Institute for Complex Adaptive Matter

New states of electronic matter created under moderate non-equilibrium conditions

Dragan Mihailovic

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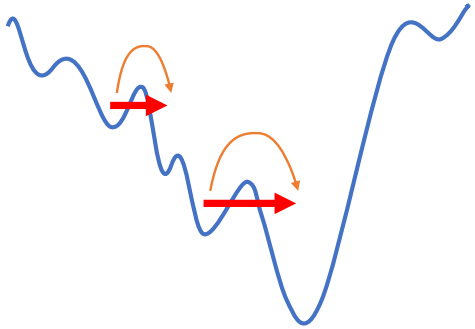
10 nm

Life exists (only) out of equilibrium

Edwin Schroedinger, What is life? (1944)



Nature Vol. 298 12 August 1982 #72 633

LETTERS TO NATURE

Is our vacuum metastable?

Michael S. Turner

Astronomy and Astrophysics Center, The University of Chicago, Chicago, Illinois 60637, USA

Frank Wilczek

Institute for Theoretical Physics, University of California, Santa Barbara, California 93106, USA

In spontaneously broken gauge theories of particle interactions there are sometimes several local minima of the effective potential. Any of these minima can serve as a vacuum in the

($V \sim 10^{84}$ cm^3), the probability that an energetically less favourable minimum has decayed during the age of the Universe is exponentially <1 so long as

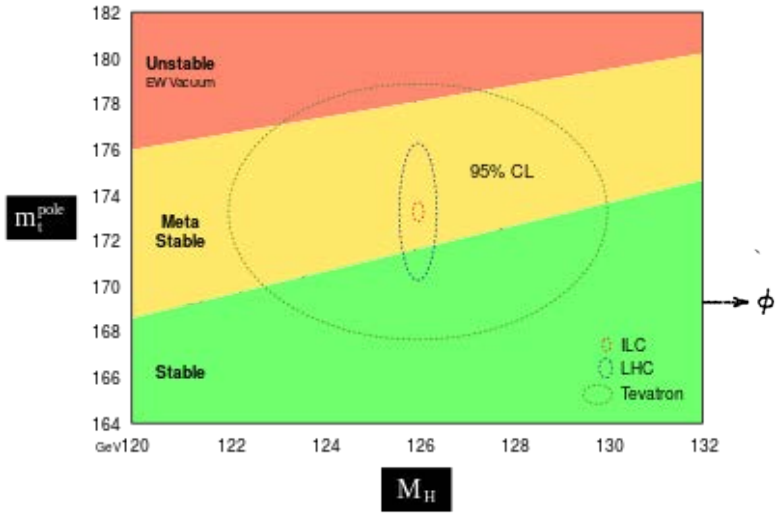
$$\varepsilon \ll 0.6\,\lambda^{-1/3} \qquad (2)$$

a condition which is easily satisfied.

We now examine the second and third issues in the context of the simplest unified model of particle interactions, the minimal SU(5) theory[7].

At zero temperature the effective potential can be written as[8,9]:

$$V(h,A) = \frac{-\mu^2}{2}\,\mathrm{tr}\,A^2 + \frac{a}{4}(\mathrm{tr}\,A^2)^2 + \frac{b}{2}\,\mathrm{tr}\,A^4 - \frac{\nu^2}{2}h^\dagger h + \frac{\lambda}{4}(h^\dagger h)^2 + \alpha(h^\dagger h)\,\mathrm{tr}\,A^2 + \beta h^\dagger A^2 h \qquad (3)$$



"False vacuum state"

Metastable states

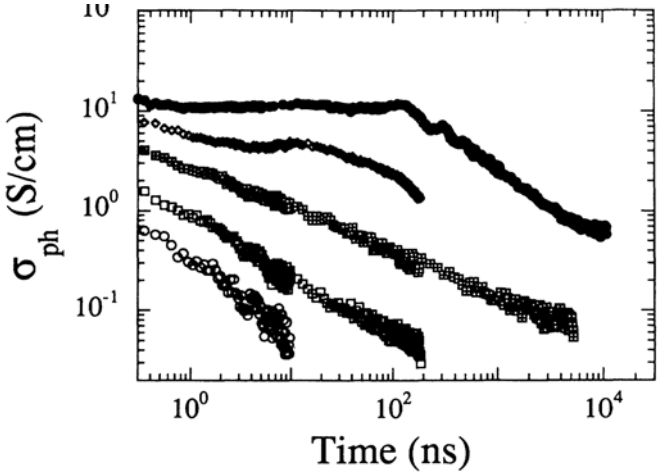
Metastability: what are the mechanisms ?

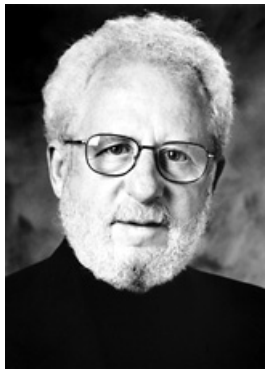
VOLUME 67, NUMBER 18 PHYSICAL REVIEW LETTERS 28 OCTOBER 1991

Transient Photoinduced Conductivity in Single Crystals of $YBa_2Cu_3O_{7-\delta}$: "Photodoping" to the Metallic State

G. Yu, C. H. Lee, and A. J. Heeger
Institute for Polymers and Organic Solids, University of California, Santa Barbara, Santa Barbara, California 93106

N. Herron and E. M. McCarron
E. I. du Pont de Nemours and Co., Inc., Central Research and Development Department, Wilmington, Delaware 19898
(Received 15 January 1991)





nitude at high excitation levels. The results are interpreted in terms of phase separation and metallic-droplet formation. A longitudinal magnetic field (≤ 5 kG) reduces both the resistivity minimum and the superlinear contribution to the transient photocurrent.

Squeezing phase diagrams towards metastability in $La_{2/3}Ca_{1/3}MnO_3$

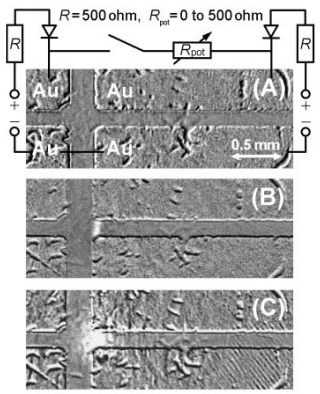
VOLUME 78, NUMBER 22 PHYSICAL REVIEW LETTERS 2 JUNE 1997

Photoinduced Insulator-to-Metal Transition in a Perovskite Manganite

K. Miyano,[1] T. Tanaka,[1] Y. Tomioka,[2] and Y. Tokura[1,2]
[1]*Department of Applied Physics, University of Tokyo, Bunkyo-ku, Tokyo 113, Japan*
[2]*Joint Research Center for Atomic Technology (JRCAT), Tsukuba 305, Japan*
(Received 3 March 1997)

Science AAAS

Visualization of the Local Insulator-Metal Transition in $Pr_{0.7}Ca_{0.3}MnO_3$
Manfred Fiebig, *et al.*
Science **280**, 1925 (1998);
DOI: 10.1126/science.280.5371.1925

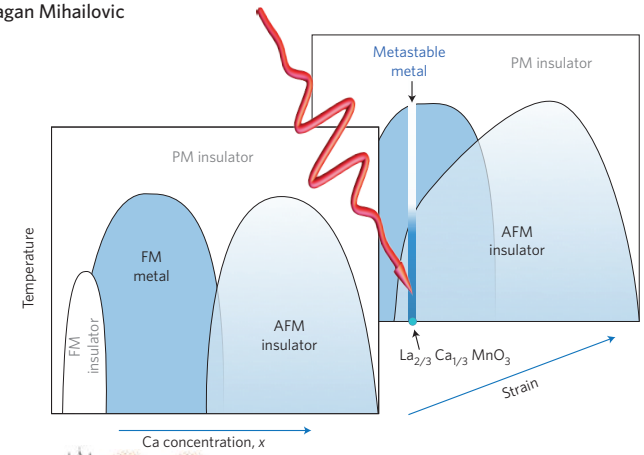


MANGANITE FILMS

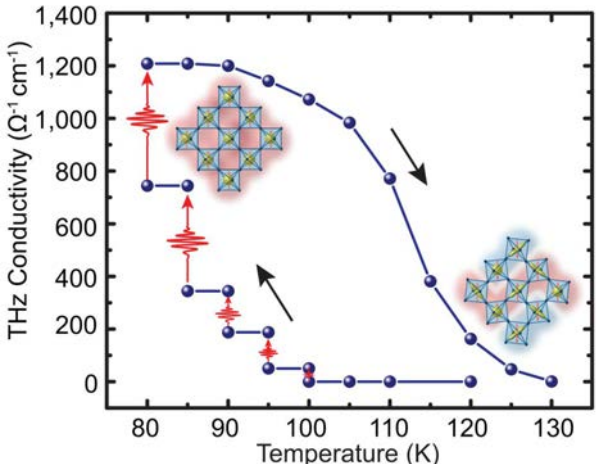
Tuning phase diagrams

Strain engineering can tune a manganite film into an antiferromagnetic insulating state whose extreme photo-susceptibility allows for the ordinary ferromagnetic metal state to then be transiently realized.

Dragan Mihailovic



Nature Materials **15,** 930–931 (2016).



Cooperative photoinduced metastable phase control in strained manganite films

Jingdi Zhang et al.

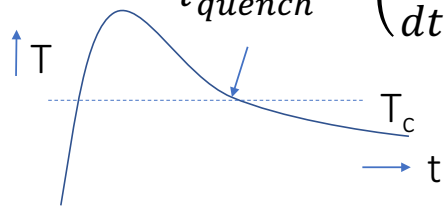
Nature Materials **15**, 956–960 (2016)

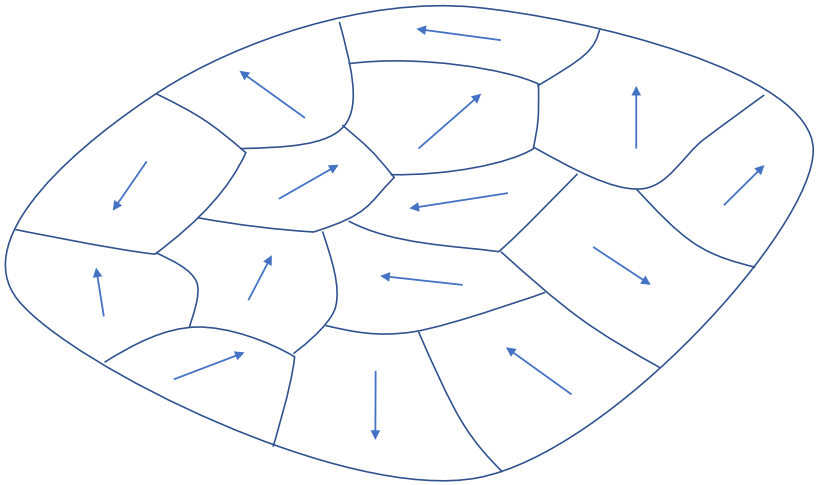
J. Phys. A: Math. Gen., Vol. 9, No. 8, 1976. Printed in Great Britain. © 1976

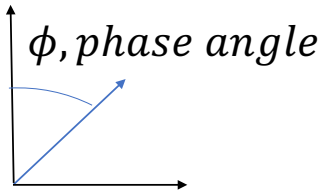
Topology of cosmic domains and strings

T W B Kibble
Blackett Laboratory, Imperial College, Prince Consort Road, London SW7 2BZ, UK

$$\tau_{quench} \sim \left(\frac{dT}{dt}\right)^{-1} T_c$$







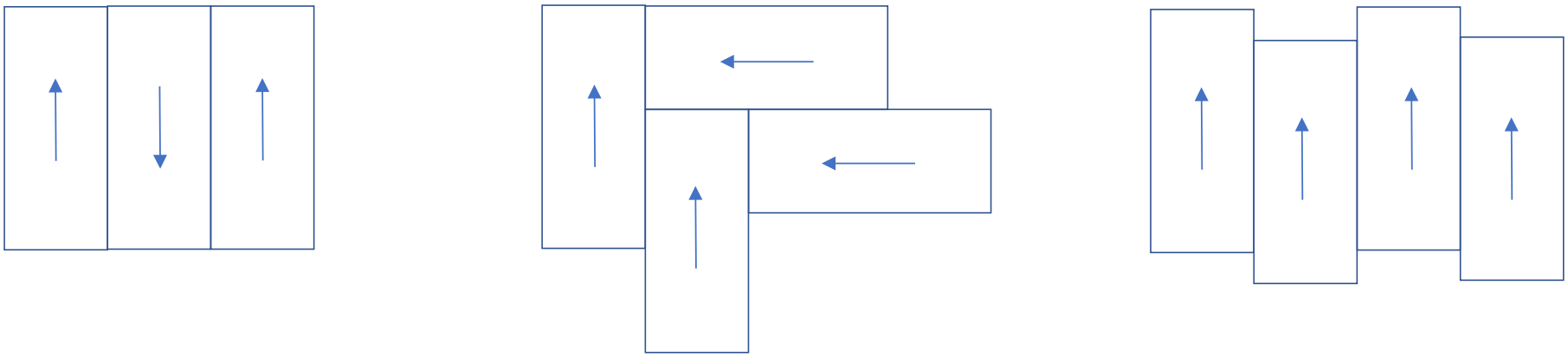
$$\Psi = Ae^{i\phi}$$

L (e.g. laser spot)

$\xi < L$

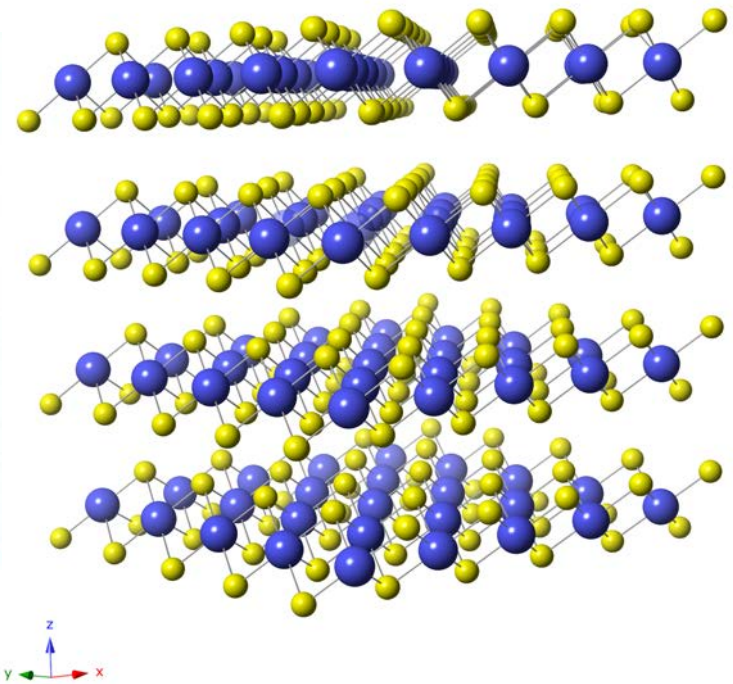
If $\tau_{quench} < L/v$, multiple domains form (Kibble-Zurek mechanism)

Just domains, or new emergent states?



1T-TaS_2

Crystals of 1T-TaS_2 grown by Petra Sutar



A system with competing Coulomb, Fermi surface instability, spin frustration and lattice strain and quantum tunneling...and more.

Experiments:

Ljupka Stojchevska

Tomaz Mertelj

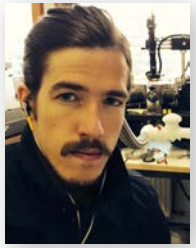
Igor Vaskivskyi

Yaroslav Gerasimenko

Venera Nasretdinova

Jan Ravnik

Ian Mihailovic



+Maksim Litskevich

Theory:

Serguei Brazovskii *(Univ. Paris Sud Orsay)*

Viktor Kabanov

Samples:

Petra Sutar *(JSI)*

Lithography:

Damjan Svetin *(JSI)*

UED, Time resolved ARPES, LEED

Hermann Durr

Patrick Kirchman

Stanford/SIMES/LCLS

SLAC:

Tyler Chase
Alex Reid
Renkai Li
Xijie Wang

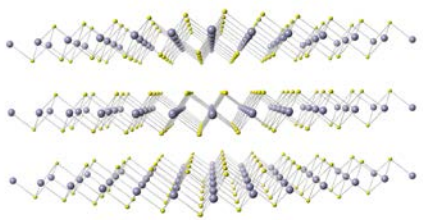
Loïc le Guyader

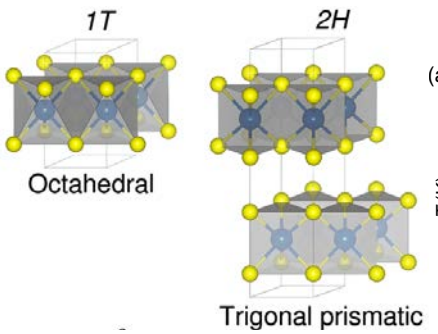
Jonathan Sobota

Equilibrium

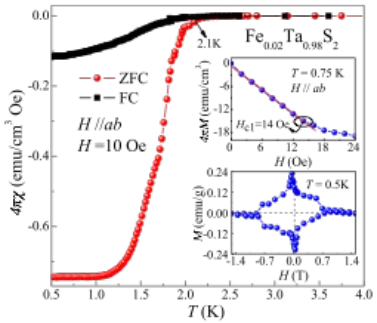
Competing phases in TaS_2: Multiple structural and charge-density-wave states, superconductivity under pressure, and/or (Fe,Cu,Na or Se…?) doping.

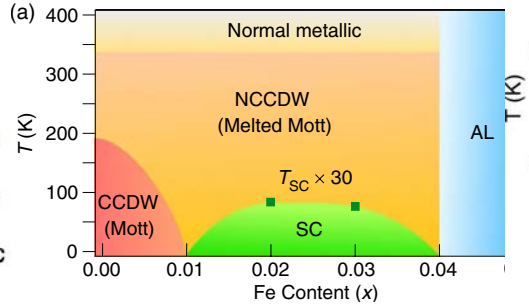
The 1T polytype of TaS_2 is metastable ($T_{c\uparrow}$=550K, $T_{c\downarrow}$=1100K)



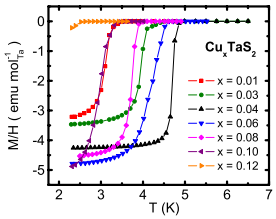


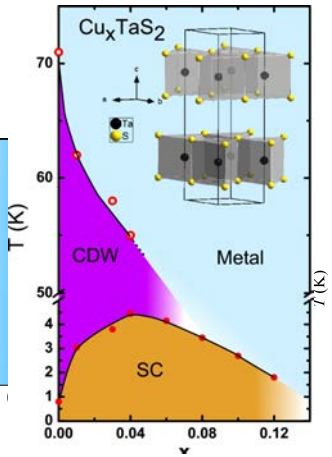
Fe : 1T-TaS_2 (substitution):





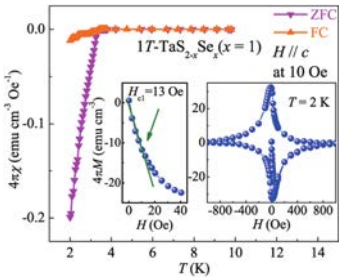
Cu : 2H-TaS_2 (substitution):

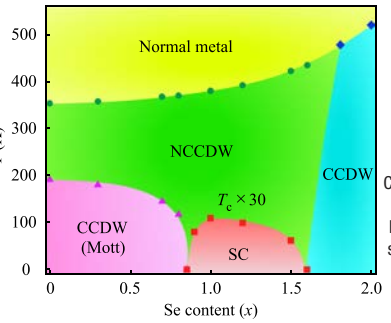




Wagner et al (PRBB 2008)

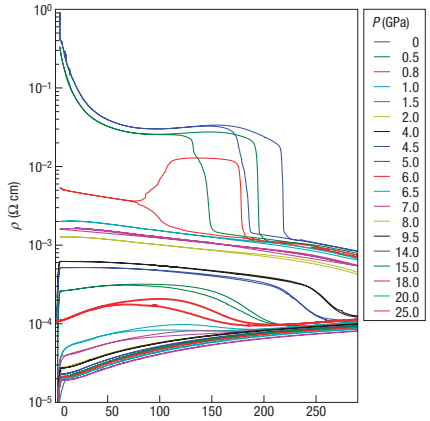
Chemical pressure, SOC: Se substitution:

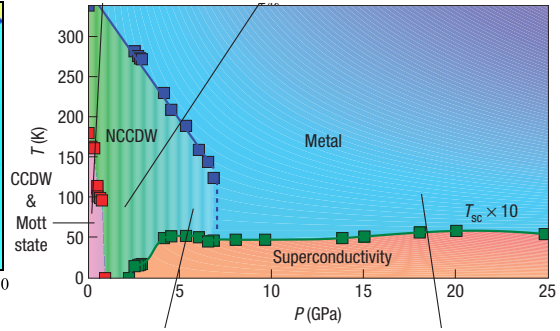




Liu et al. APL 2013

Physical pressure: 1T-TaS_2

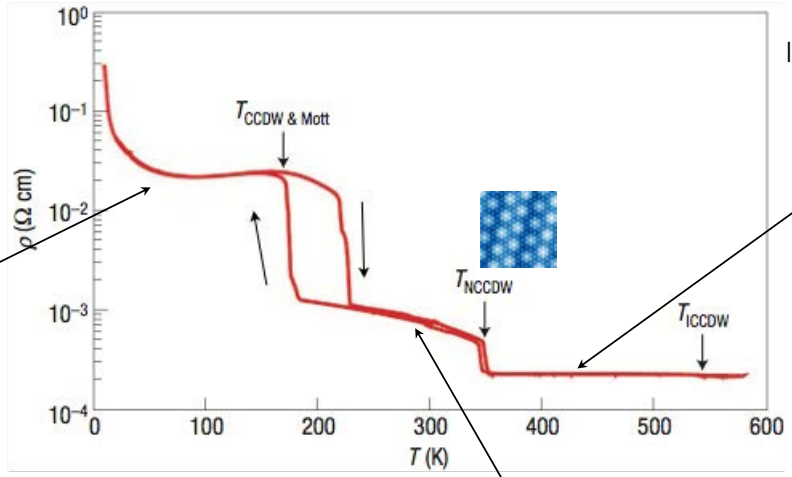




Sipos et al (Nat.Mat. 2008)

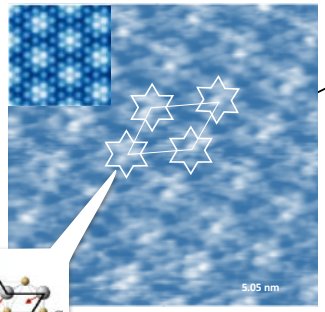
Resistivity of *1T*-TaS_2 through CDW phases under equilibrium conditions

Sipos et al, *Nature Materials* **7,** 960–965 (2008).

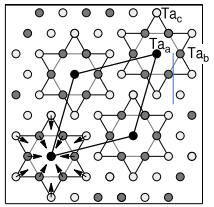


Mott state

STM : Commensurate CDW

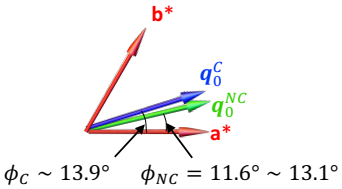


Polaron

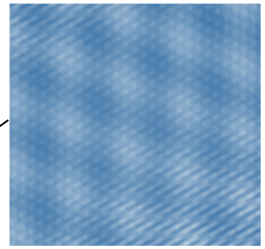


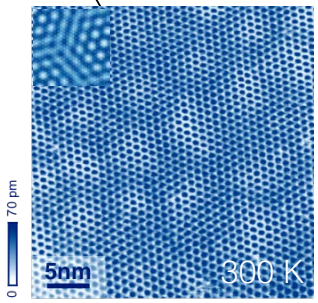
K. Rossnagel, JPCM **23** 213001 (2011)

Ordering vector directions



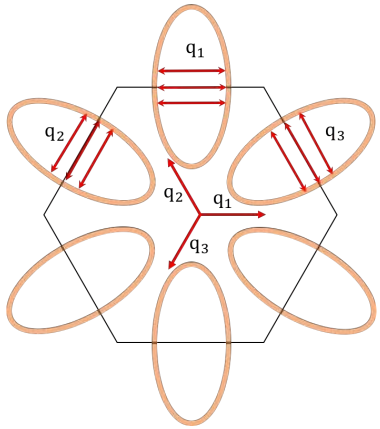
Incommensurate CDW





STM : Commensurate CDW

Stretched honeycomb structure with domain walls in between (Nearly Commensurate CDW phase)

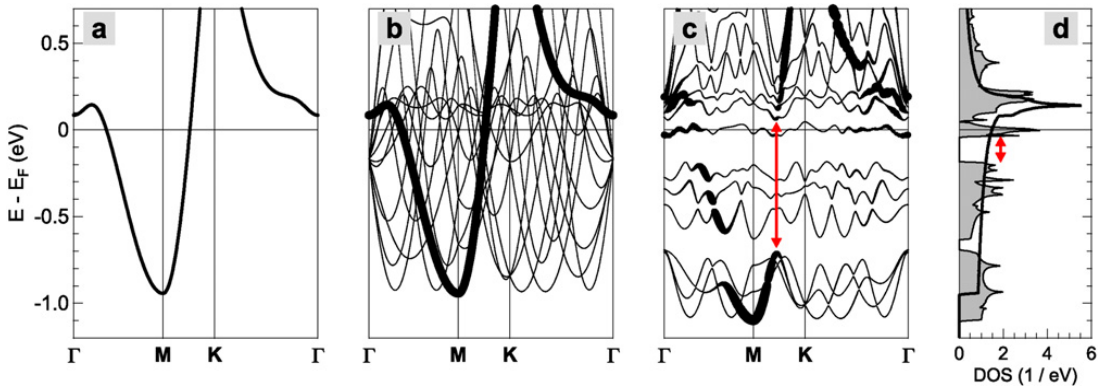


Fermi surface nesting

Tosatti, E. & Fazekas, P. On the nature of the low-temperature phase of 1T-TaS2. *J. Phys. Colloques* **37,** C4–165–C4–168 (1976).

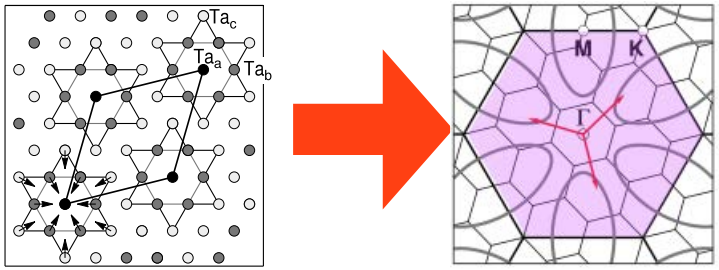
The electronic structure of *1T*-TaS_2 in the low temperature **commensurate** state

1T-TaS_2 ($T>T_{c0}$) → + C CDW ($\sqrt{13} \times \sqrt{13}$) → + Spin-orbit c.



Unit cell of $\sqrt{13} \times \sqrt{13}$ superlattice

Rossnagel et al (2011)



B.Z. of 1T-TaS_2 ($T>T_{c0}$)

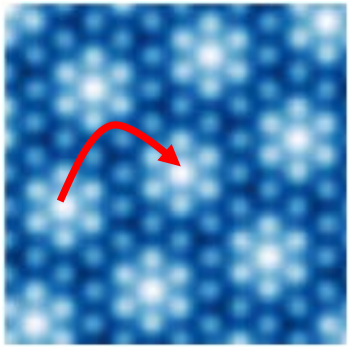
Rossnagel, K. *J Phys-Condens Mat* **23,** 213001 (2011).

Mott physics?

JOURNAL DE PHYSIQUE *Colloque C4, supplément au n° 10, Tome 37, Octobre 1976, page C4-165*

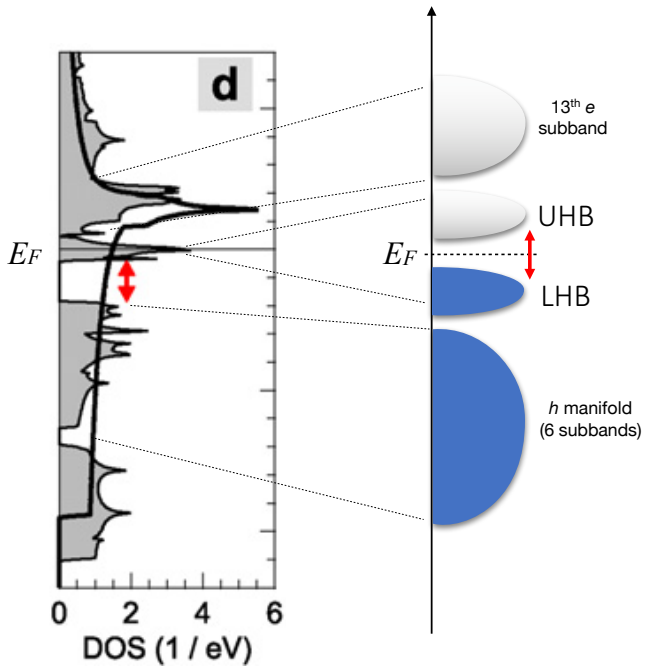
ON THE NATURE OF THE LOW-TEMPERATURE PHASE OF 1T-TaS_2

E. TOSATTI
Istituto di Fisica, Universitá degli Studi Roma, Italy
and P. FAZEKAS
Central Research Institute for Physics, Budapest, Hungary

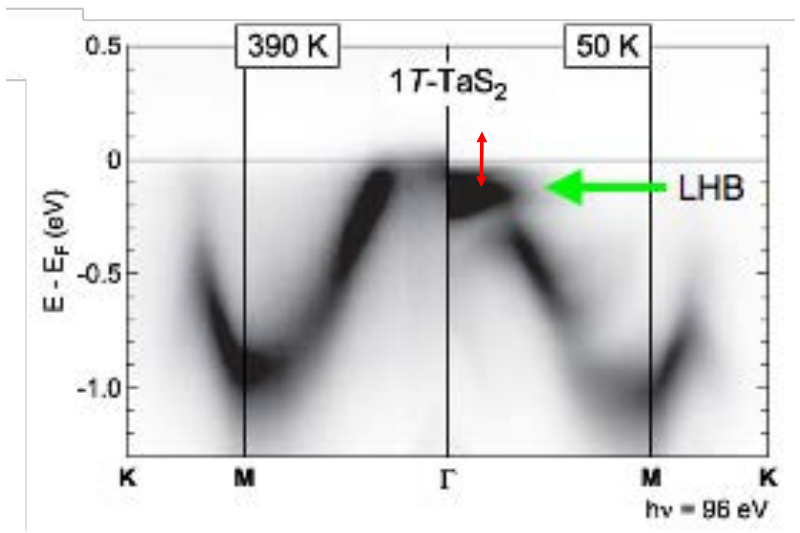


½-filled band

+ Coulomb repulsion
(Mott state)



Gap by ARPES:



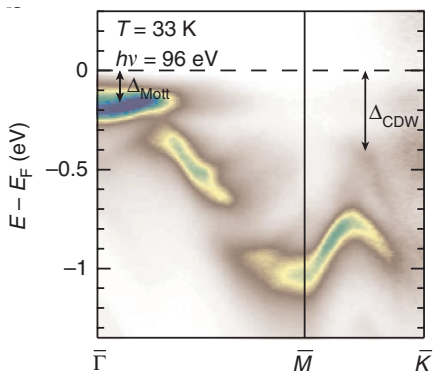
Rossnagel, K. *J Phys-Condens Mat* **23,** 213001 (2011).

What is the spin ground state of 1T-TaS_2?

Charge

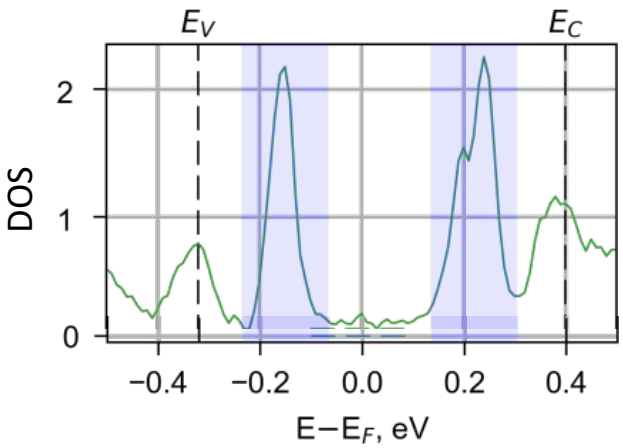
ARPES

Hellmann et al, Nat. Comm. 2012



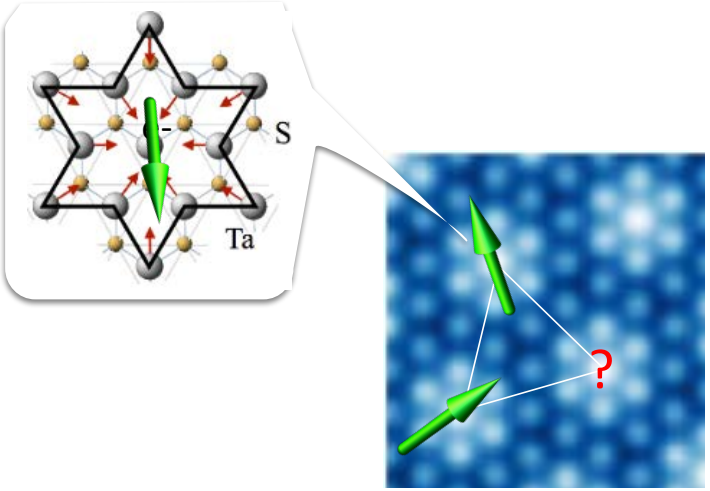
STM

Gerasimenko, 2017

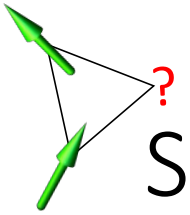


Charge gap $\simeq 0.3$ eV

Spin?



Polaron (CDW) lattice



Spins on triangular lattices

Materials Research Bulletin
Volume 8, Issue 2, February 1973, Pages 153-160

Resonating valence bonds: A new kind of insulator?

P.W. Anderson [a, b]

Show more

https://doi.org/10.1016/0025-5408(73)90167-0

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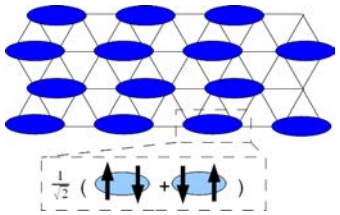
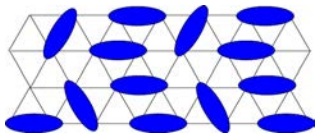
Abstract

The possibility of a new kind of electronic state is pointed out, corresponding roughly to Pauling's idea of "resonating valence bonds" in metals. As observed by Pauling, a pure state of this type would be insulating; it would represent an alternative state to the Néel antiferromagnetic state for S = 1/2. An estimate of its energy is made in one case.



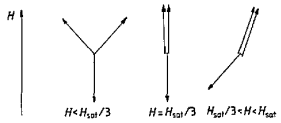
?



J. Phys.: Condens. Matter 3 (1991) 69–82.

Quantum theory of an antiferromagnet on a triangular lattice in a magnetic field

A V Chubukov and D I Golosov
Institute for Physical Problems, USSR Academy of Sciences, 117334 ul. Kosygina 2, Moscow, USSR



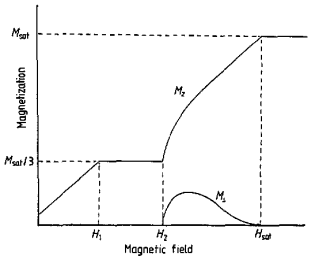
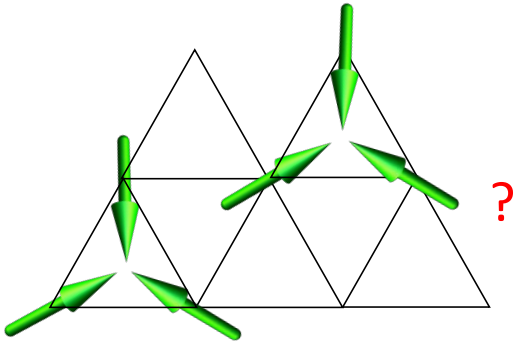


Figure 5. The field dependence of the longitudinal and transverse magnetizations in 2D classical Heisenberg AFM with easy-axis anisotropy on the triangular lattice (see equations (27) and (28)).

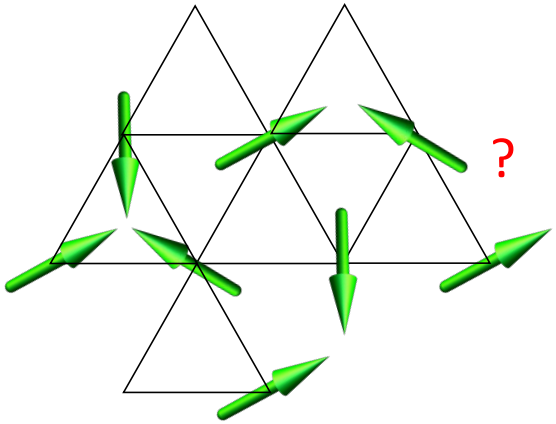
VOLUME 82, NUMBER 19 PHYSICAL REVIEW LETTERS 10 MAY 1999

Long-Range Néel Order in the Triangular Heisenberg Model

Luca Capriotti,[1] Adolfo E. Trumper,[1,2] and Sandro Sorella[1]



?

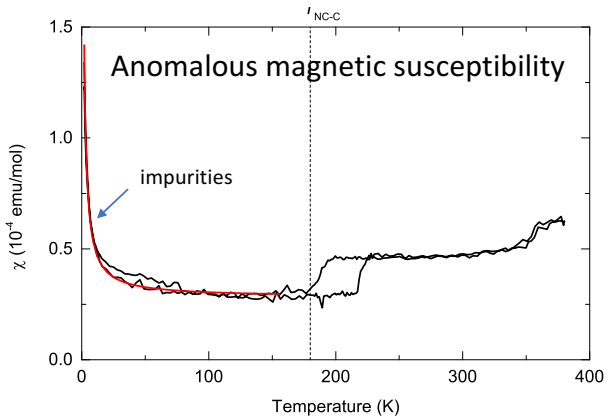


?

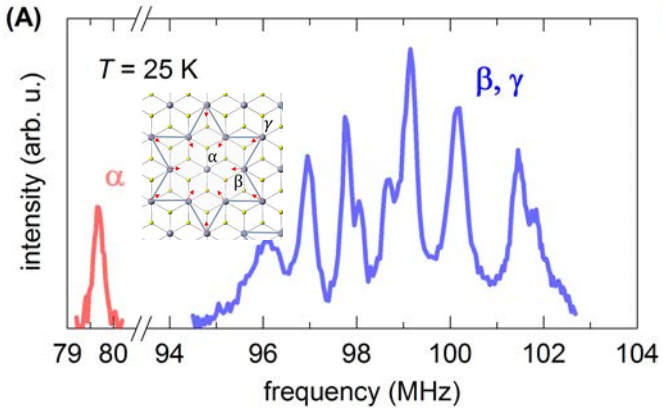
nature physics
ARTICLES
PUBLISHED ONLINE: 31 JULY 2017 | DOI: 10.1038/NPHYS4212

A high-temperature quantum spin liquid with polaron spins

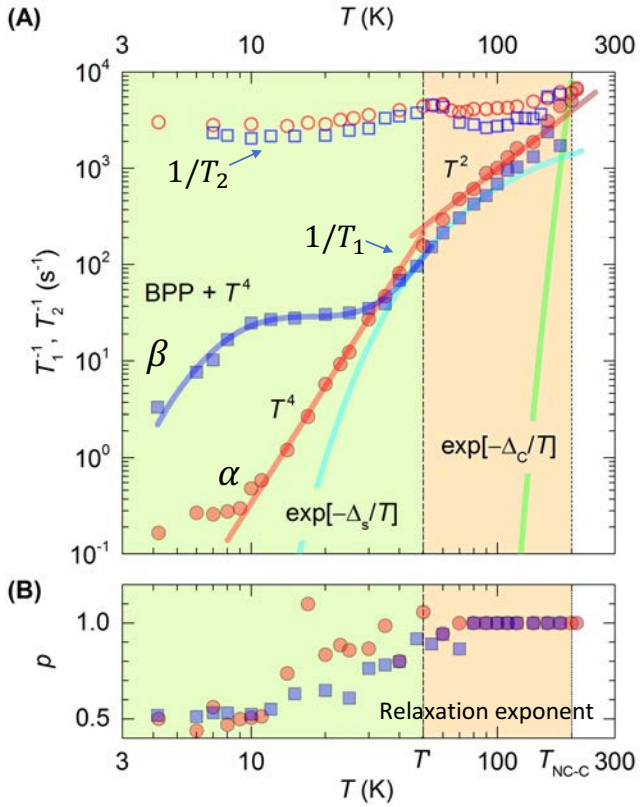
Martin Klanjšek[1], Andrej Zorko[1], Rok Žitko[1], Jernej Mravlje[1], Zvonko Jagličić[2,3], Pabitra Kumar Biswas[4], Peter Prelovšek[1,5], Dragan Mihailovic[1,5] and Denis Arčon[1,5]*



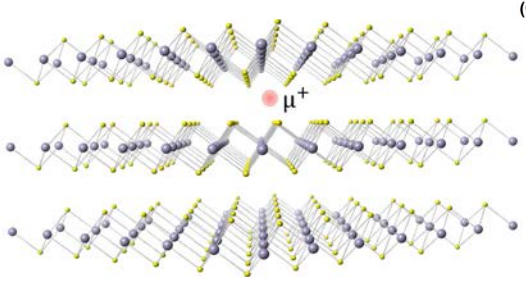
NQR fequencies, by site in a polaron cluster

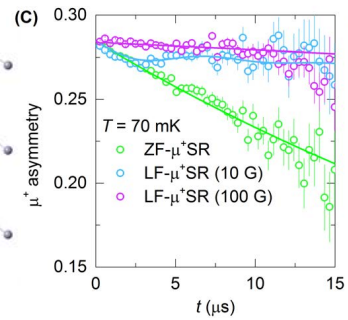


NQR Spin relaxation is gapless!

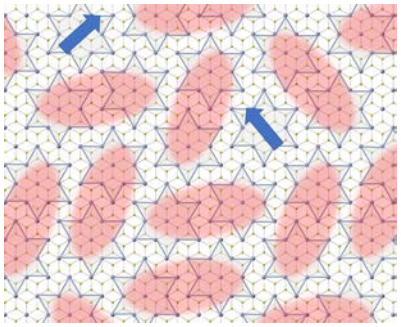


μSR shows no magnetic order down to 70mK





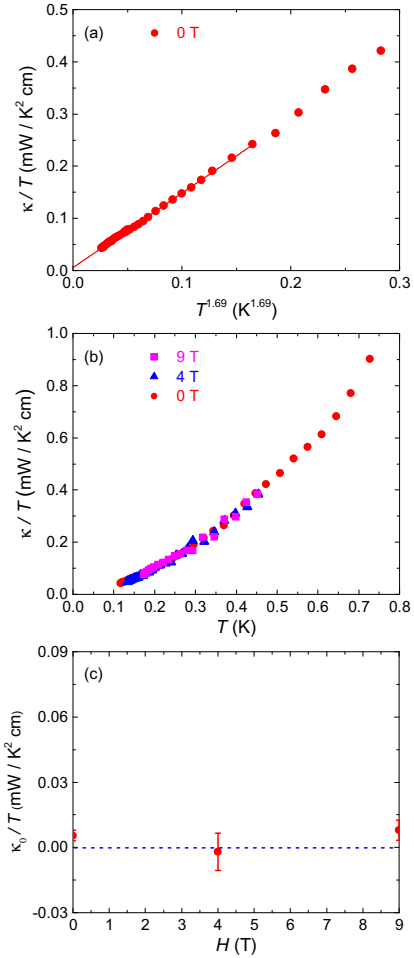
RVB state with defects (impurities)?



PHYSICAL REVIEW B **96**, 081111(R) (2017)

Heat transport study of the spin liquid candidate 1*T*-TaS_2

Y. J. Yu,[1] Y. Xu,[1] L. P. He,[1] M. Kratochvilova,[2,3] Y. Y. Huang,[1] J. M. Ni,[1] Lihai Wang,[4] Sang-Wook Cheong,[4] Je-Geun Park,[2,3] and S. Y. Li[1,5,*]



Linear term in κ is independent of H

PNAS

1T-TaS_2 as a quantum spin liquid

K. T. Law[a] and Patrick A. Lee[b,1]

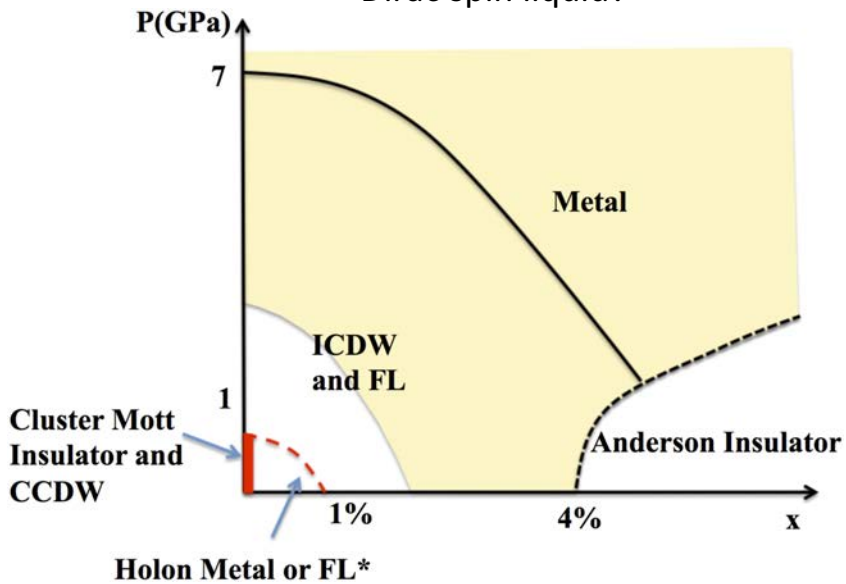
[a]Department of Physics, Hong Kong University of Science and Technology, Hong Kong, China; and [b]Department of Physics, Massachusetts Institute of Technology, Cambridge MA 02139

Contributed by Patrick A. Lee, May 26, 2017 (sent for review April 24, 2017; reviewed by Steven A. Kivelson and N. Phuan Ong)

1T-TaS_2 is unique among transition metal dichalcogenides in that it is understood to be a correlation-driven insulator, where the unpaired electron in a 13-site cluster experiences enough correlation to form a Mott insulator. We argue, based on existing data,

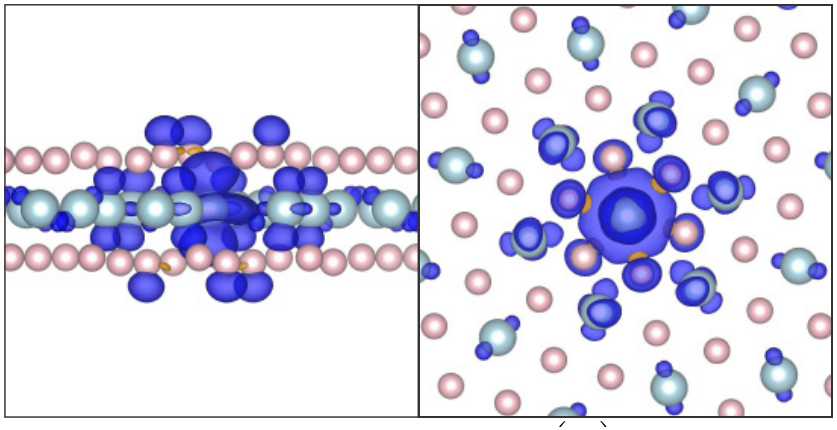
Mott insulator. This fact was pointed out by Fazekas and Tosatti (8) in 1976. Band calculations show that band folding creates a cluster of bands near the Fermi surface. Rossnagel and Smith (9) found that, due to spin–orbit interaction, a very narrow band

Dirac spin liquid?



The magnetic state of C-1T-TaS_2

$$W_{\perp} \gg U_{in-plane}$$



Interlayer magnetic coupling

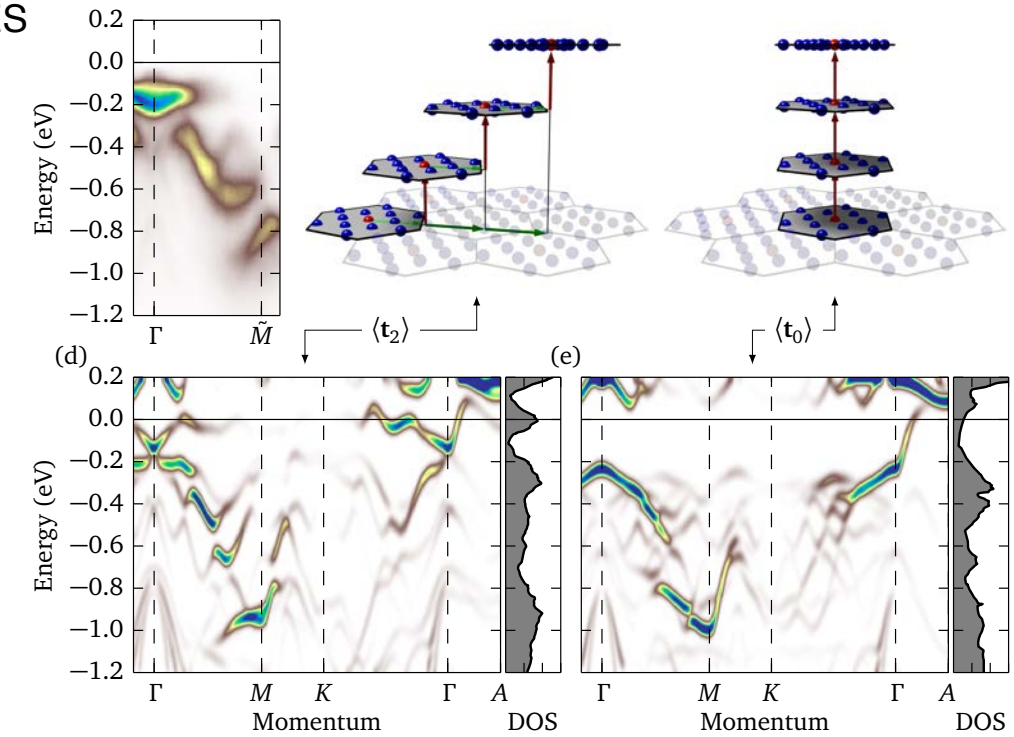
delocalized across the SD. The monolayer compounds are predicted to be Mott insulators with a $S = 1/2$ degree of freedom in each unit cell of the CDW structure, while the bilayers form a singlet state with a tunable optical gap. ?

Darancet, Millis & Marianetti.
Three-dimensional metallic and two-dimensional insulating behavior in octahedral tantalum dichalcogenides. ***PRB* 90,** 045134 (2014).

Out-of-plane orbitronics?

ARPES

LDA

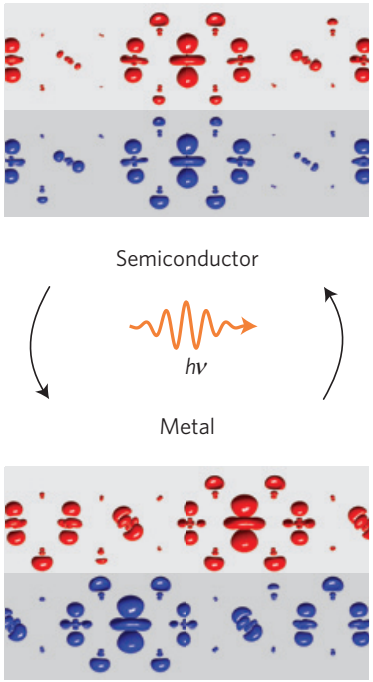


LETTERS
PUBLISHED ONLINE: 16 MARCH 2015 | DOI: 10.1038/NPHYS3267
nature physics

Nat Phys 11, 328–331 (2015).

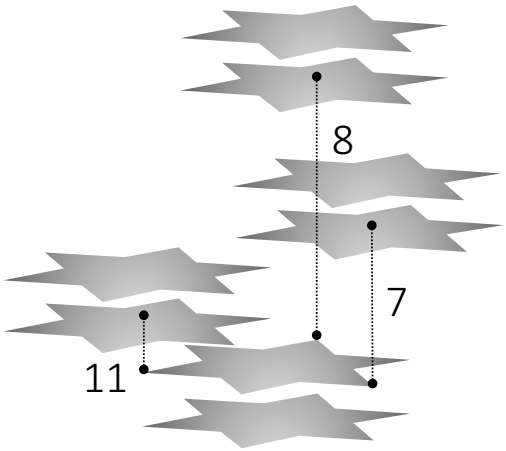
Orbital textures and charge density waves in transition metal dichalcogenides

T. Ritschel[1,2]*, J. Trinckauf[1], K. Koepernik[1], B. Büchner[1,2], M. v. Zimmermann[3], H. Berger[4], Y. I. Joe[5], P. Abbamonte[5] and J. Geck[1]*



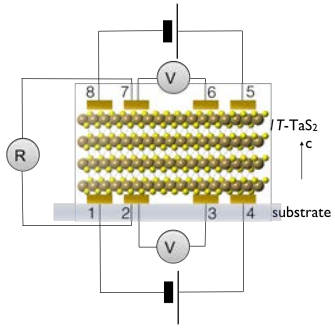
Experiment: c-axis stacking and transport

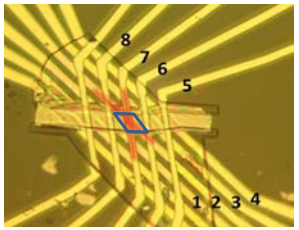
Helical C CDW order from HR-TEM:

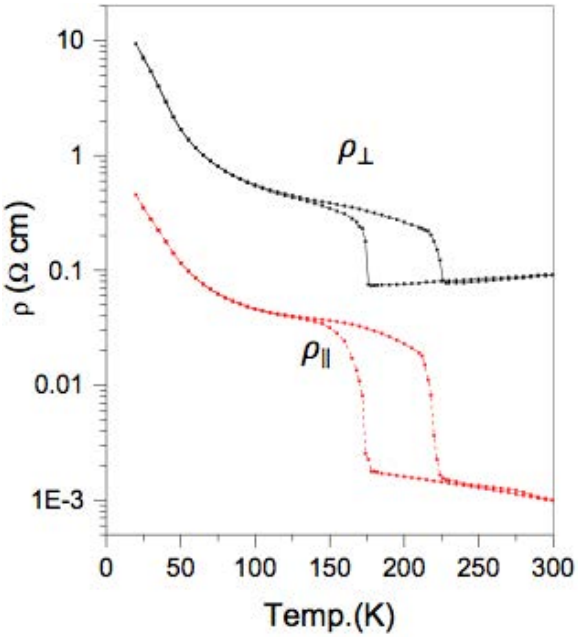


Ishiguro, T. & Sato, H.
Phys. Rev. B (1991)

Resistivity along c axis:



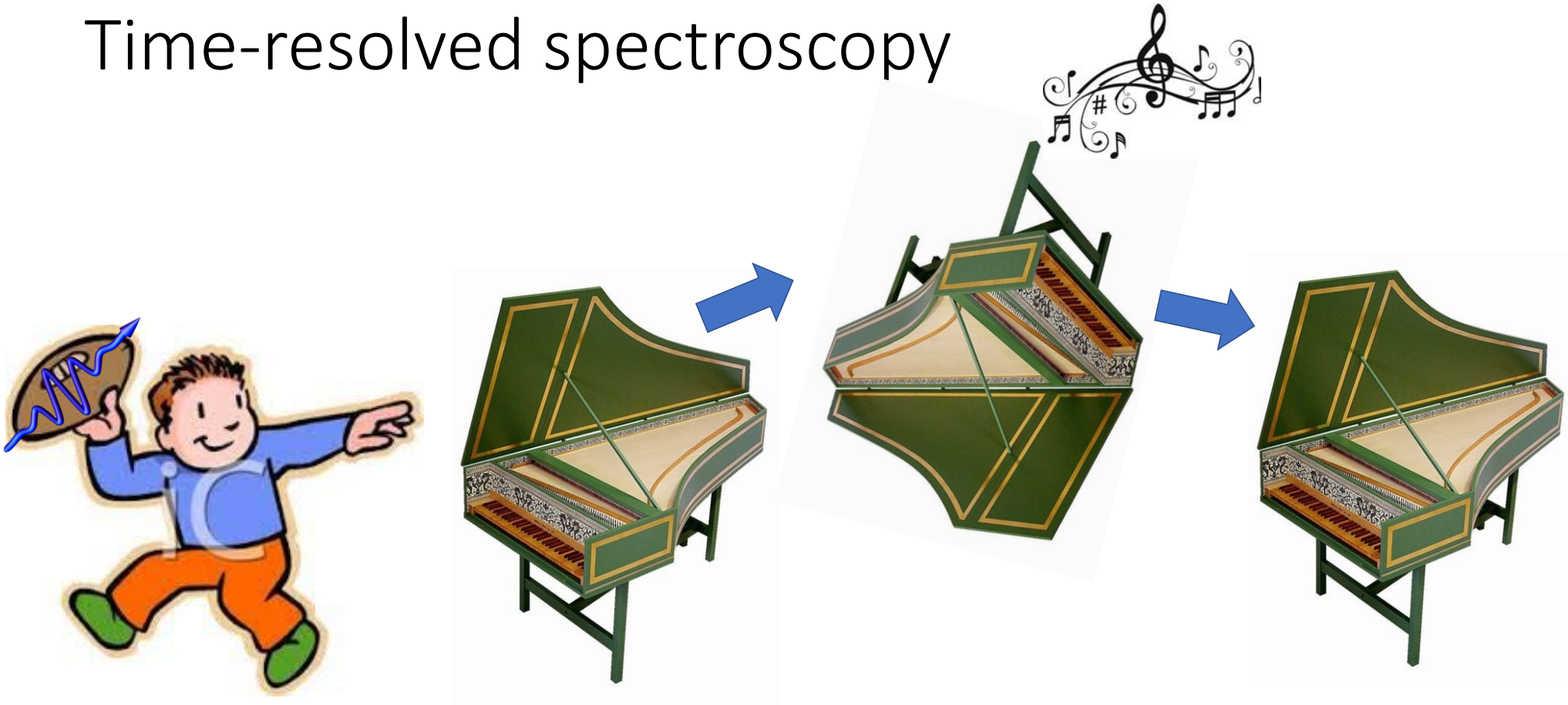




Svetin et al., Sci Rep **7,** 46048 (2017).

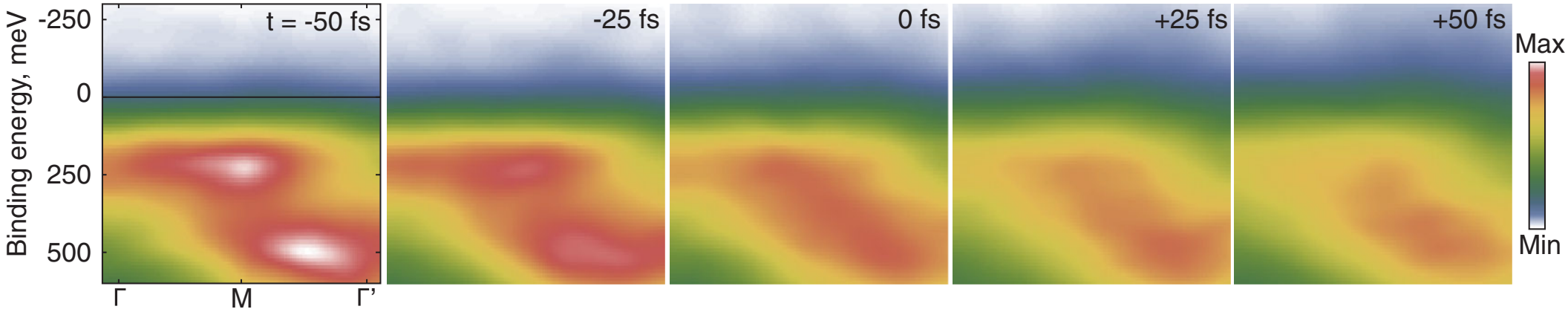
Measurements are not consistent with predicted band picture for out-of-plane transport

Time-resolved spectroscopy



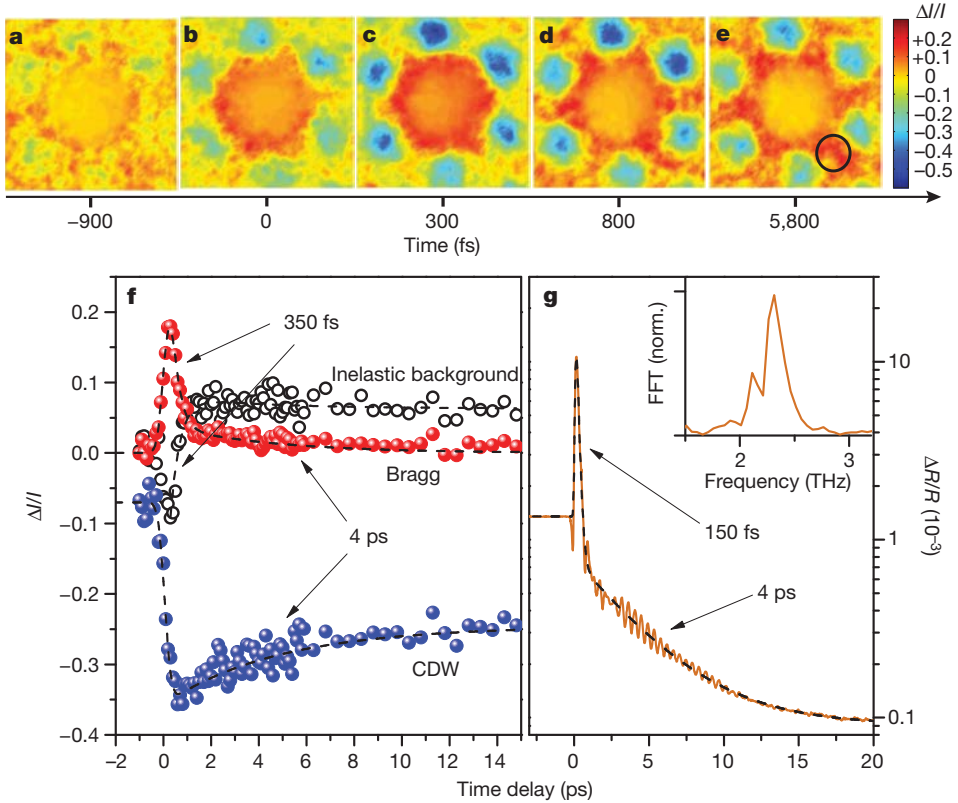
“Melting of Mott gap and CDW”

Time-resolved ARPES



J. C. Petersen et al, PRL, **107**, 177402 (2011).

Snapshots of cooperative atomic motions in the optical suppression of charge density waves at the N-IC transition

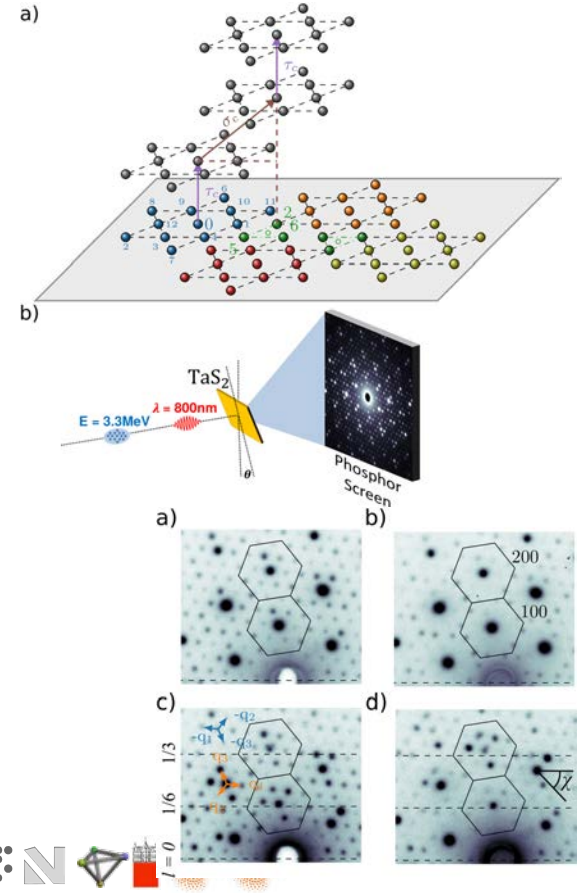


Eichberger, M. et al. Snapshots of cooperative atomic motions in the optical suppression of charge density waves. **Nature** 468, 799–802 (2010).

STRUCTURAL DYNAMICS **4**, 044020 (2017)

Stacking order dynamics in the quasi-two-dimensional dichalcogenide 1 *T*-TaS_2 probed with MeV ultrafast electron diffraction

L. Le Guyader,[1,2,a)] T. Chase,[1,3] A. H. Reid,[1] R. K. Li,[1] D. Svetin,[4] X. Shen,[1] T. Vecchione,[1] X. J. Wang,[1] D. Mihailovic,[4] and H. A. Dürr[1,b)]



C→NC Dynamics :

Melting of C phase:

$$\tau^{C}_{melt} \simeq 0.3 \text{ ps}$$

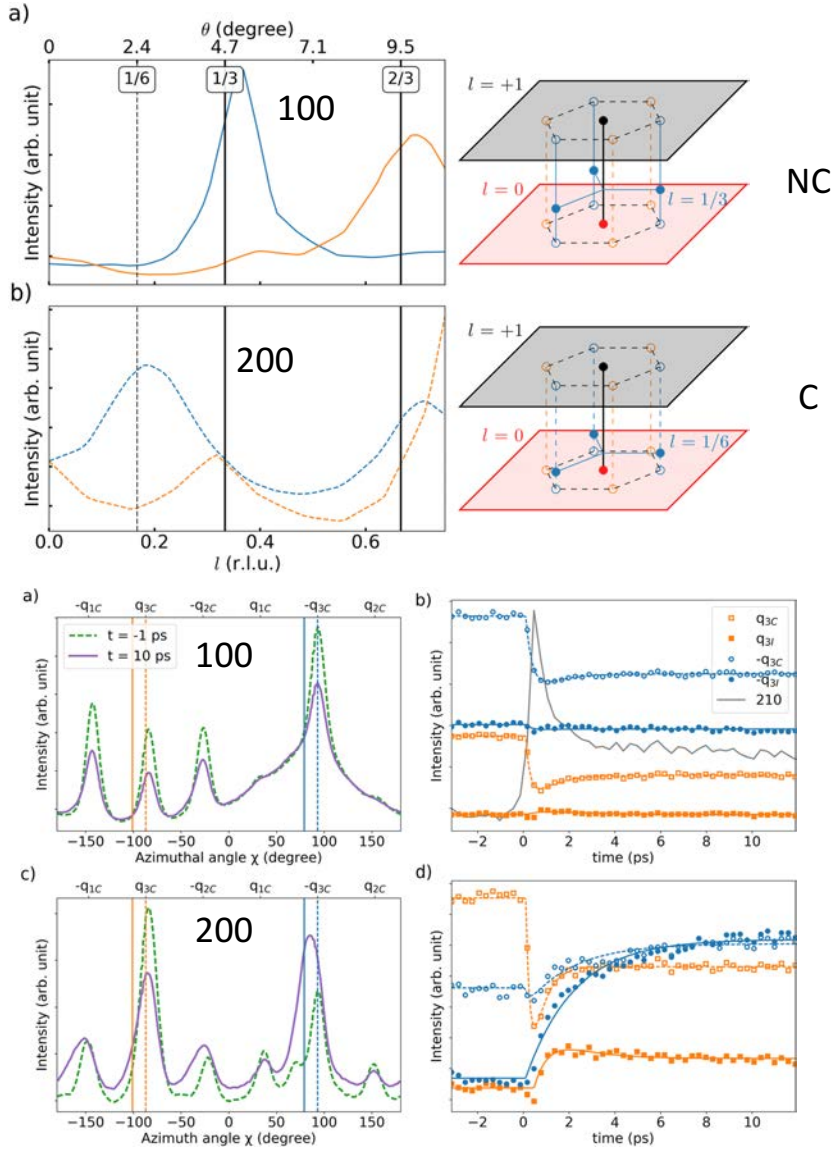
Re-stacking in NC phase:

$$\tau^{NC}_{restoration} \simeq 2 \text{ ps}$$

Dynamical (re)stacking

$$T = 140K,$$
$$\Phi = 3\ mJ/cm^2$$

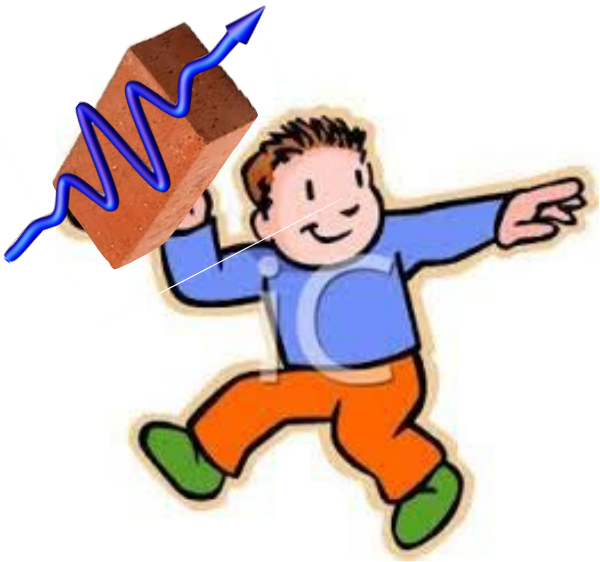
Equilibrium stacking



NC

C

Emergent states…

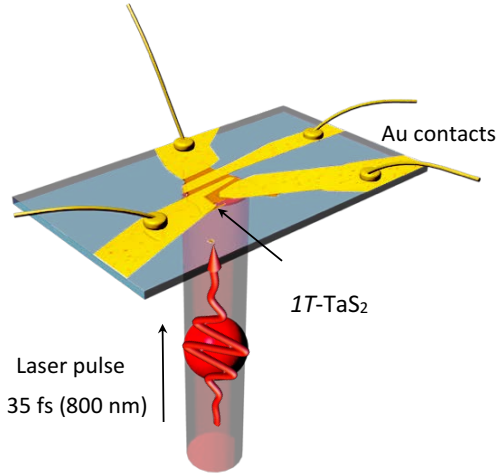


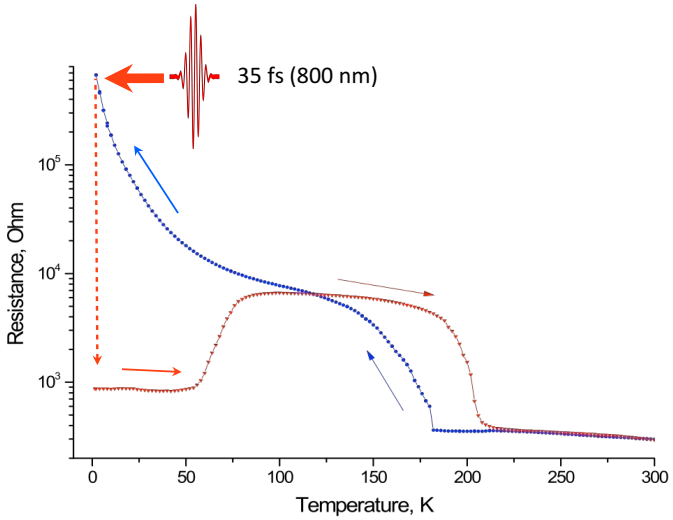
…created under non-equilibrium conditions

Switching to a hidden emergent state in *1T*-TaS_2

Igor Vaskivskyi

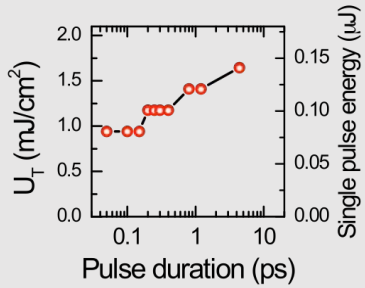
1T-TaS_2 single crystal, ~100 nm thick.
Au contacts by laser lithography (LPKF LDI).





Resistance switching after a 35 fs laser pulse

Threshold:



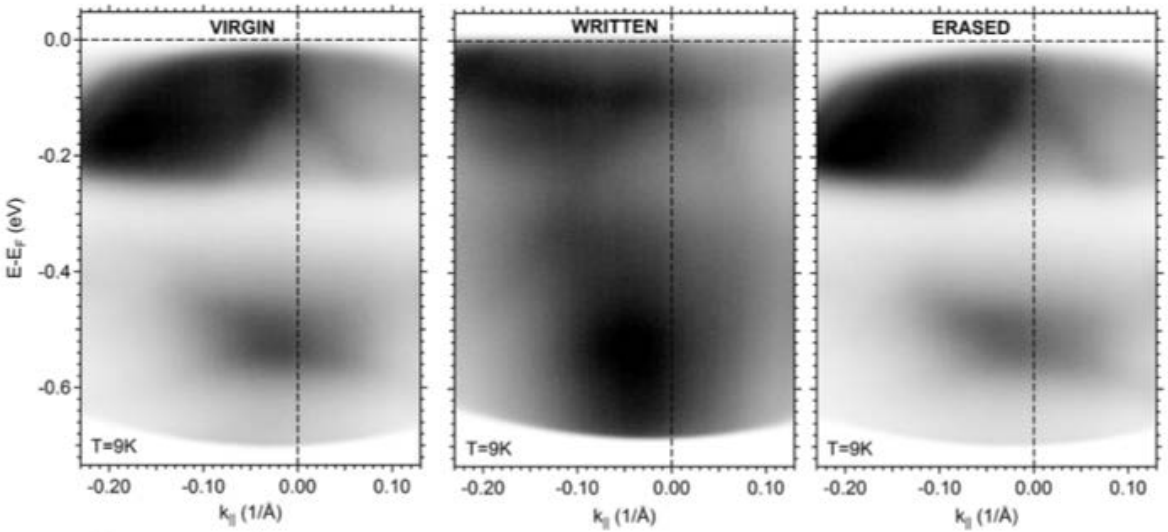
L Stojchevska et al. Science 2014;344:177-180

Time resolved ARPES on 1T-TaS_2

Patrick Kirchmann

Low Temperature ARPES of Switched 1T-TaS_2 Overview

SLAC

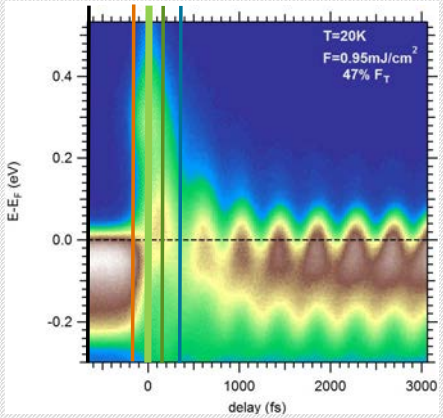


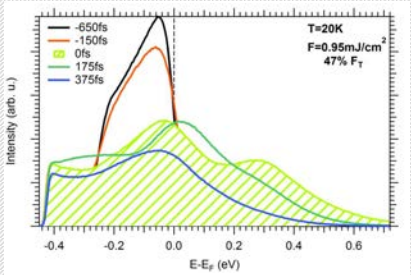
Mott-gapped "VIRGIN" C-state

H-state WRITE: single >2mJ/cm^2 pulse changes

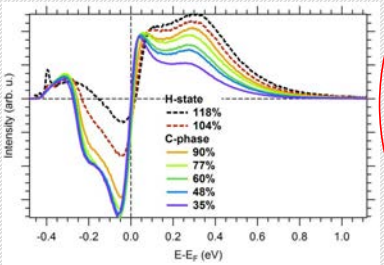
C-state ERASE: multiple ~1μJ/cm^2 pulses or

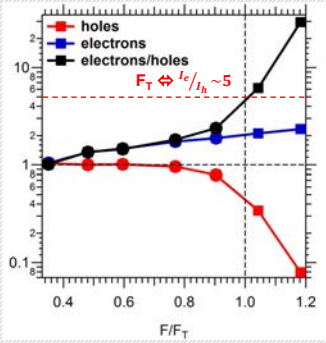
Time resolved ARPES



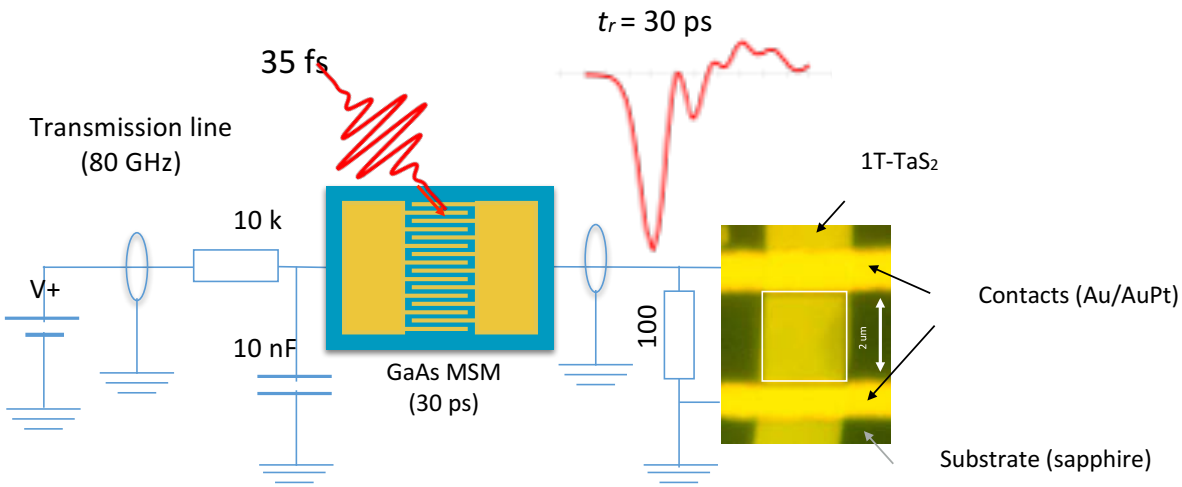


Fluence dependence:





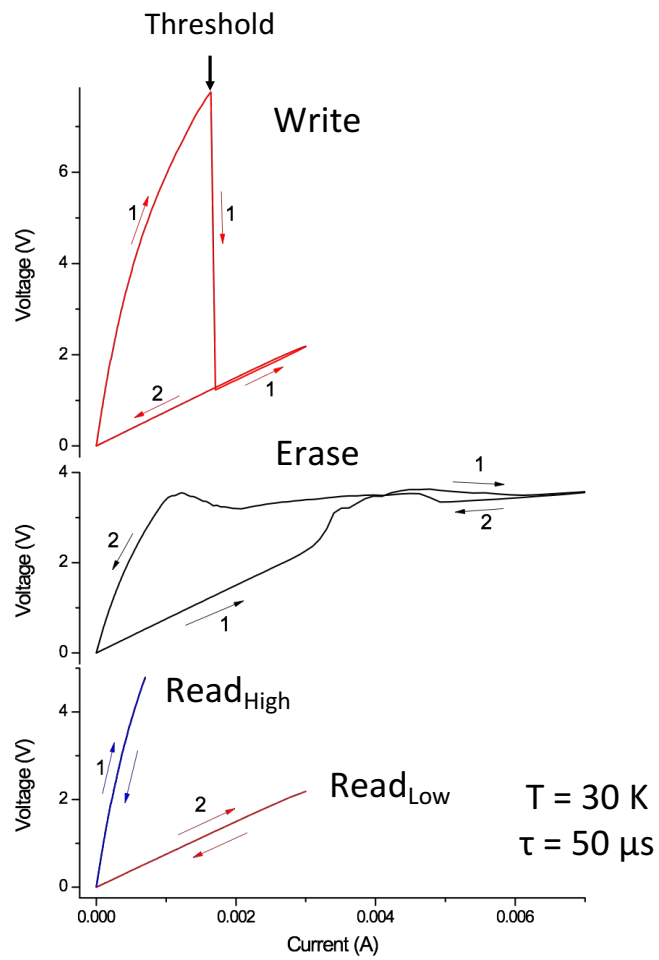
Switching to the H state using a 30 ps current pulse



Vaskivskyi, I. *et al. arXiv* **cond-mat.mes-hall,** (2014), **Nat. Comm.** (**7,** 11442 2016)
I.V., I.A.M and D.M., UK, US, EU *patents*, 2014

CDW memory operations

I-V measured in pulsed mode
(10 µs pulses)



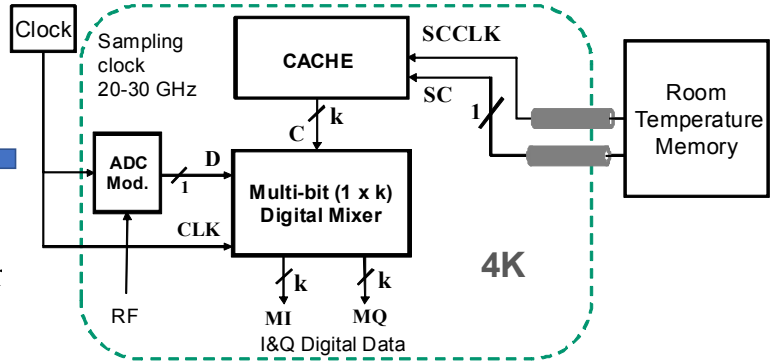
Vaskivskyi, I. *et al. arXiv* **cond-mat.mes-hall,** (2014)
Nat. Comm. (**7,** 11442 2016)

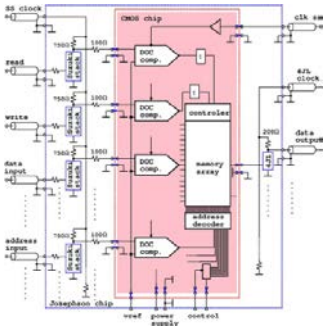
CMOS memory for flux quantum logic (2016)

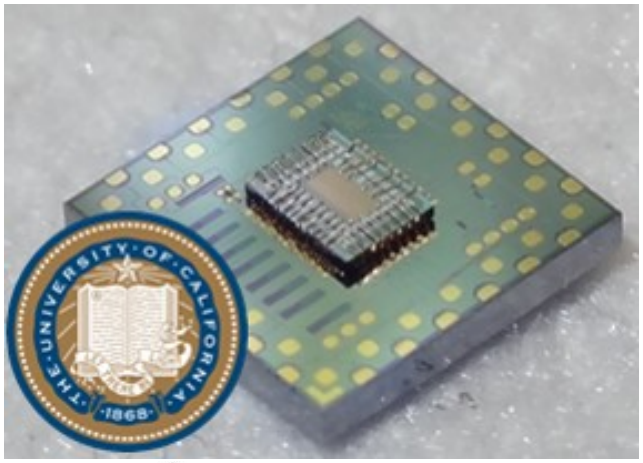
HYPRES
The Digital Superconductor Company™

Hybrid Semiconductor-Superconductor
Fast-Readout Memory for Digital RF Receiver

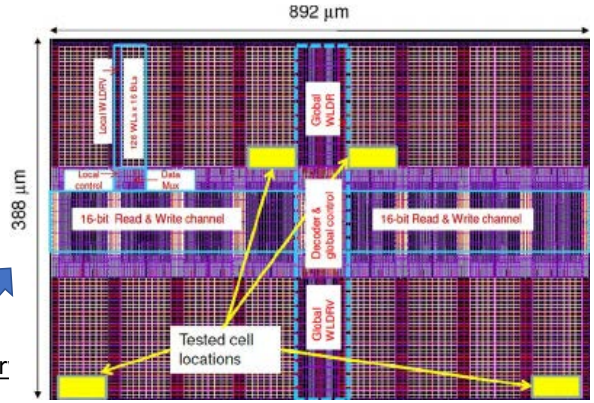
O. A. Mukhanov, *Senior Member, IEEE,* A. F. Kirichenko, T. V. Filippov, S. Sarwana







JJ+cryoCMOS



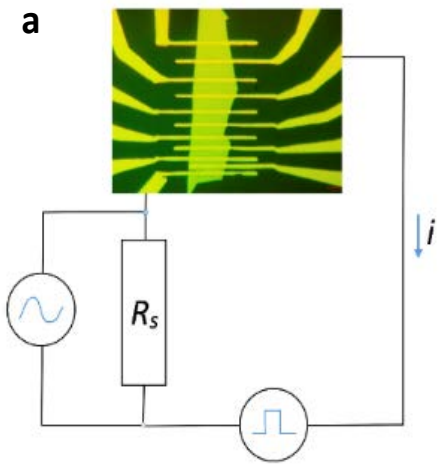
Possible solutions to low temperature memor

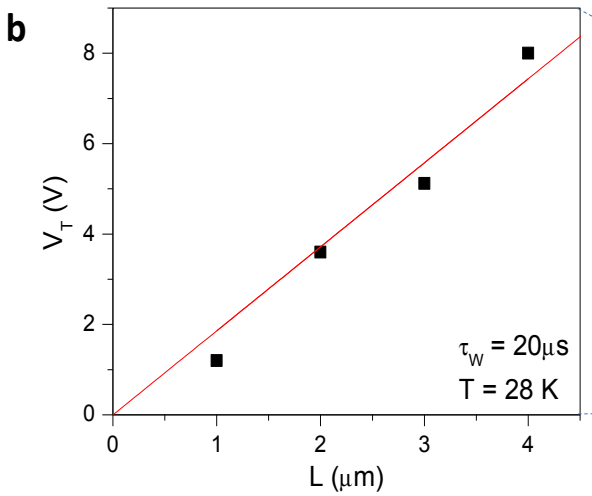
64-kb hybrid Josephson-CMOS 4 Kelvin RAM with 400 ps access time and 12 mW read power.
T. Van Duzer et al., IEEE TAS, Vol. 23, No. 3, 1700504 (4pp), 2013;

Design guidelines for Suzuki stacks as reliable high-speed Josephson voltage drivers
T. Ortlepp et al., Supercond. Sci. Technol. Vol. 26, 035007 (12pp), 2013.

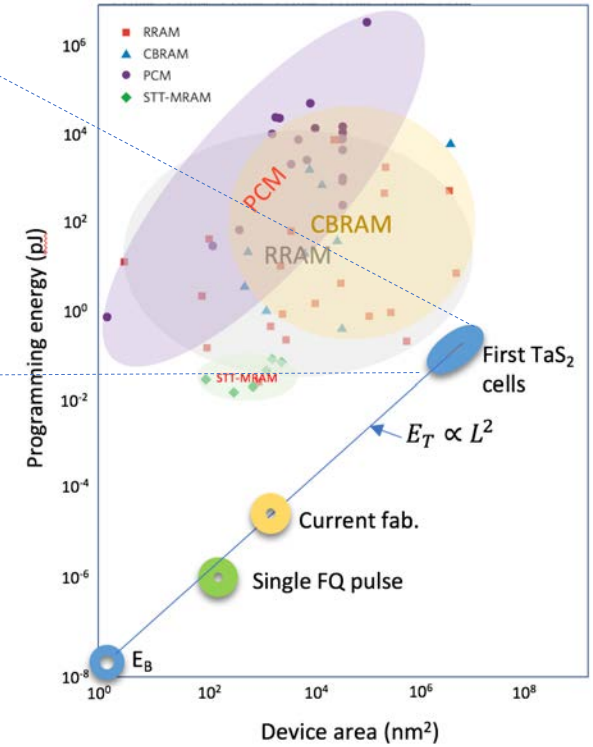
Scaling of switching energy with device size

Wong & Salahuddin, S.
Nat. Nanotech. **10,** 191–194 (2015).





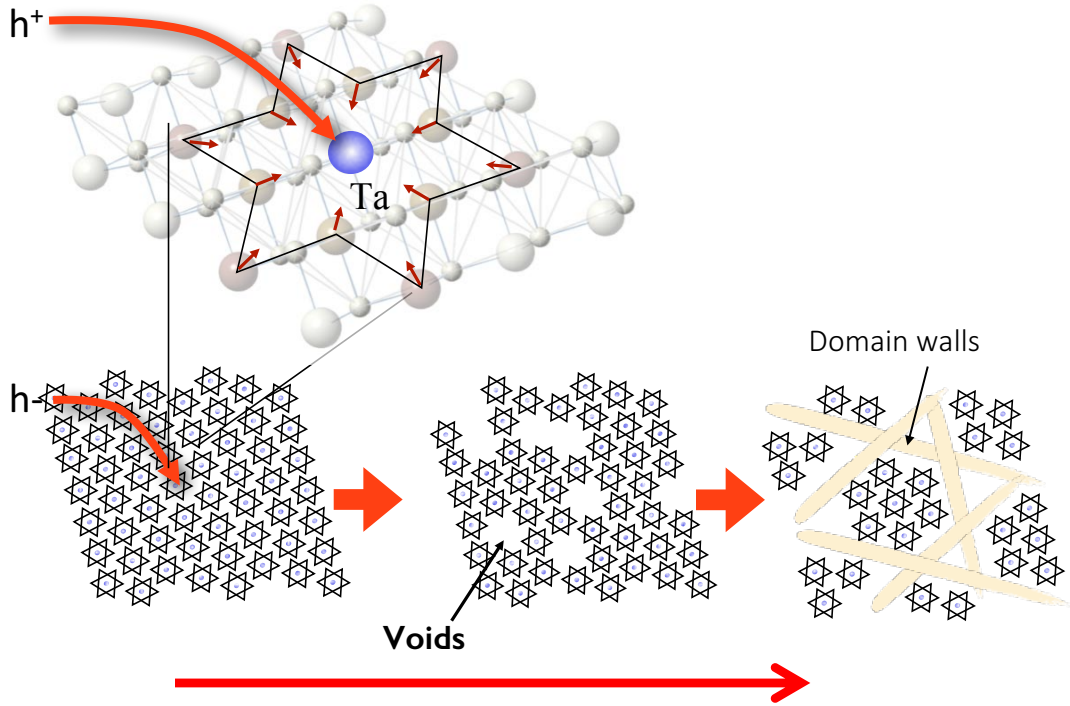
Switching energy, $E_S \propto Q$

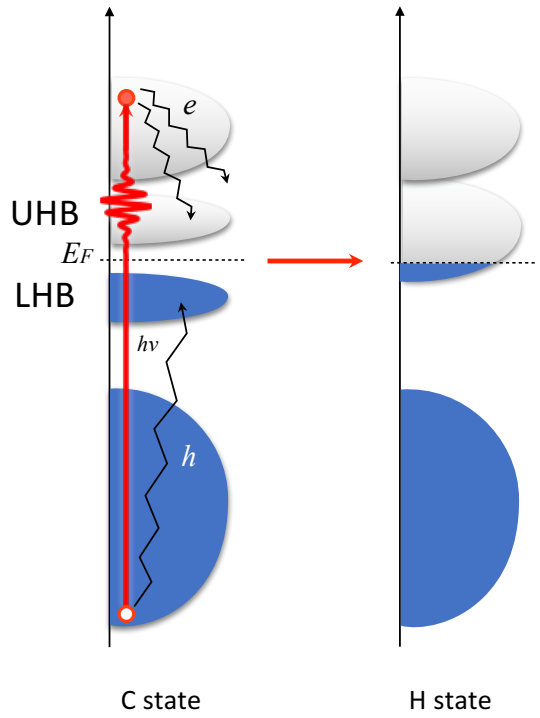


Vaskivskyi, I. *et al.* **Nat. Comm.** (**7,** 11442 2016)

Photo”doping” and formation of a textured state

The photo-hole annihilates a polaron, creating a void.





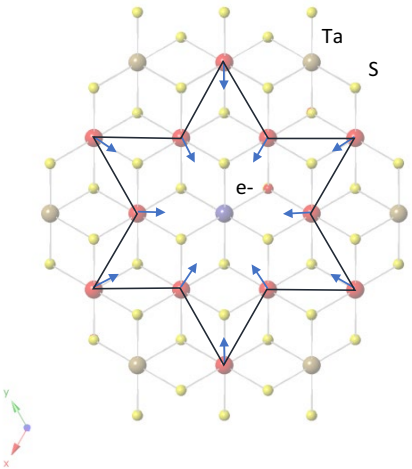
Conversion from polaronic to itinerant (extended) states

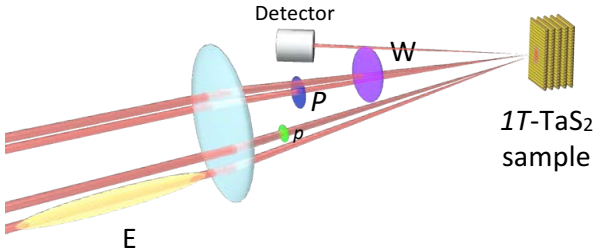
A (non-homotopic) topological transformation

Stojchevska et al. Science (2014) 344, 177-180

Frequency switching of the collective mode

Collective amplitude mode



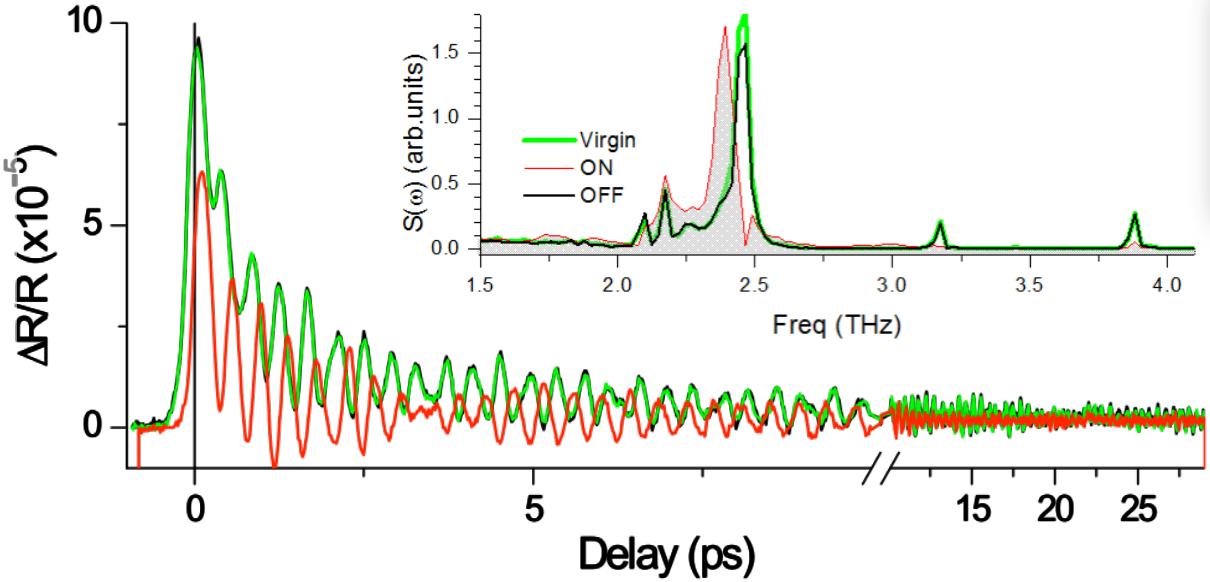


1T-TaS_2 sample

W = 50 fs "write"
E = 50 ps "erase"
P = "pump" (50 fs)
p = "probe" (50 fs)

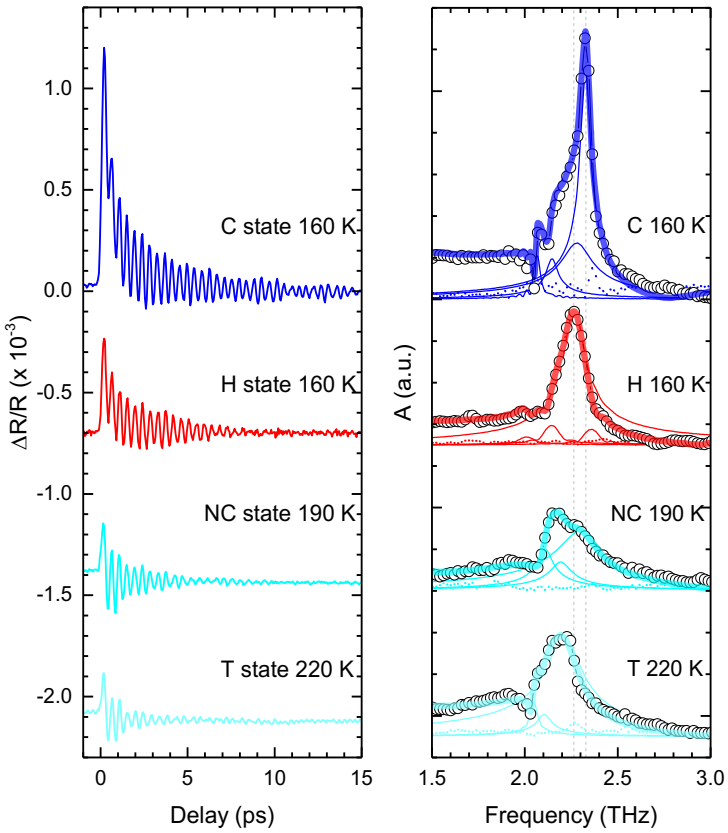
Ljupka Stojchevska

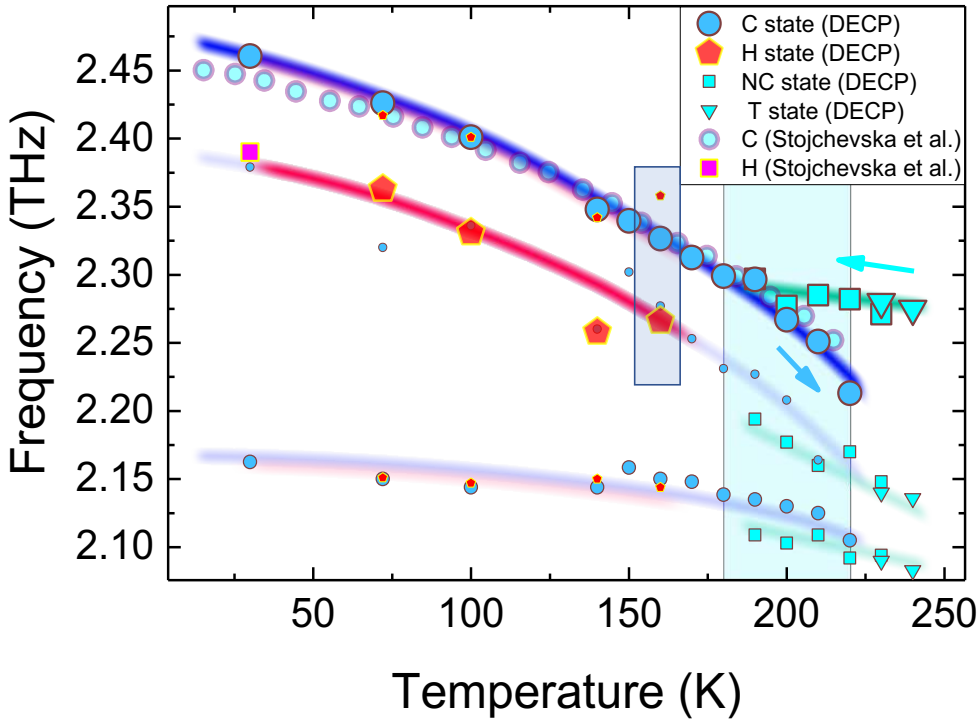
Primoz Kusar



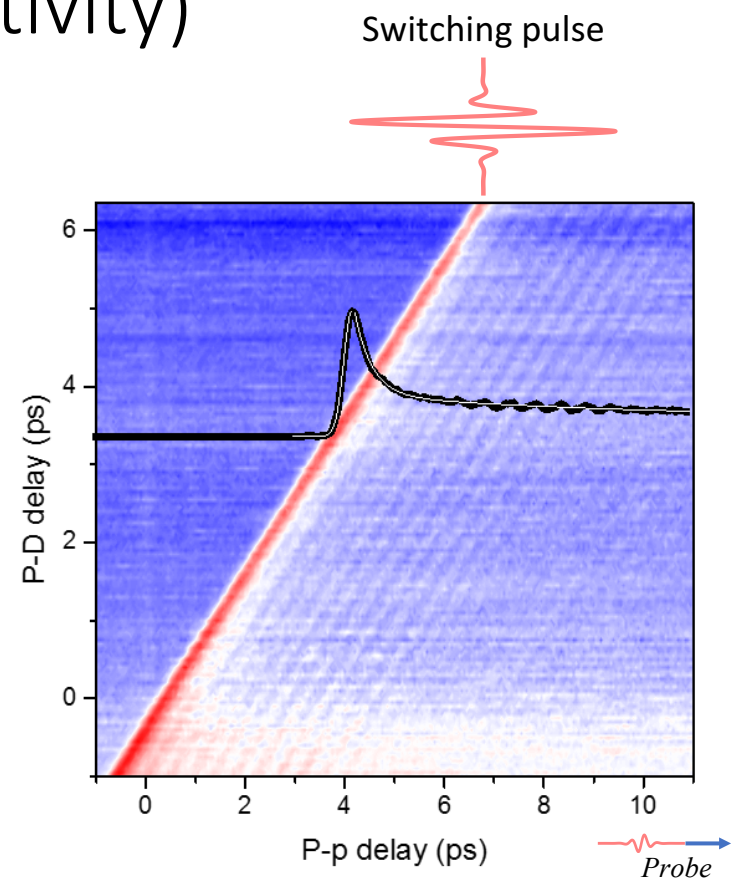
L Stojchevska et al. Science 2014;344:177-180

C, H and NC have distinct mode frequencies





Electronic transition to the H state (measured by reflectivity)



Reflectivity

C state destruction time

$\tau_{rise} = 150 \pm 10\ fs$

Formation time of the H state:

Electronic relaxation

$\tau_{fast} = 300 \pm 20\ fs$

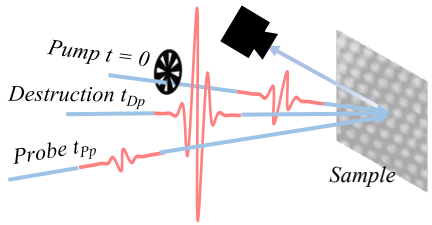
Slow(er) relaxation (domains?)

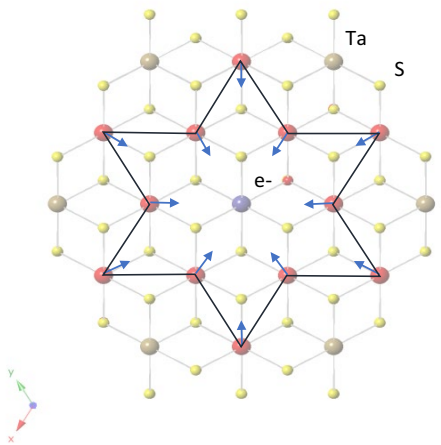
$\tau_{slow} = 4.78 \pm 0.4\ ps$

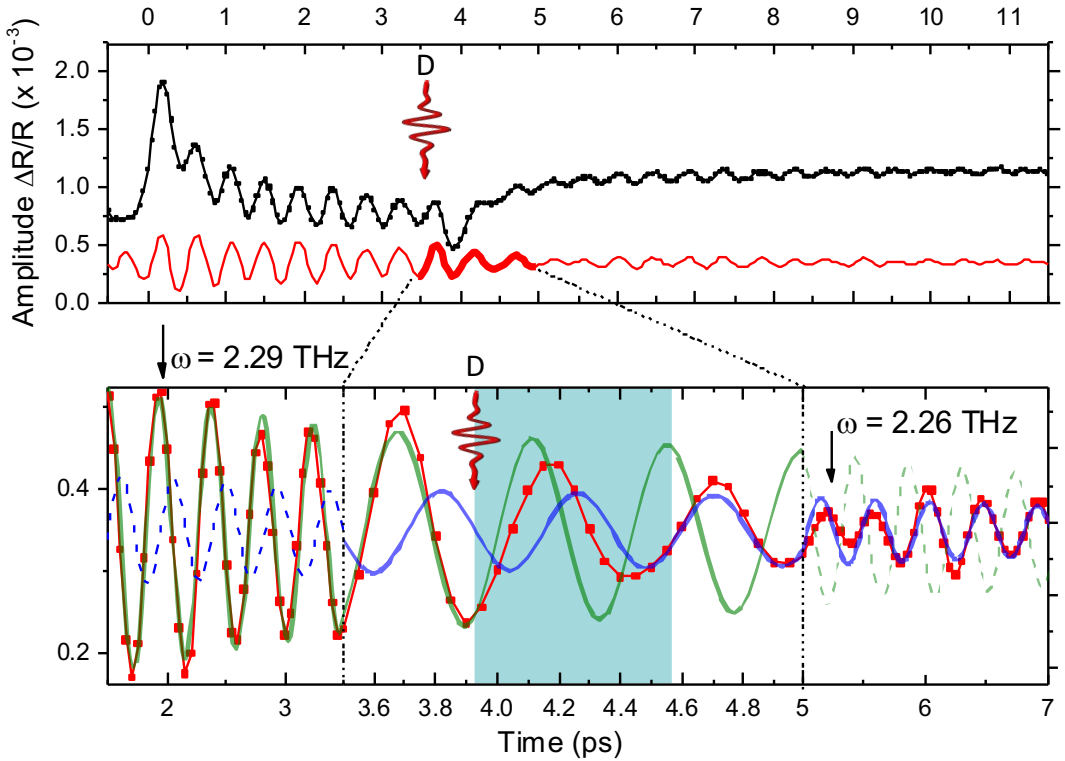
$$\tau_{electronic} < 2\pi/\omega_{phonon}$$

The lattice response: how long does it take?

J. Ravnik

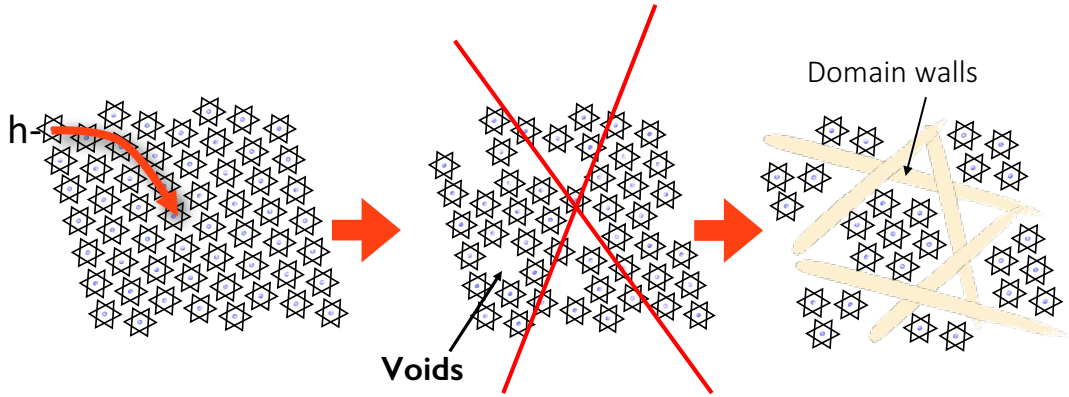






The I-M transition takes place in < 400fs.

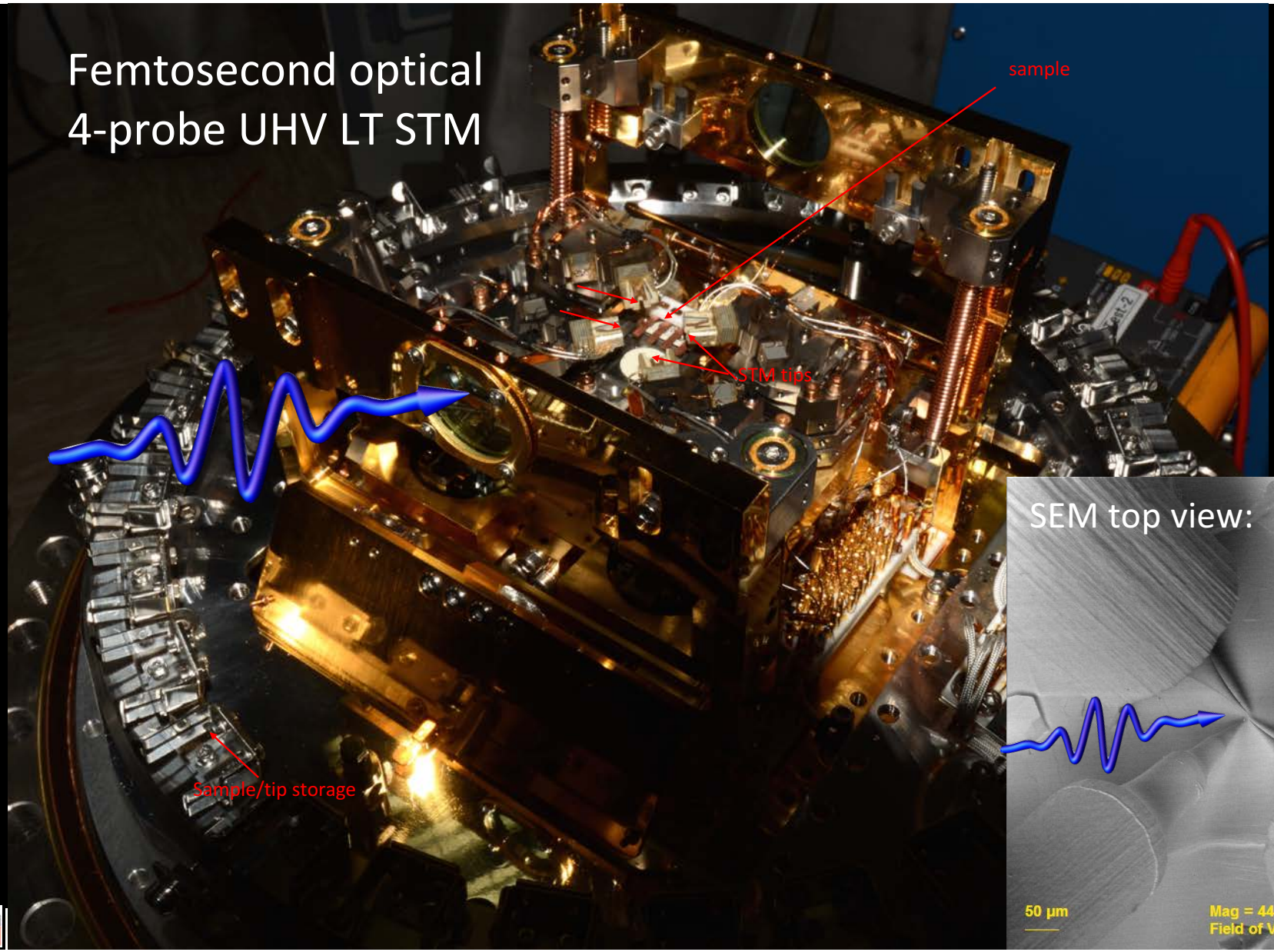
The transition time implies no diffusive processes are involved

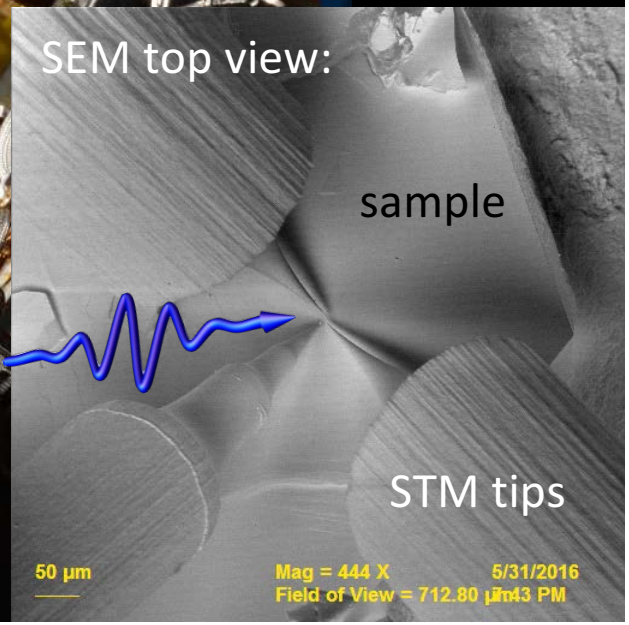


~400 fs

Topological transition

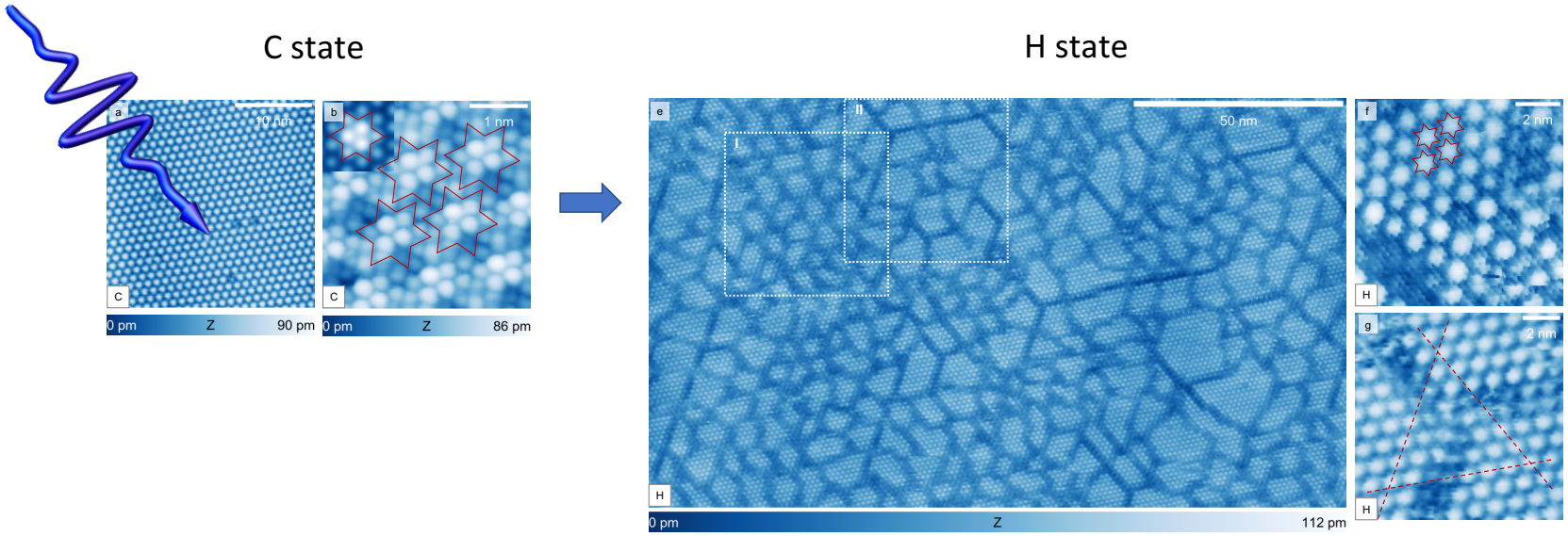
Femtosecond optical 4-probe UHV LT STM





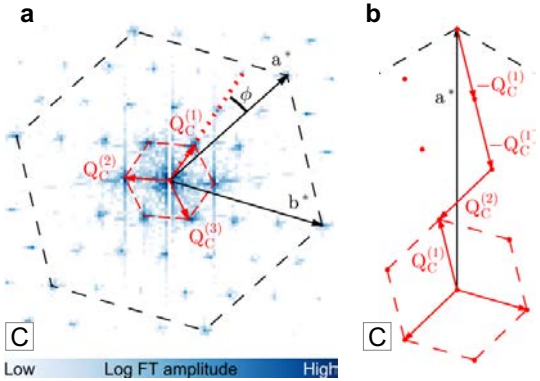
STM after a single 30 fs optical pulse

Yaroslav Gerasimenko

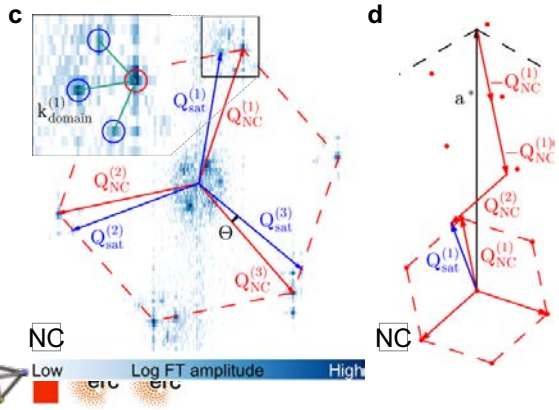


Fine structure of the H state

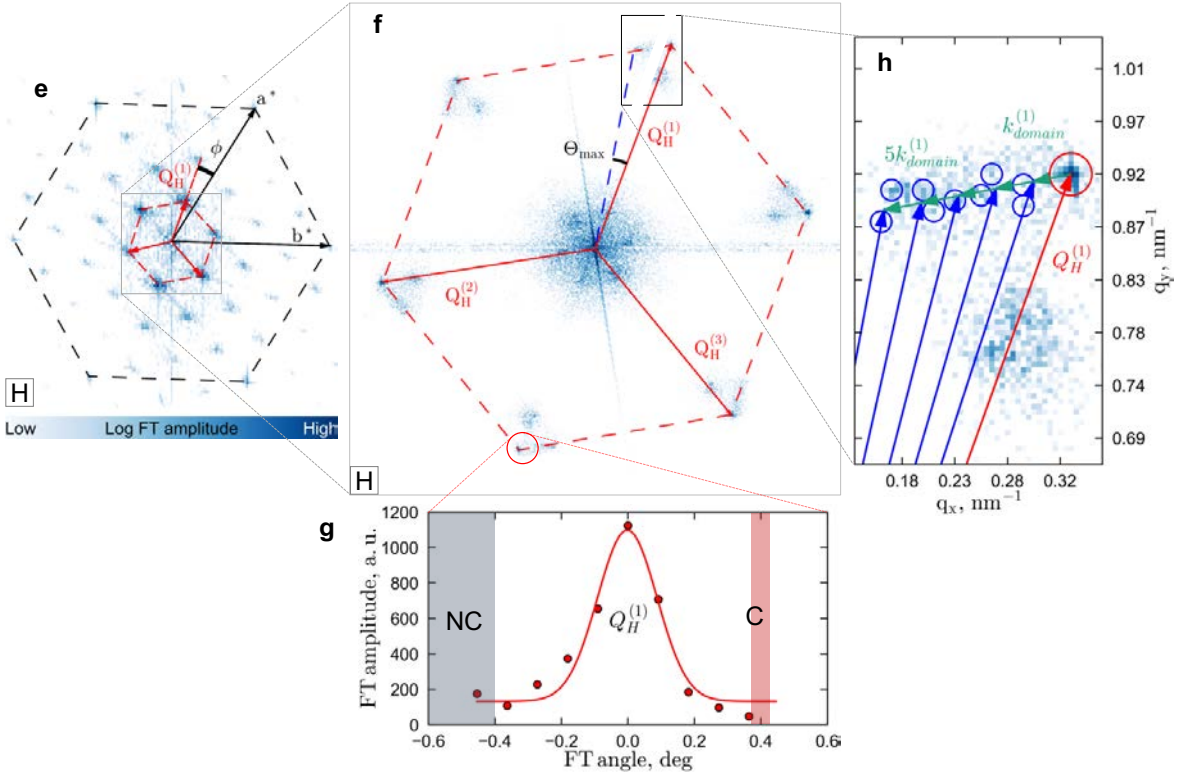
Commensurate state



Nearly Commensurate state



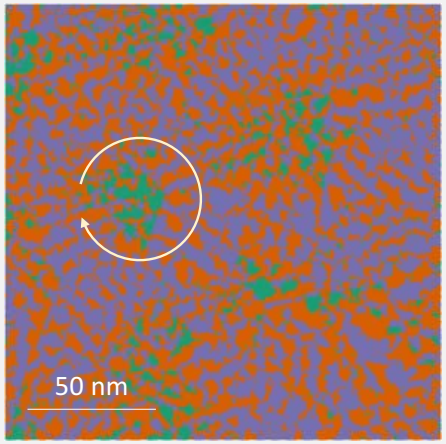
Hidden state



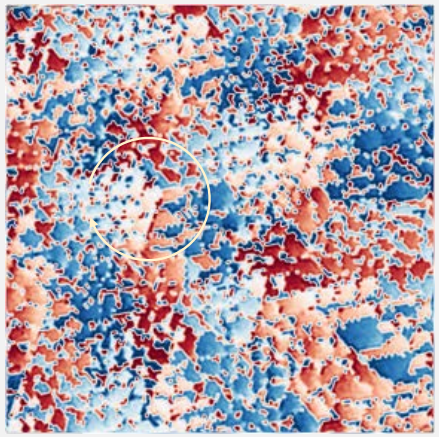
Moiré strain patterns

H state:

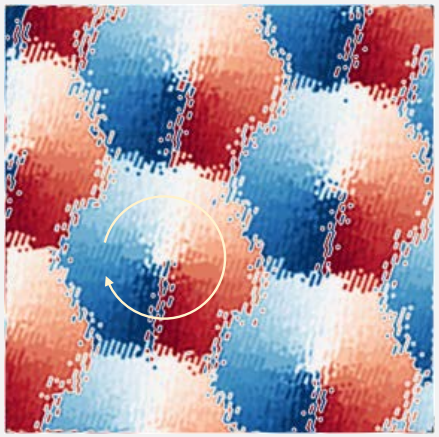
Displacement (relative to C)



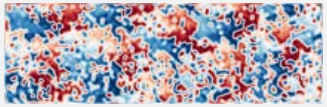
Angle of displacement (relative to C)

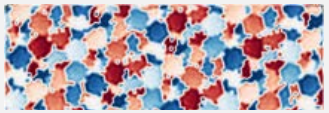


Reconstructed angle (relative to C)



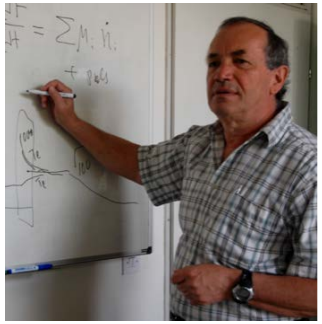
NC state: (same scale)





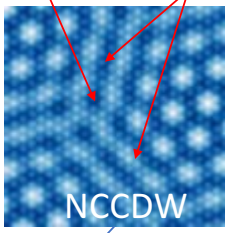
The free-energy landscape of 1T-TaS_2

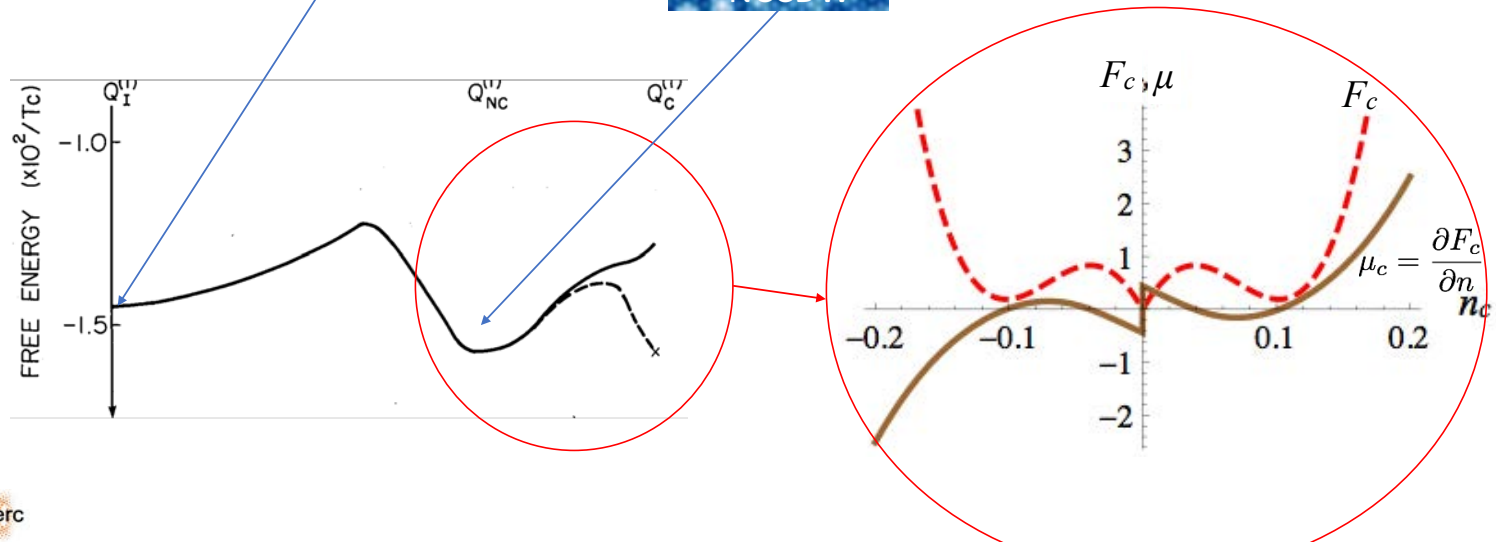
Serguei Brazovskii, (2013) , McMillan (1975), Nakanishi and Shiba (1977)



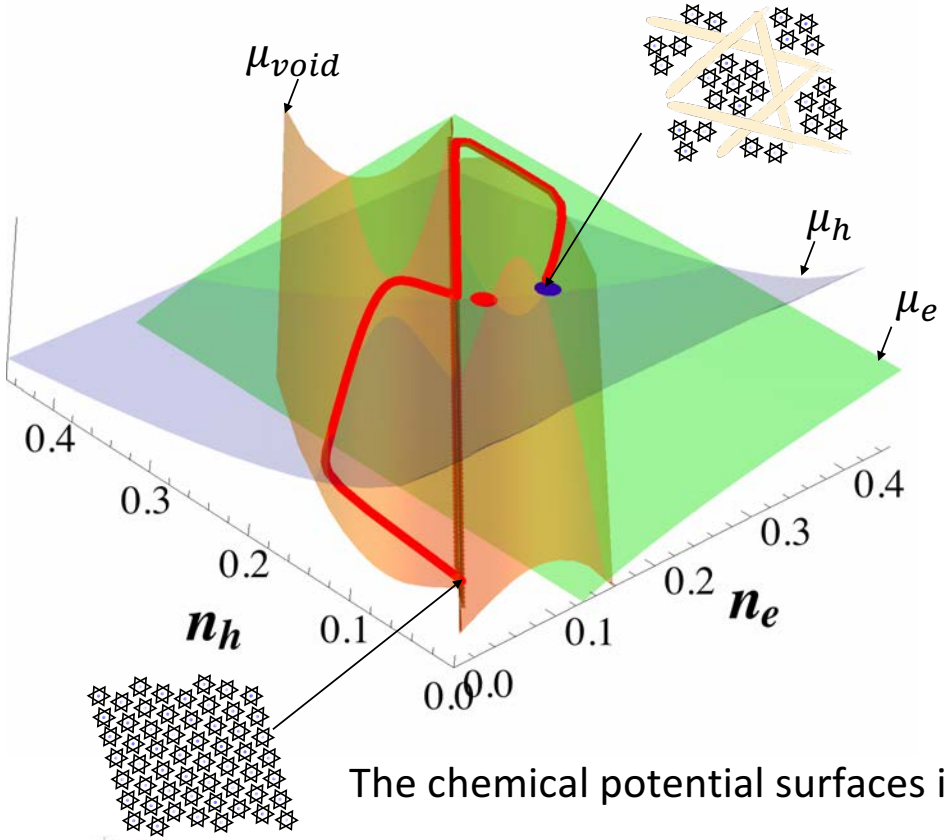
Serguei Brazovskii

$$F_c(n_c) = E_{DW}(\underbrace{C_0|n_c| + C_1|n_c|e^{-1/(\xi|n_c|)}}_{\text{M - IC transition (MacMillan, 1975)}} - \underbrace{C_2\xi n_c^2}_{\text{Intersection of DW}} + \underbrace{C_4\xi^3 n_c^4}_{\text{Repulsion between DWs}})$$





Topologically protected q-vector



The chemical potential surfaces in Q-space

STABILITY CONDITIONS:

The system is stable <u>in the H state</u> when:

$$\mu_h - \mu_e = \mu_{void}$$

Subject to charge conservation:

$$n_e - n_h = n_{voids}$$

$$(Q_e - Q_h = Q_H)$$

The topologically protected wavevector:

$$Q_H = \pi \, n_{void}$$

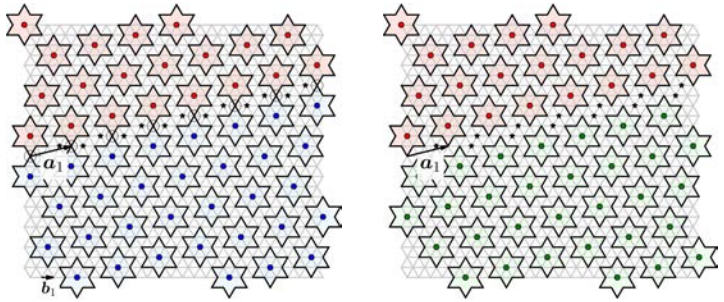
Stojchevska et al. Science (2014) 344, 177-180

Domain wall with fractional charge

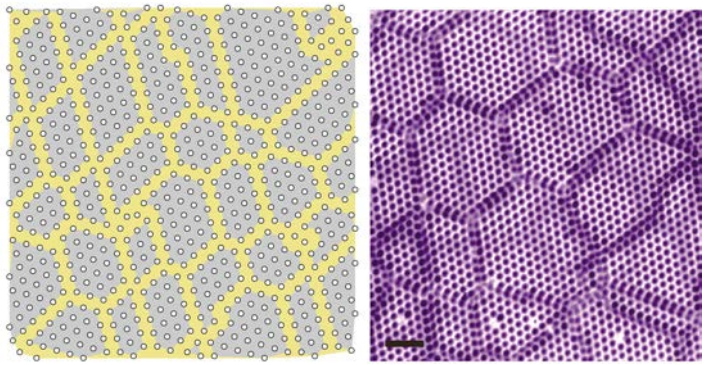
Karpov, P. & Brazovskii, S. *arXiv.org* **cond-mat.str-el,** (2017).

$$H = \sum_{i,j} U_{ij} n_i n_j,$$

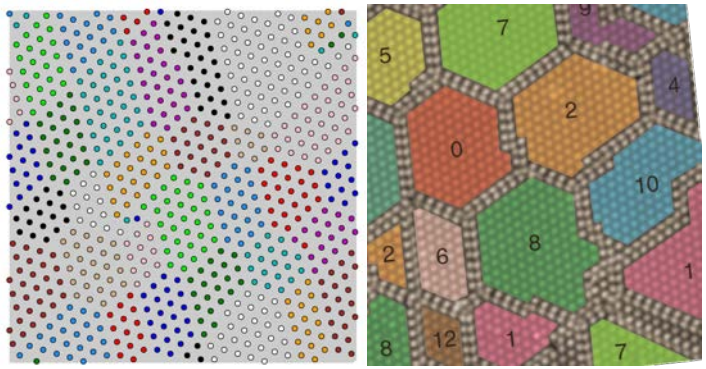
$$U_{ij} = e^2 \frac{\exp(-r/l_s)}{|\mathbf{r}_i - \mathbf{r}_j|} \equiv U_0 \frac{a \exp(-\frac{r-a}{l_s})}{|\mathbf{r}_i - \mathbf{r}_j|},$$



(a) $q = +e/13$ (b) $q = +2e/13$



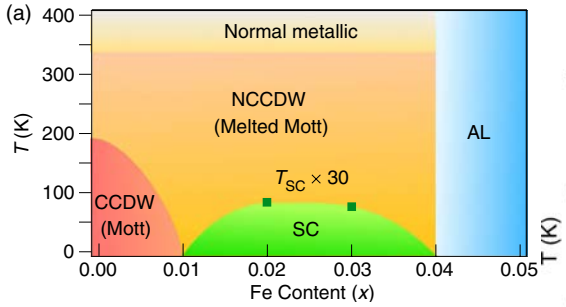
(a) $N_v = 17$ voids, $\nu_v = 2.3\%$ – domain walls (b)



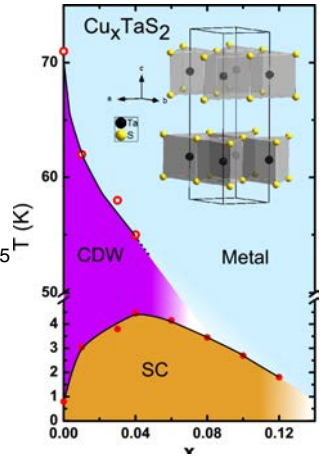
(c) $N_v = 17$ voids, $\nu_v = 2.3\%$ (d)

Equilibrium phases

Fe : 1T-TaS_2 (substitution):

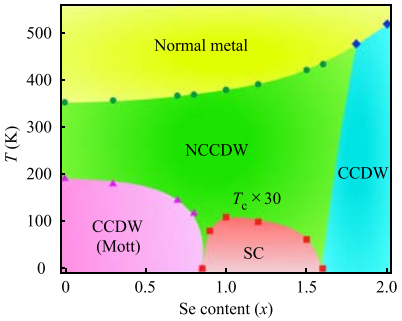


Cu : 2H-TaS_2 (substitution):



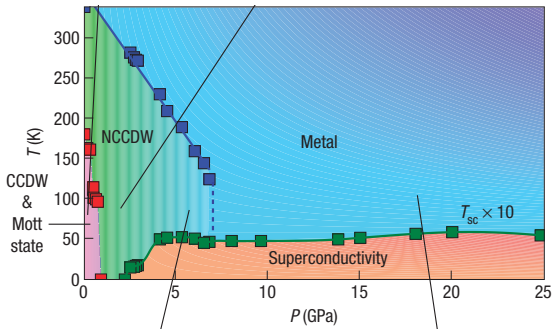
Liu et al. APL 2013

Chemical pressure: Se substitution:



Wagner et al (PRBB 2008)

Physical pressure: 1T-TaS_2



Sipos et al (Nat.Mat. 2008)

Emergent transient phases