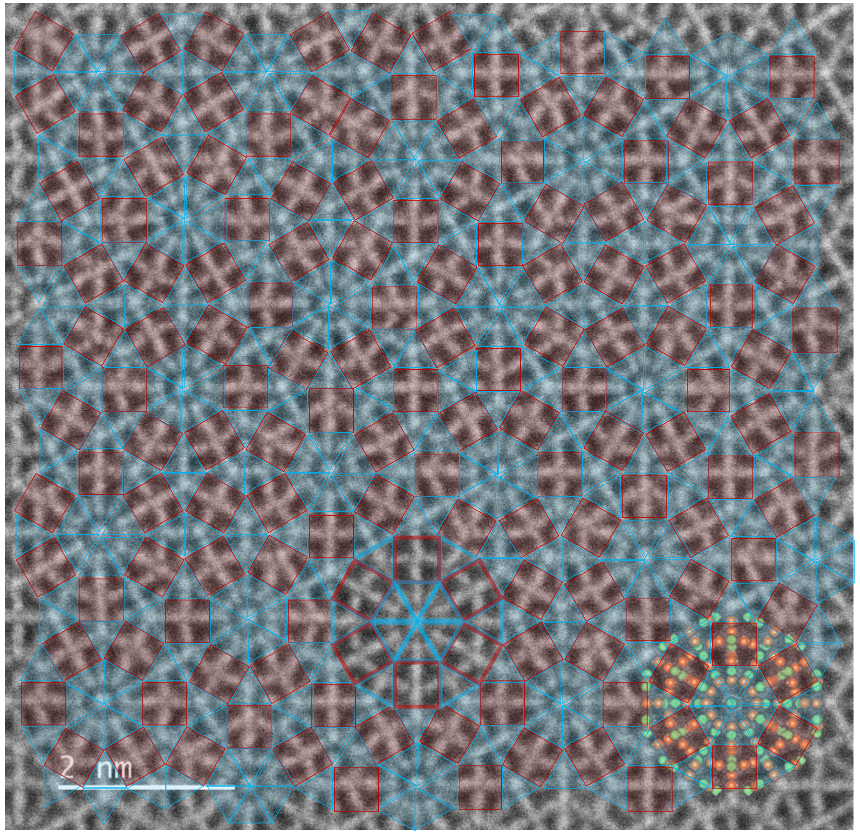
Selective Area Diffraction

dd-$Ta_{1.6}Te$ (QC) | $Ta_{181}Te_{112}$ (AC) | $Ta_{97}Te_{60}$ (AC)



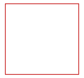
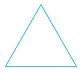
Dodecagonal Atomic Clusters: Building Block



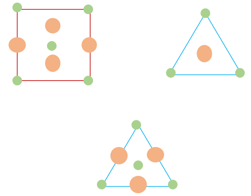


Ta
Te

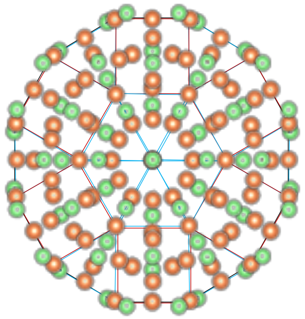
Basic building blocks:
(prototiles)


&


Decoration with atoms:



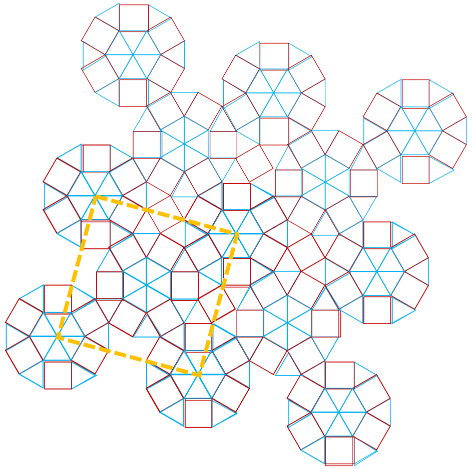
Dodecagonal building blocks

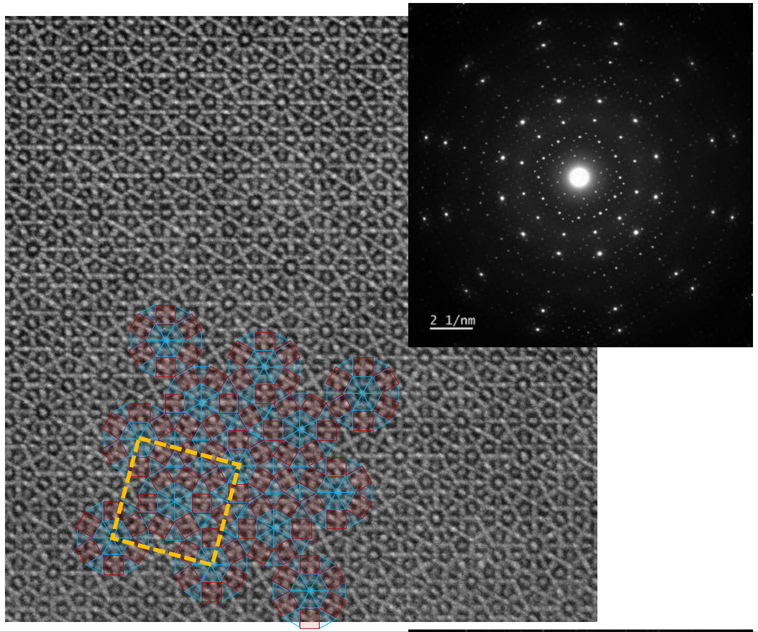


High-Angle Annular Dark-Field Scanning Transmission Electron Microscopy (HAADF STEM)

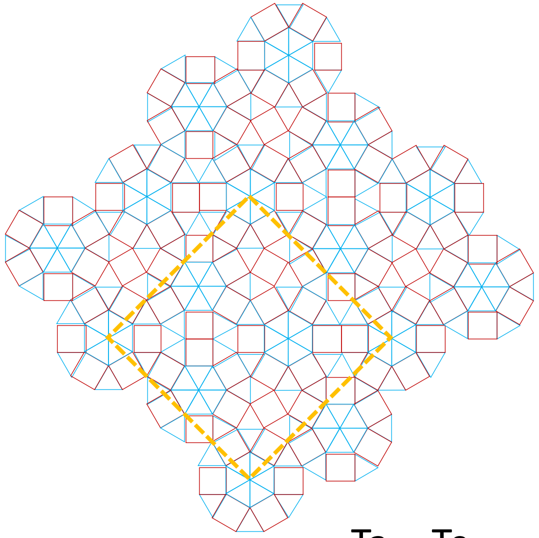
Approximants and Quasicrystals

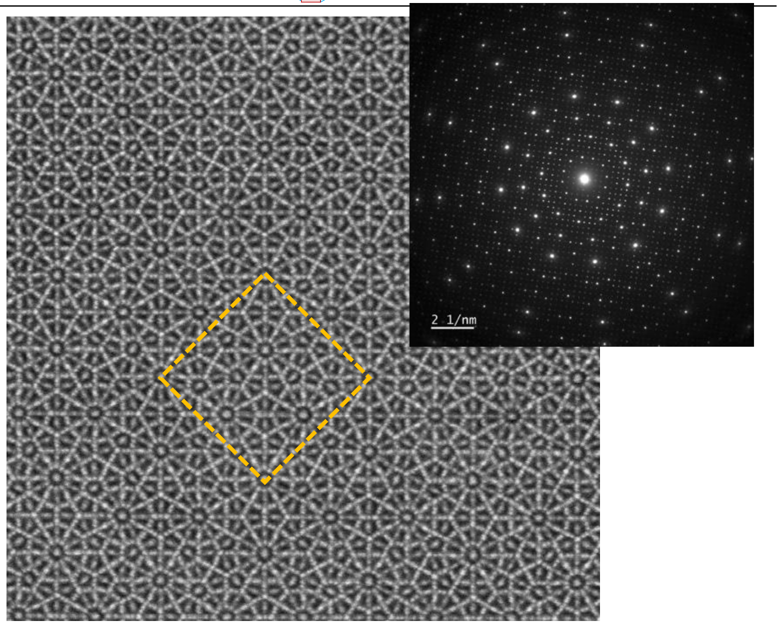
Approximant tiling





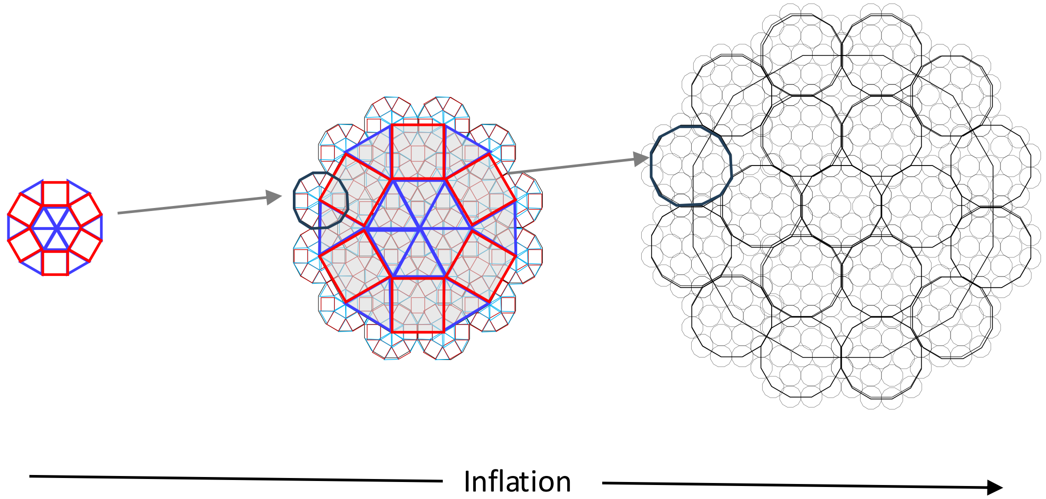
$Ta_{97}Te_{60}$





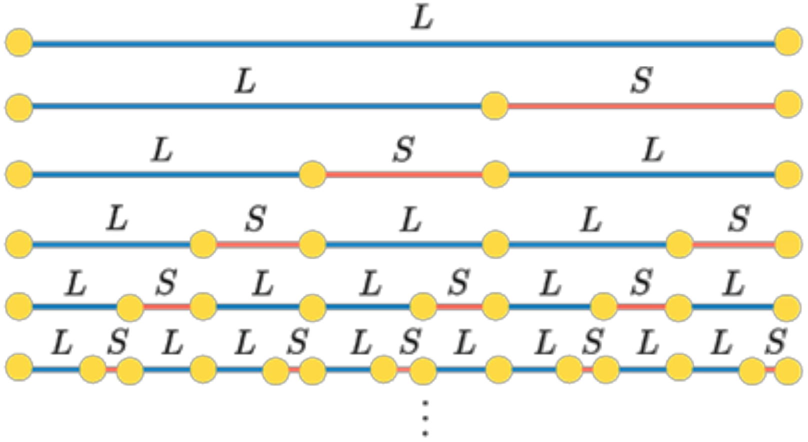
$Ta_{181}Te_{112}$

Quasicrystal Tiling

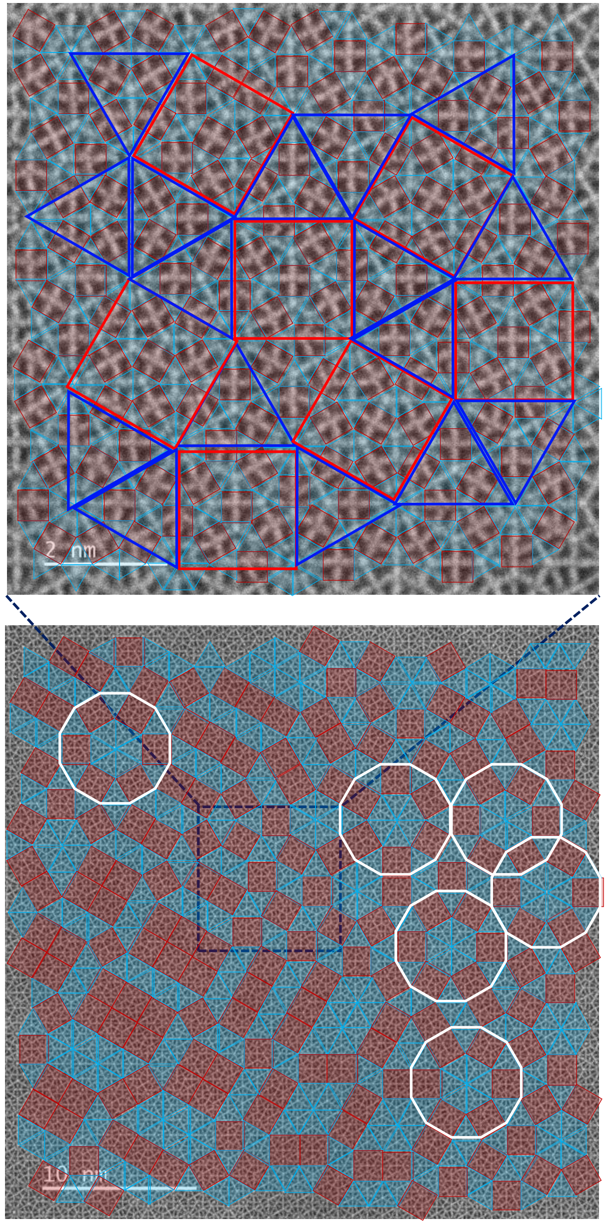


Fibonacci Chain

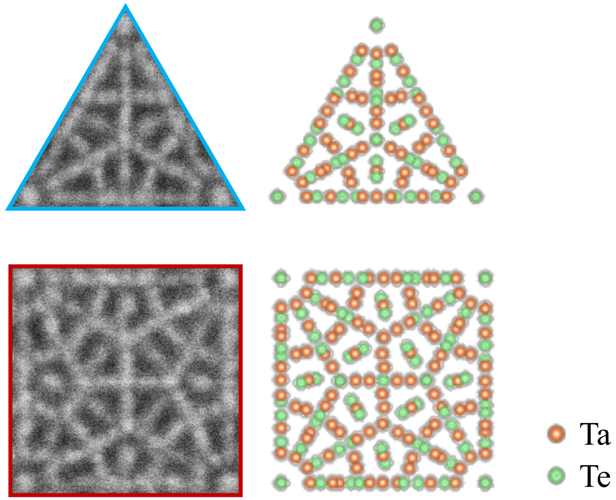
$L \rightarrow L+S; \quad S \rightarrow L$



Inflation Rules: Second Order Tiling



Second Order Tiles

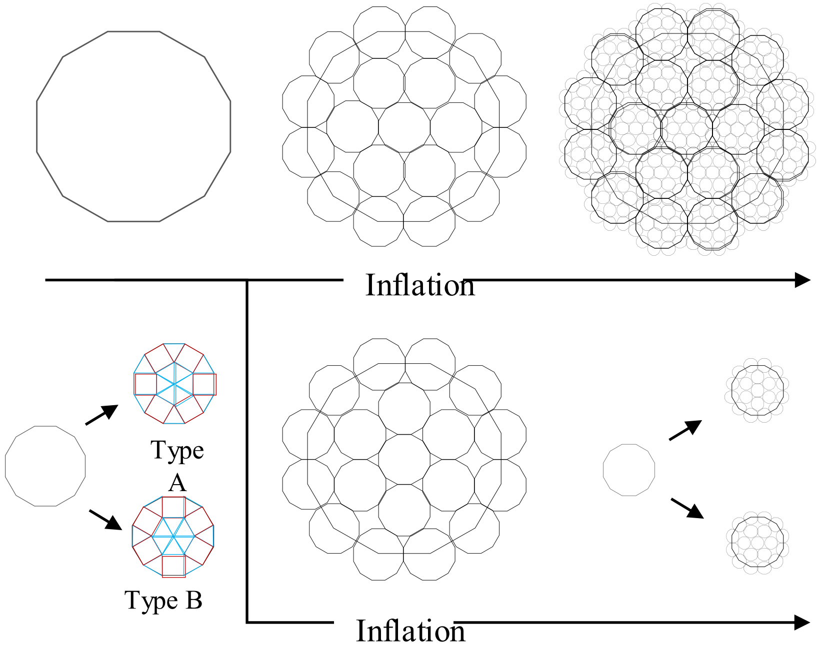


- Tiling can be done using larger squares and triangles with a scaling factor of $2 + \sqrt{3}$.

- Beyond the second-order tiling, it is not possible to proceed in the same way. The inflation rule does not apply.

Failure of the inflation rule after second order tiling!

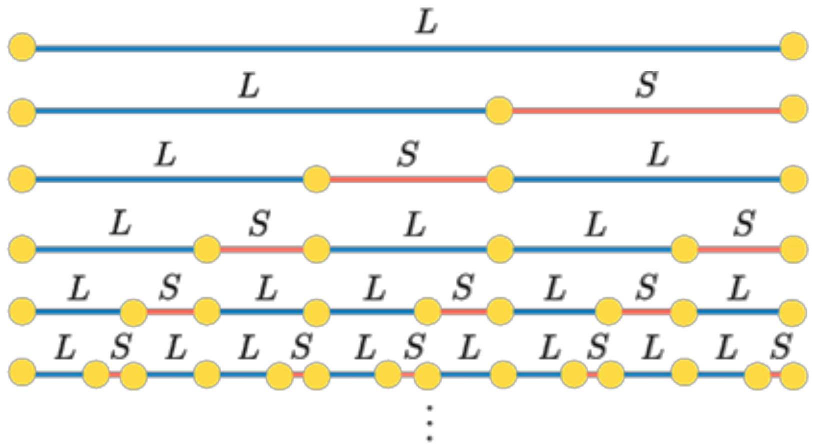
Ambiguity of Substitution Rule

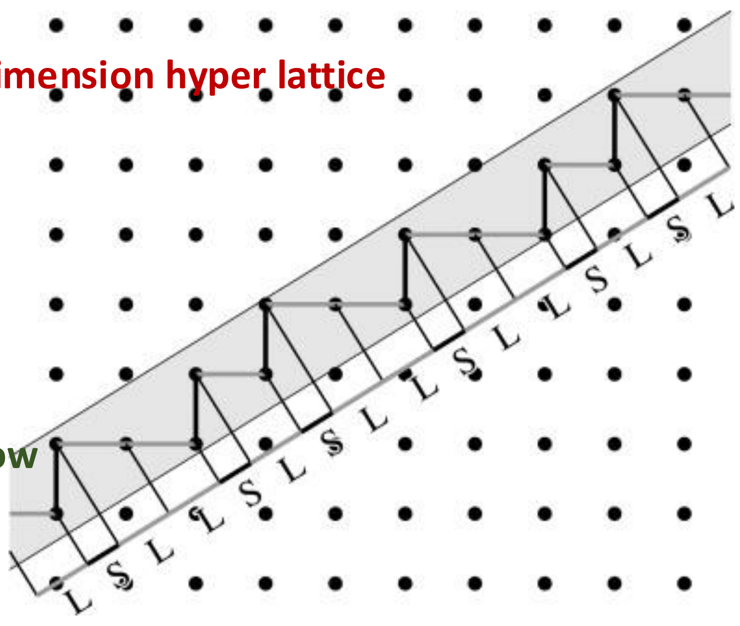


Many possible inflation possibility…

Cut and Projection Method: Fibonacci Chain

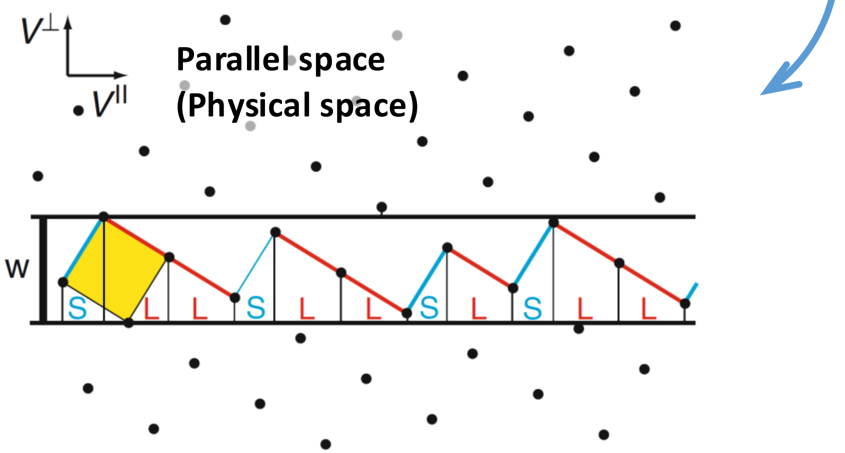
Fibonacci Chain $L \rightarrow L+S; \quad S \rightarrow L$





Slope:

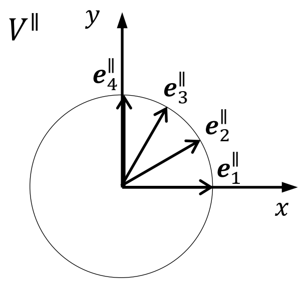
$$\tau = \frac{1+\sqrt{5}}{2}$$



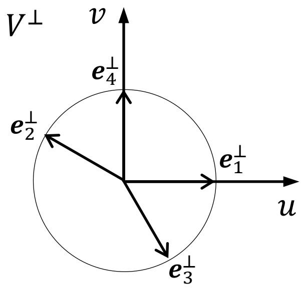
A system without periodicity in real space can exhibit periodicity in higher-dimensional space.

Physical Space and Perpendicular Space: Toward Higher Dimension

The dodecagonal square-triangle tiling can be generated from 4D lattice.



2D physical Space



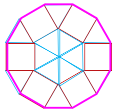
2D Perpendicular

4D basis vectors

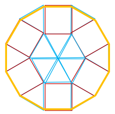
$$e_i = (e_i^{\parallel}, e_i^{\perp})$$

$$e_i^{\parallel} = R_{12}^{(i-1)} e_1^{\parallel}$$

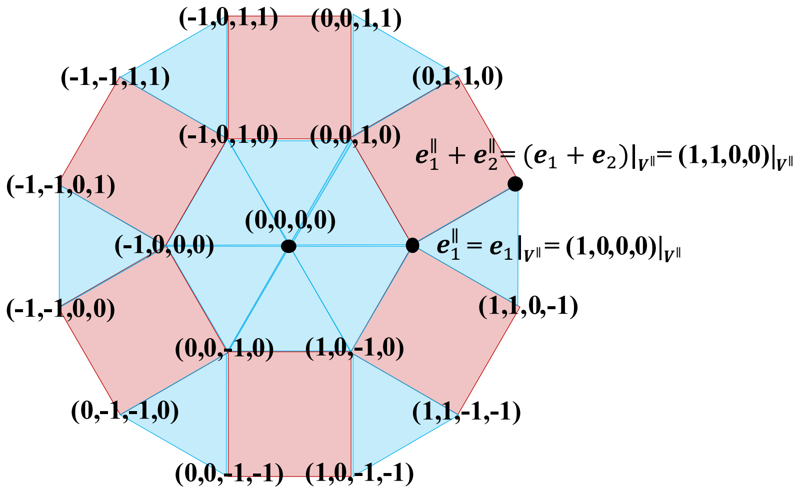
$$e_i^{\perp} = R_{12}^{5(i-1)} e_1^{\perp}$$



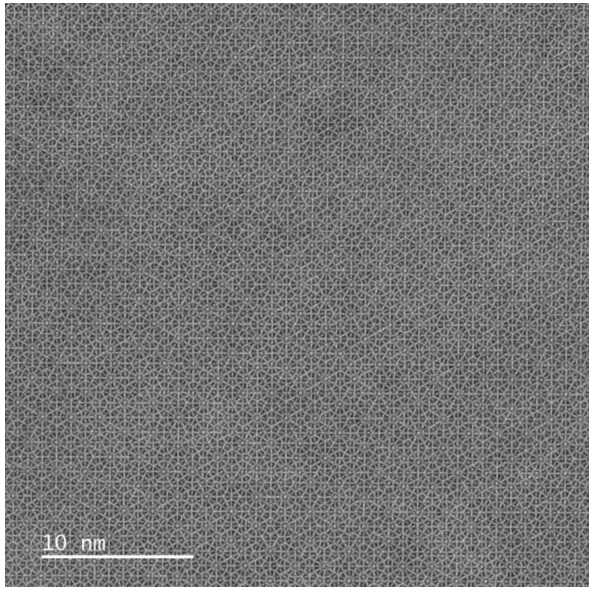
$\#(A) = 219$

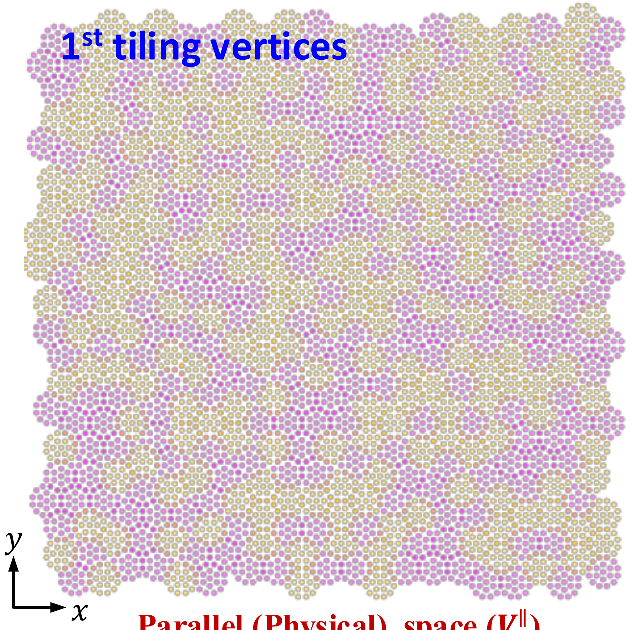


$\#(B) = 211$



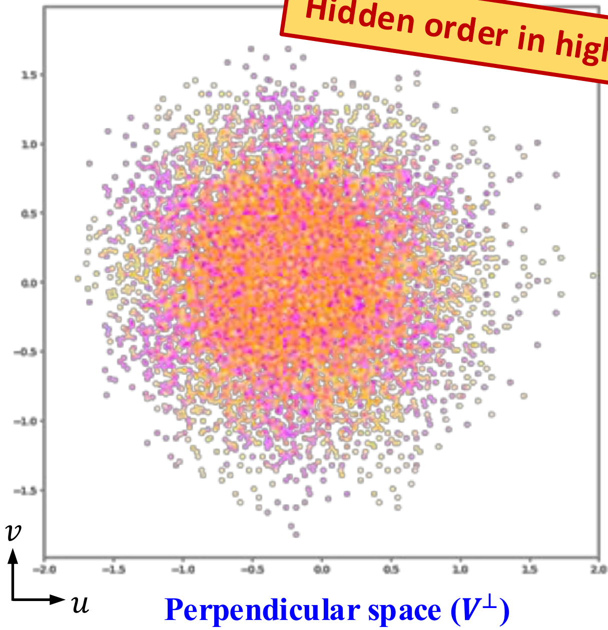
Coordination with basis $e_i (i = 1,2,3,4)$





Parallel (Physical) space ($V^{\parallel}$)

Hidden order in higher dimension!



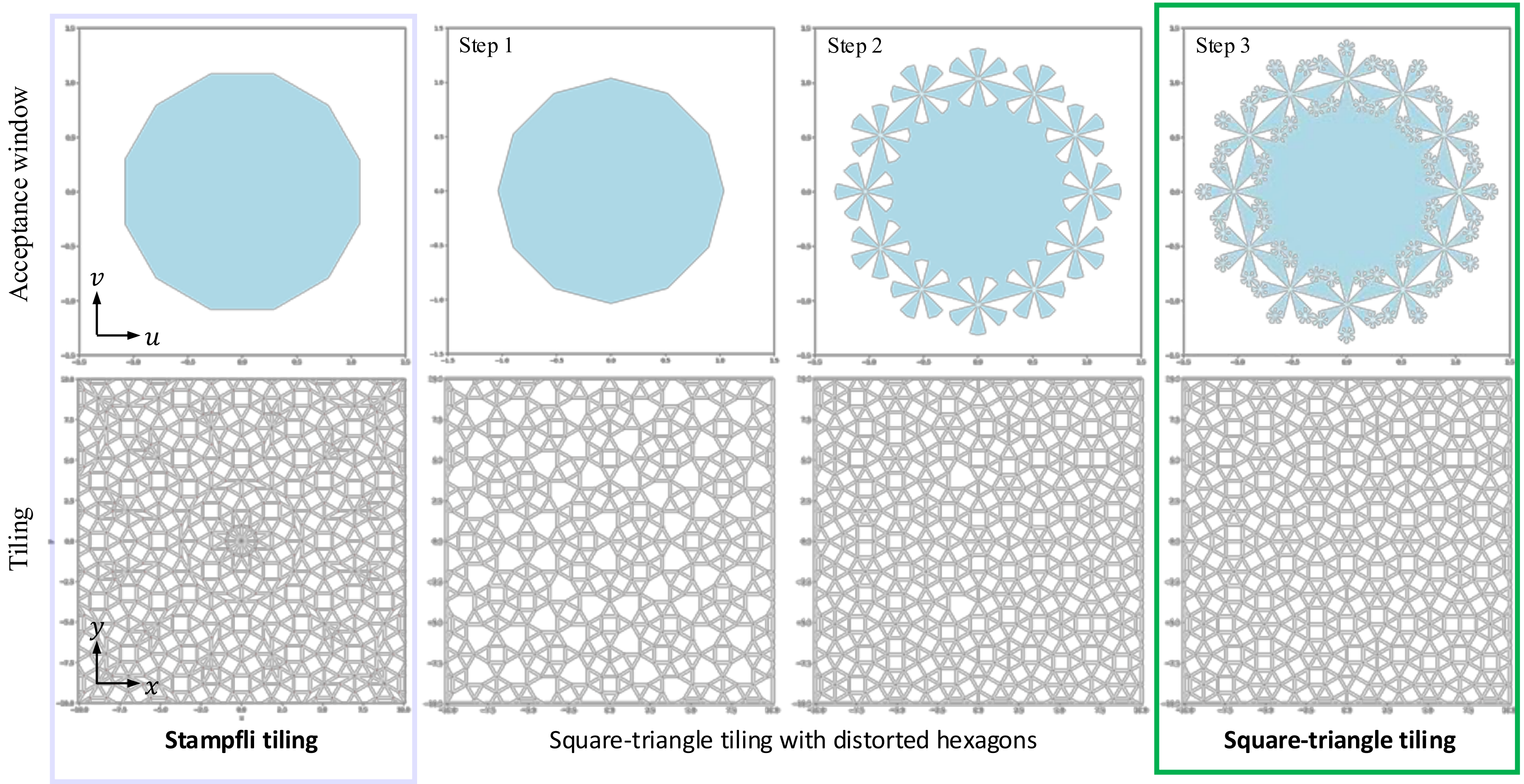
Perpendicular space ($V^{\perp}$)

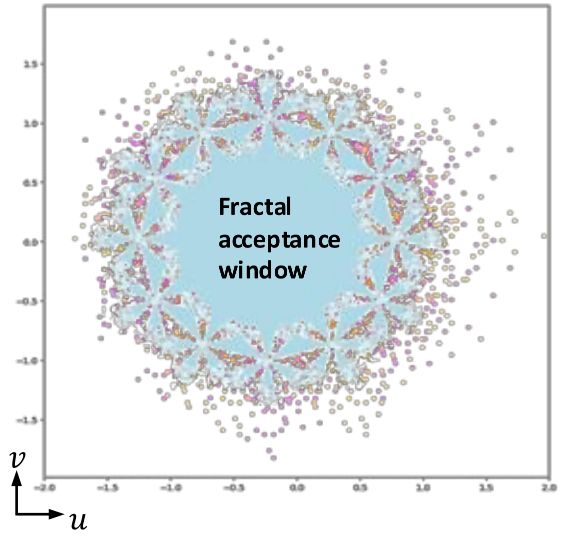
Cut and Projection Method: Fractal Acceptance Window

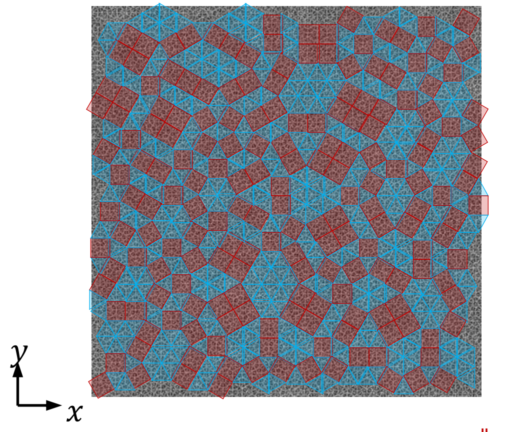
Dodecagonal Quasicrystal Tiling: 4D Hyper Lattice projection with acceptance windows

Inspired by Baake, et. al., Physica A 191 554 (1992)

Fractal acceptance window works reasonable but not perfect. Disorder in quasicrystal?





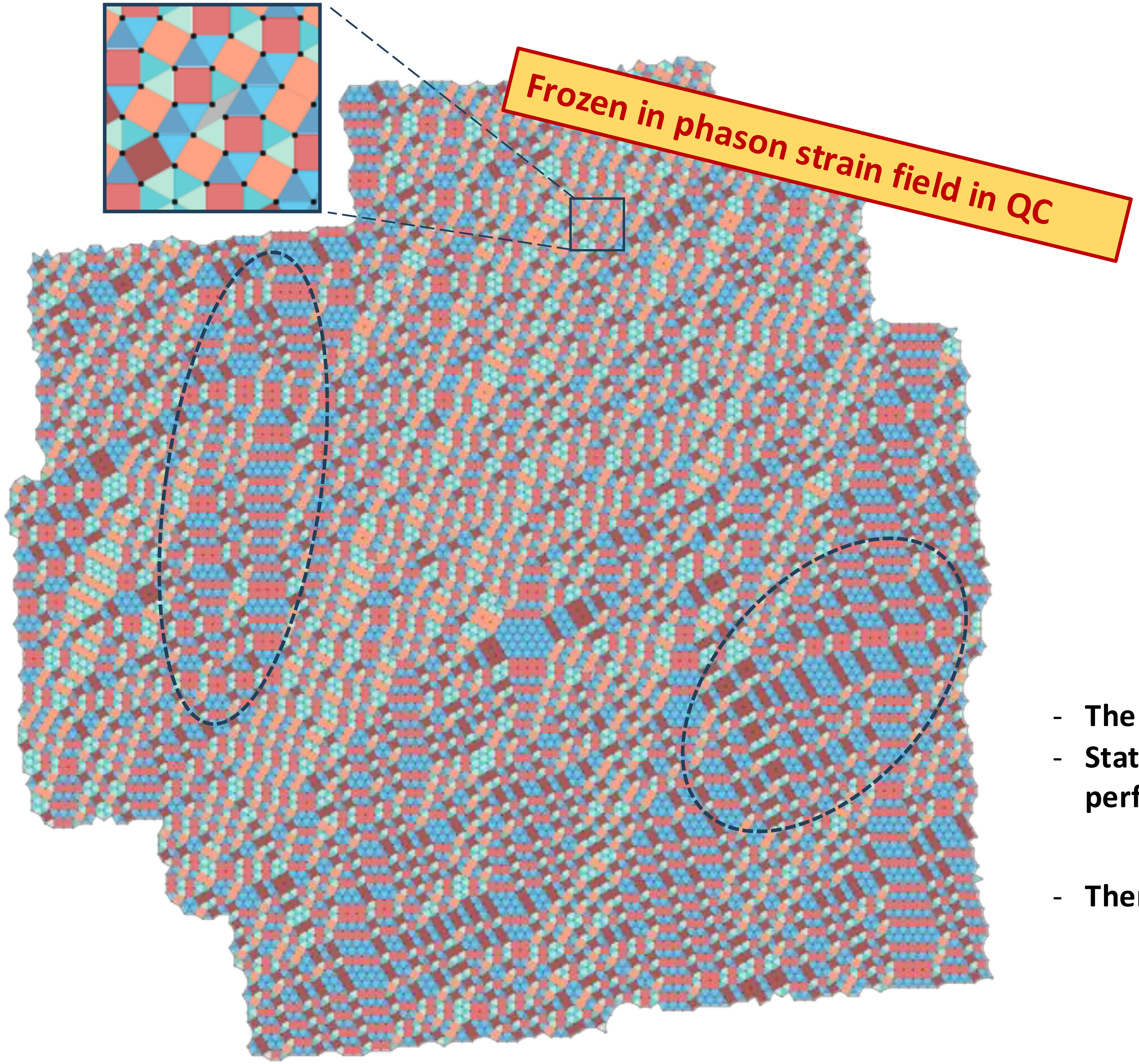


Parallel (Physical) space ($V^{\parallel}$)

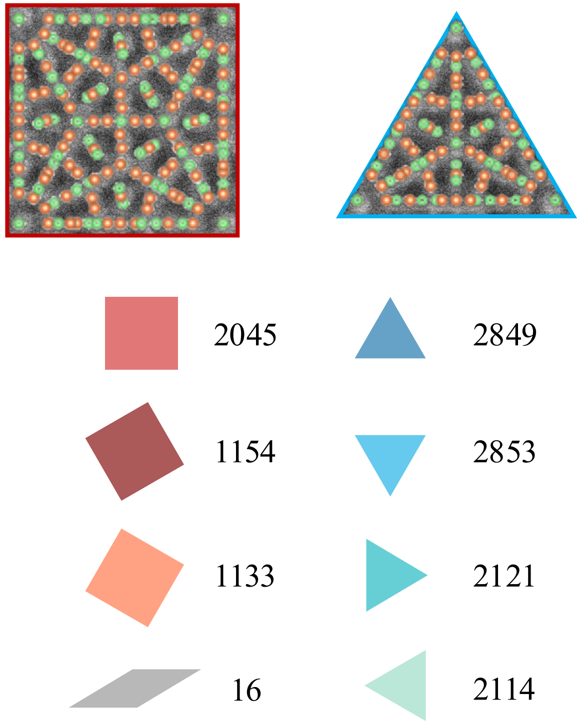
A fractal-shaped acceptance window is required to eliminate skinny rhombi in the Stampfli tiling.

Second Order Tiling and Phason Flip Disorders Quasicrystals

- There exist defects that require rhombus tiles.



Second order tiles

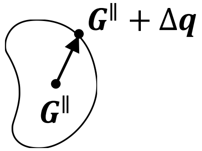


- **The distribution of second-order tiles is not isotropic.**
- **Statistically, it is not maximally entropic and does not exhibit perfect 12-fold symmetry.**
- **There exist domains composed of only specific types of tiles.**

Temperature Dependent Diffraction: Phonon versus Phason Elastic Constants

$$\boldsymbol{G} = \boldsymbol{G}^{\parallel} + \boldsymbol{G}^{\perp}$$

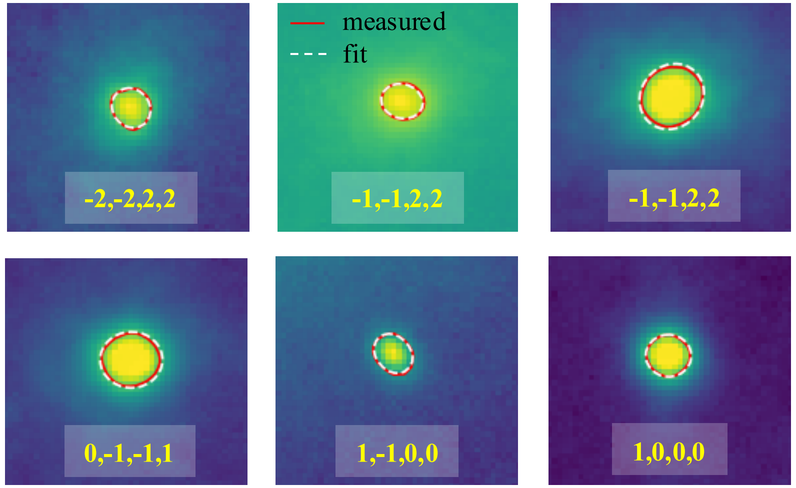
$$I(\boldsymbol{G}^{\parallel} + \Delta\boldsymbol{q}) \propto \exp\left[-\frac{1}{2}\Delta\boldsymbol{q}^{T} C^{-1}(\boldsymbol{G})\Delta\boldsymbol{q}\right]$$



$$C = C_{\text{phonon}} + C_{\text{phason}} + C_{\text{coupling}}$$

$$= C_{\text{phonon}}(\boldsymbol{G}^{\parallel}, \text{T}) + C_{\text{phason}}(\boldsymbol{G}^{\perp}) + C_{\text{coupling}}(\boldsymbol{G}^{\parallel}, \boldsymbol{G}^{\perp}, \text{T})$$

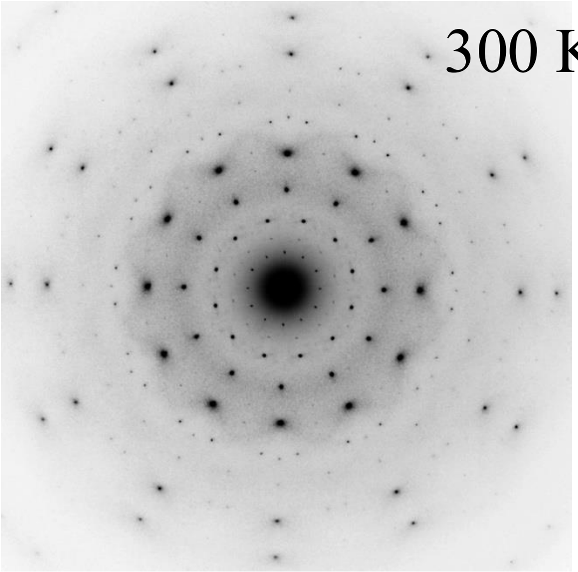
- If the phason contribution is temperature-independent, temperature-dependent diffraction patterns can be used to isolate the peak broadening caused by phonon fluctuations and phonon–phason coupling.

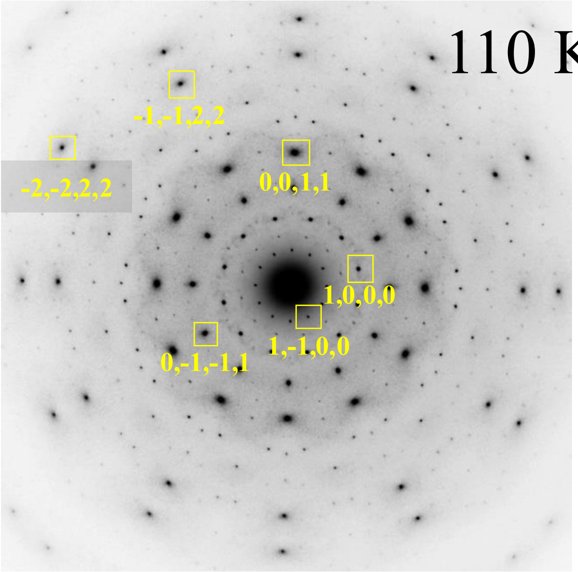


$$\begin{pmatrix} K_{tt} & K_{tr} & K_{ts} \\ K_{tr} & K_{rr} & K_{rs} \\ K_{ts} & K_{rs} & K_{ss} \end{pmatrix} = \begin{pmatrix} 1.042 & 0.002 & -0.167 \\ 0.002 & 1.012 & -0.013 \\ -0.167 & -0.013 & 0.948 \end{pmatrix} \xRightarrow{\text{12-fold}} \begin{pmatrix} K_{tt} & 0 & 0 \\ 0 & K_{tt} & 0 \\ 0 & 0 & K_{ss} \end{pmatrix}$$

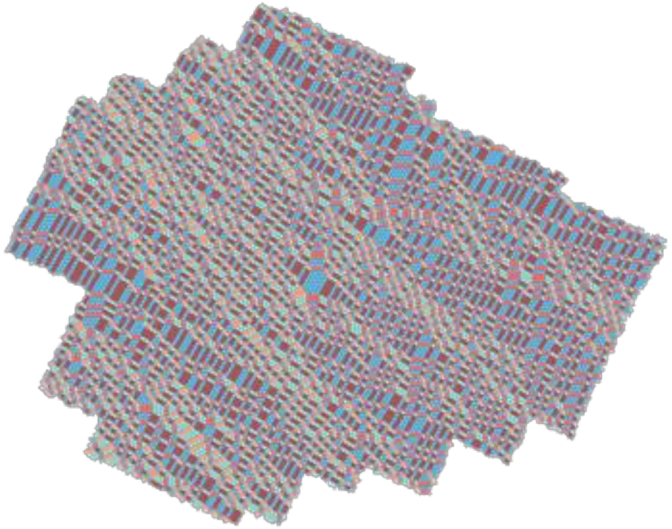
$$K_{tt} \approx K_{rr} > K_{ss} \gg K_{tr}, K_{ts}, K_{rs}$$

- The phason elastic constant becomes comparable to that of the 12-fold quasicrystalline phase.

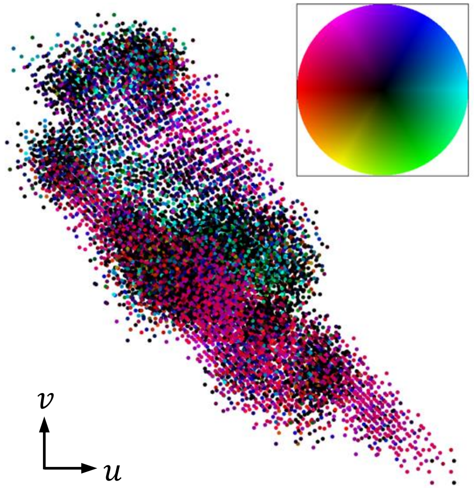




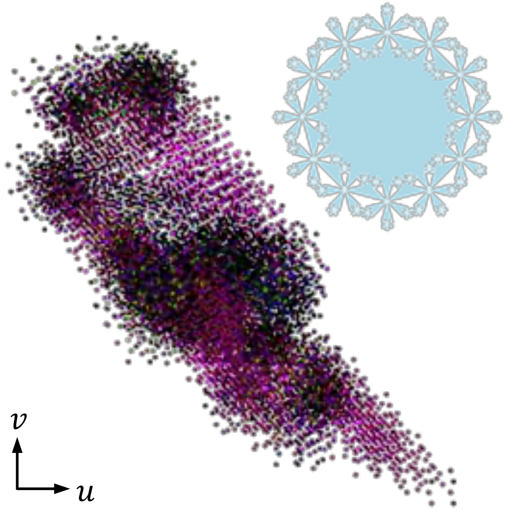
Phason Strain Field in Second Order Tiling



phason strain field in perpendicular space

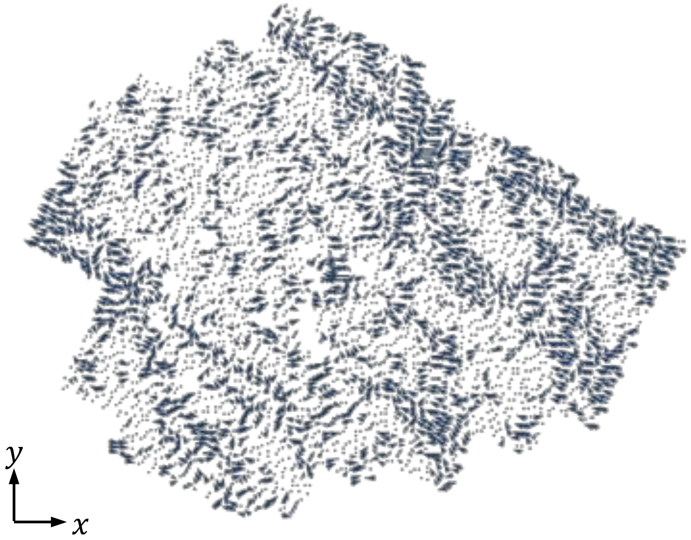


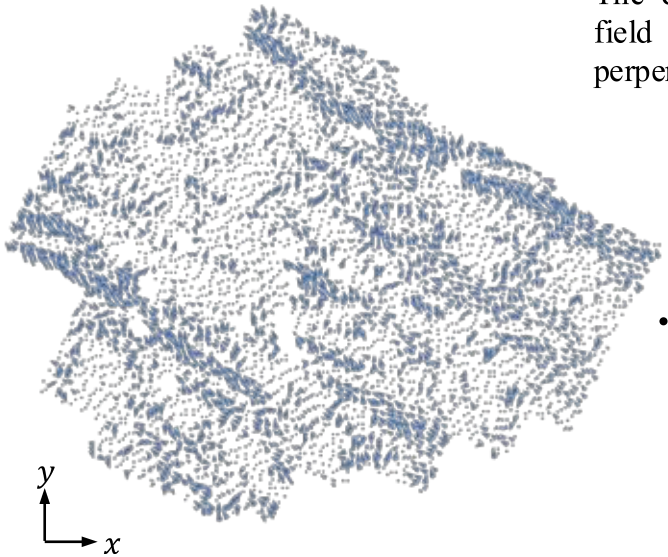
$\boldsymbol{U}_x$ field $\quad (U = [\boldsymbol{U}_x, \boldsymbol{U}_y])$



$\boldsymbol{U}_y$ field

phason strain field in real space



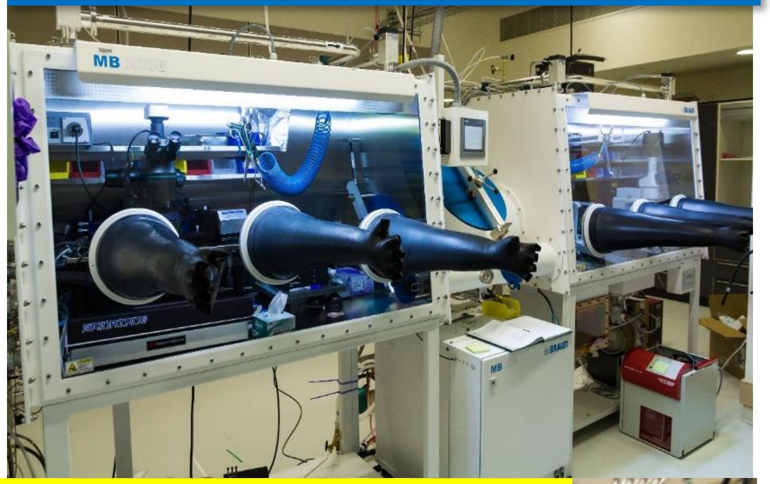


- The direction of the local phason strain field follows the elongation observed in perpendicular space.

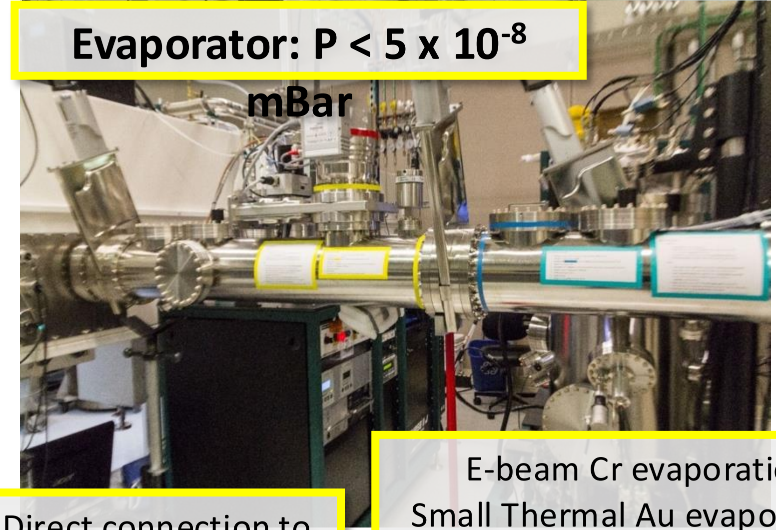
- Larger local phason strain is observed in approximant-like domains, while QC-like domains retain relatively small phason strain.

Device Fabrication

Glovebox: H_2O Concentration 5 - 40 ppb



Evaporator: P < 5 x 10^{-8} mBar

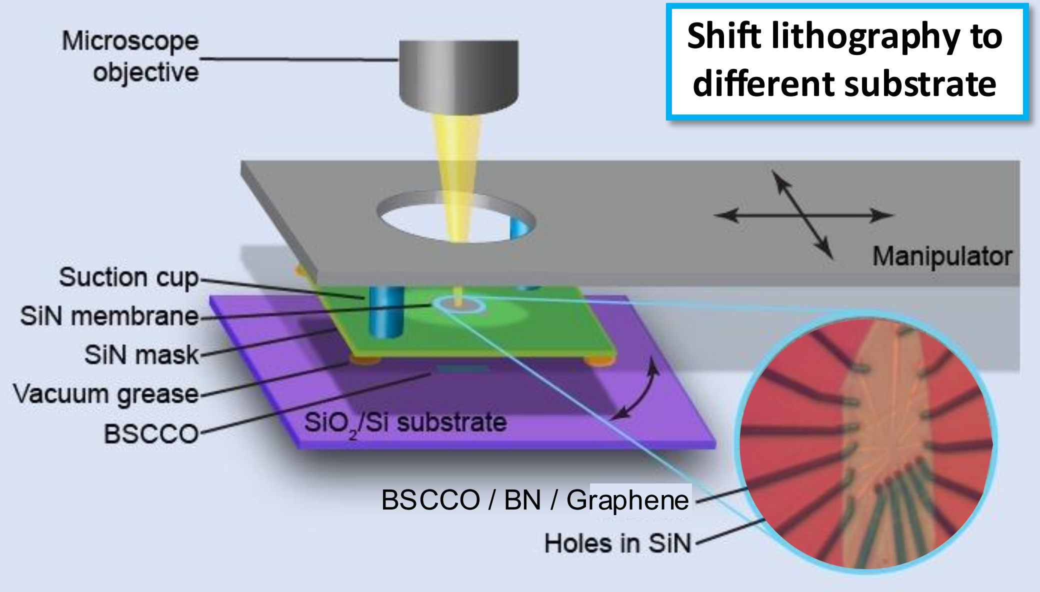


Direct connection to argon glovebox

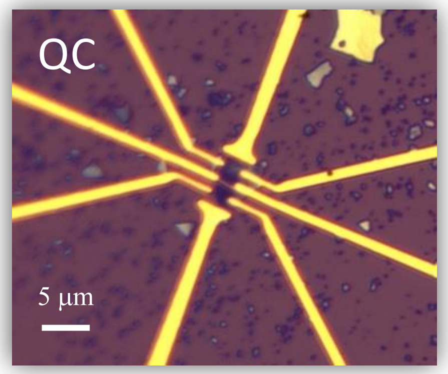
E-beam Cr evaporation source
Small Thermal Au evaporation source
Argon ion milling
Thermoelectric cooled sample stage

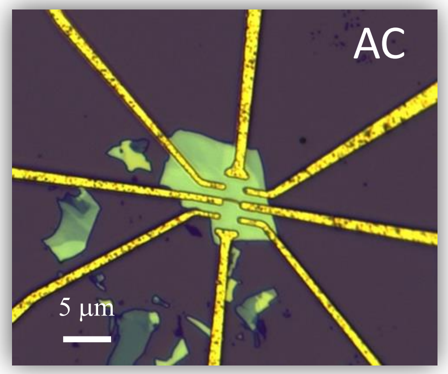
- *Stacking*
- *Selection*
- ✓ *Deposition*
- ✓ *Etch*
- ✓ *Packaging*

Shift lithography to different substrate



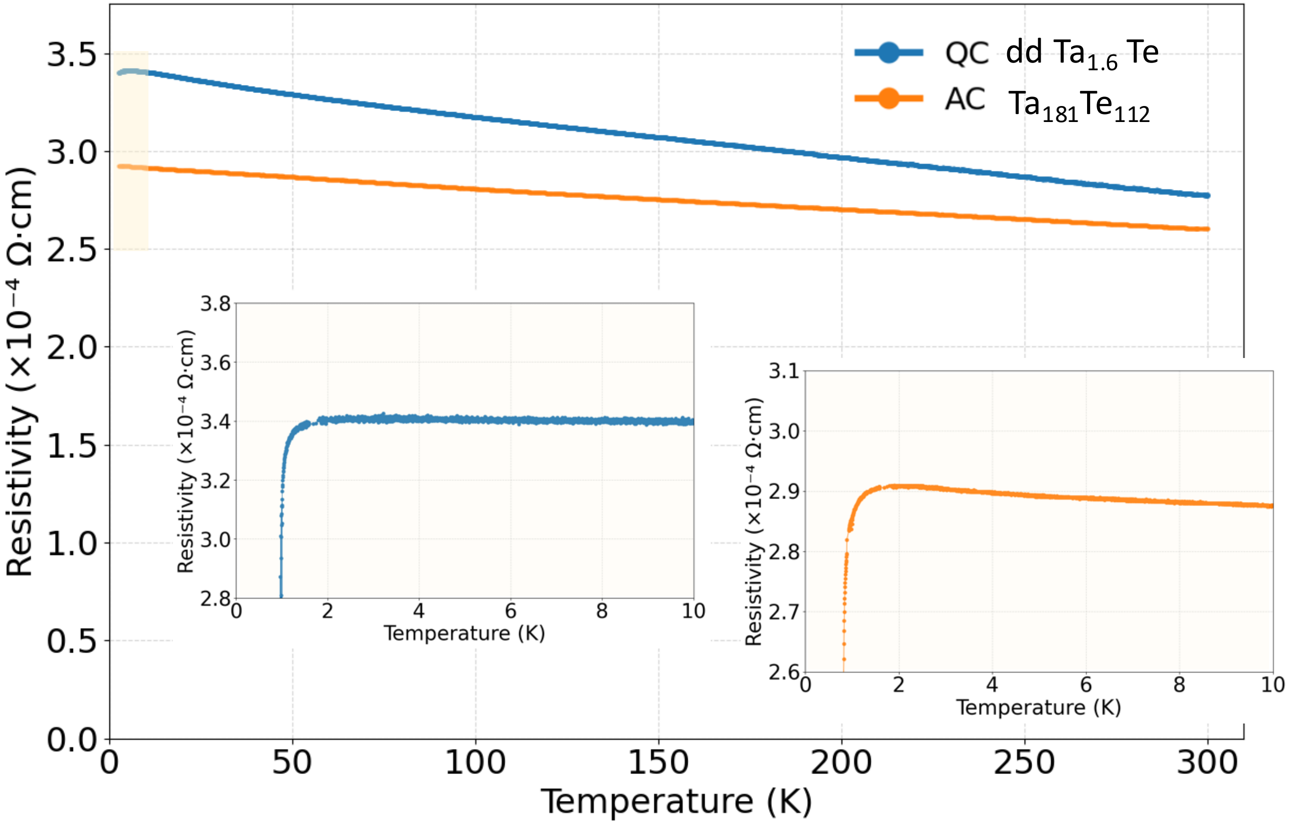
Technique advantages:
No air exposure
No heating
No solvent, no residue.



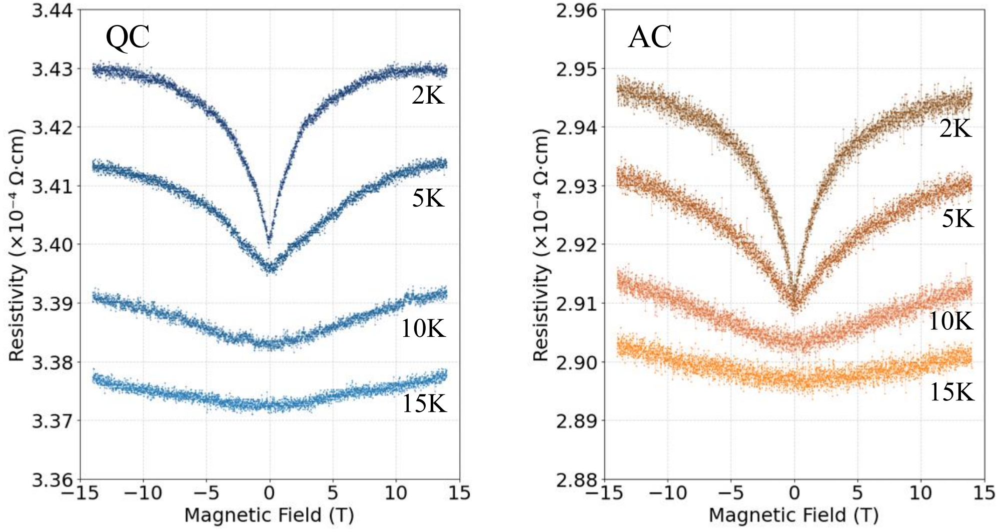


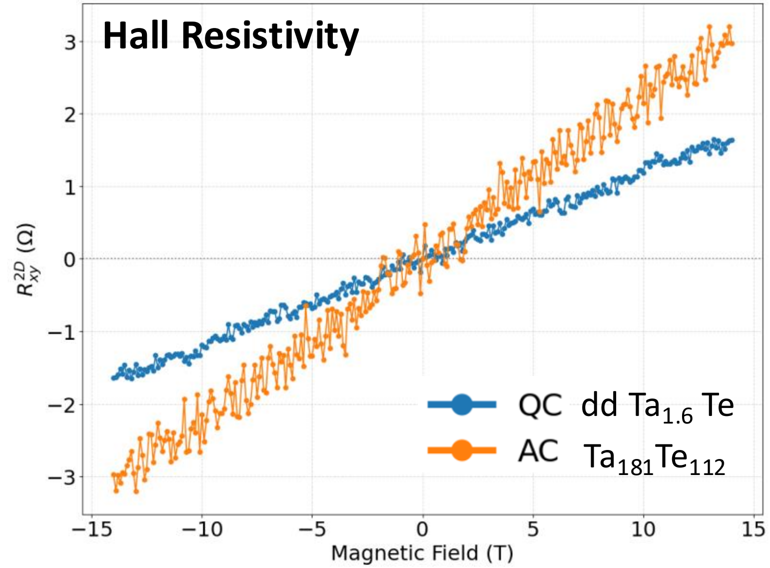
Electrical Transport Characteristics

Temperature Dependent Resistivity



Magneto Resistivity





QC: dd $Ta_{1.6}$ Te

$t\sim10\ nm, L\sim6\mu m, W\sim4\mu m$

$n = 3.506 \times 10^{22}\ cm^{-3}$

$\mu = 0.525\ cm^2/Vs$

$\boldsymbol{k_F l = 3.544}$

AC: $Ta_{181}Te_{112}$

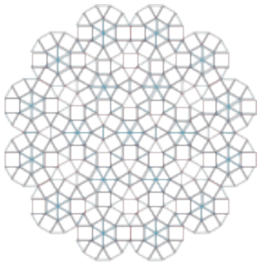
$t\sim150\ nm, L\sim8\mu m, W\sim5\mu m$

$n = 5.944 \times 10^{21}\ cm^{-3}$

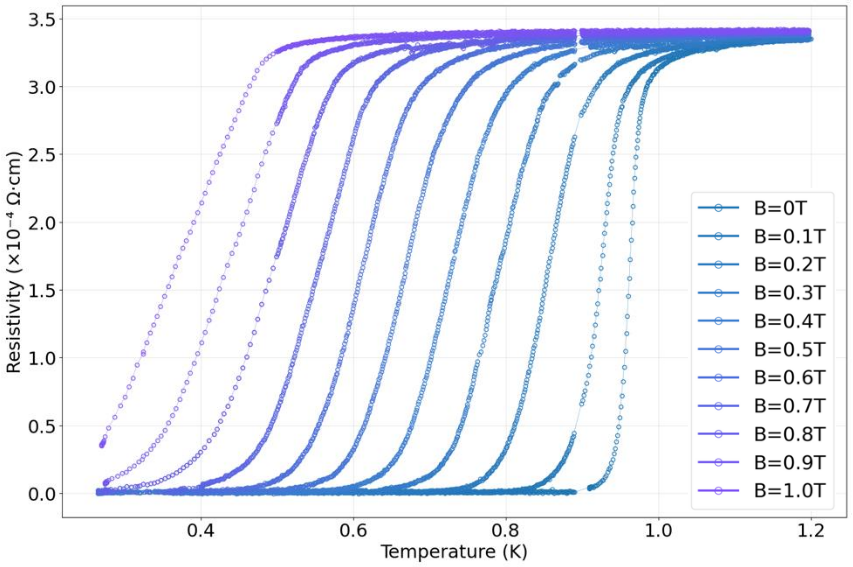
$\mu = 3.619\ cm^2/Vs$

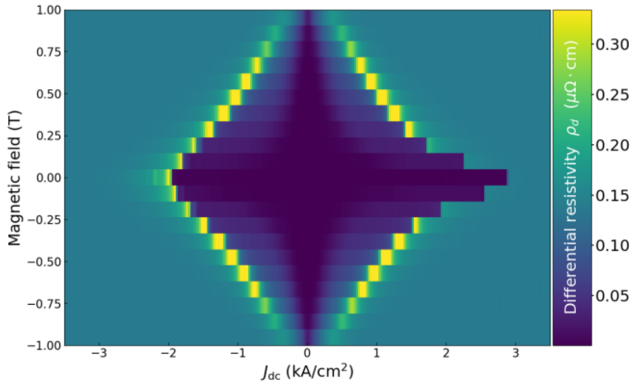
$\boldsymbol{k_F l = 7.481}$

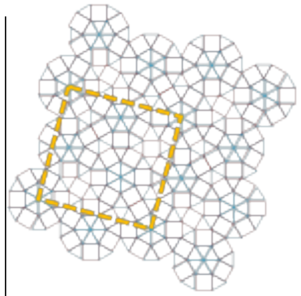
Superconductivity in QC and AC



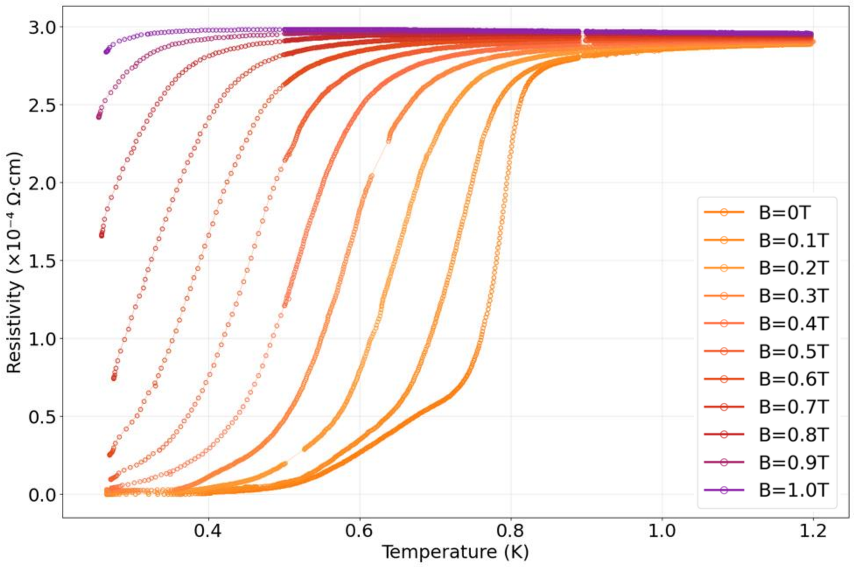
QC ($Ta_{(3+2\sqrt{3})/4}Te$)





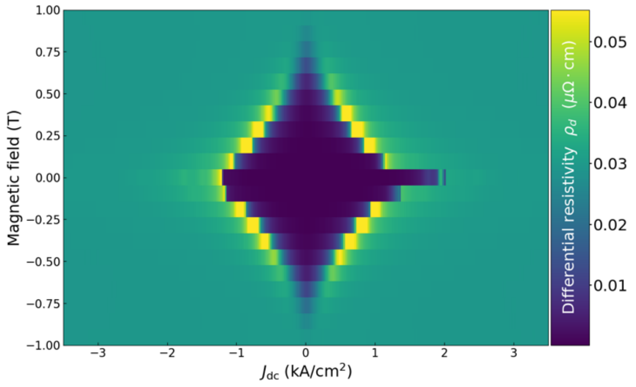


AC ($Ta_{181}Te_{112}$)



$T_c^{(QC)} > T_c^{(AC)}$

$\xi^{(QC)} < \xi^{(AC)}$

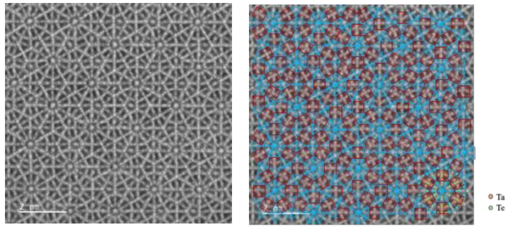


Coherence length $\xi = \sqrt{\dfrac{\Phi_0}{2\pi H_{c2}(T)}} \approx 18.2$ nm (T = 0.3K)

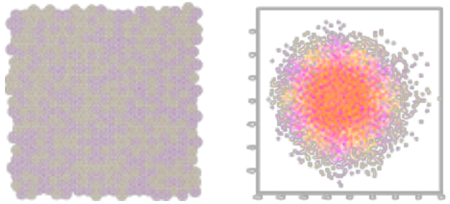
- Superconducting Tc of QC TaTe: ~0.9 K
- Superconductivity has also been observed in the approximant crystal.

Summary and Outlook

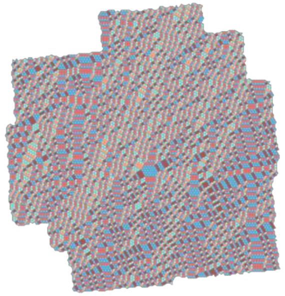
- Atomically thin 2D QC and AC TaTe samples were analyzed using TEM.



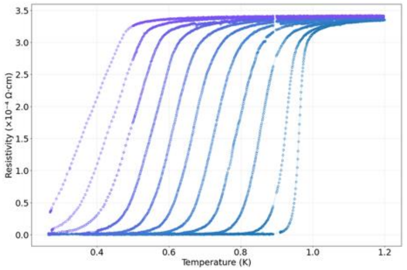
- Information from higher-dimensional space was utilized in the analysis.

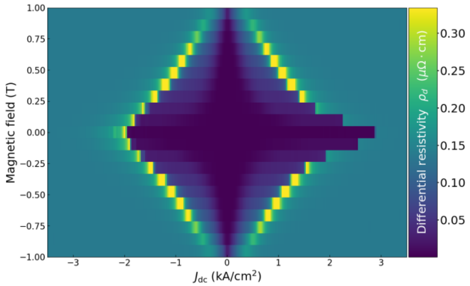


- Quasicrystals contain phason strain disorders



- Superconductivity in QC and AC has been observed

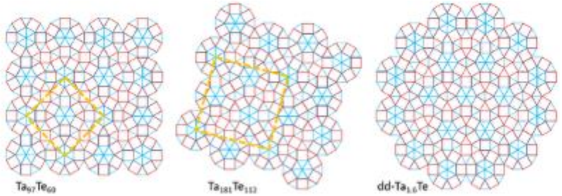




Outlook

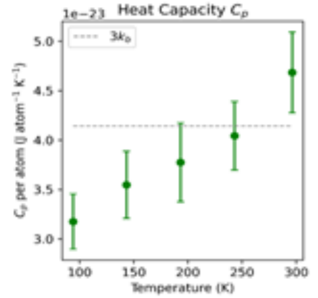
Can we control phason disorder in 2D dd-quasicrystals?

Systematic studies of ACs and QC in the 2D limits



Thermodynamic measurement: sensing higher dimensional effect

Breakdown of Dulong-Petit law in QC specific heat



Quasicrystal moire