Amsterdam Summer Workshop on Low-D Quantum Condensed Matter 2015

Ultrafast Switching to a Stable Hidden Quantum State in an Electronic Crystal

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Jožef Stefan International Postgraduate School

European Research Council
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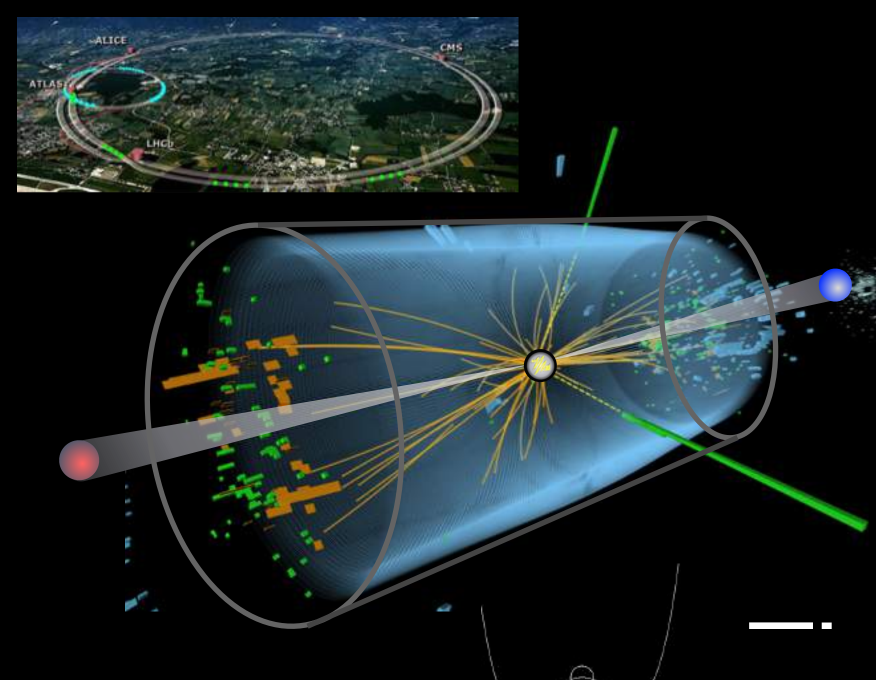
Transitions…in time

Stock market crashes



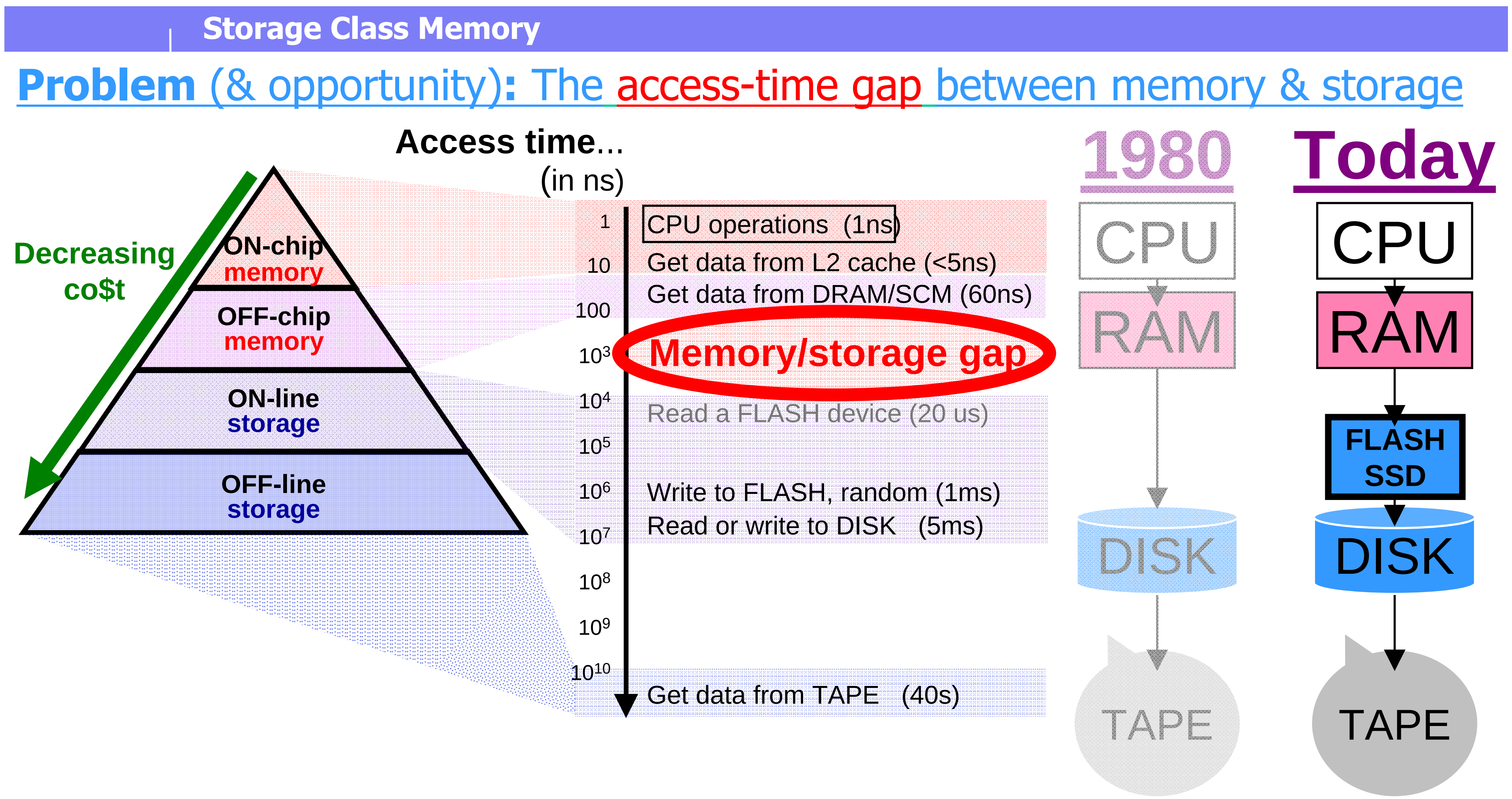
Elementary particle collisions

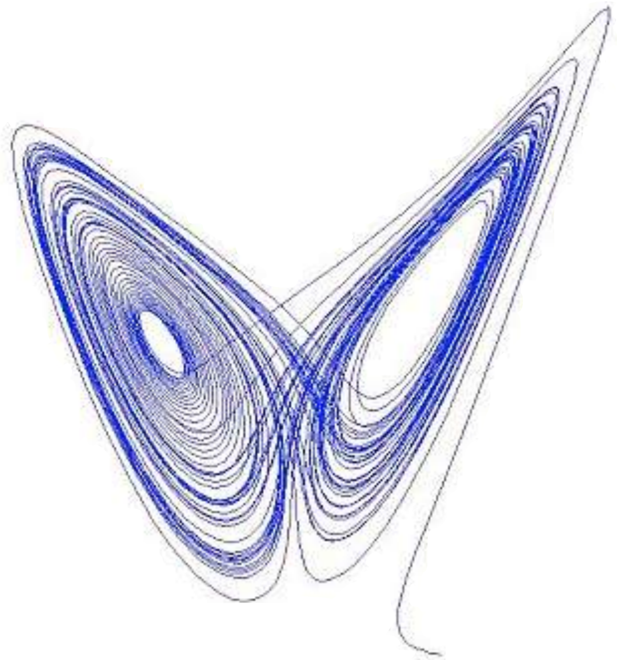
The Big Bang - hidden universes

D.M. What can physics tell us about stock market crashes, TEDx, Dec. 2013

The  challenge

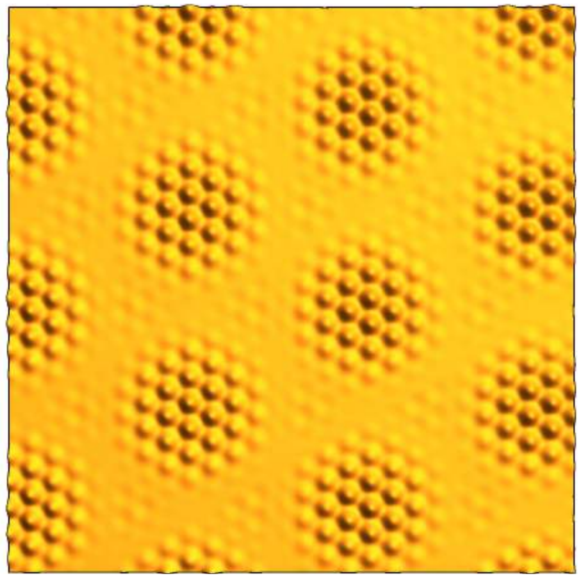
G. W. Burr, IBM, 2013





- Our aim is to investigate trajectories of systems though symmetry-breaking transitions under **<u>nonequilibrium conditions</u>**, in real time.

Why CDWs?



CDW in 1T-TaS_2

Bardeen, J. Classical versus quantum models of charge-density-wave depinning in quasi-one-dimensional metals. *Phys. Rev. B* **39,** 3528–3532 (1989).

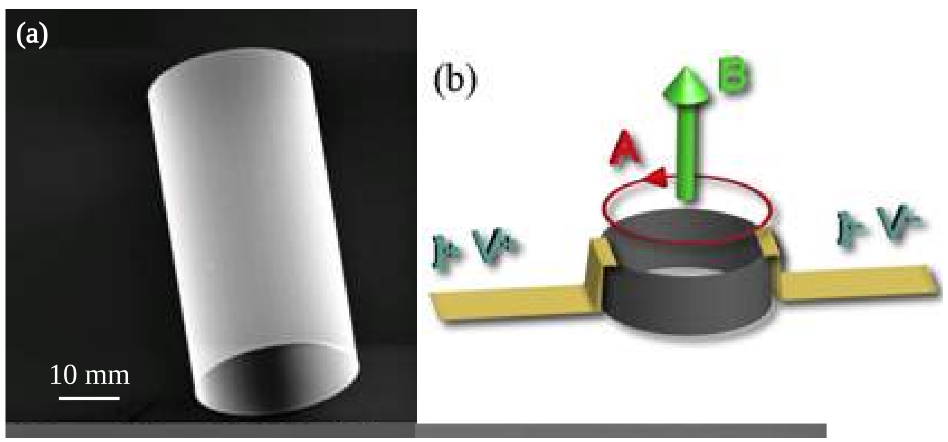
John Bardeen: CDWs are macroscopic quantum states

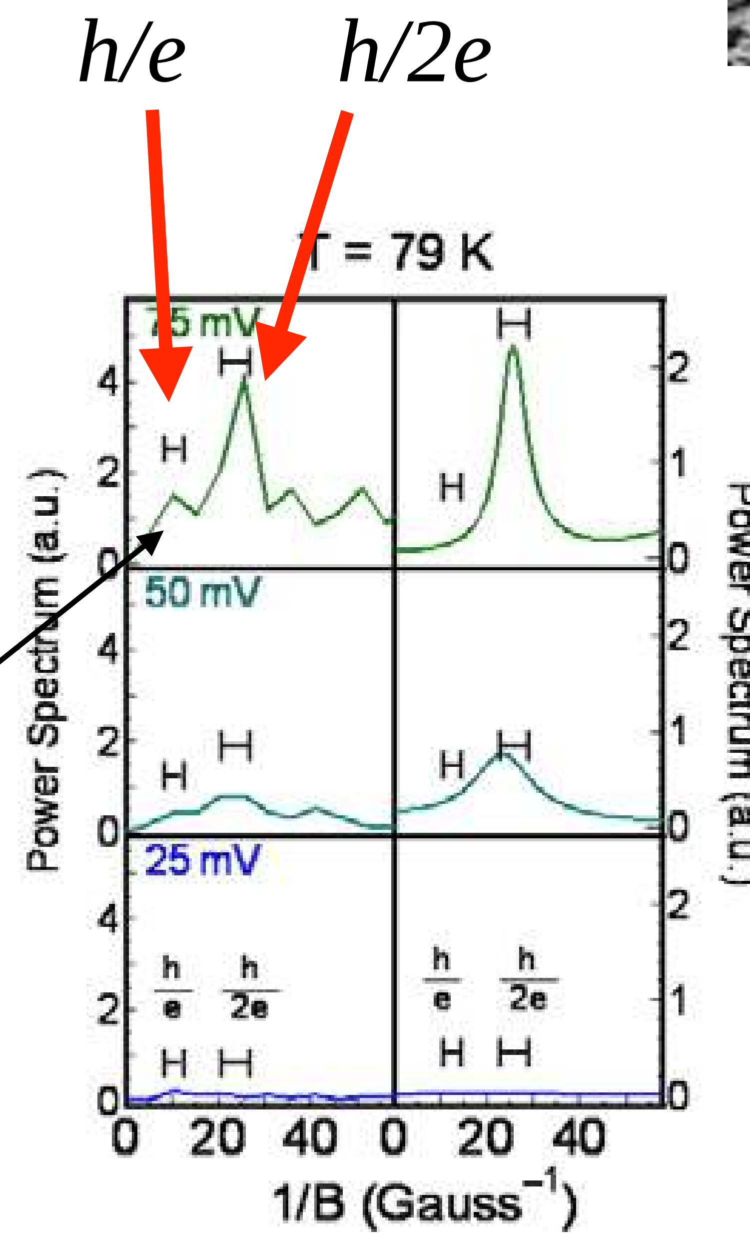
EPL, **97** (2012) 57011
doi: 10.1209/0295-5075/97/57011

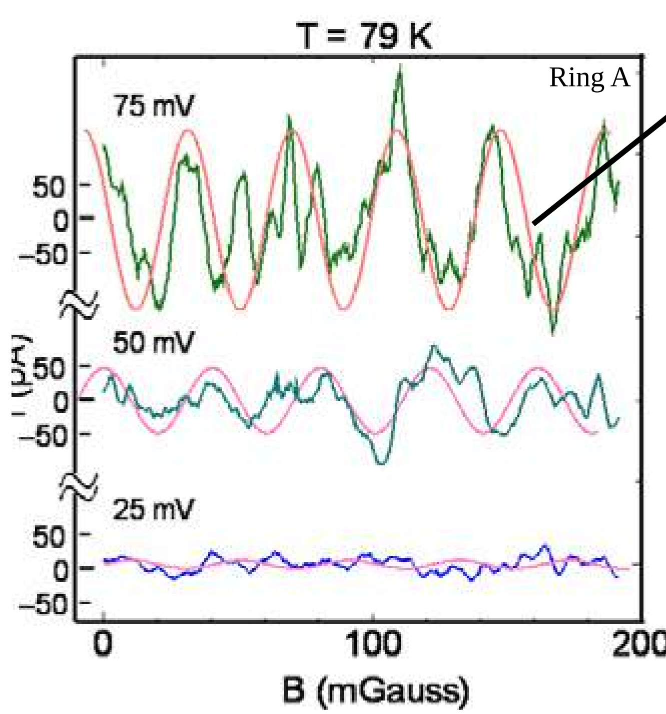
Aharonov-Bohm effect in charge-density wave loops with inherent temporal current switching

M. Tsubota[1], K. Inagaki[1,2], T. Matsuura[1] and S. Tanda[1,2]

S. Tanda







Optical experiments: (at JSI)

Ljupka Stojchevska

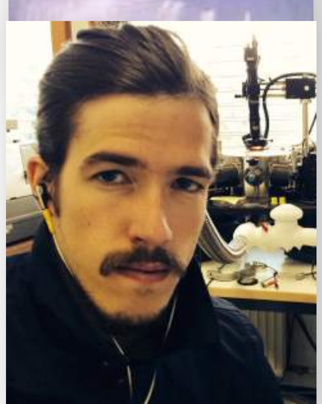
Igor Vaskivskyi

Tomaz Mertelj

Jan Gospodaric
Primoz Kusar
Roman Yusupov

Current switching experiments

Ian Mihailovic
(IJS, FE-Uni-Lj)



Samples+:

I. Fisher *(Stanford)*

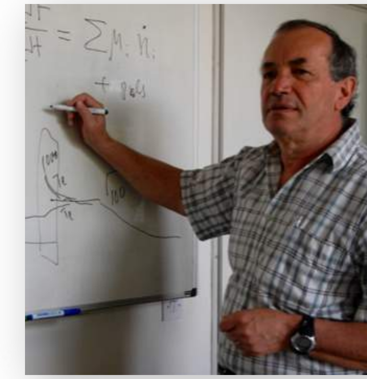
P. Sutar *(JSI)*

Lithography:

D.Svetin *(JSI)*

Theory

Serguei Brazovskii
(Univ. Paris Sud Orsay)



Time resolved ARPES

Patrick Kirchman
ZX Shen *(Stanford)*

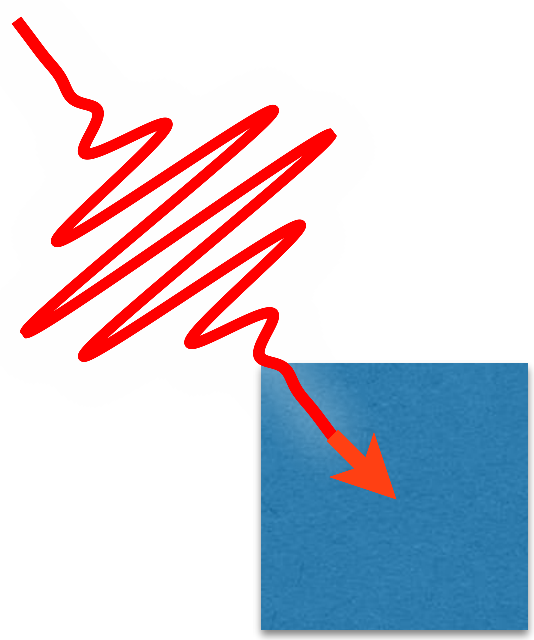
European Research Council
Established by the European Commission

PHYSICAL REVIEW B VOLUME 4, NUMBER 7 1 OCTOBER 1971

Destruction of Superconductivity by Laser Light

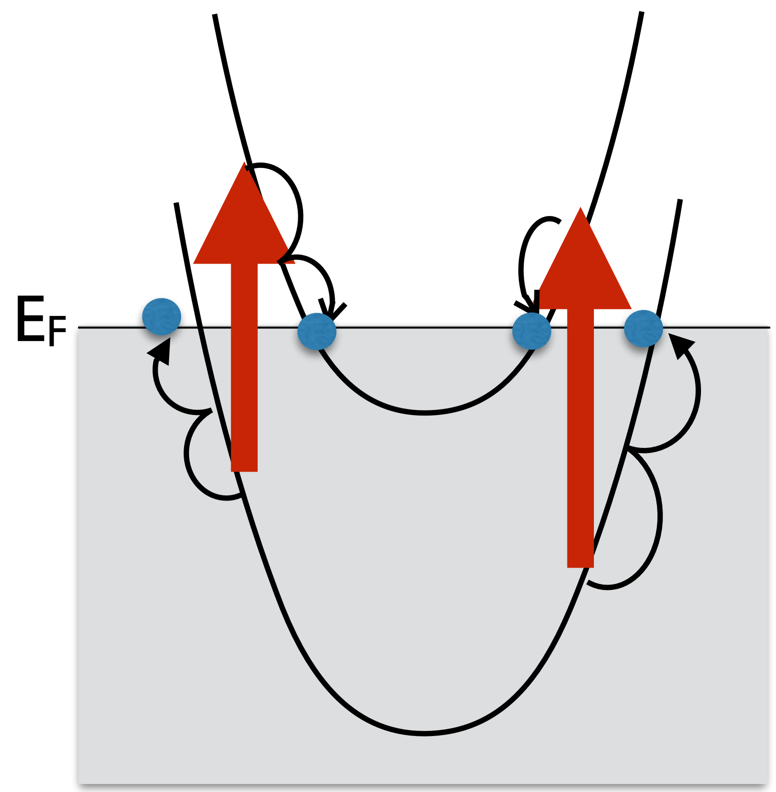
L. R. Testardi
Bell Telephone Laboratories, Murray Hill, New Jersey 07974
(Received 27 January 1971)

Superconductivity is destroyed by laser light in Pb films of thickness comparable to the optical penetration depth δ and less than the superconducting coherence length ξ. Thermal effects, which have been independently determined, cannot account for this. For films of thickness greater than δ and ξ, only the thermal effect is observed. In a proposed explanation it is shown that the electron gas may be heated from 3 to 18 °K above the lattice temperature by the light absorption in these experiments.



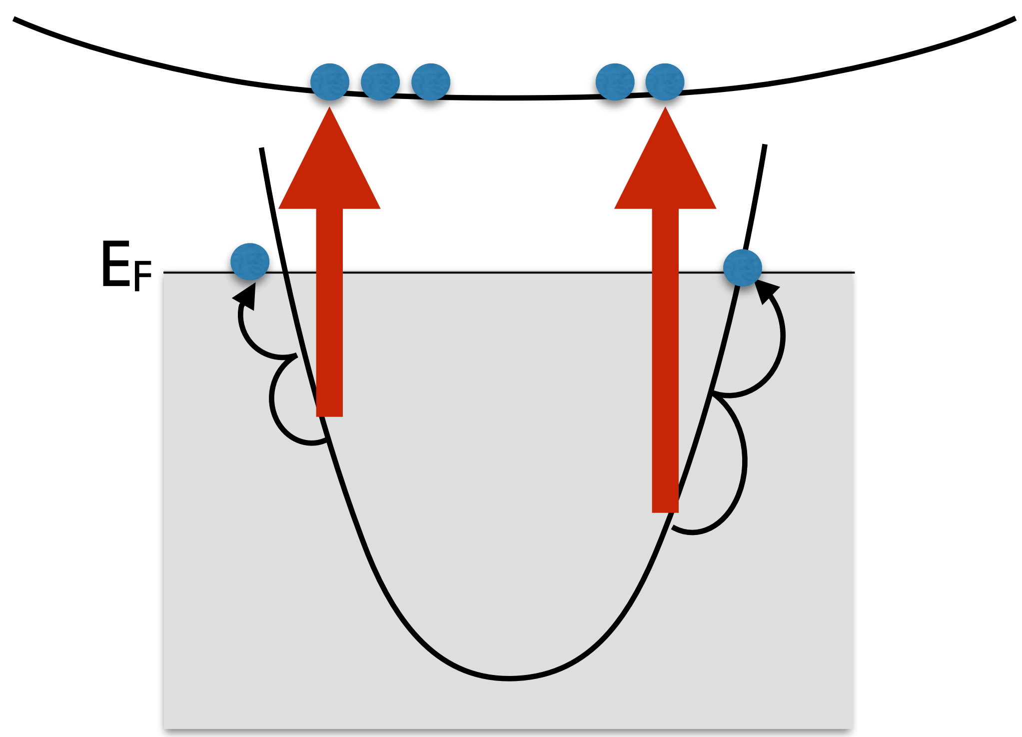
heat.

The electron relaxation process



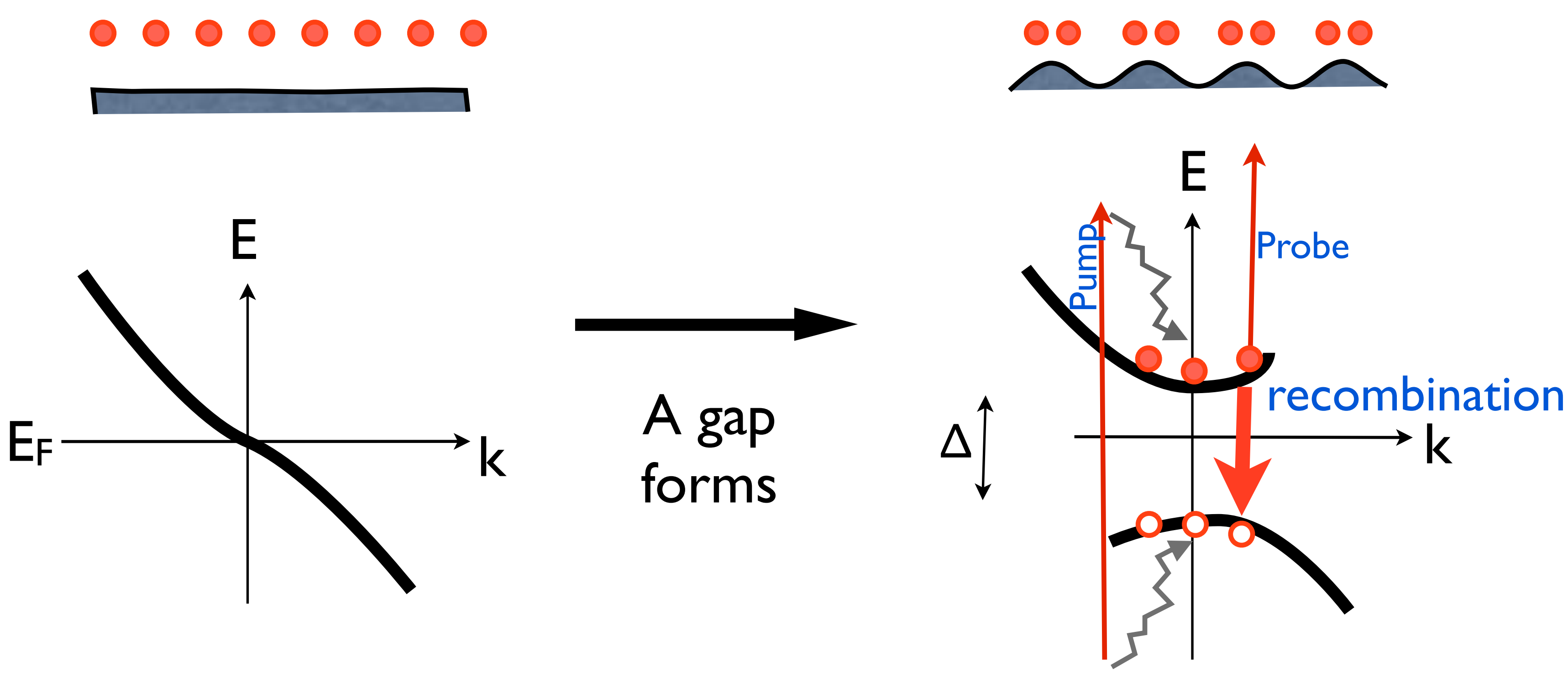
Just heating ($T_e^*=T_L^*=\ldots$).
No doping.

The importance of *e-h* asymmetry for reaching photoinduced hidden states

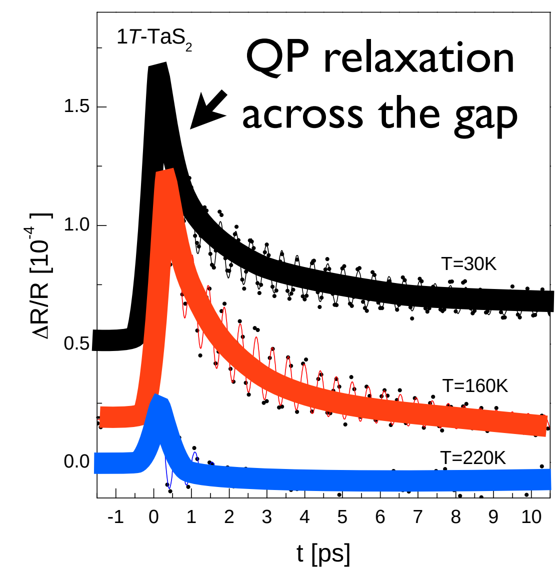


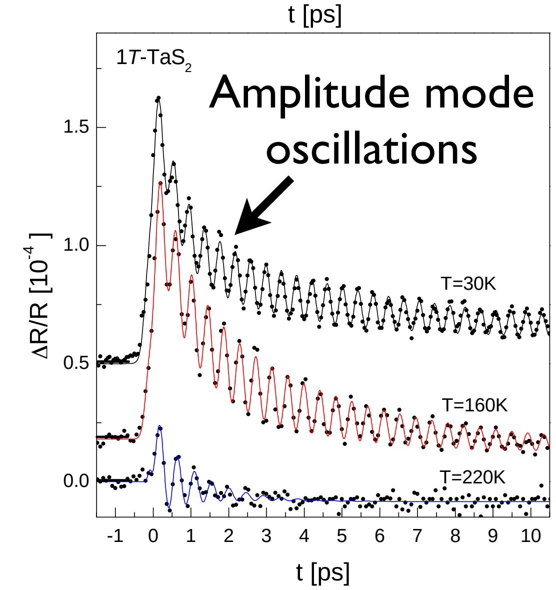
The observable elementary excitations with femtosecond spectroscopy

1. Quasiparticle (fermionic) excitations (detect the presence of a gap)



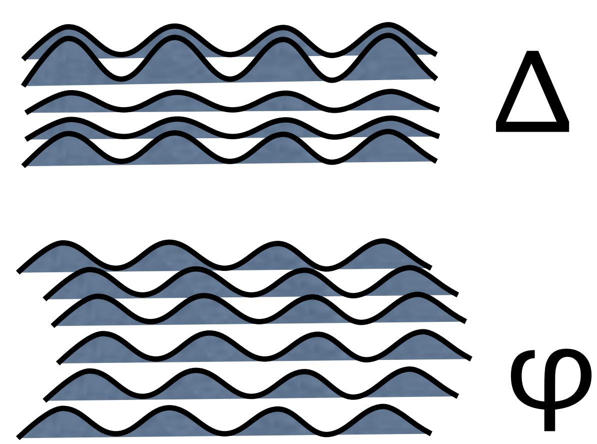
EXAMPLE:





2. Collective mode (bosonic) excitations

The amplitude and phase modes



$$\Psi = \Delta e^{i\varphi}$$

Demsar et al., PRL **83**, 800 (1999).
Demsar, J.,et al, PRB **66**, (2002).
Demsar, et al., PRB **82**, 4918 (1999).
Kusar et al, PRL 101, 227001 (2008)
Yusupov et al.,PRL **101**, 246402 (2008).

Leibnitz

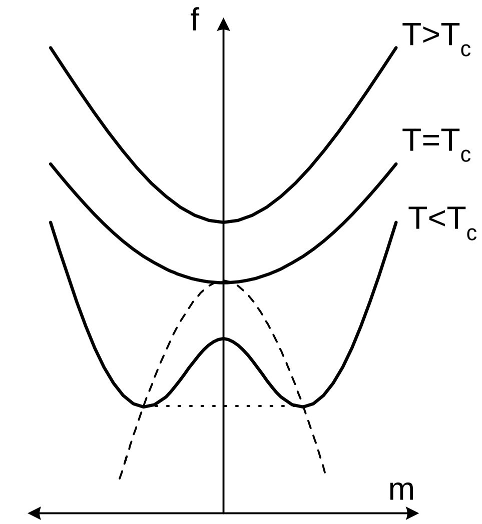
A theory should be more simple than the observations it is trying to describe.

(It should have fewer parameters)

The non-linear energy functional

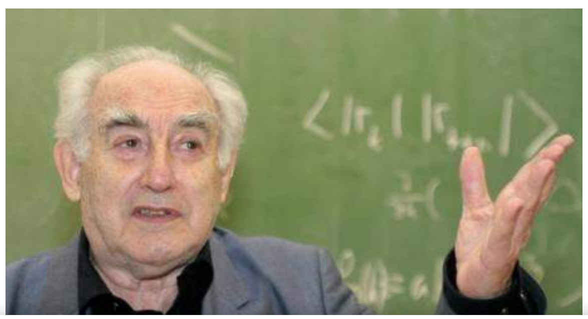
The Landau non-linear energy functional originally written to describe a **structural phase transition**:

$$F = \alpha\Psi^2 + \beta\Psi^4 + H\Psi \quad \text{where} \quad \alpha = \alpha_0(T - T_c)$$



The Ginzburg-Landau equation for a **superconductor**:

$$F = F_0 + \alpha|\psi|^2 + \frac{\beta}{2}|\psi|^4 + \frac{1}{2m}|(-i\hbar\nabla - 2e\mathbf{A})\psi|^2 + \frac{|\mathbf{B}|^2}{2\mu_0}$$



Complex order parameter

$$\Psi = \Delta\, e^{i\phi}$$

LUME 13, NUMBER 16 PHYSICAL REVIEW LETTERS 19 OCTOBER 1964

BROKEN SYMMETRIES AND THE MASSES OF GAUGE BOSONS

Peter W. Higgs
Tait Institute of Mathematical Physics, University of Edinburgh, Edinburgh, Scotland

(Received 31 August 1964)

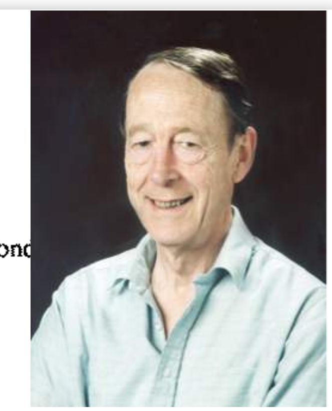
Lagrangian density, includes K.E.

$$L(\varphi) = \partial_\mu\varphi^*\partial^\mu\varphi - \alpha\varphi^*\varphi - \frac{\beta}{2}|\varphi^*\varphi|^2$$

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, Lond



2. The phase transition

Although our discussion will be quite general, for illustrative purposes it is convenient to have a specific example in mind. Let us consider an N-component real scalar field ϕ with a Lagrangian invariant under the orthogonal group $\mathrm{O}(N)$, and coupled in the usual way to $\frac{1}{2}N(N-1)$ vector fields represented by an antisymmetric matrix B_μ. We can take

$$L = \tfrac{1}{2}(D_\mu\phi)^2 - \tfrac{1}{8}g^2(\phi^2-\eta^2)^2 + \tfrac{1}{8}\mathrm{Tr}(B_{\mu\nu}B^{\mu\nu}) \qquad (1)$$

with

$$D_\mu\phi = \partial_\mu\phi - eB_\mu\phi$$
$$B_{\mu\nu} = \partial_\nu B_\mu - \partial_\mu B_\nu + e[B_\mu, B_\nu].$$

Time-dependent GL equation

Serguei Brazovskii, 2010

The energy of the system can be described in terms of a time-dependent Ginzburg-Landau functional†:

$$F = \alpha\Psi^2 + \beta\Psi^4 + H\Psi$$

where instead of the usual temperature dependence (T - T_c), the *first* term is time-dependent:

$$\alpha = [1 - \frac{T_e(t, \mathbf{r})}{T_c}]$$

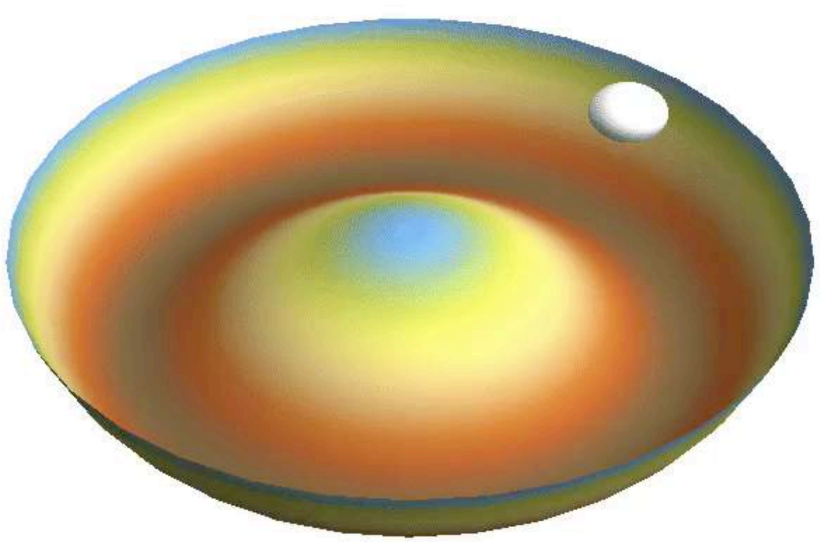
"Ergodicity"

The equation of motion is obtained via the Euler-Lagrange theorem :

$$\boxed{\frac{1}{\omega_0^2}\frac{\partial^2}{\partial t^2}A + \frac{\alpha}{\omega_0}\frac{\partial}{\partial t}A - (1-\eta)A + A^3 - \xi^2\frac{\partial^2}{\partial z^2}A = 0}$$

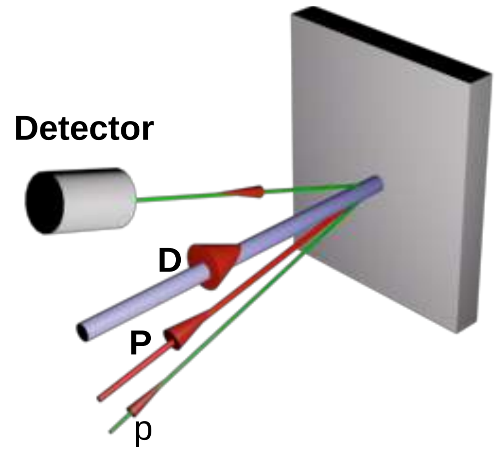
The order parameter, $\psi(t) = A(t)e^{i\phi(t)}$

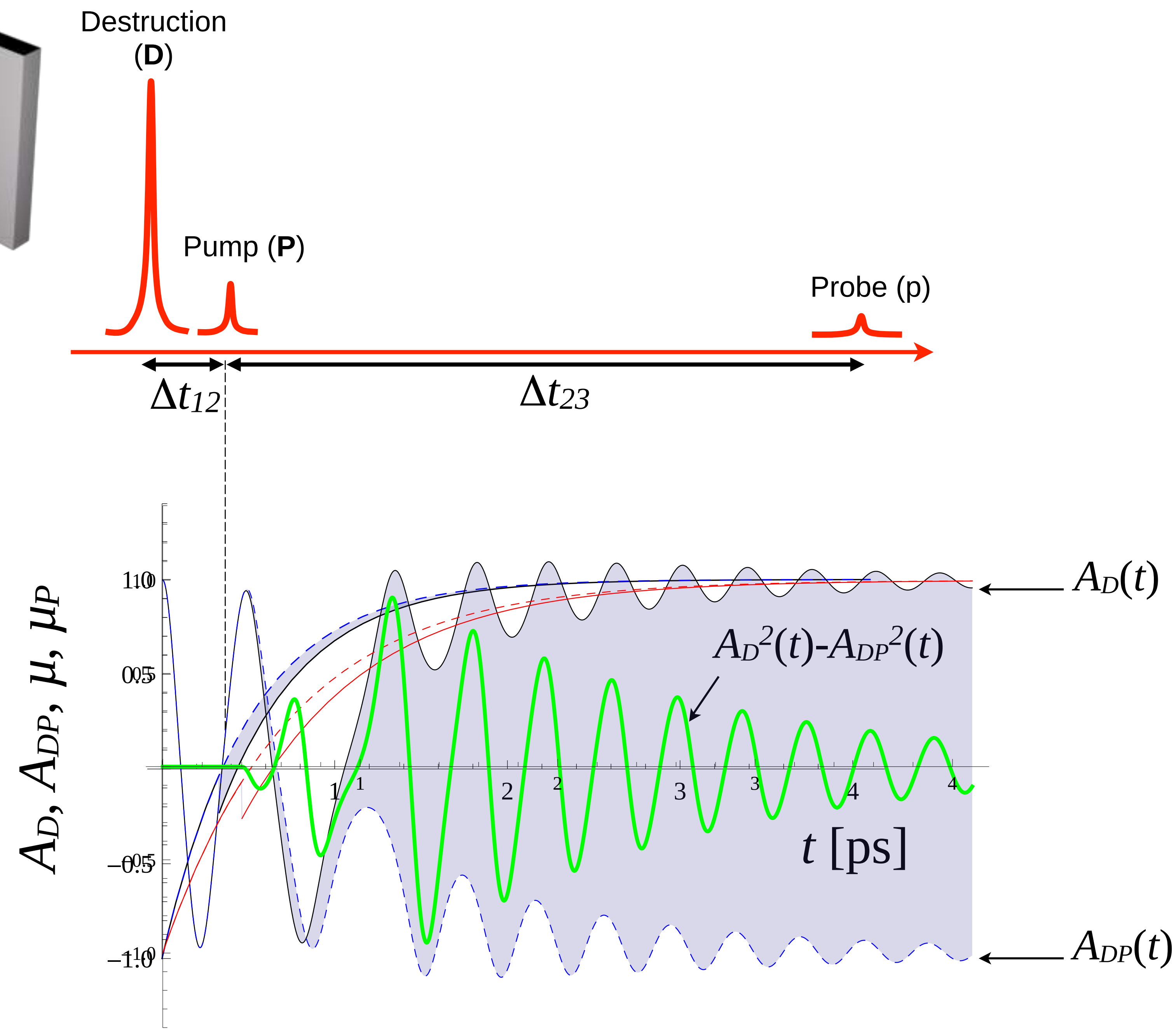
† Phase fluctuations are assumed to be slow.

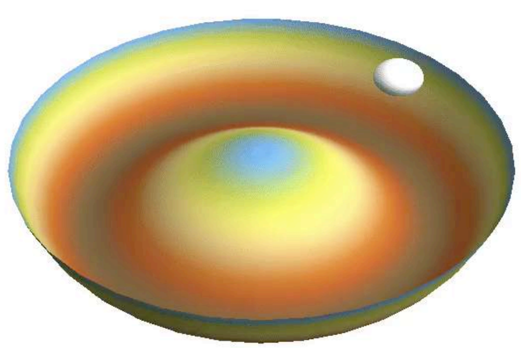


Yusupov, R. *et al.* Coherent dynamics of macroscopic electronic order through a symmetry breaking transition. *Nat Phys* **6,** 681–684 (2010).

The predicted optical response of the collective mode

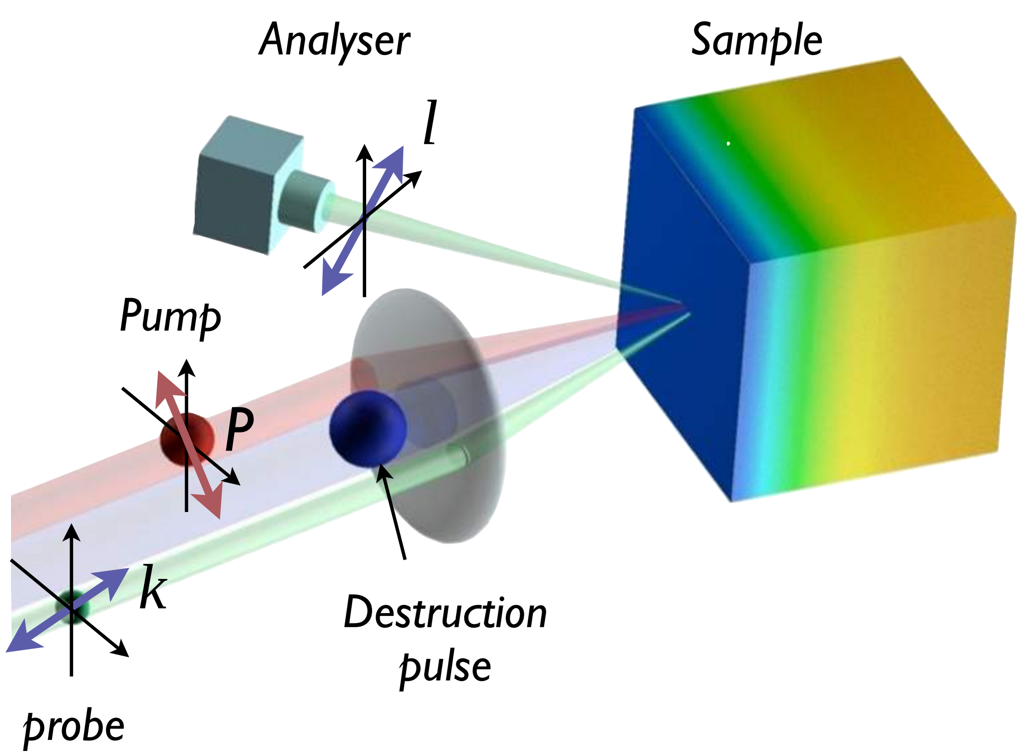




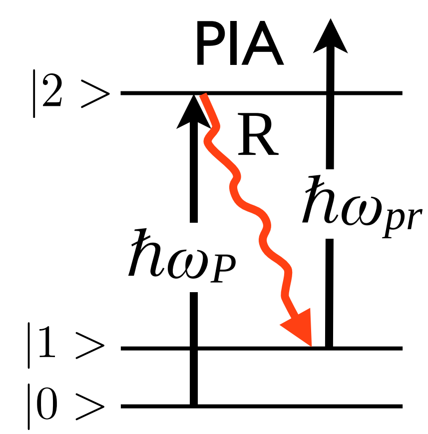


The reflectivity, $\Delta R(t) \propto \left(\frac{\partial R}{\partial \epsilon}\right) \Delta\epsilon \propto \int [A^2_{DP}(t, \mathbf{r}, \Delta t_{12}) - A^2_D(t, \mathbf{r})] e^{-z/\lambda} d^3\mathbf{r}.$

The response of the probe in all-optical experiments



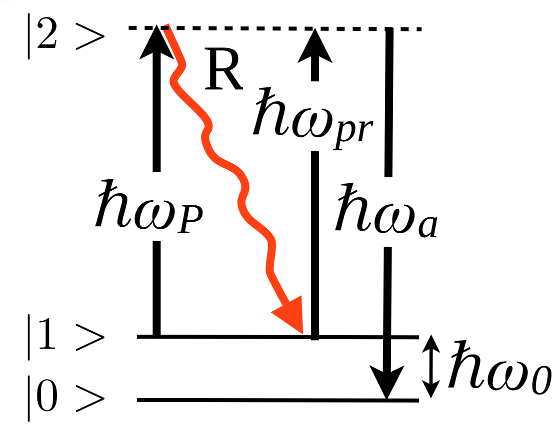
1. Photoinduced absorption (PIA):



The polarisation selection rules are determined by the dielectric tensor

1. Kabanov, V., Demsar, J., Podobnik, B. & Mihailovic, D. *Phys Rev B* **59,** 1497–1506 (1999).
2. Dvorsek, D. *et al.* *Phys Rev B* **66,** 020510 (2002).
3. Mihailovic, D., et al,., *J Phys-Condens Mat* **25,** 404206 (2013).

2. Coherent Raman-like (CRS) process:

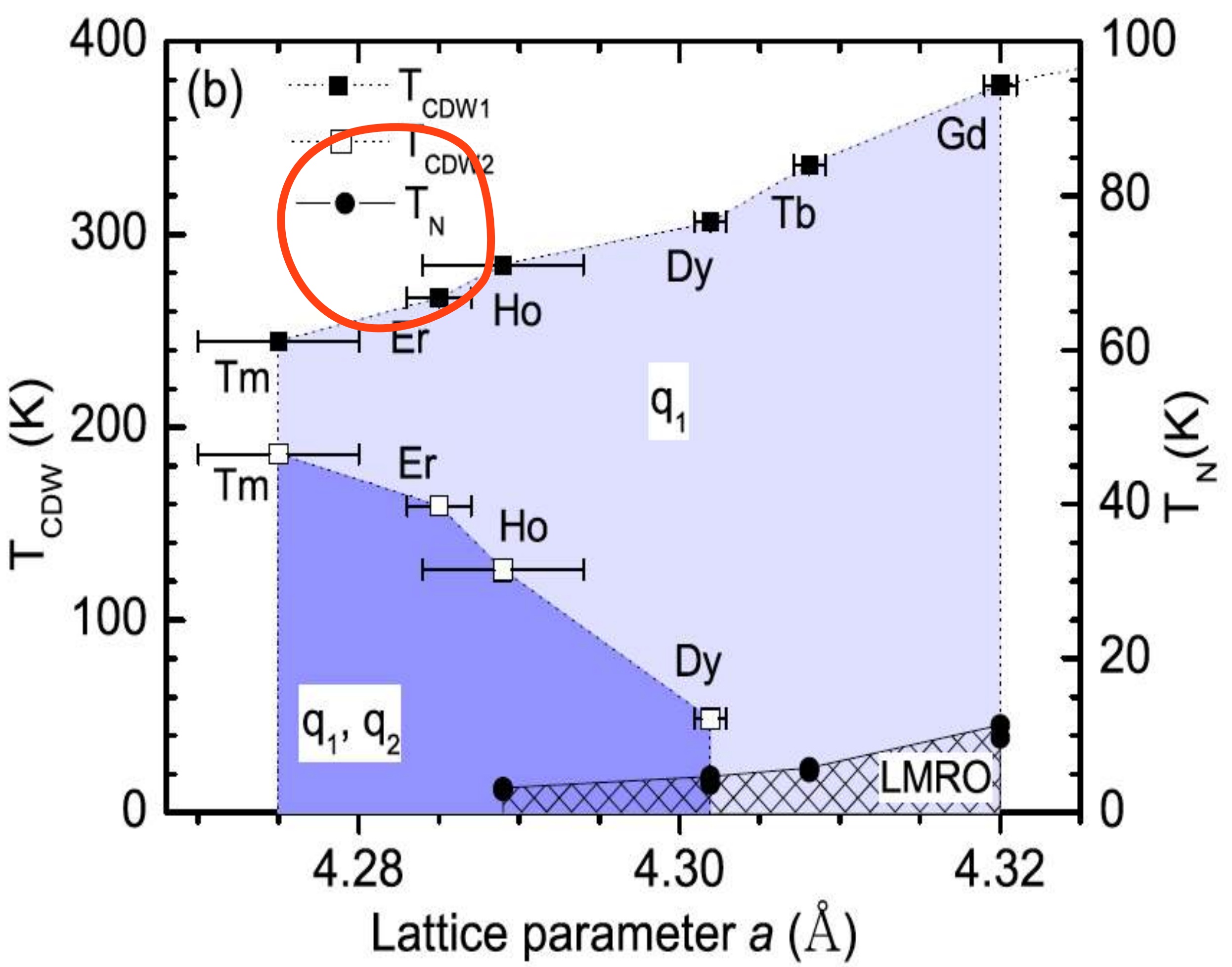


The polarisation selection rules are governed by the Raman tensor χ_{kl}

1. Garrett, G., Albrecht, T., WHITAKER, J. & Merlin, R. *Phys Rev Lett* **77,** 3661–3664 (1996).
2. Stevens, T. E., Kuhl, J. & Merlin, R. *Phys Rev B* **65,** 144304 (2002).

CRS and PIA probe processes can be distinguished by polarisation selection rules

CDWs: the tri-tellurides

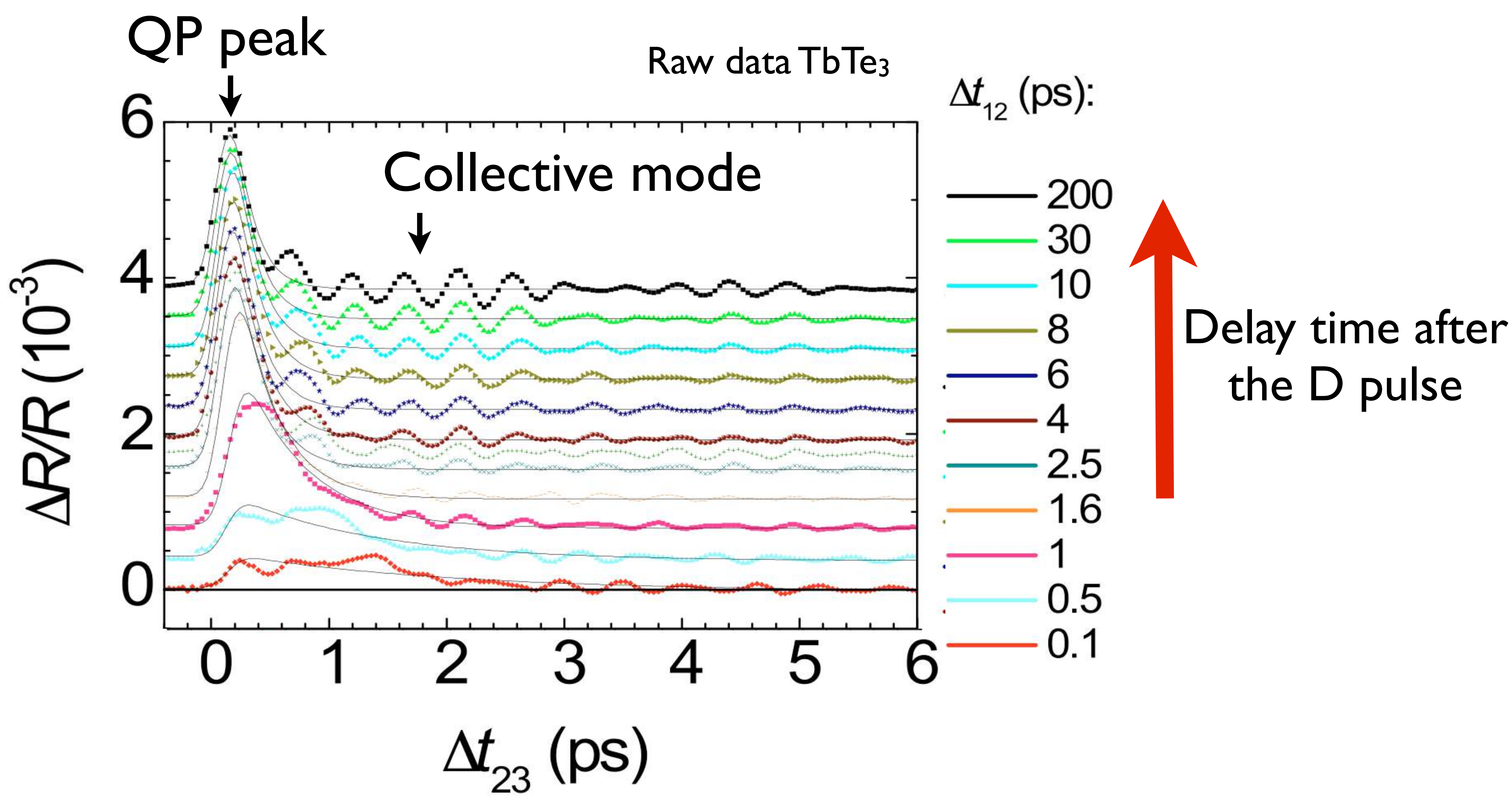


Yusupov, R. V., Mertelj, T., Chu, J. H., Fisher, I. R. & Mihailovic, D. Single-Particle and Collective Mode Couplings Associated with 1-and 2-Directional Electronic Ordering in Metallic RTe3 (R=Ho,Dy,Tb). **101,** 246402 (2008).

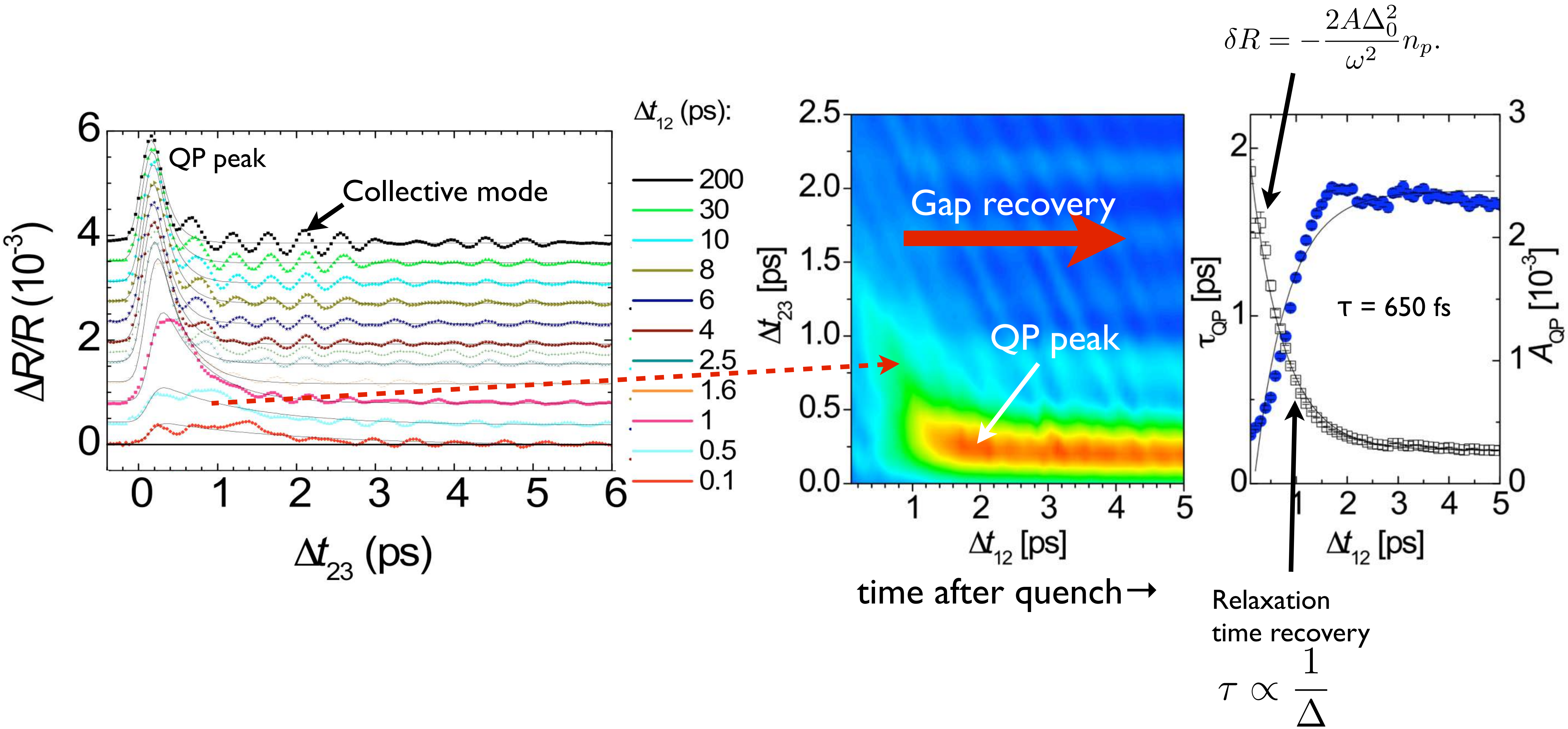
- The tritellurides are layered, quasi 2-dimensional metals with an orthorhombic (pseudo-tetragonal) crystal structure Cmca (D_{2h})
- They exhibit a purely electronically driven 2nd order incommensurate **CDW transition** at T_{c1} = 230~330K
- An **AFM state** exists at low T_N, some compounds exhibit another transition at low T_{c2}.
- A **Superconducting** transition exists with Tc = 3.5 K under a pressure of 75 kbar.

DiMasi '94,'95, Fisher '05,'08

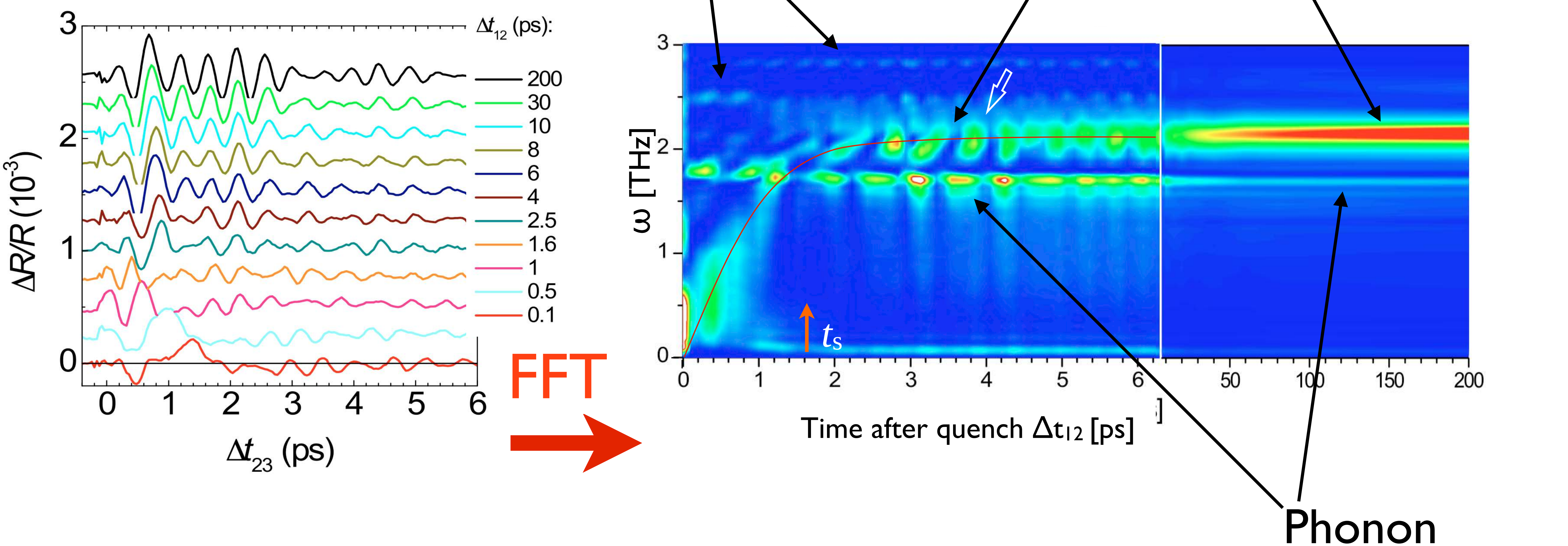
The transient reflectivity ΔR/R after a quench at Δt_{12}=0 in $TbTe_3$



Quasi-particle (Fermion) evolution through t_c: gap recovery

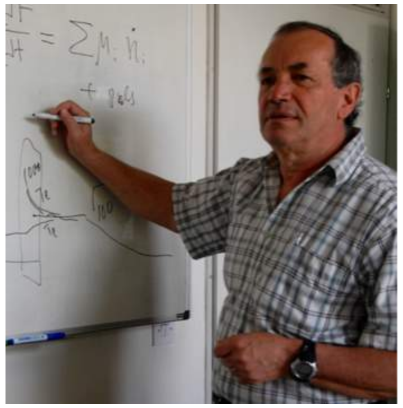


The collective mode (boson) spectrum as a function of time after quench



The most obvious feature:
oscillations of intensity of the collective mode

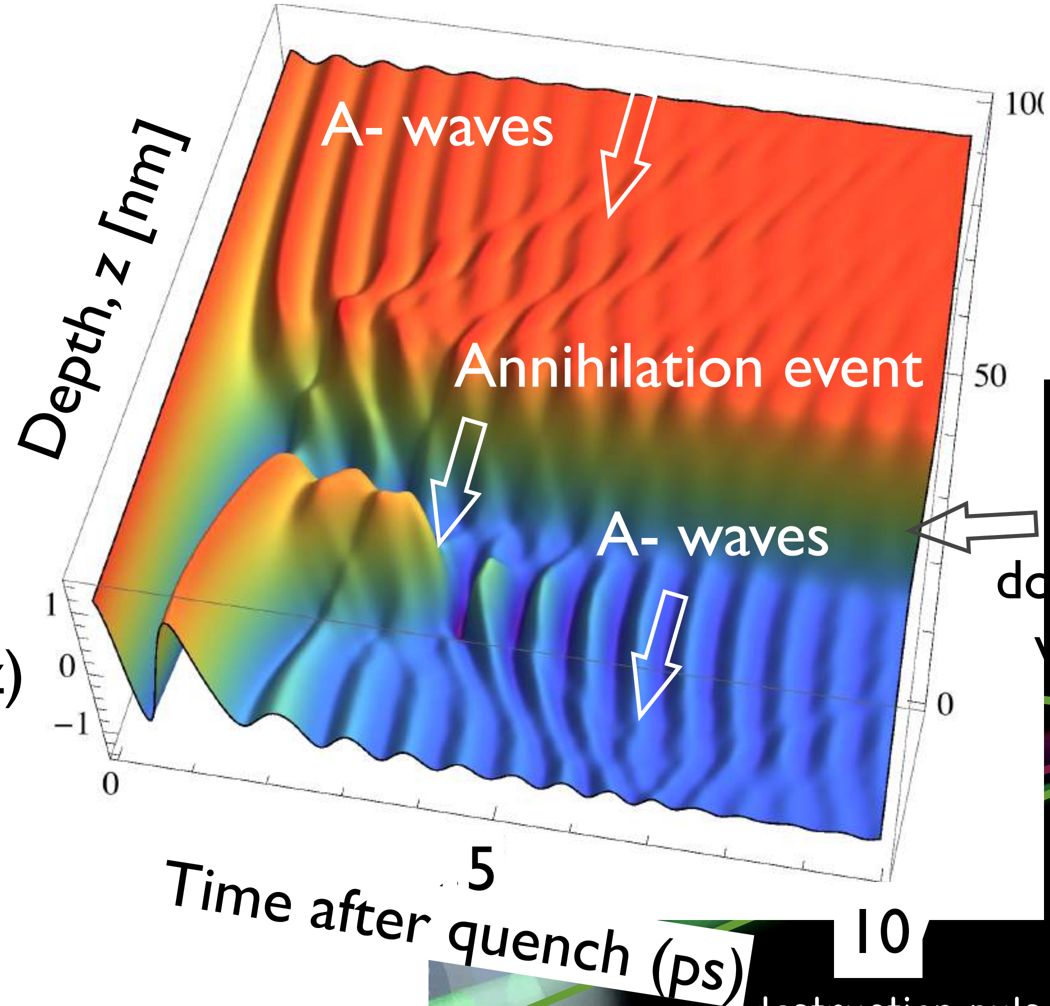
Order parameter calculation for an inhomogeneous system

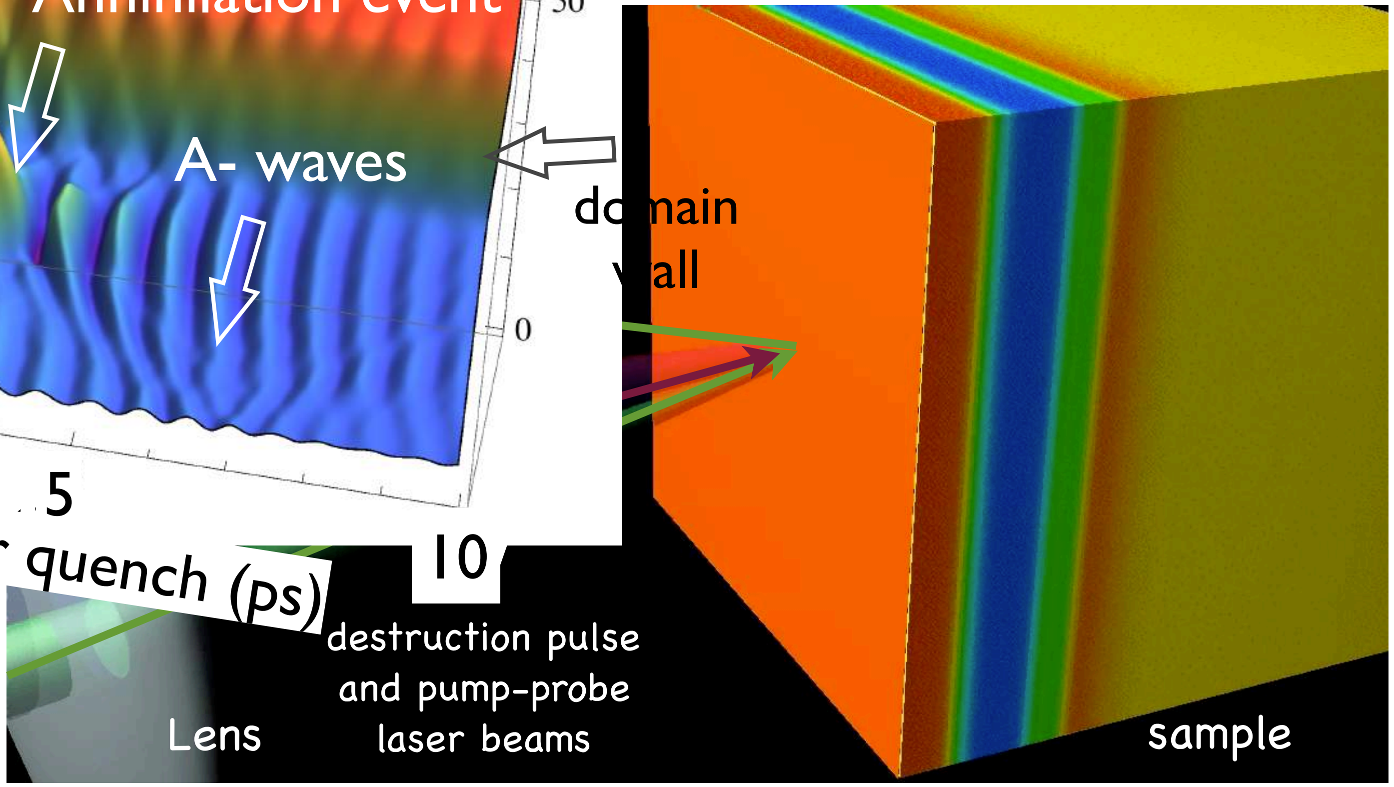


The eq. of motion:

$$\frac{1}{\omega_0^2}\frac{\partial^2}{\partial t^2}A + \frac{\alpha}{\omega_0}\frac{\partial}{\partial t}A - (1-\eta)A + A^3 - \xi^2\frac{\partial^2}{\partial z^2}A = 0$$

Calculated *A(z,t)* after quench:

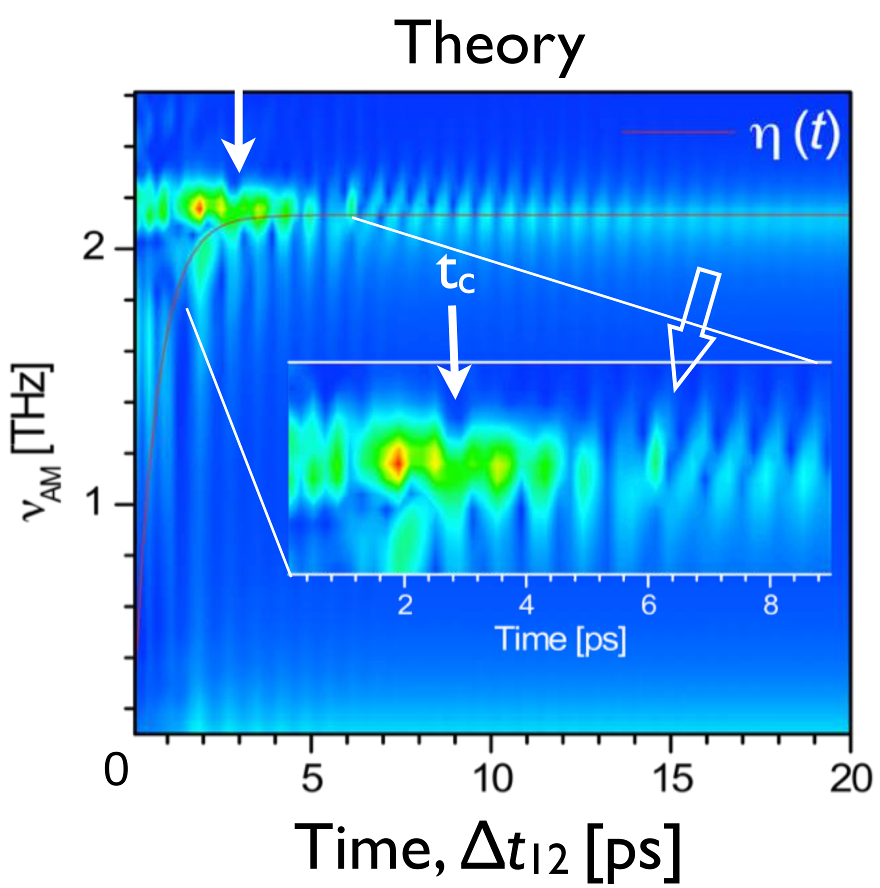


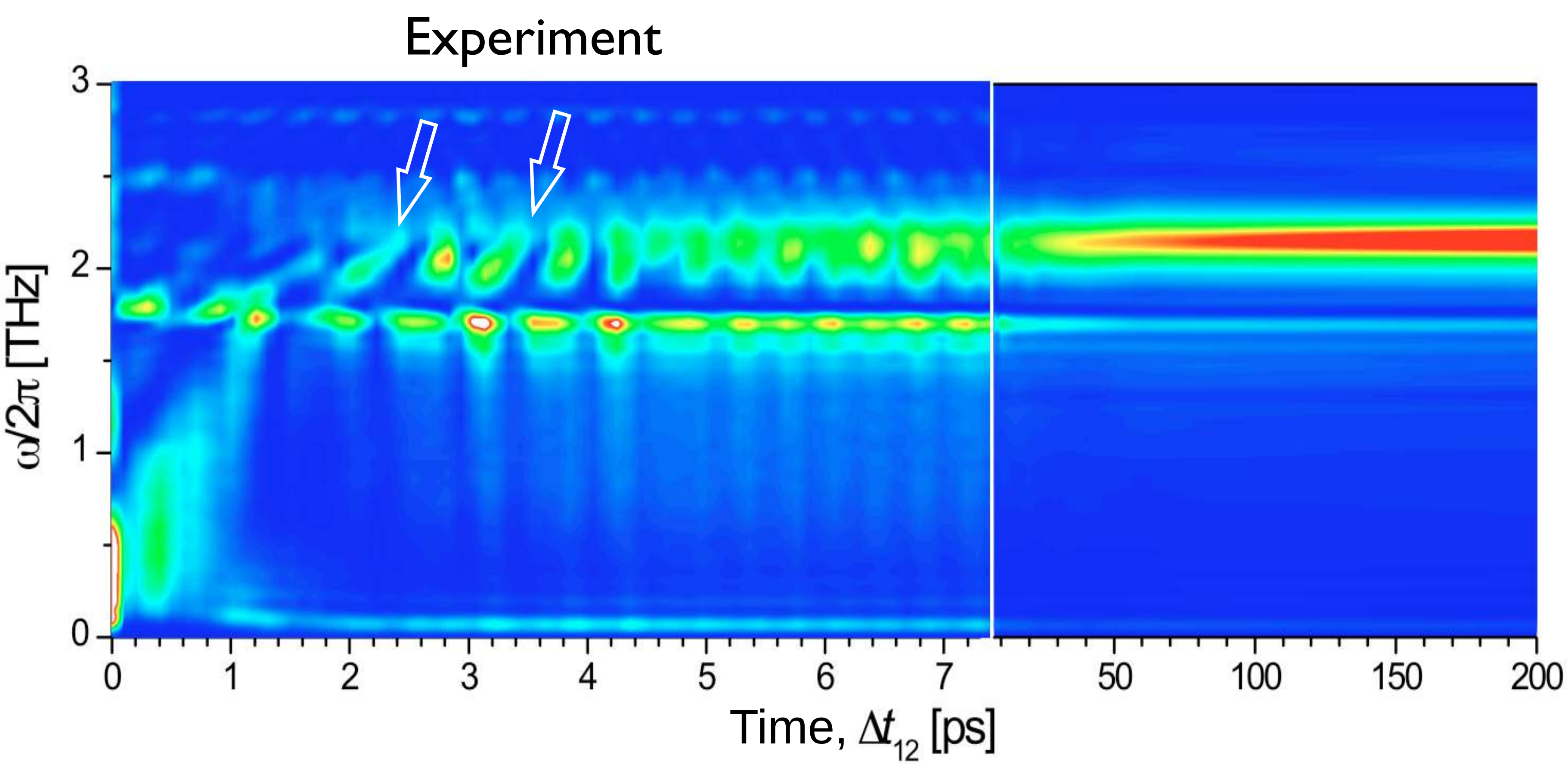


Experimental parameters:

$\tau_{QP} = 650$ fs
$\omega_0/2\pi = 2.18$ THz
$\eta = 2$
$\alpha = 0.1$

Order parameter dynamics: theory vs. experiment

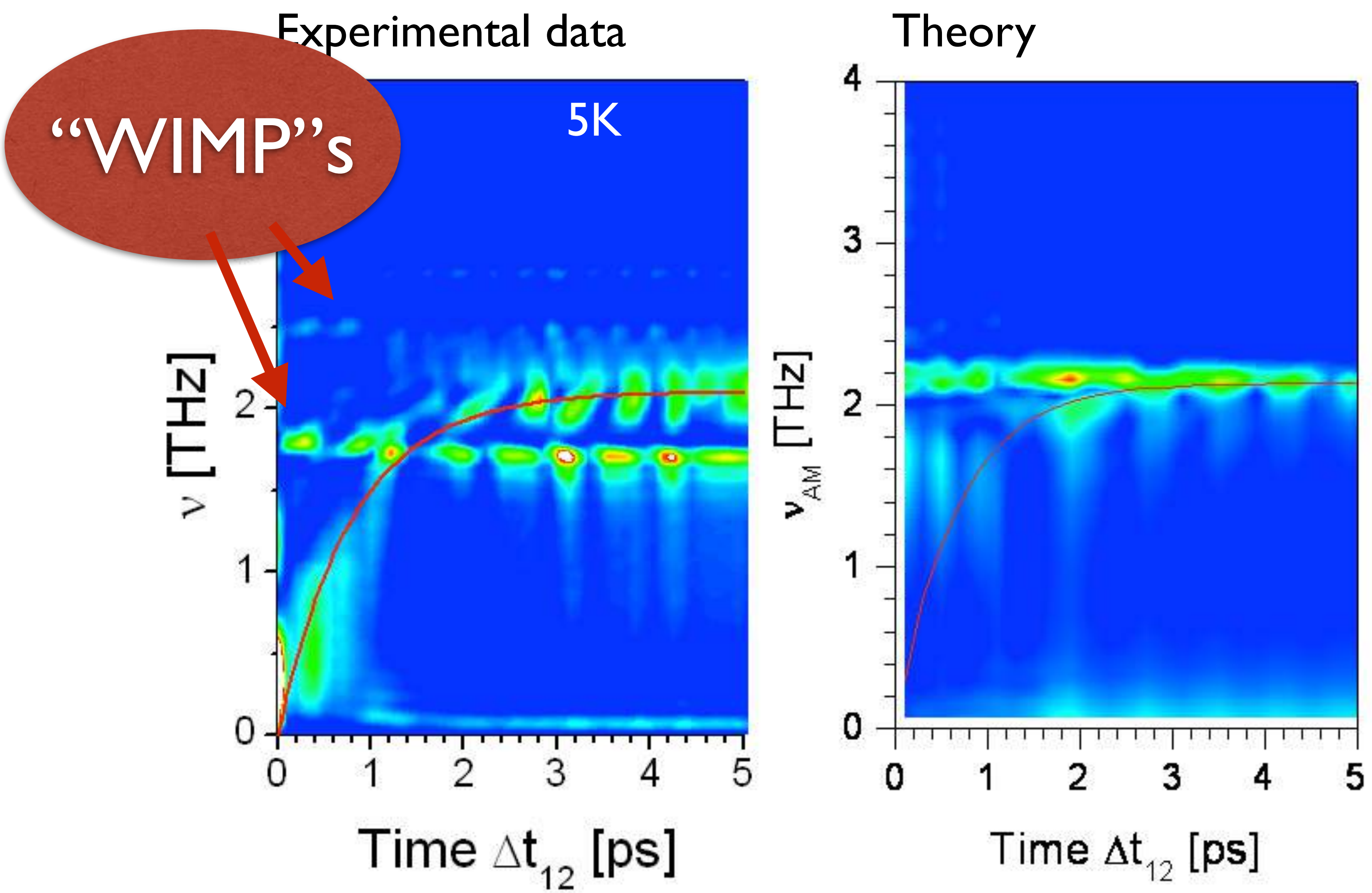




Theory predictions:	Experimental observations
Oscillations of Δ or |Ψ|	Intensity oscillations
Critical slowing down	
(Collective mode softening)	Softening of ω
Domain annihilation	
Ψ field (Higgs) waves	Distortions in ω-t spectra

Critical dynamics near t_c.

Pre-transition behaviour: non-ergodic processes

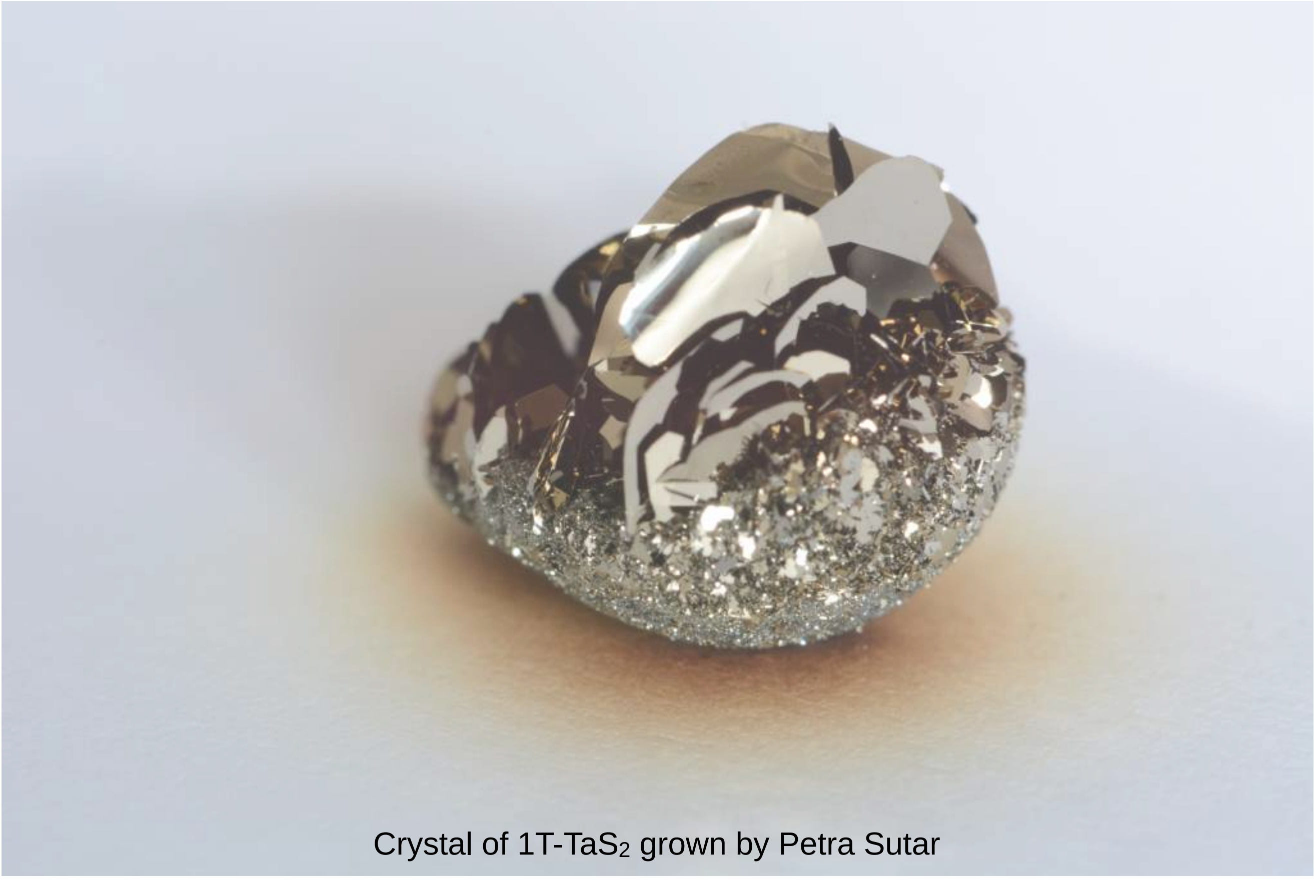


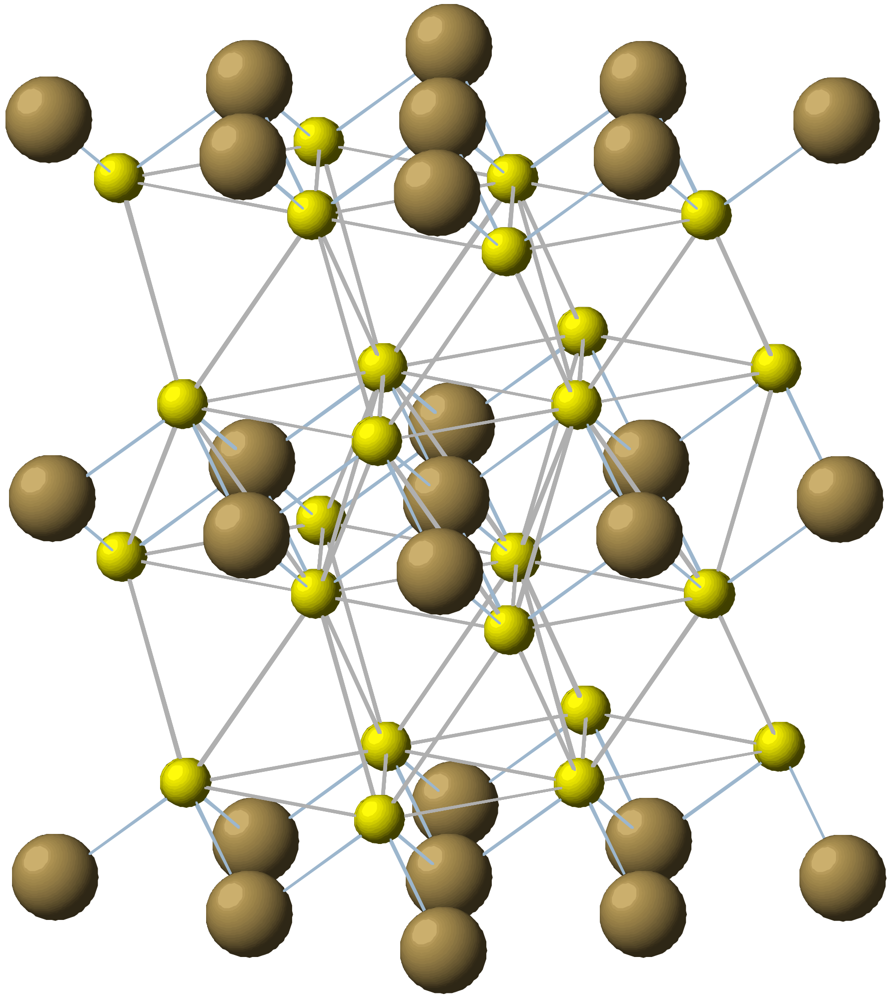
Phonons: WIMPs from previous eons

"By analogy with the PM, dark matter excitations may be thought of as remnant excitations from before *the SBT* (the Big Bang)."

Mihailovic, D., Mertelj, T., Kabanov, V. V. & Brazovskii, S. Coherent topological defect dynamics and collective modes in superconductors and electronic crystals. *J Phys-Condens Mat* **25,** 404206 (2013). (Invited paper by T.W.Kibble)

The trajectory to a <u>hidden</u> state in *1T*-TaS_2





1T-TaS_2

What is a hidden state?

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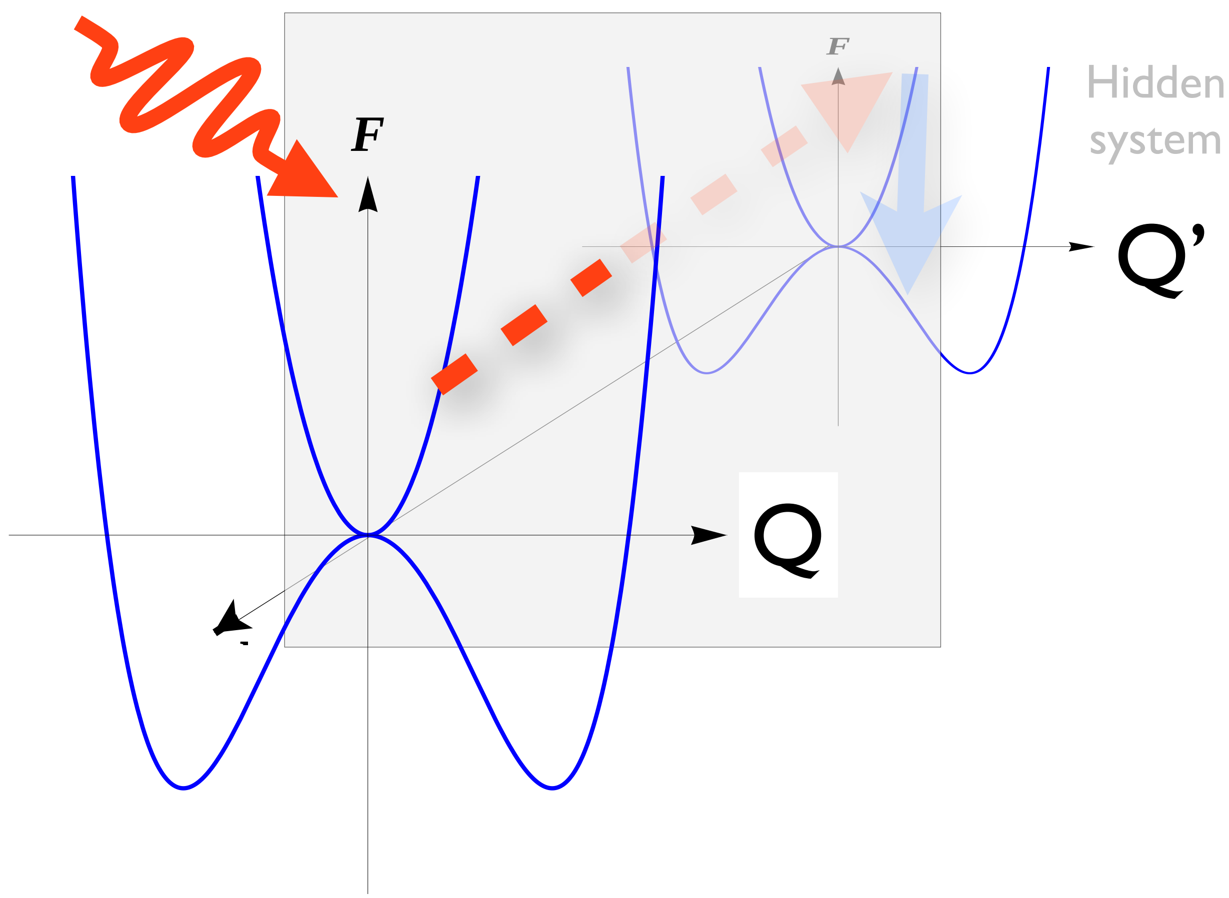
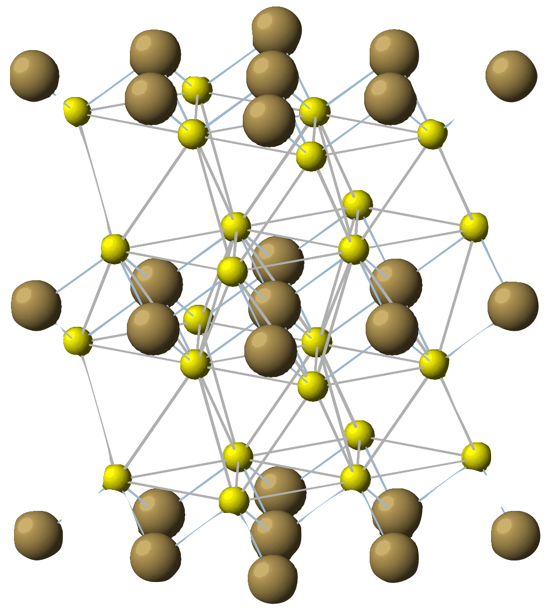
Hidden states of matter

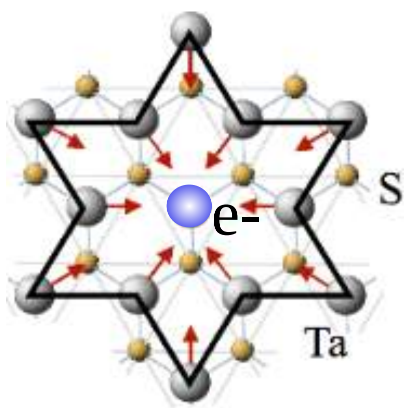
From Wikipedia, the free encyclopedia

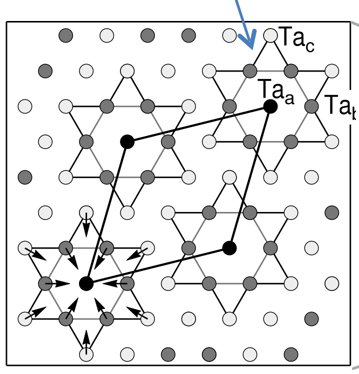
A **hidden state of matter** is a state of matter which cannot be reached under ergodic conditions, and is therefore distinct from known thermodynamic phases of the material.[1] Examples exist in condensed matter systems, and are typically reached by the non-ergodic conditions created through laser photo excitation.[2][3] Short-lived hidden states of matter have also been reported in crystals using lasers. Recently a persistent hidden state was discovered in a crystal of Tantalum(IV) sulfide (TaS_2), where the state is stable for days at low temperatures.[4] A hidden state of matter is not to be confused with hidden order, which exists in equilibrium, but is not immediately

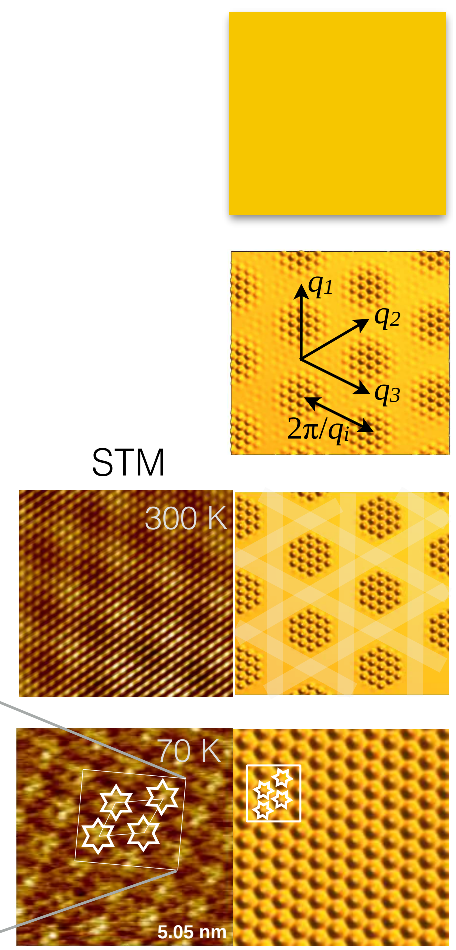


CDWs in 1T-TaS_2









I.Vaskivskyi, 2015 (unpub)

T (K)

Metal

540 K

IC-CDW

340 K

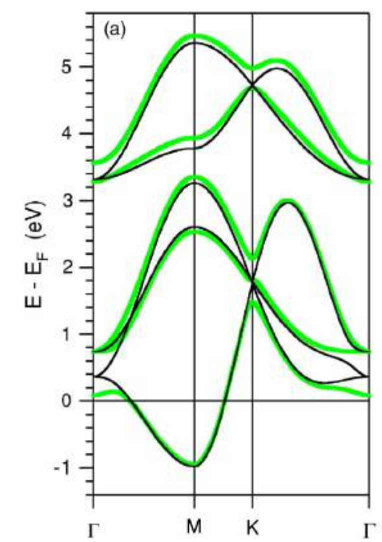
NC-CDW

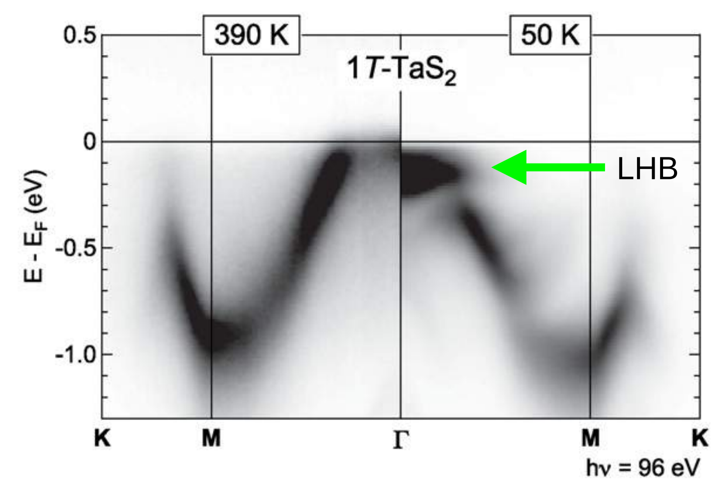
180 K

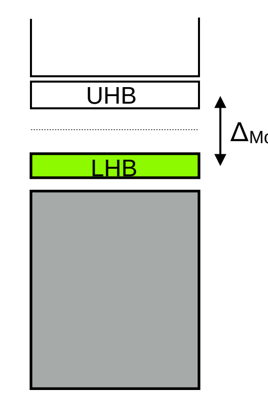
1st order

Mott state (C-CDW)

Kai Rossnagel, 2012





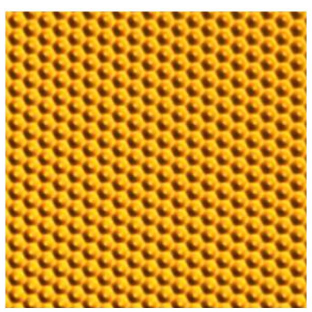


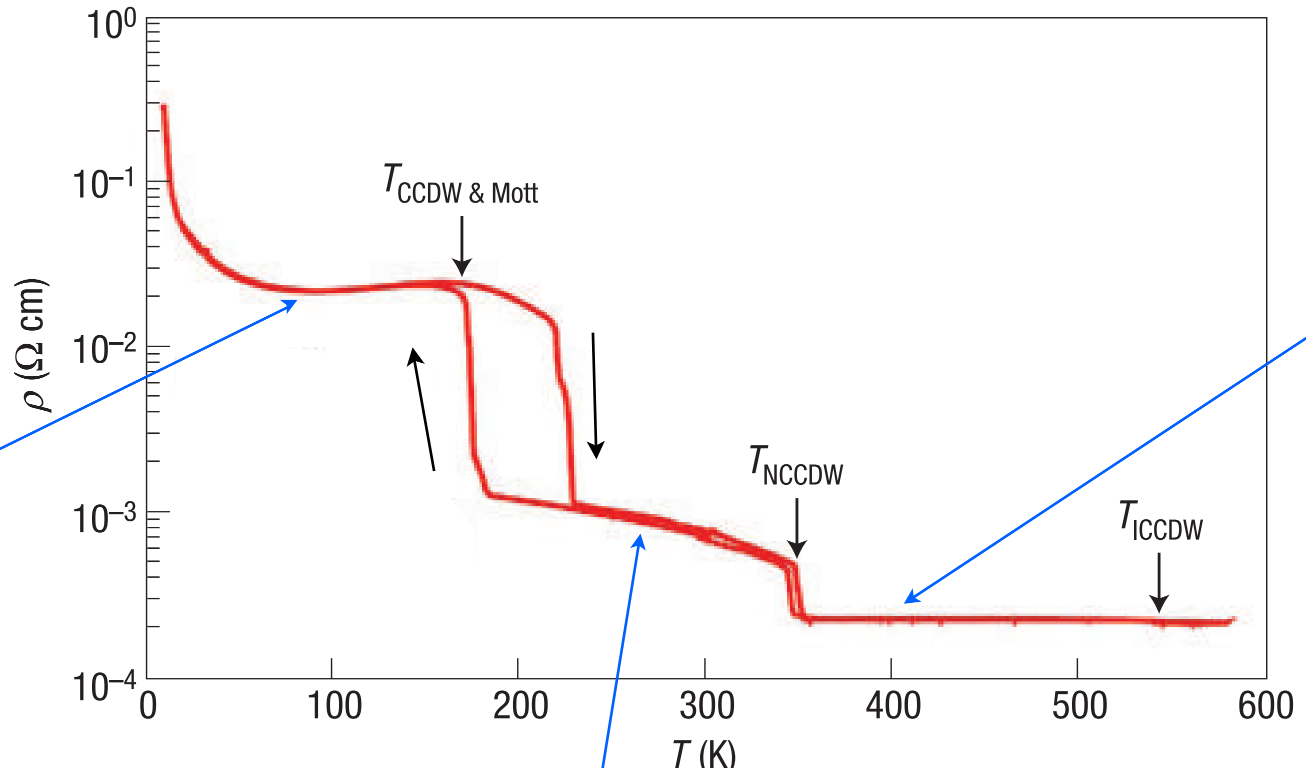
A system with competing Coulomb, Fermi surface instability and lattice strain

Resistivity of $1T$-TaS_2 under equilibrium conditions

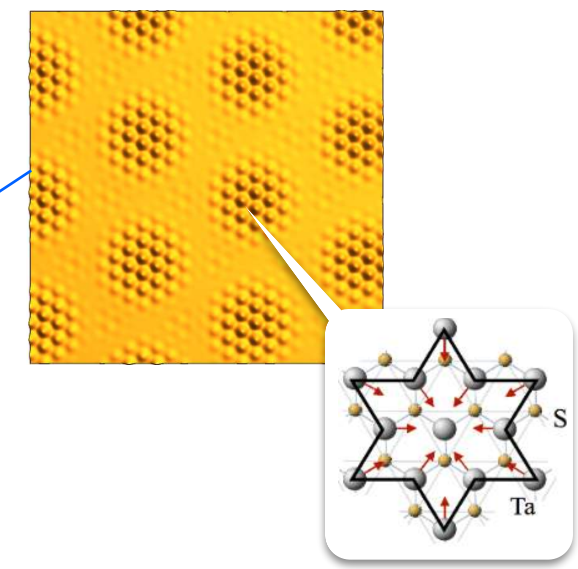
Mott state

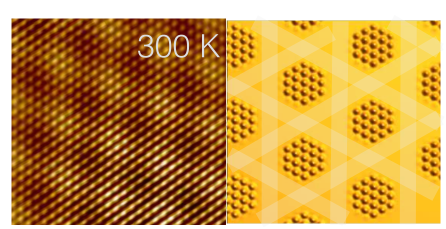
(Commensurate CDW)





Incommensurate CDW

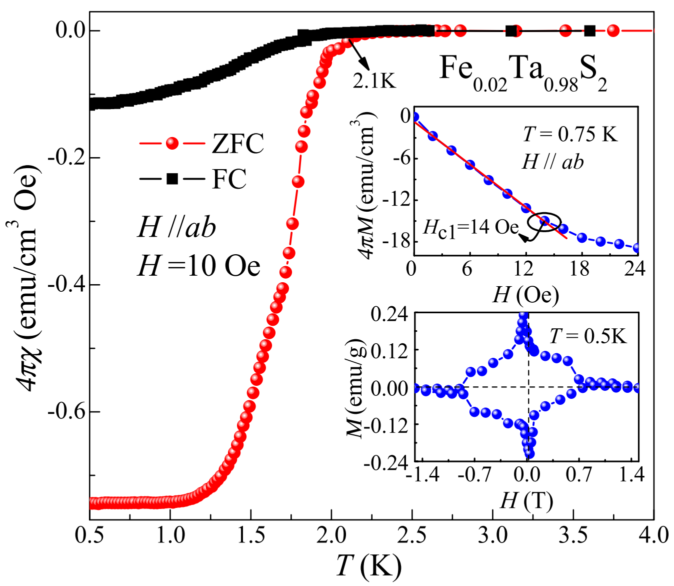


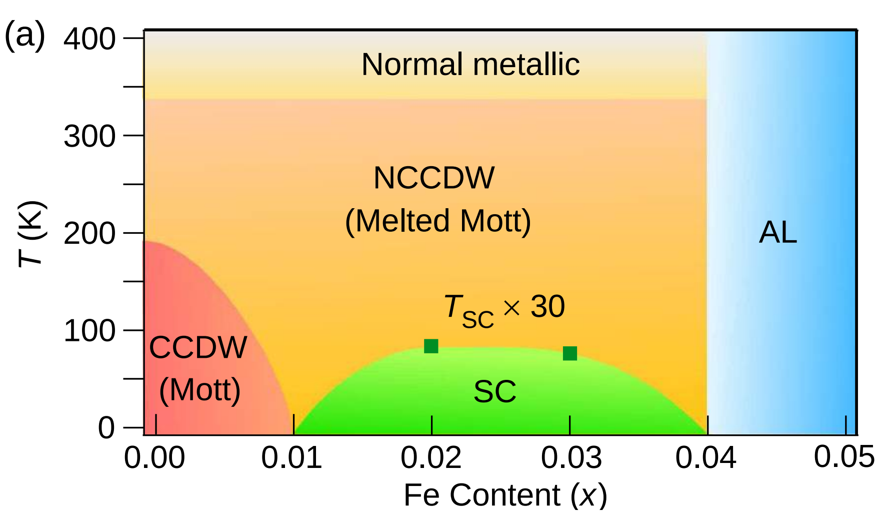


Stretched honeycomb structure with domain walls in between (Nearly Commensurate CDW phase)

Competing orders in 1T-TaS_2: Superconductivity under pressure, or Fe, or Se doping.

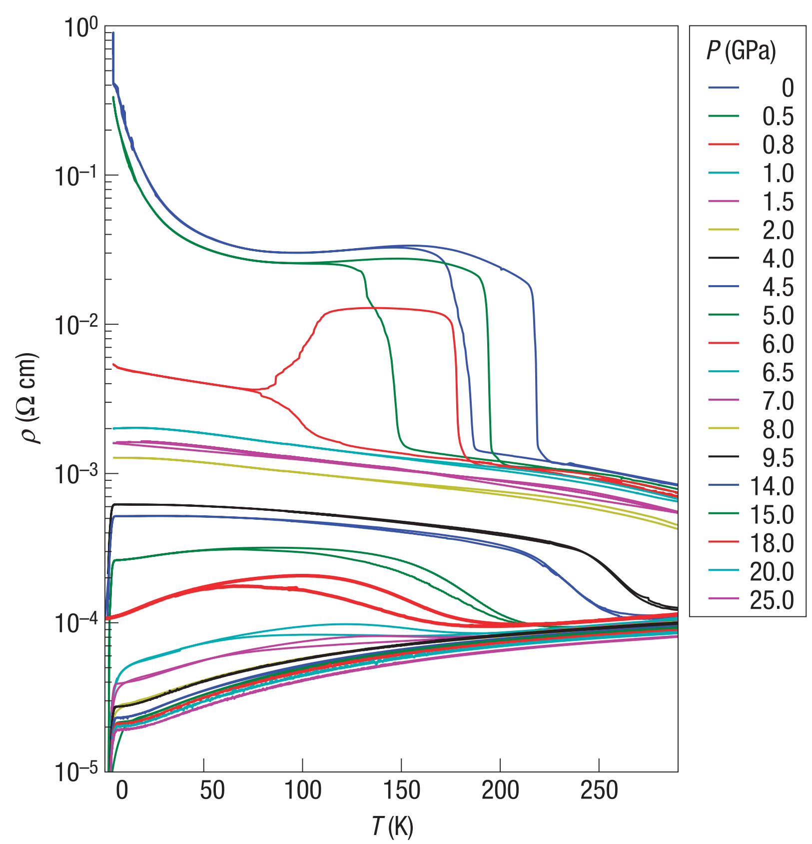
Fe doping:

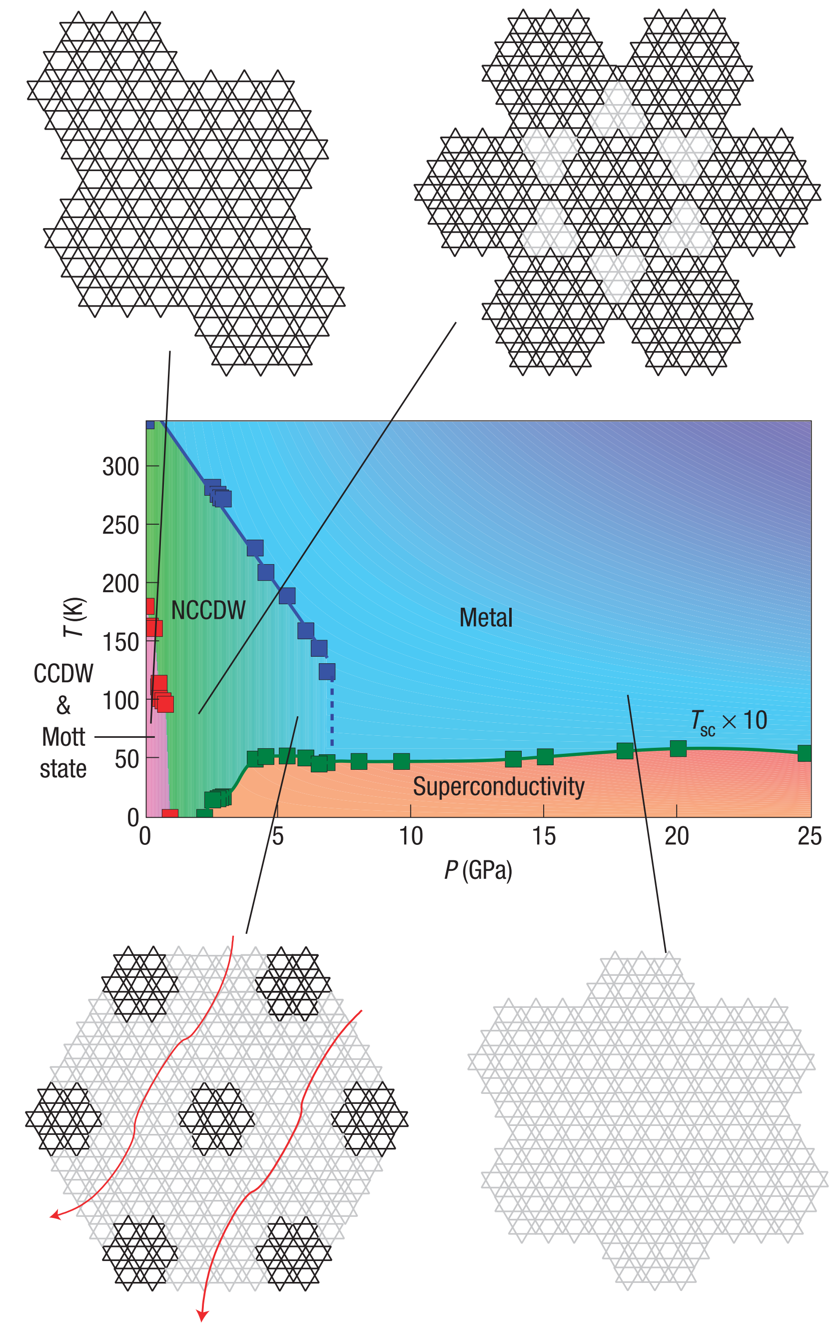




Li et al. EPL 2012

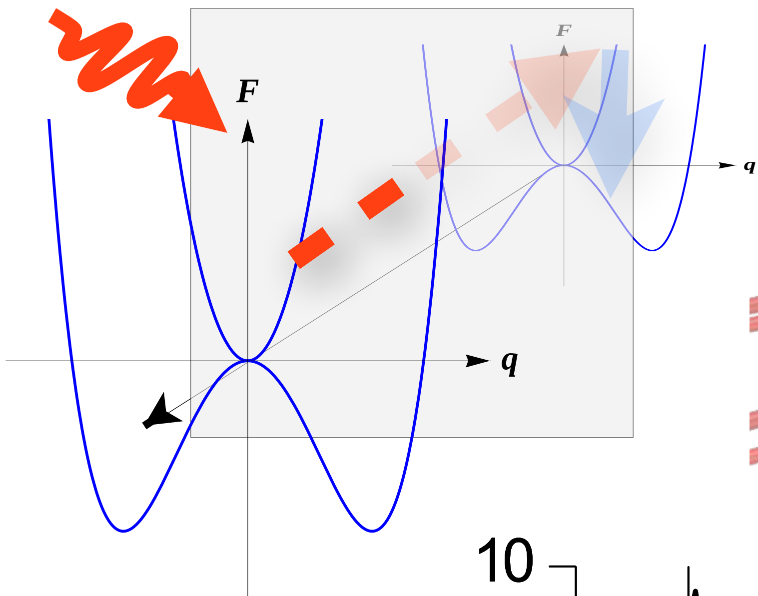
Pressure:

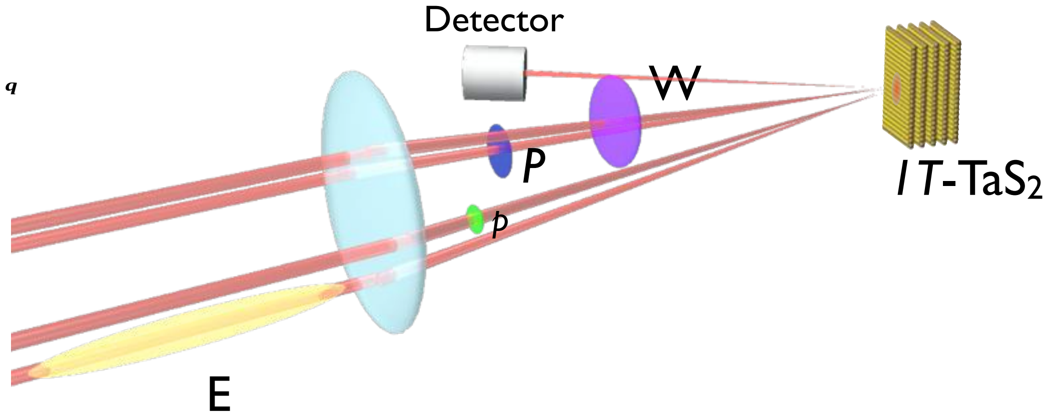




Sipos et al (Nat.Mat. 2008)

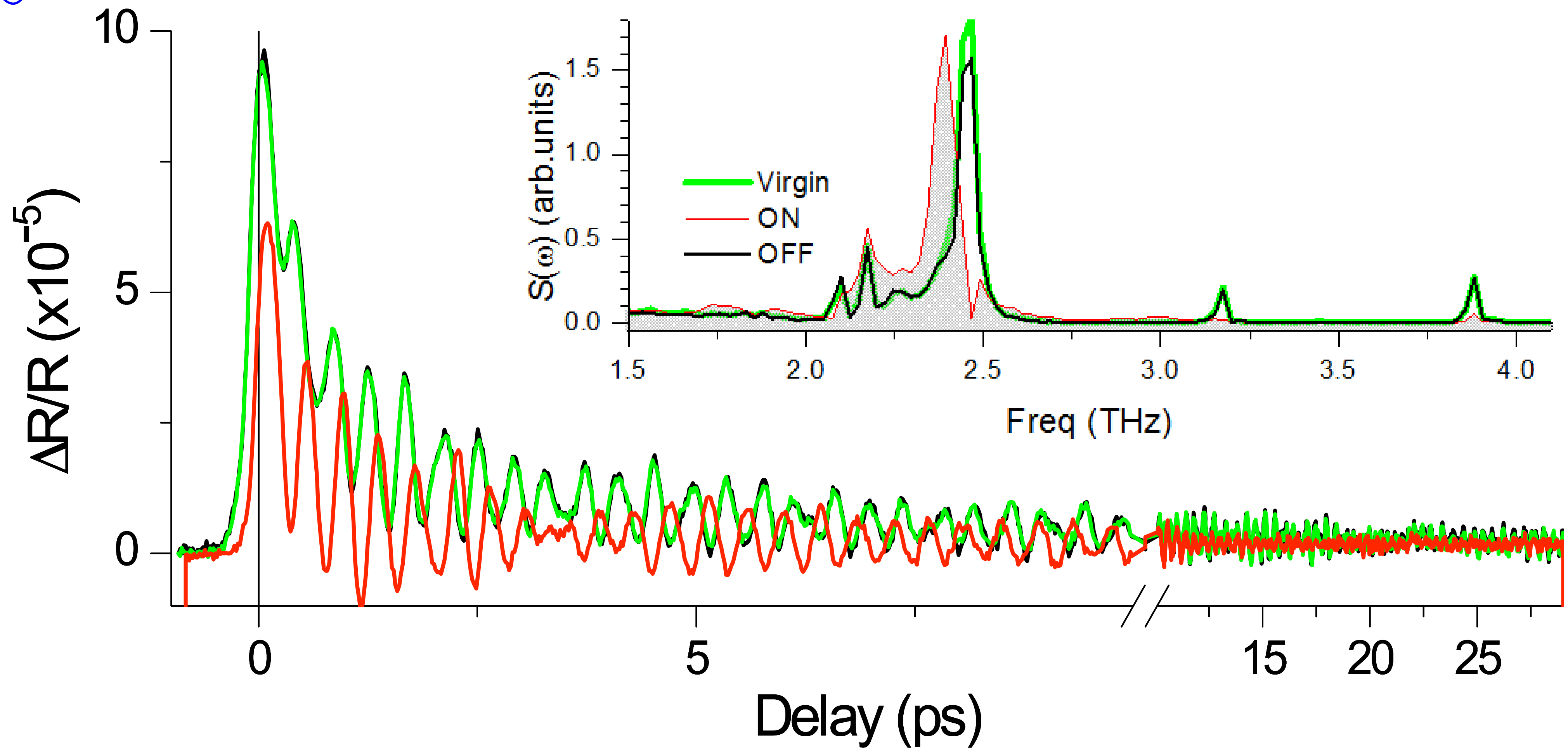
1T-TaS_2: Collective mode switching





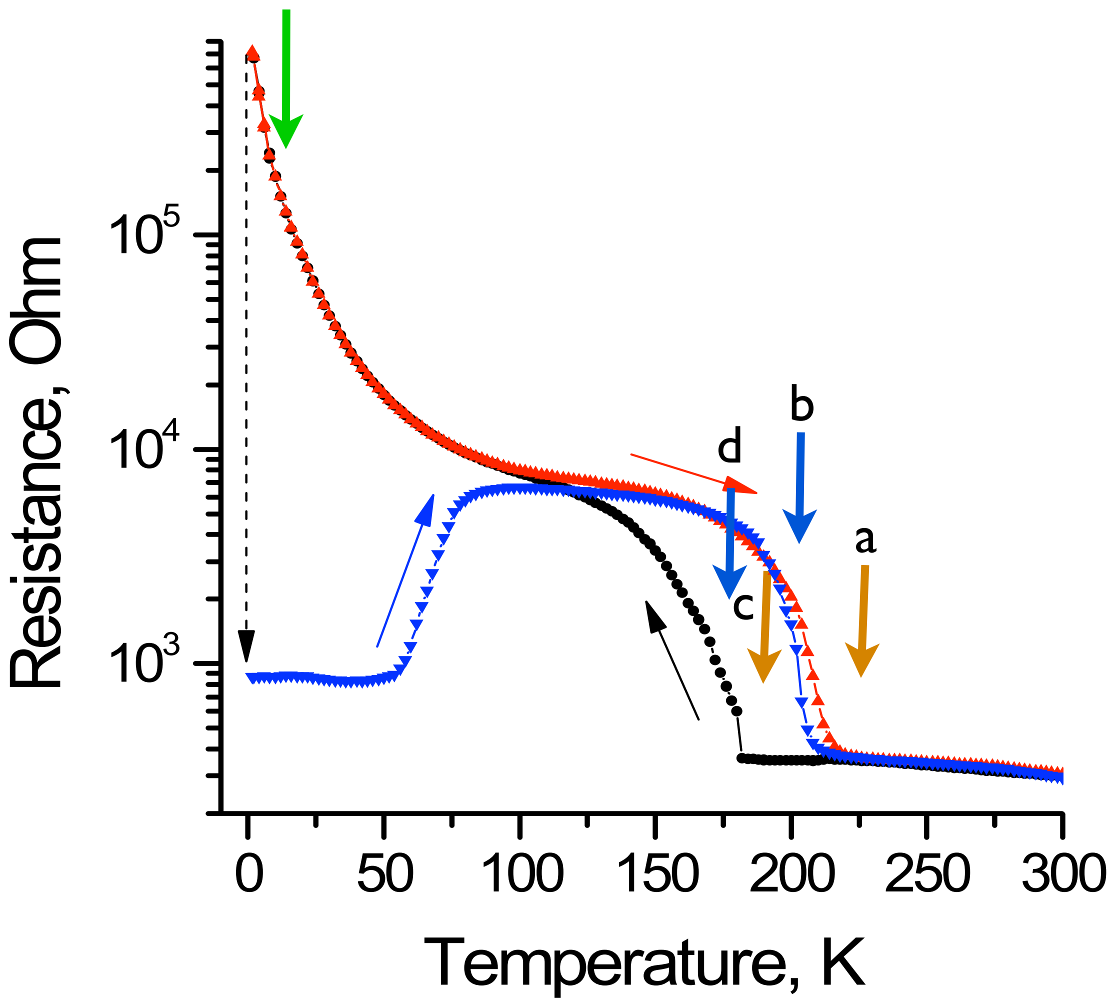
W = 50 fs "write"
E = 50 ps "erase"
P = "pump" (50 fs)
p = "probe" (50 fs)

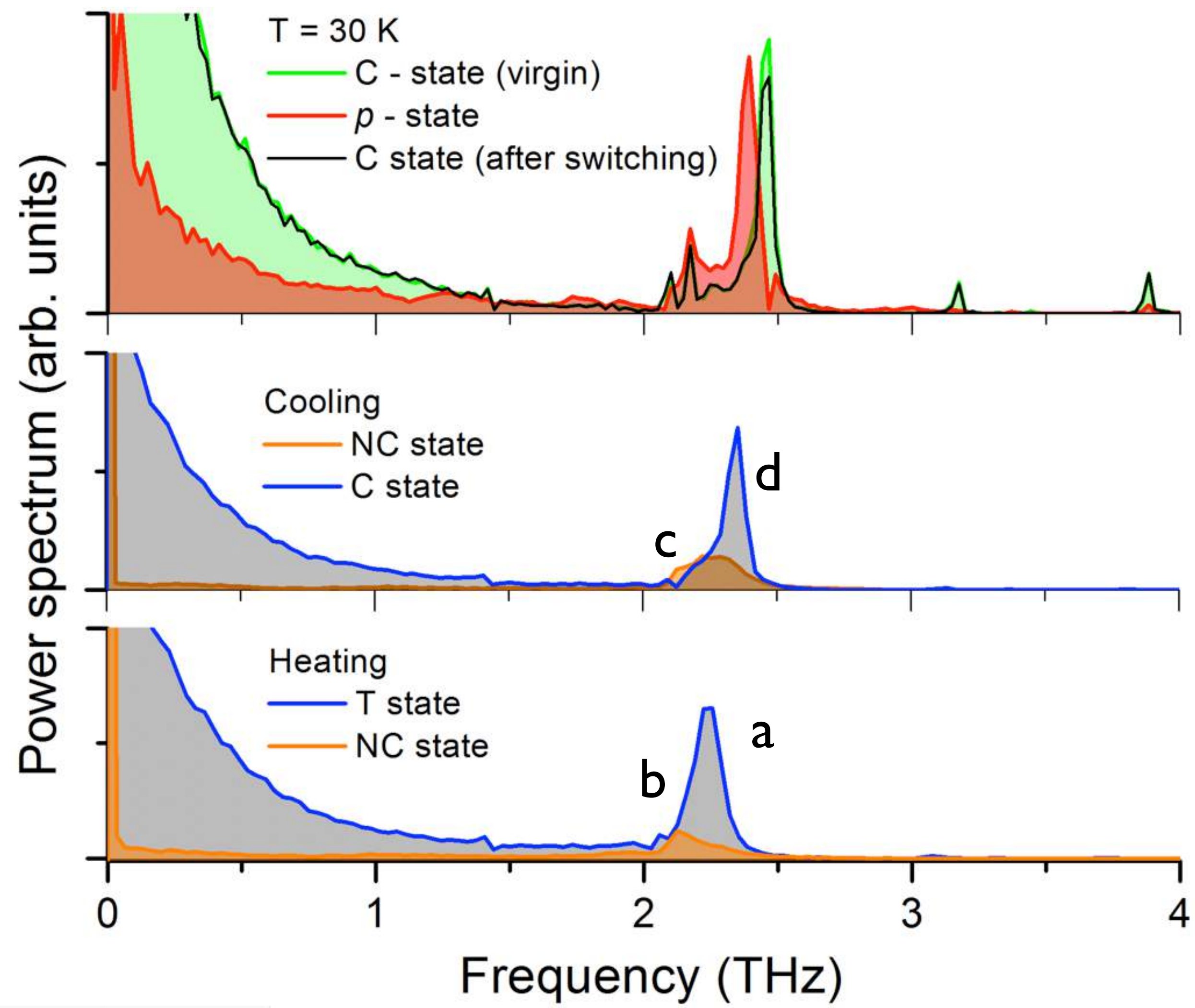
Ljupka Stojchevska



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

Is the hidden state spectrum similar to any known equilibrium state?

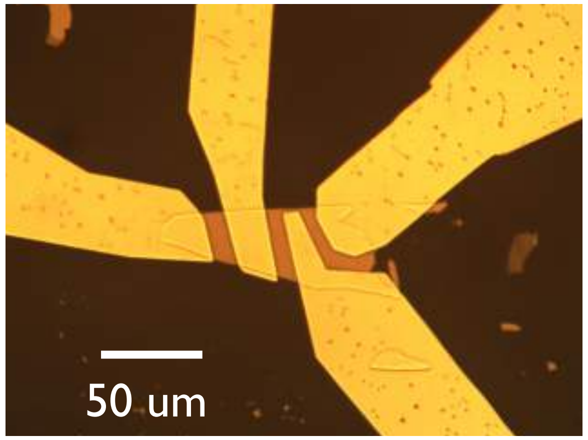




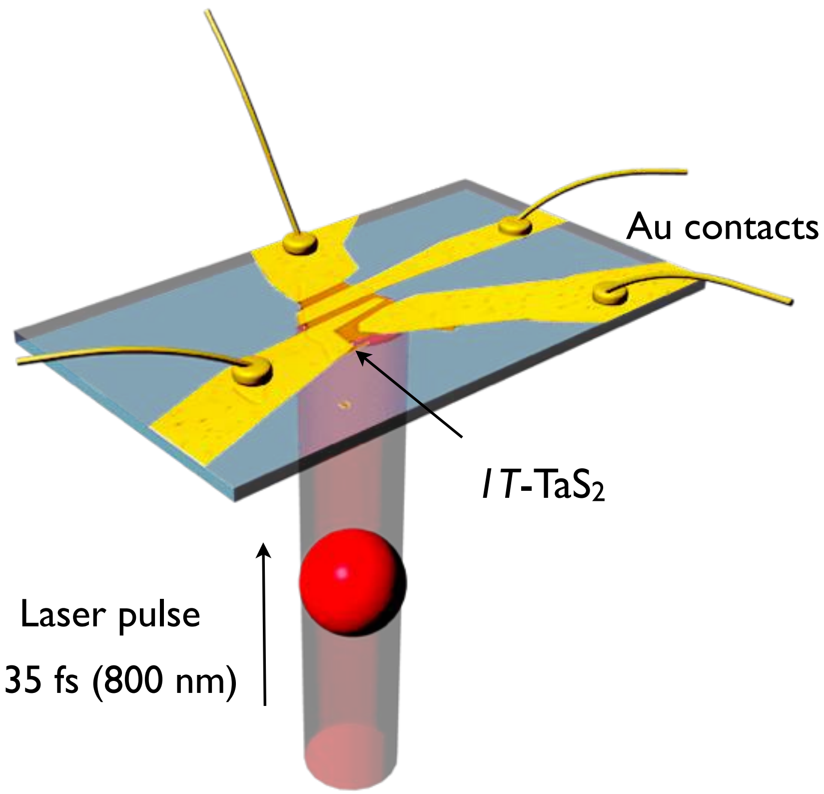
No!

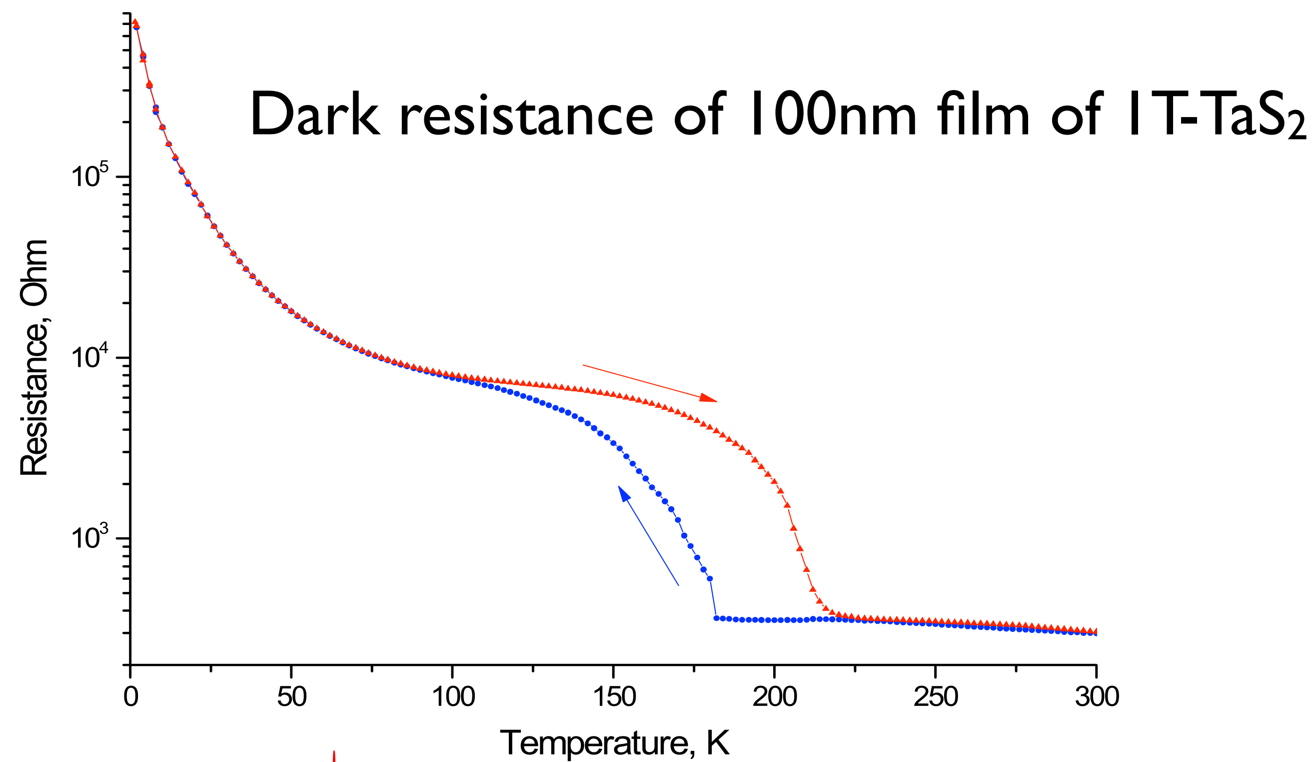
Switching to a hidden state in *1T*-TaS_2 : Resistance change after a (single) 35 fs pulse

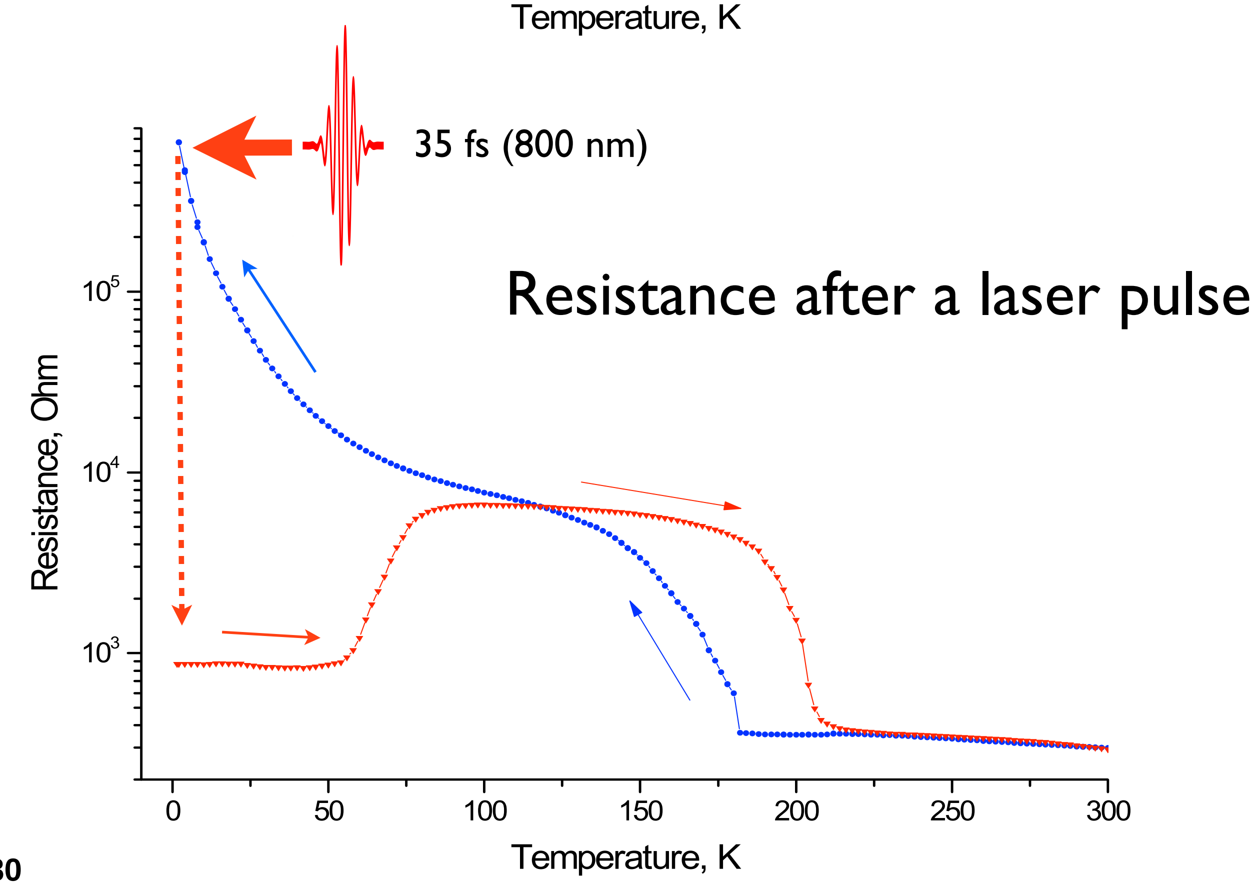
Igor Vaskivskyi



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



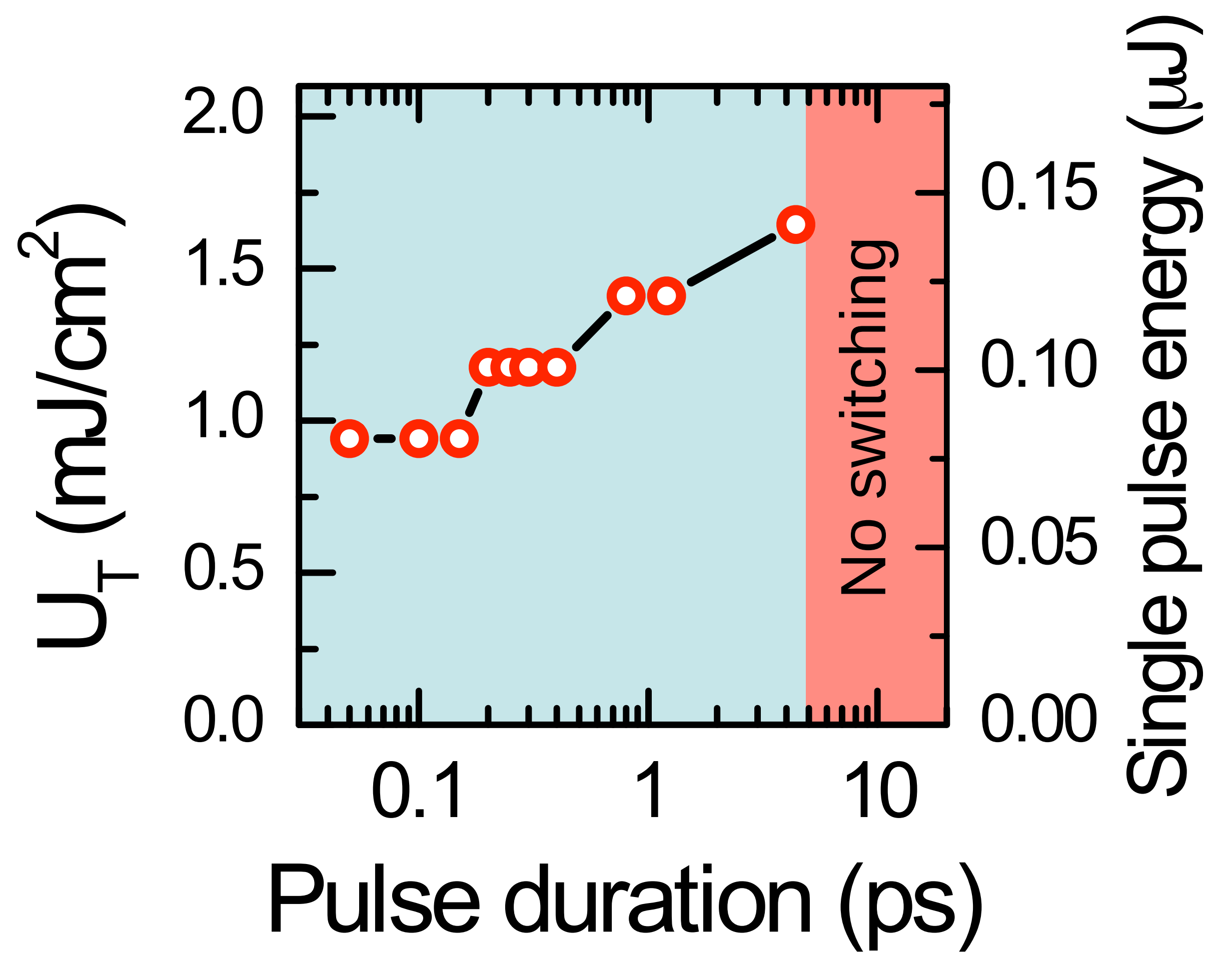




Science AAAS

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

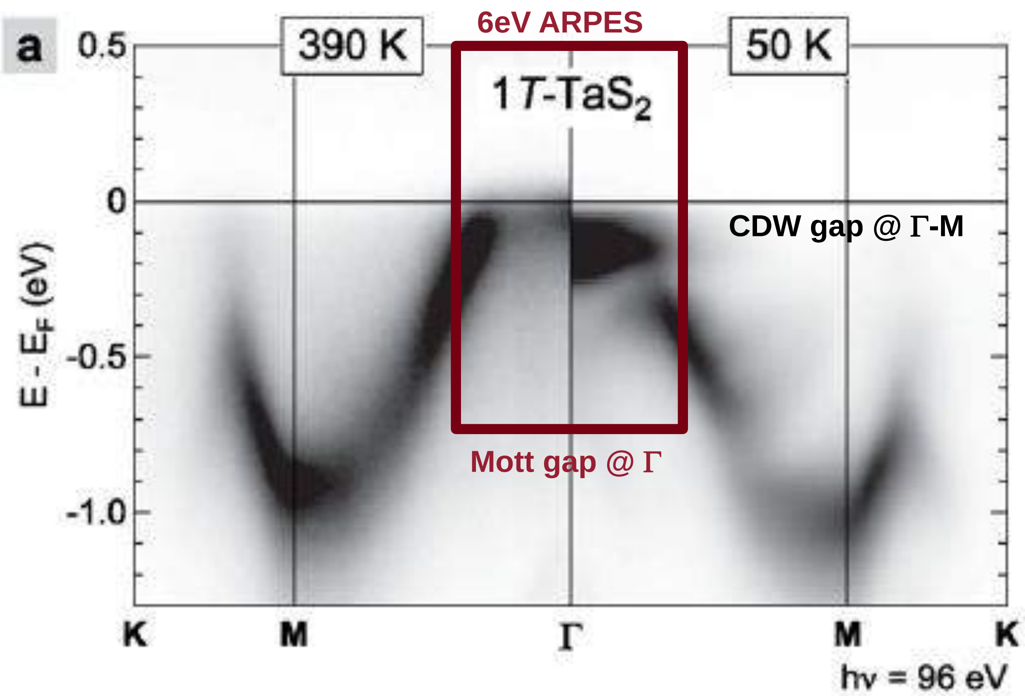
Switching only occurs for short pulses $\tau_L < 4$ ps



Switching in ARPES

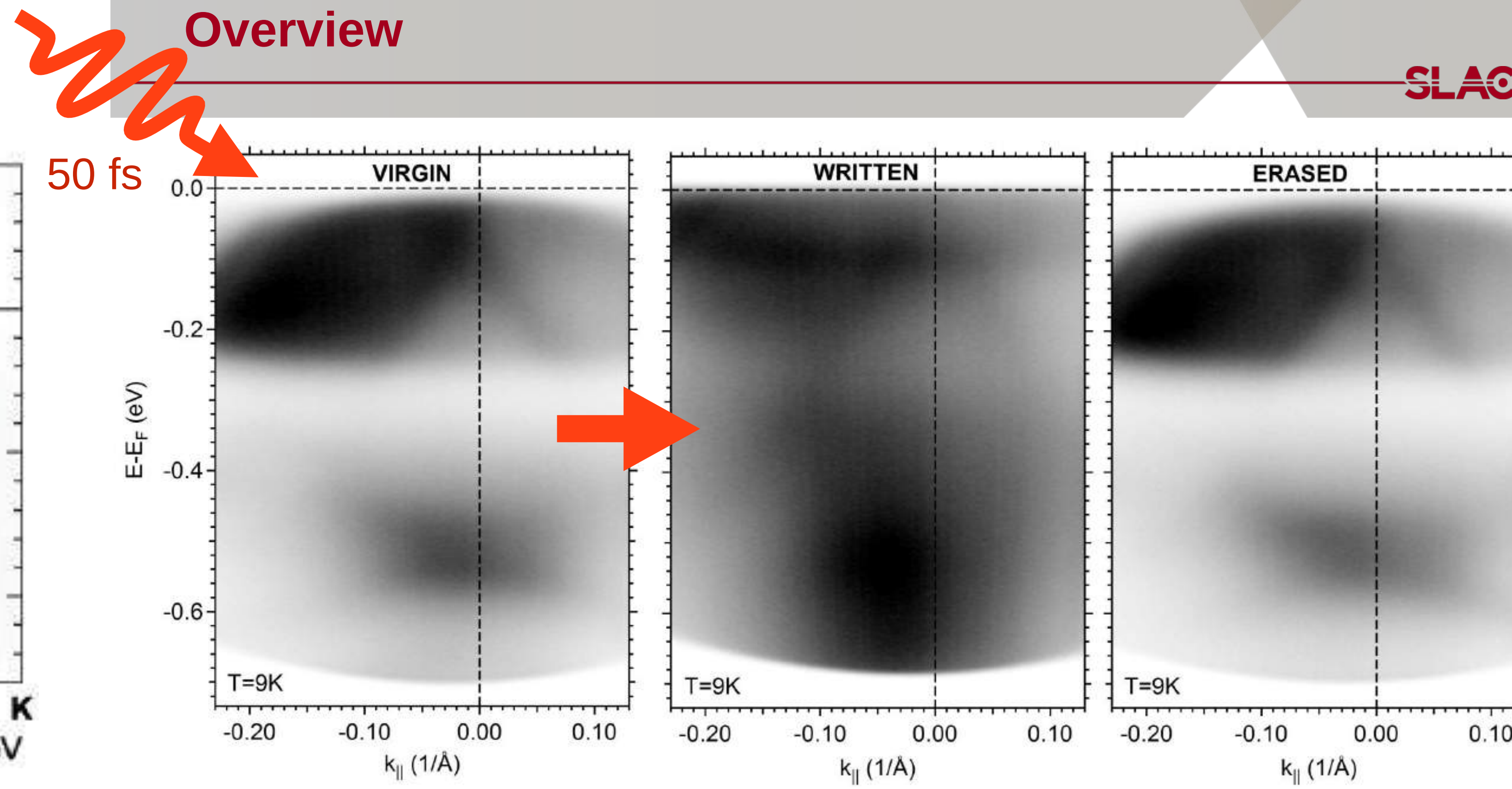
Patrick Kirchmann

"Normal" ARPES:



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

SLAC SIMES

U.S. DEPARTMENT OF ENERGY Office of Science

erc

Landau theory of the CDWs in $1T$-TaS_2

McMillan, PRB 1975

The Landau free energy: $$F = F_1 + F_2 + F_3 \ .$$

The order parameter (the charge density):

$$\rho(\vec{r}) = \rho_0(\vec{r})[1 + \alpha(\vec{r})]$$

Where $\alpha(\vec{r}) = \mathrm{Re}[\psi_1(\vec{r}) + \psi_2(\vec{r}) + \psi_3(\vec{r})]$

$$F_1 = \int d^2r[a(\vec{r})\alpha^2 - b(\vec{r})\alpha^3 + c(\vec{r})\alpha^4 + d(\vec{r})(|\psi_1\psi_2|^2 + |\psi_2\psi_3|^2 + |\psi_3\psi_1|^2)]$$

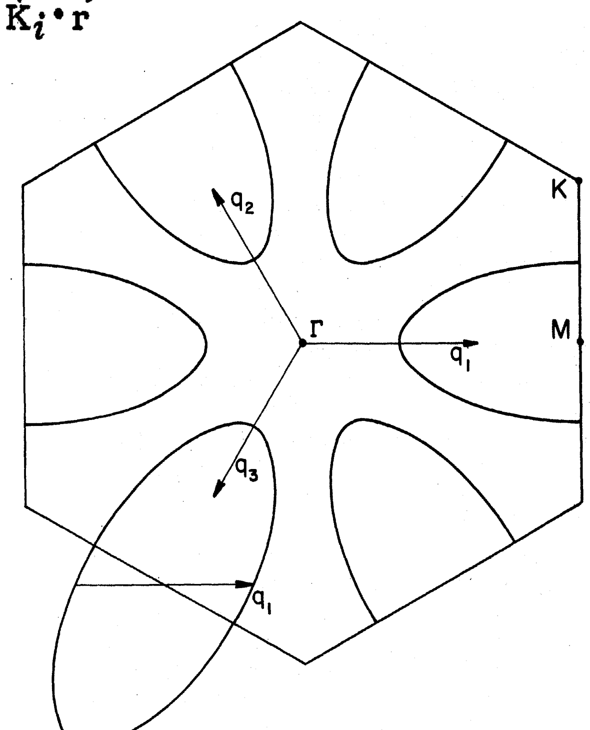
$$c(\vec{r}) = c_0 + c_1 \sum_i e^{i\vec{K}_i\cdot\vec{r}}$$

Impurity potential $$F_2 = \int d^2r\, U(\vec{r})\rho_0(\vec{r})\alpha(\vec{r})$$

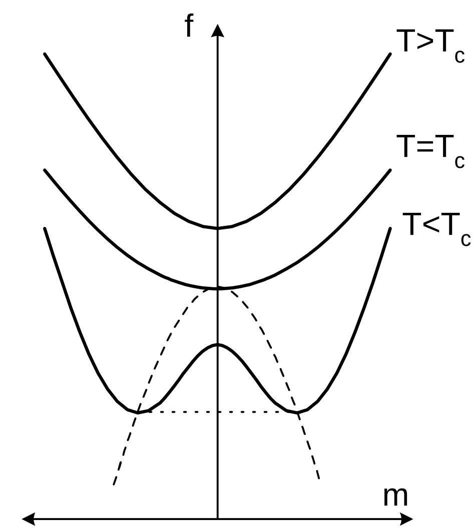
Gradient terms: $$F_3 = \int d^2r\left[e(\vec{r})\sum_i |(\vec{q}_i\cdot\vec{\nabla} - iq_i^2)\psi_i|^2 + f(\vec{r})\sum_i |\vec{q}_i\times\vec{\nabla}\psi_i|^2\right],$$

where

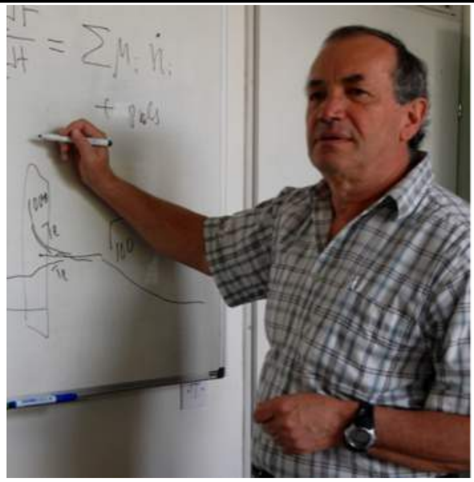
$$|\vec{q}_1| = |\vec{q}_2| = |\vec{q}_3| = 2\pi/\lambda \ ,$$



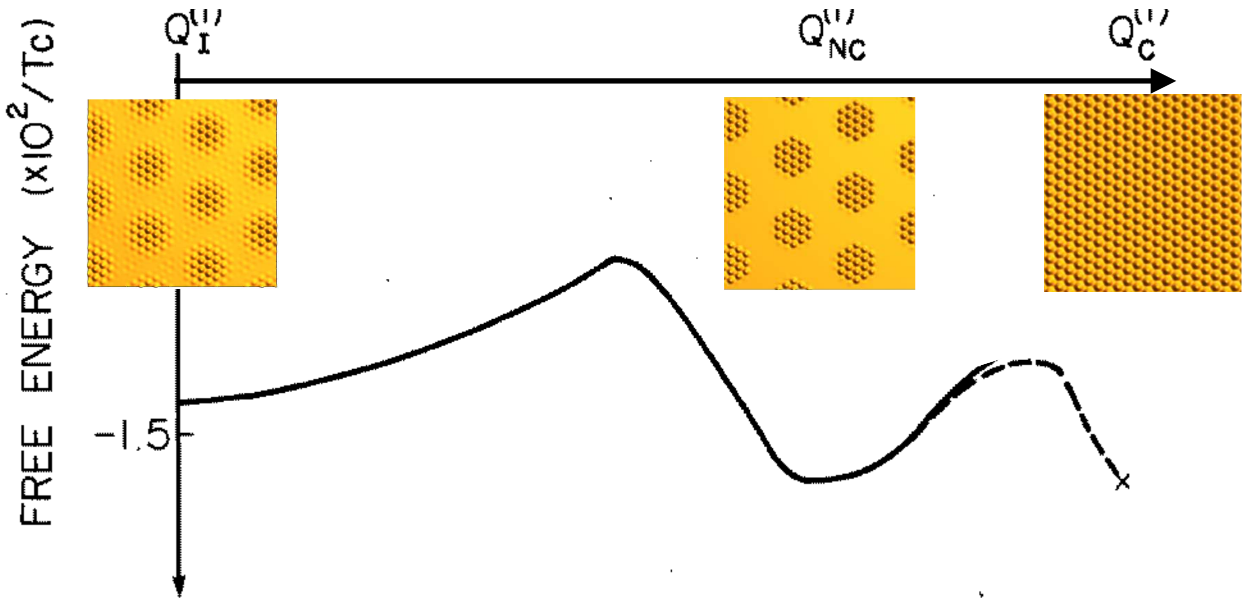
The Fermi Surface of 1T-TaS_2 in the undistorted phase



The free energy of $1T$-TaS_2 with domain walls (the NC state)



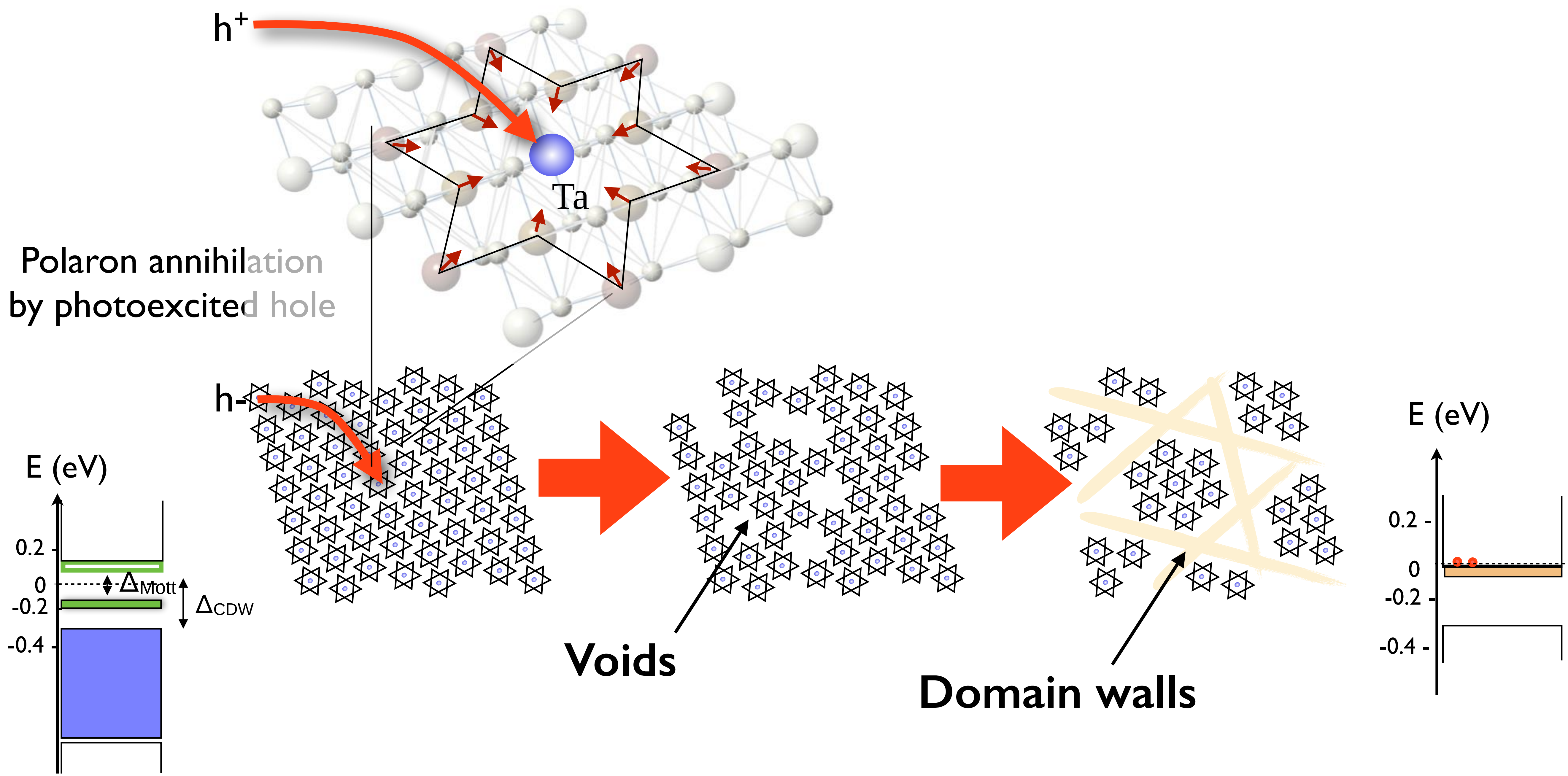
Free energy: $$F_c(n_c) = E_{DW}(\underbrace{C_0|n_c| + C_1|n_c|e^{-1/(\xi|n_c|)}}_{\text{C - IC transition (MacMillan, 1975)}} \underbrace{-C_2\xi n_c^2}_{\text{Intersection of DW}} + \underbrace{C_4\xi^3 n_c^4}_{\text{Repulsion between DW crossings}})$$



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

Microscopic mechanism: Photo"doping" and subsequent ordering of voids

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



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

Macroscopic quantum tunneling?

<u>Condition for MQT</u>:

The <u>potential</u> must change faster than the system can tunnel

Timescales:

The charge rearrangement in the H state occurs on a timescale:

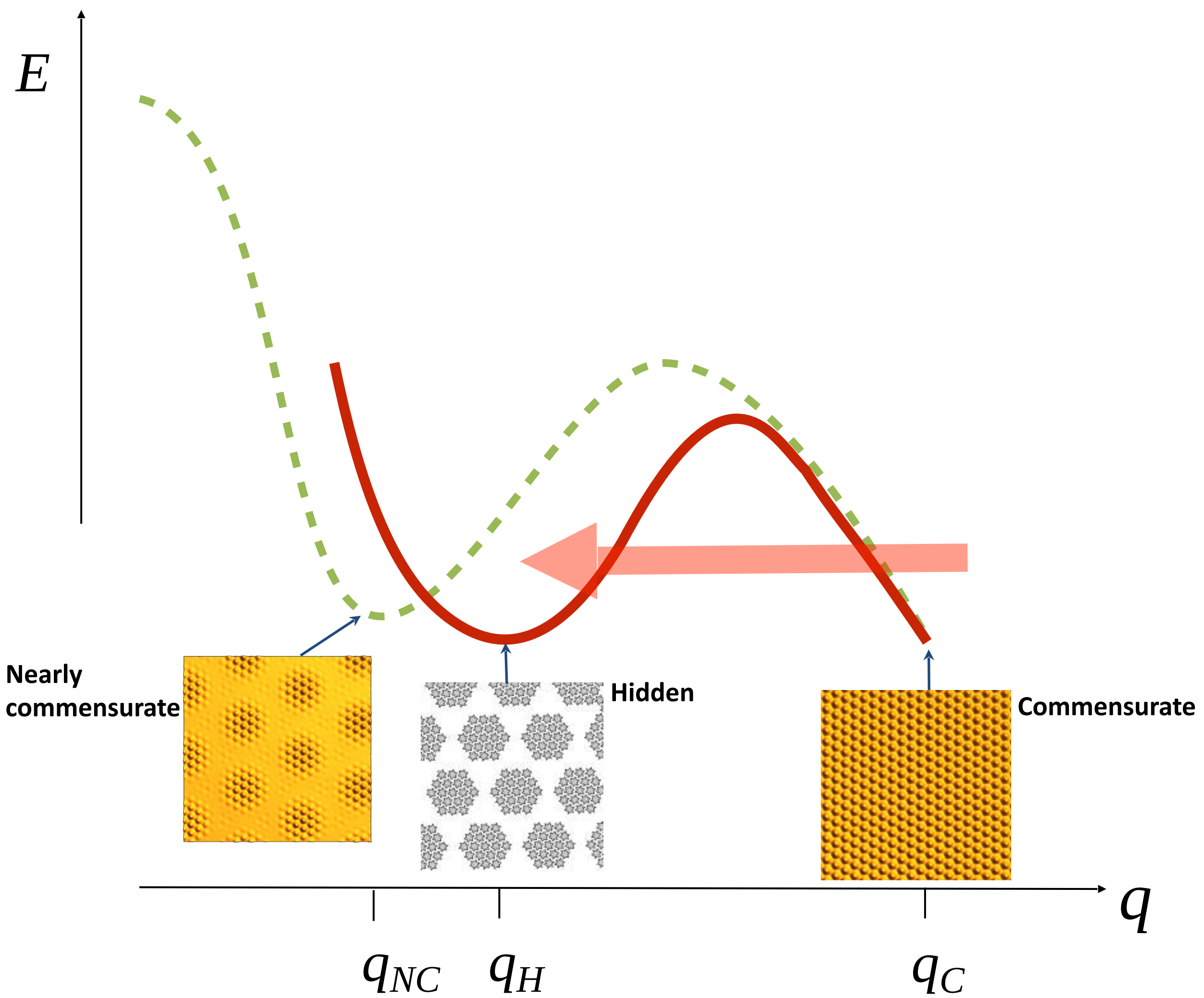
$$t_{charge} \sim \hbar/E_{Mott} \simeq 13\text{fs}$$

The polaron-forming ions reach new equilibrium positions on a timescale:

$$t_{ions} \leq \frac{1}{4}T_{AM} \simeq 110\text{fs}$$

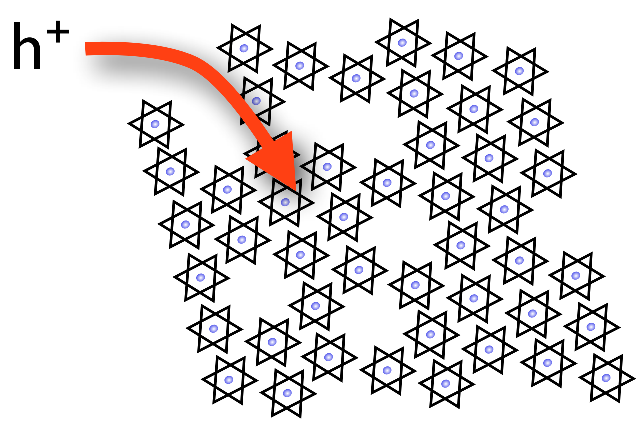
Tunneling time:

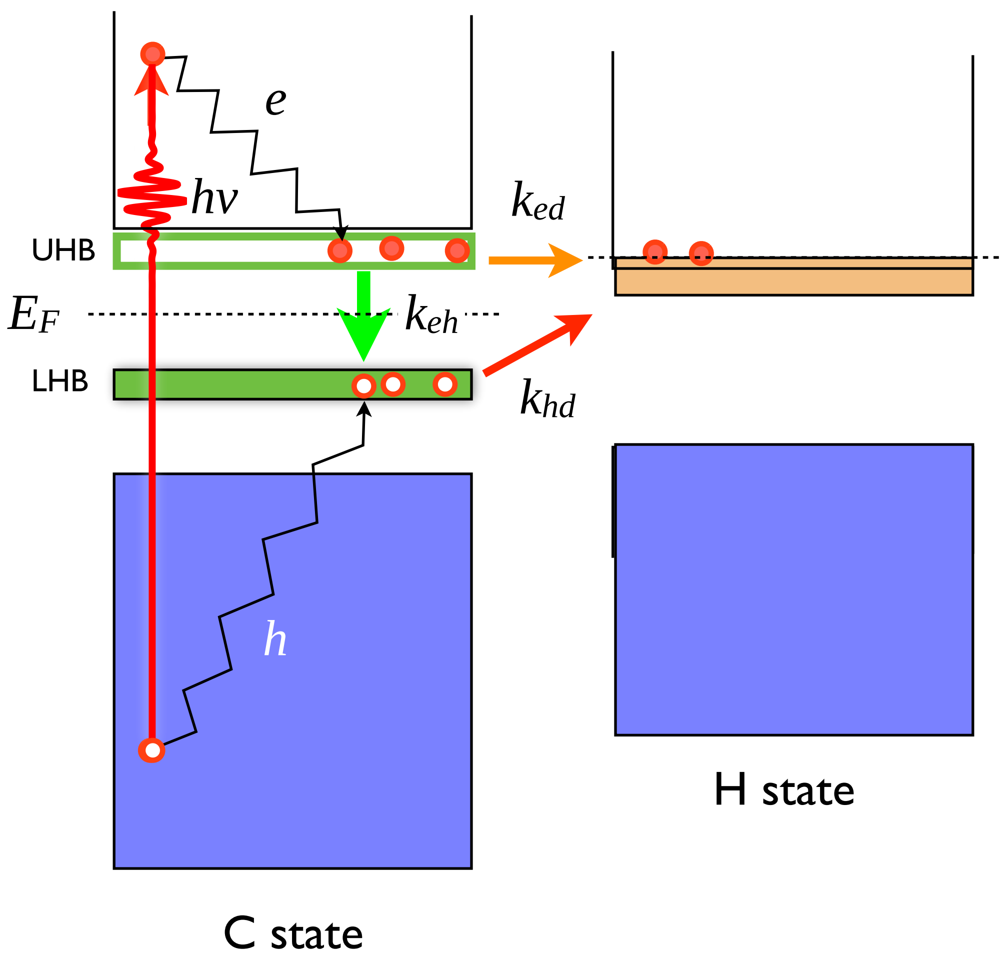
$$t_{tunneling} \sim O(1)T_{AM} \simeq 500\text{fs}$$



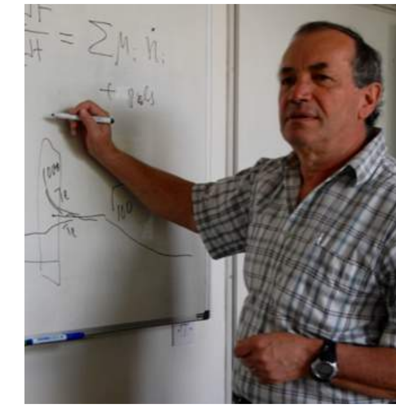
Conclusion: the MQT condition is fulfilled

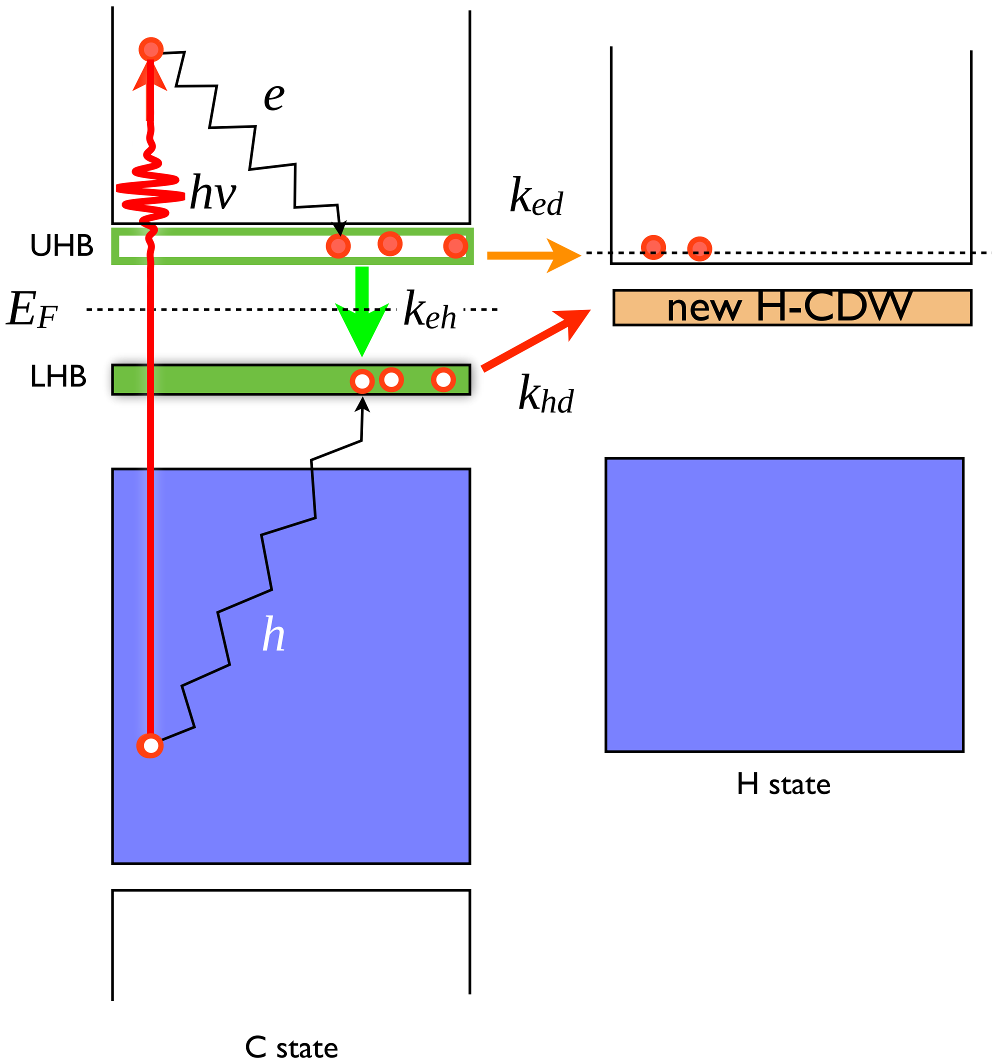
Kinetics cannot be described in a rigid band approximation





Electron and *hole* relaxation kinetics beyond the rigid band approximation: <u>photodoping in terms of a time-dependent shift of the chemical potential</u>





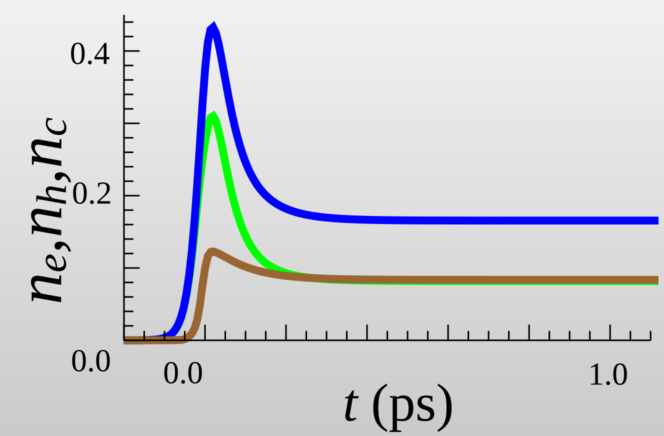
The kinetic equations for the electrons and holes:

$$\frac{dn_h}{dt} = -k_{eh} n_e n_h (\mu_e + \mu_h) - k_{hd} n_h (\mu_h - \mu_d) \ + P(t)$$

$$\frac{dn_e}{dt} = -k_{eh} n_e n_h (\mu_e + \mu_h) - k_{ed} n_e (\mu_e + \mu_d) \ + P(t)$$

μ_i are time-dependent, and *P(t)* is the laser pulse

Numerical solution:



Subject to conservation of charge

$$n_e - n_h = n_v - n_i = n_d.$$

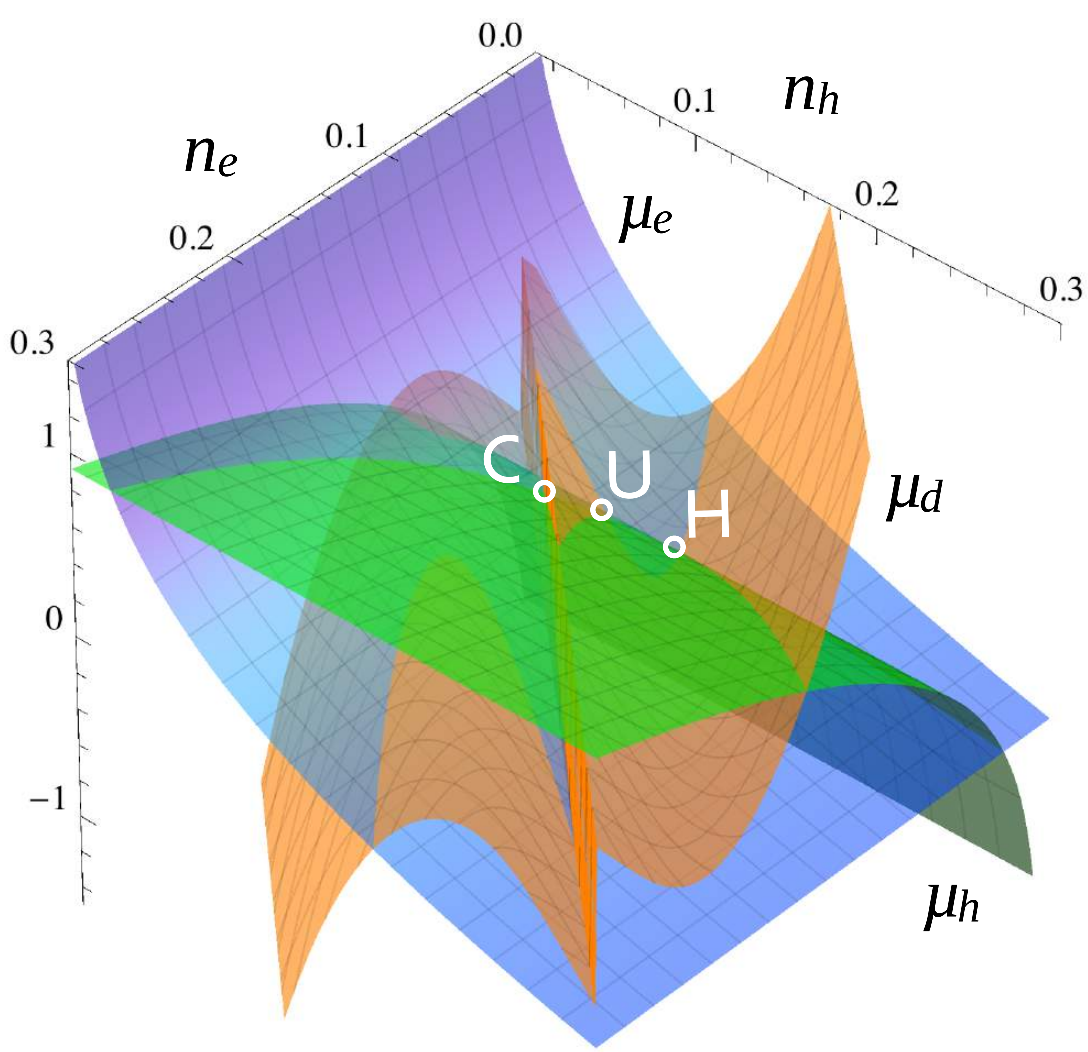
Chemical potential surfaces

The chemical potentials: $\mu_i = \dfrac{\partial F_i(n_i)}{\partial n_i}$

for electrons, holes: $\mu_{e,h}(n) = \Delta_{e,h} + k_B T \ln(e^{n_{e,h}/(k_B T N_{e,h})} - 1)$

for the condensate:

$$\mu_c(n_c) = E_{DW}(C_1(1 + \frac{1}{\xi|n_c|})e^{-1/(\xi|n_c|)} + C_0 - 2C_2\xi|n_c| + 4C_4(\xi|n_c|)^3)sign(n_c)$$



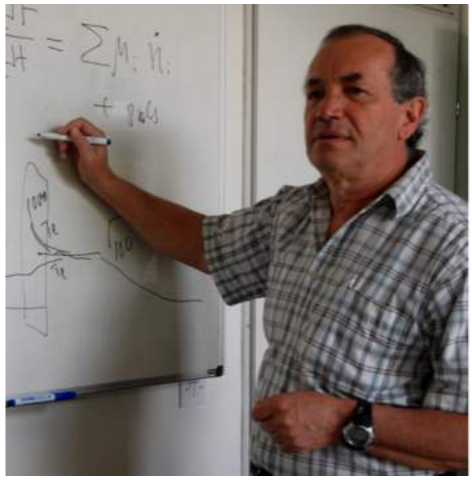
The system is stable in equilibrium when:

$$\mu_e = \mu_h = \mu_c$$

where $n_c = n_h - n_e$

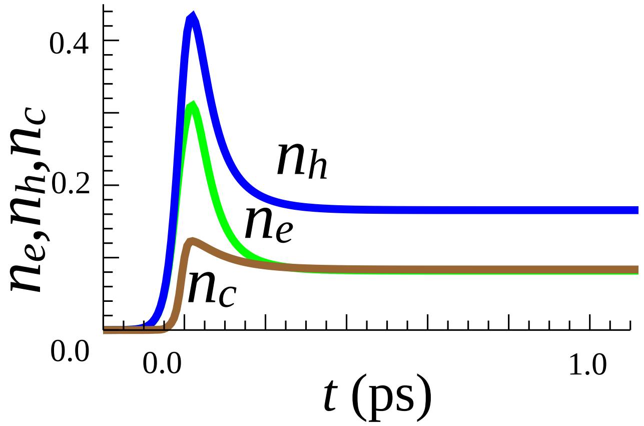
L Stojchevska et al. Science 2014;344:177-180

Calculated trajectory

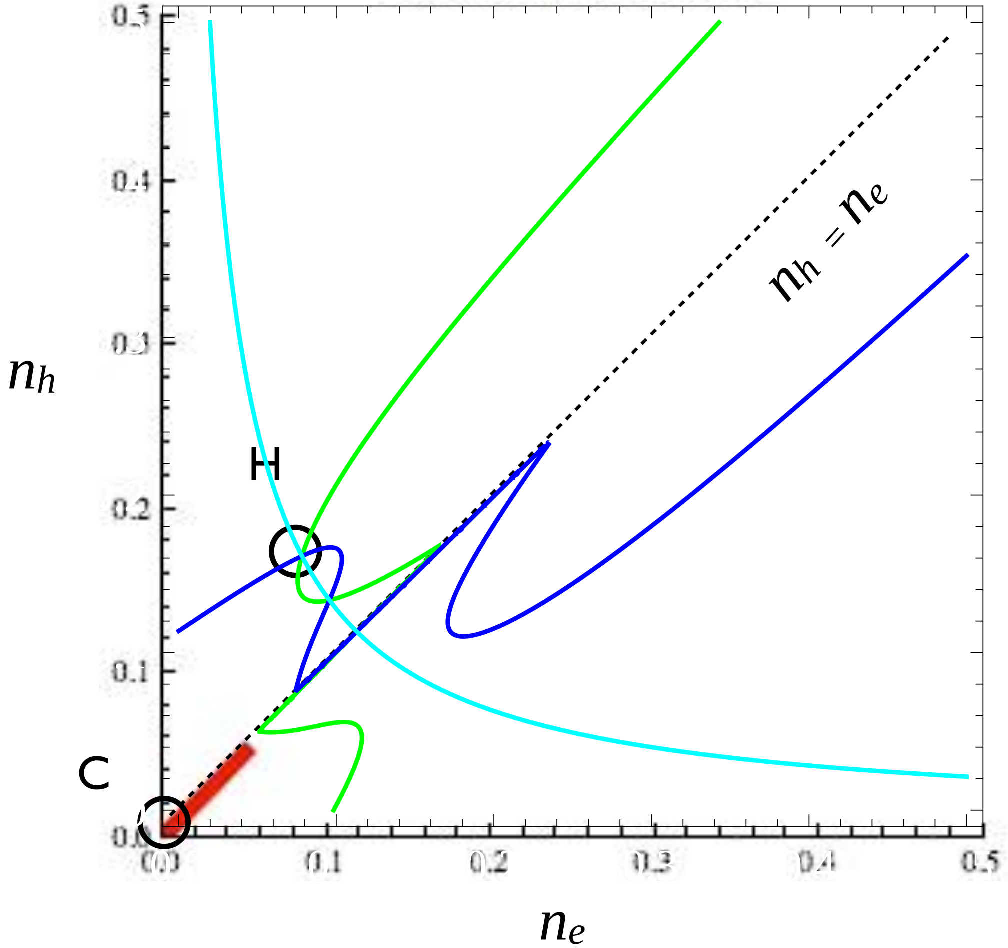


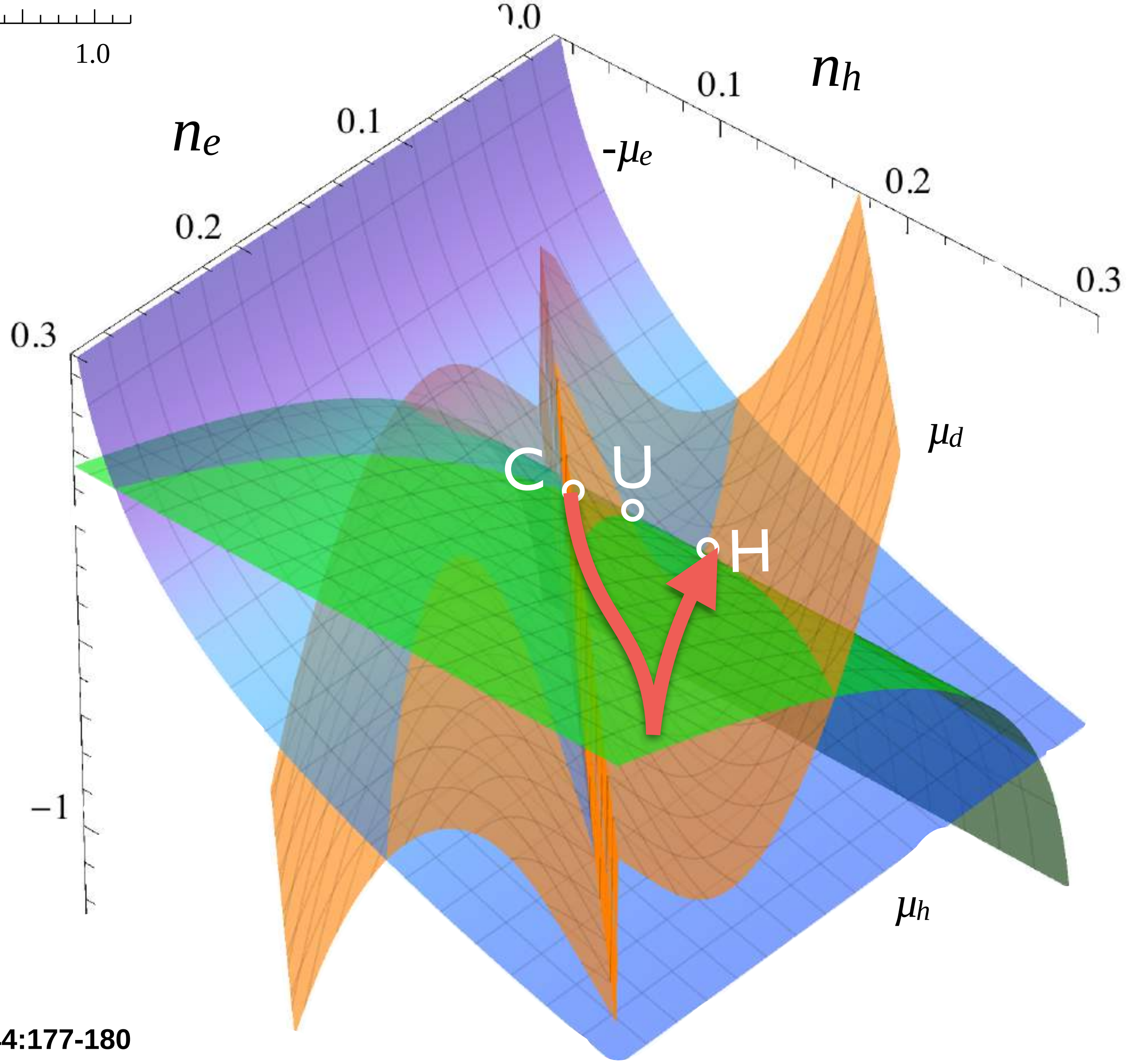
Laser pulse energy **above** threshold ($U_W > U_T$):

The time evolution of n_e and n_h:



System trajectory (parametric plot)

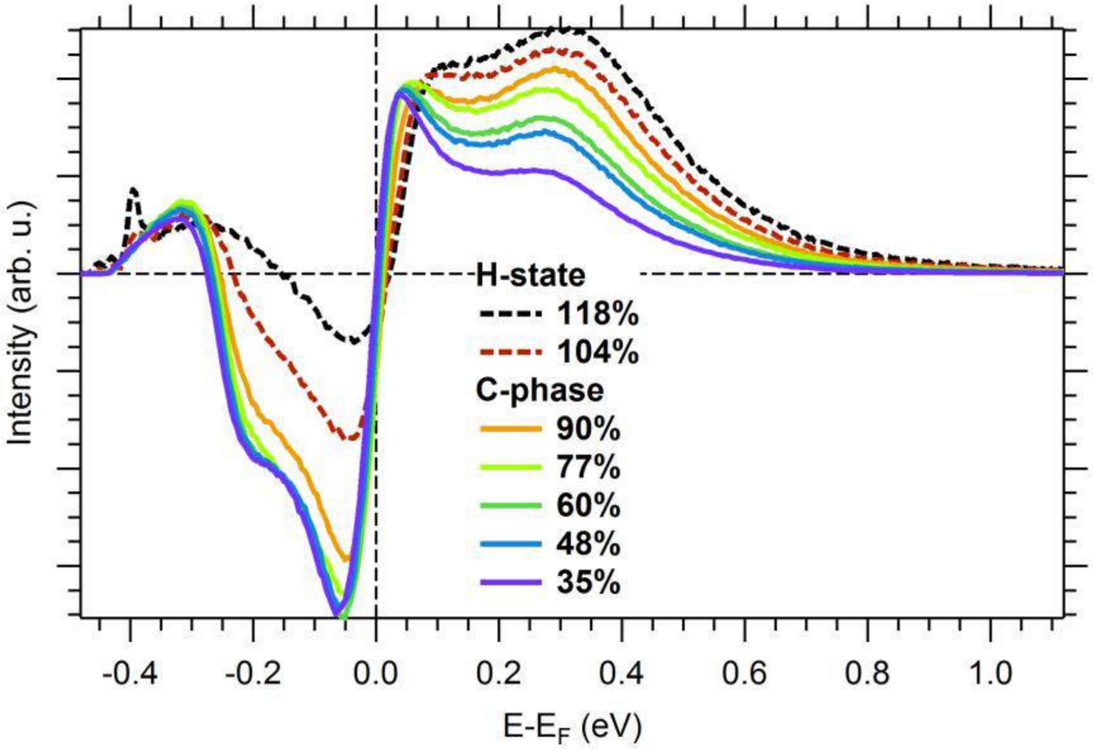


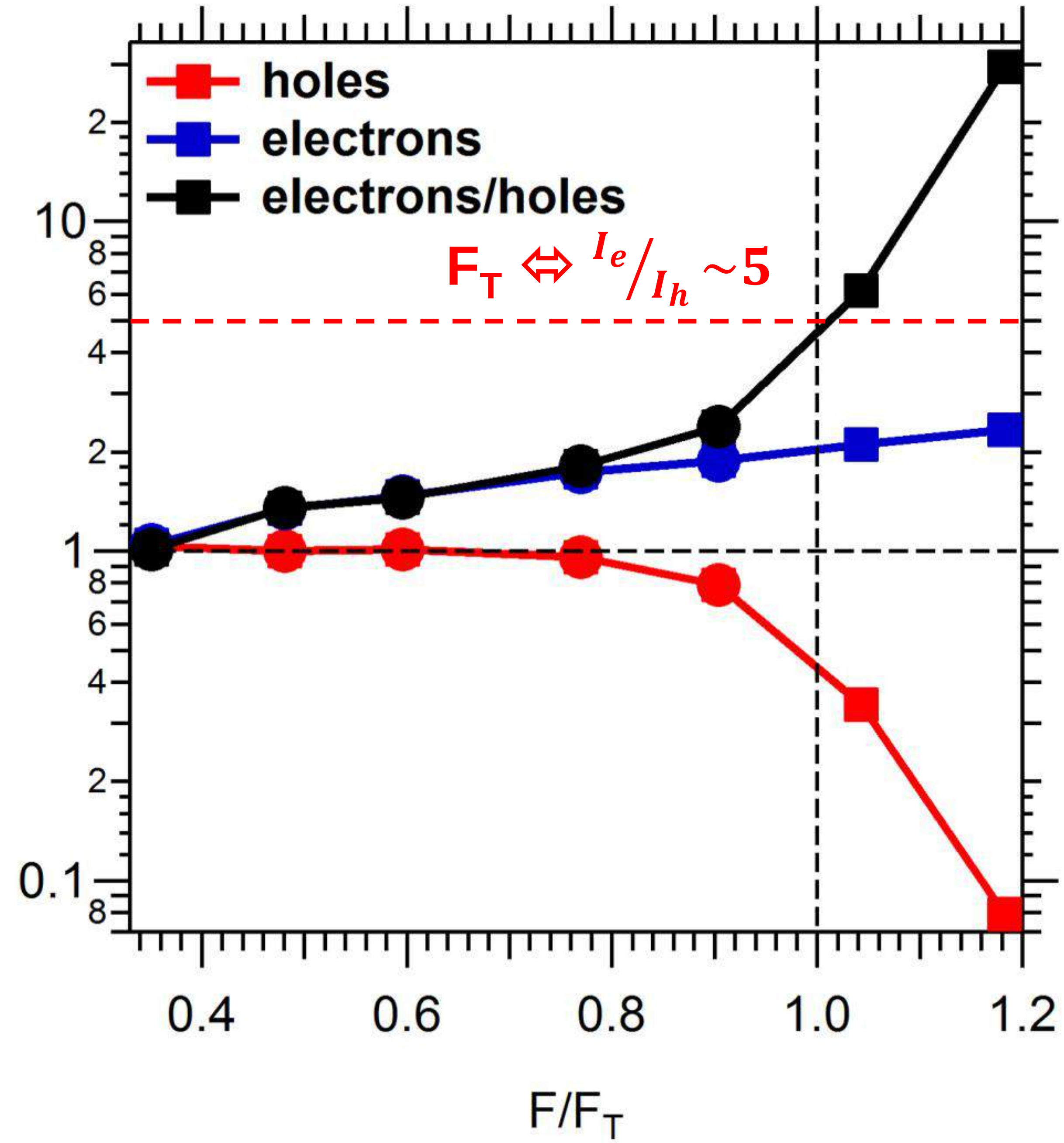


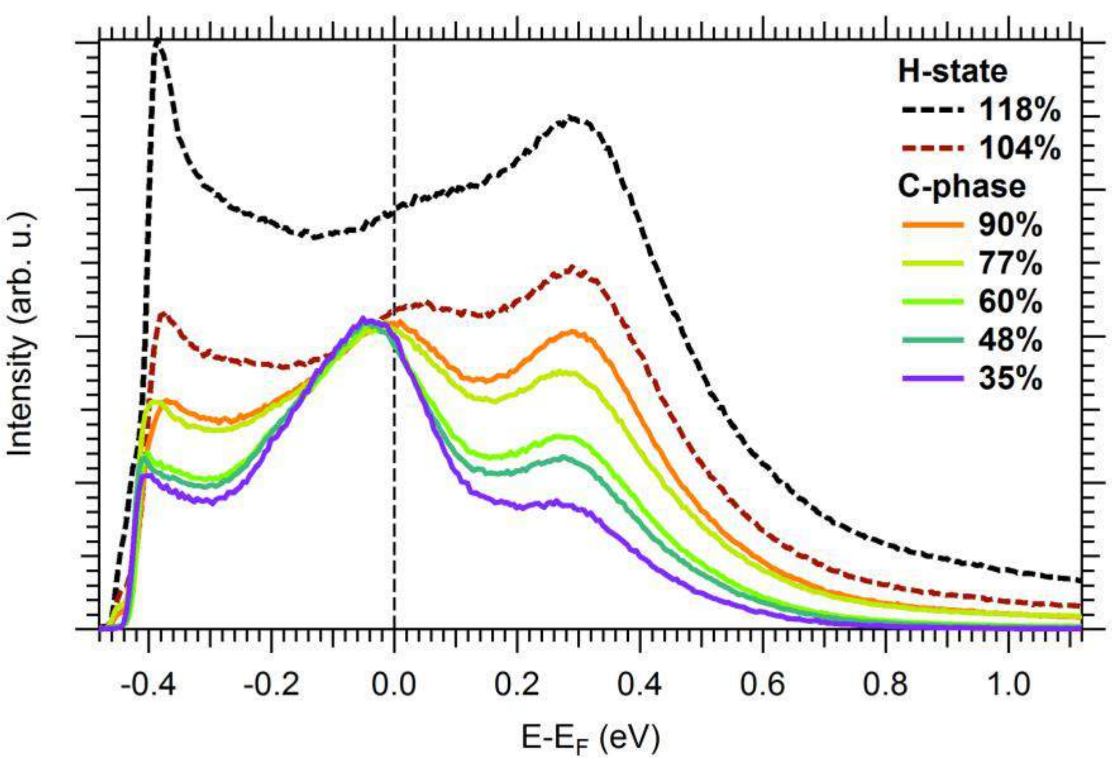
L Stojchevska et al. Science 2014;344:177-180

e-h Imbalance
C-Phase Near Threshold / H-State above Threshold

SLAC







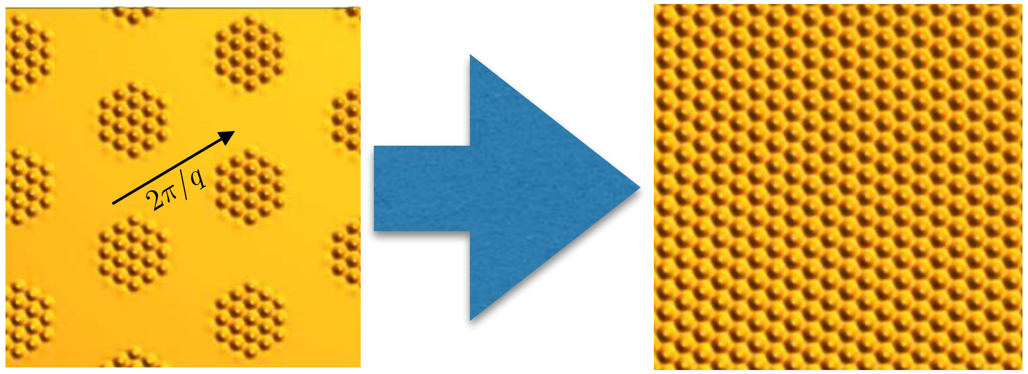
trARPES suggest that e-h imbalance drives switching

k-number conservation: A topological protection mechanism

The <u>modulation</u> wave vector

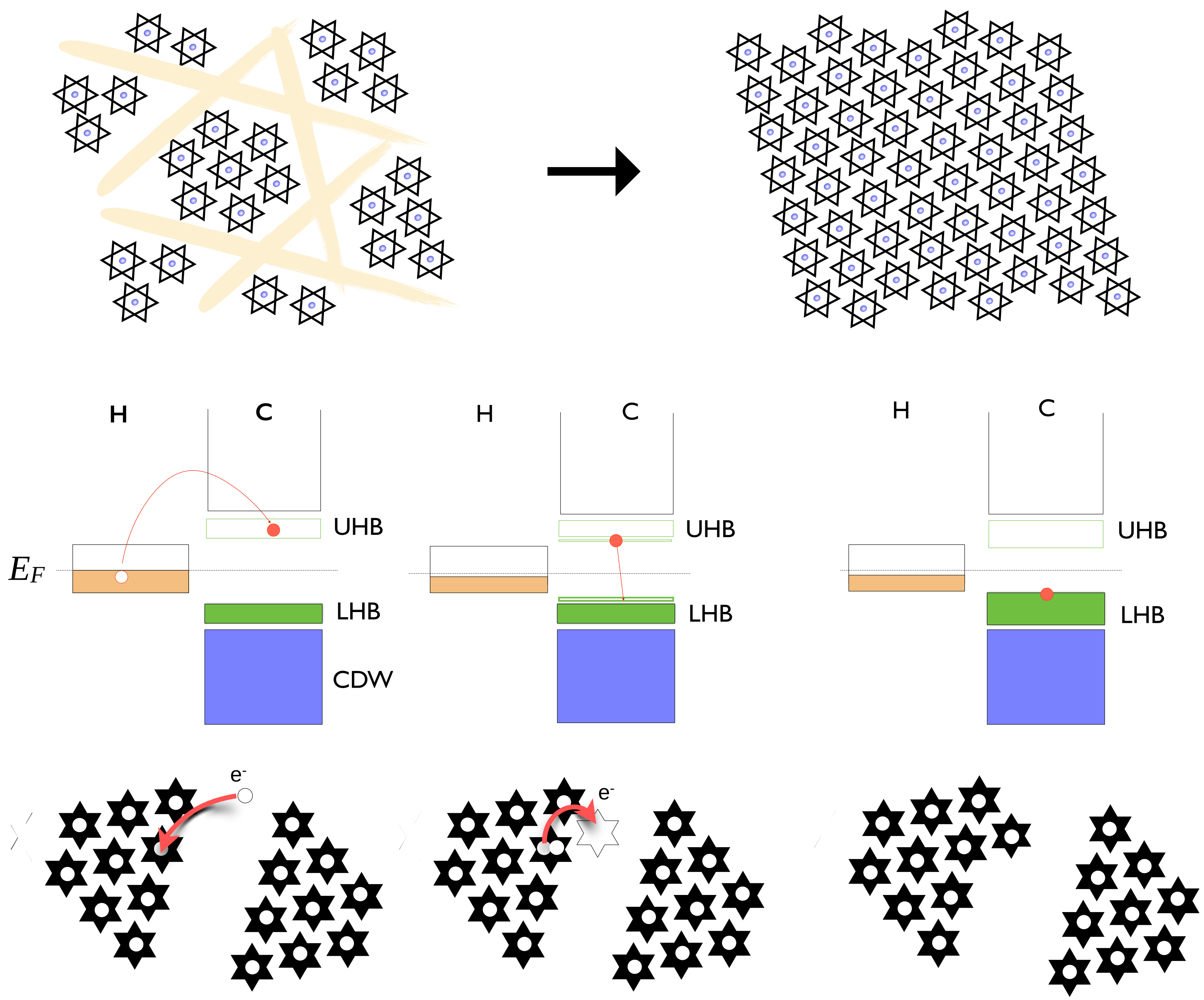
$$\delta q = n/\pi$$

n is the number of void sites



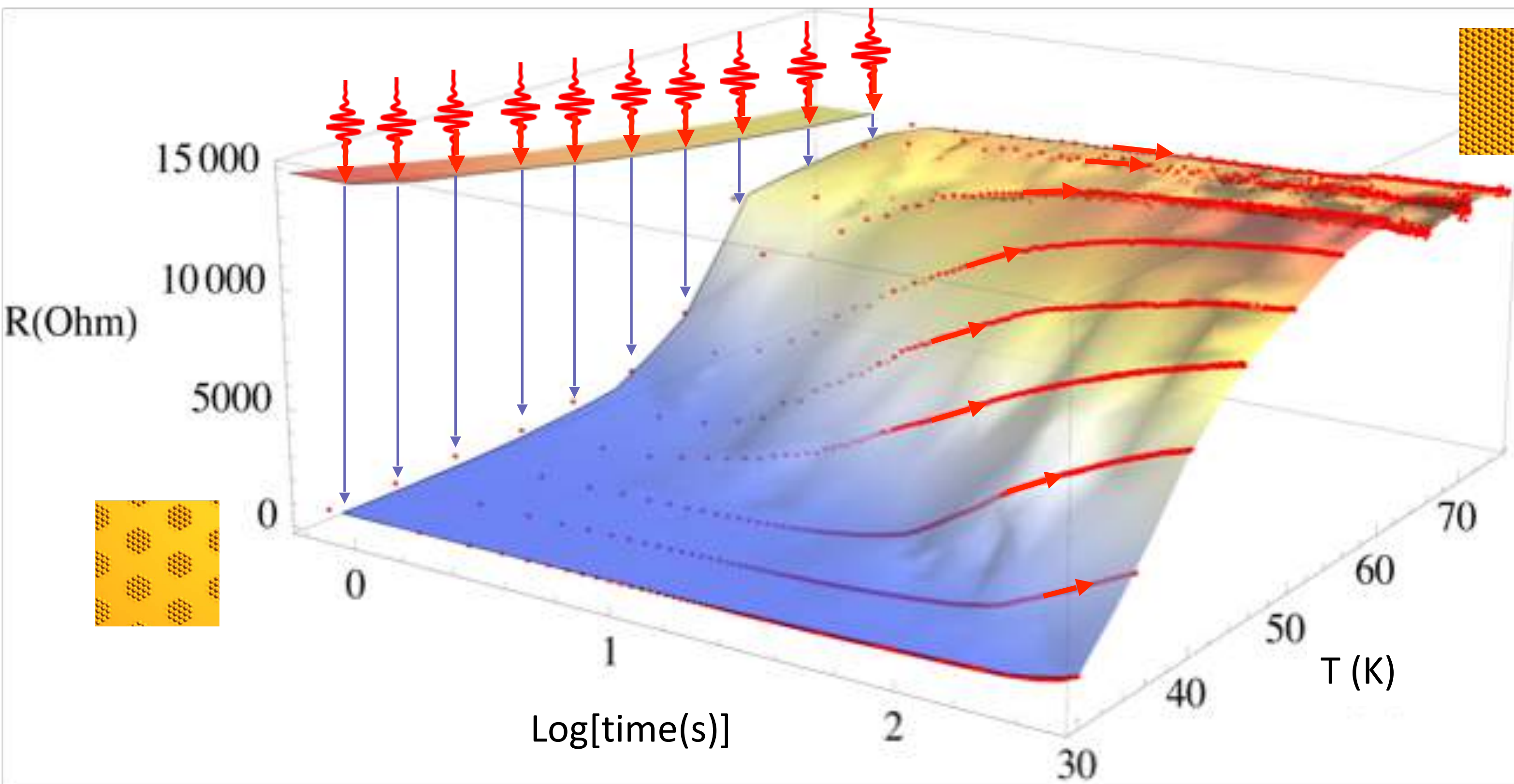
The H and C states are topologically distinct

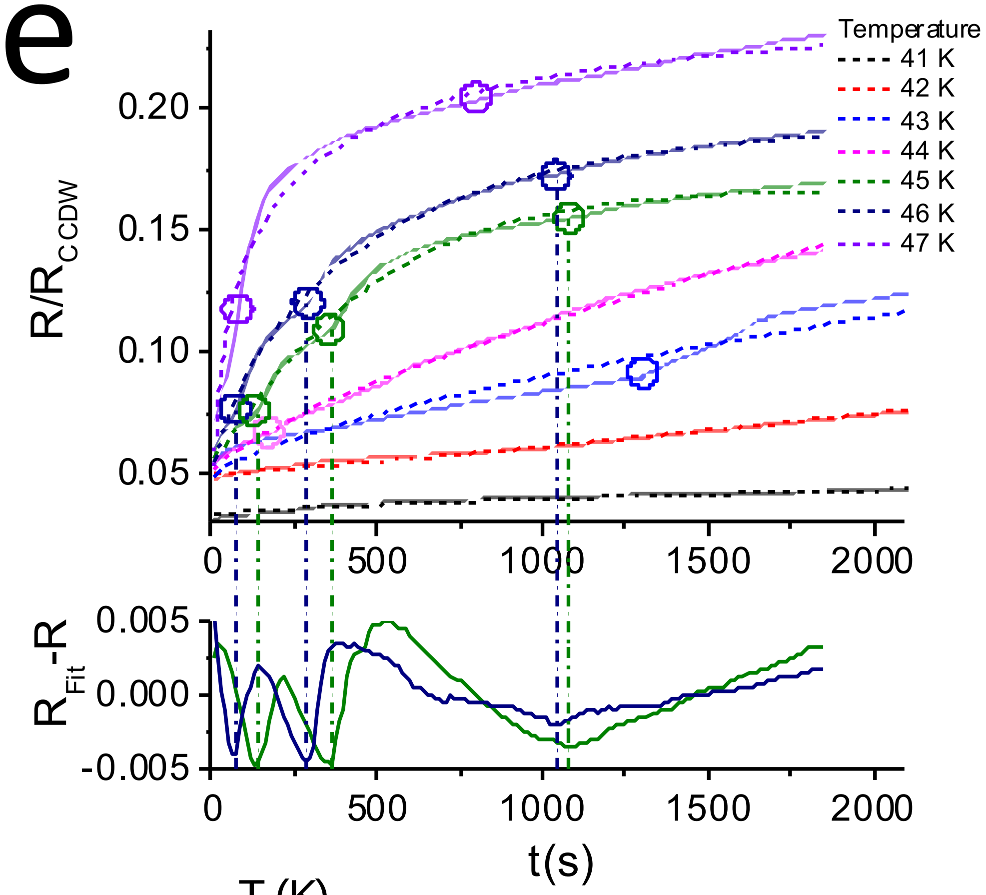
The relaxation mechanism (microscopic)



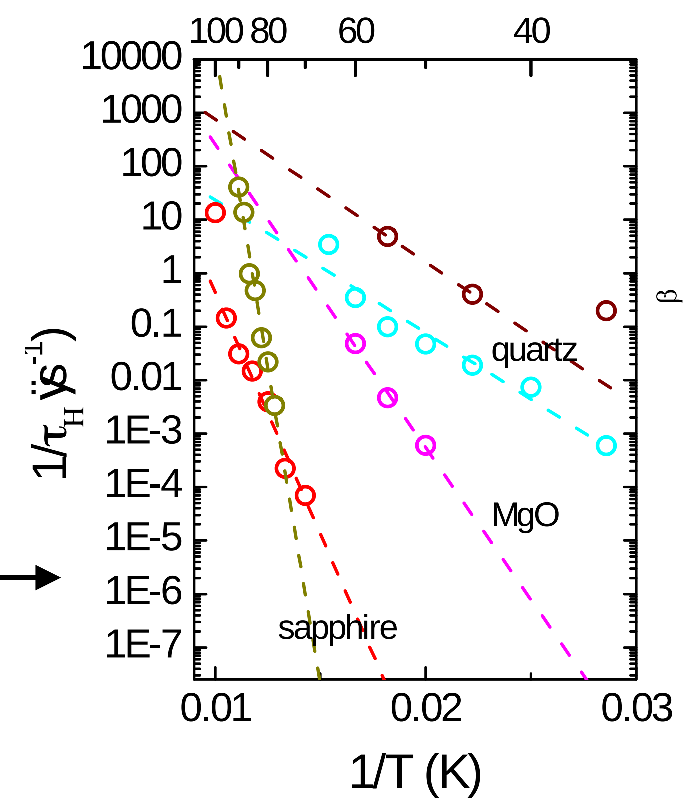
I.Vaskivskyi et al., Science Advances (2015)

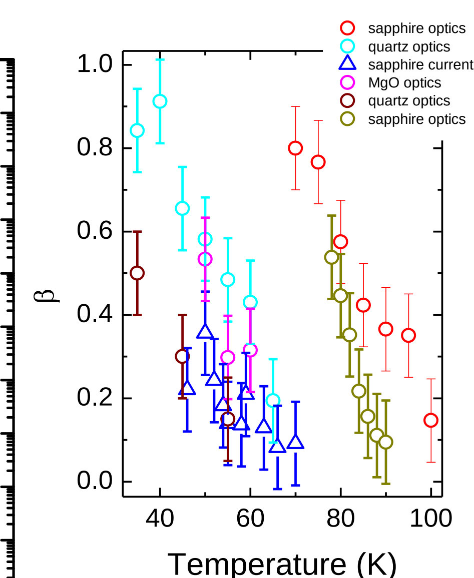
Relaxation of the H state





$$R = R_0[1 - exp(-(t/\tau_H)^{\beta})]$$



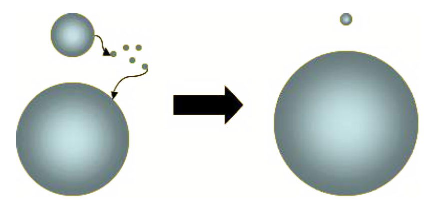


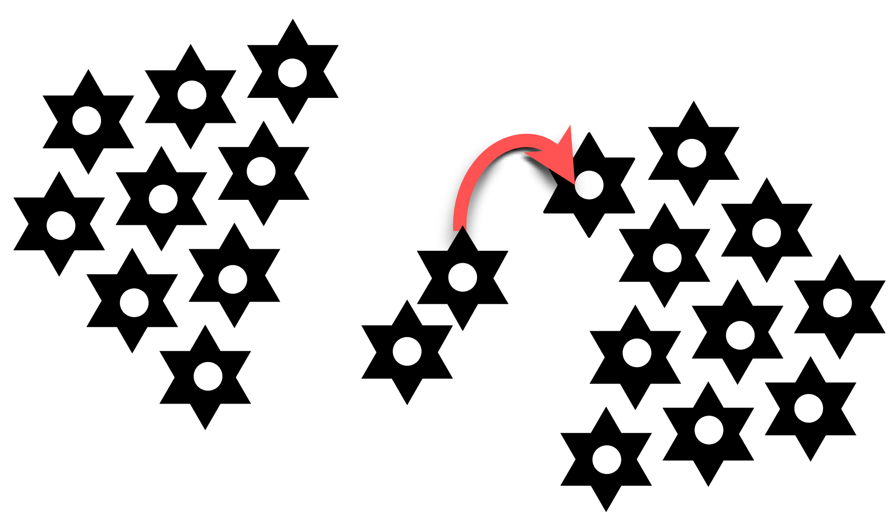
At low T (<10K) the lifetime is longer than the age of the Universe →

I.Vaskivskyi et al., Science Advances (2015)

Relaxation H→C (2).

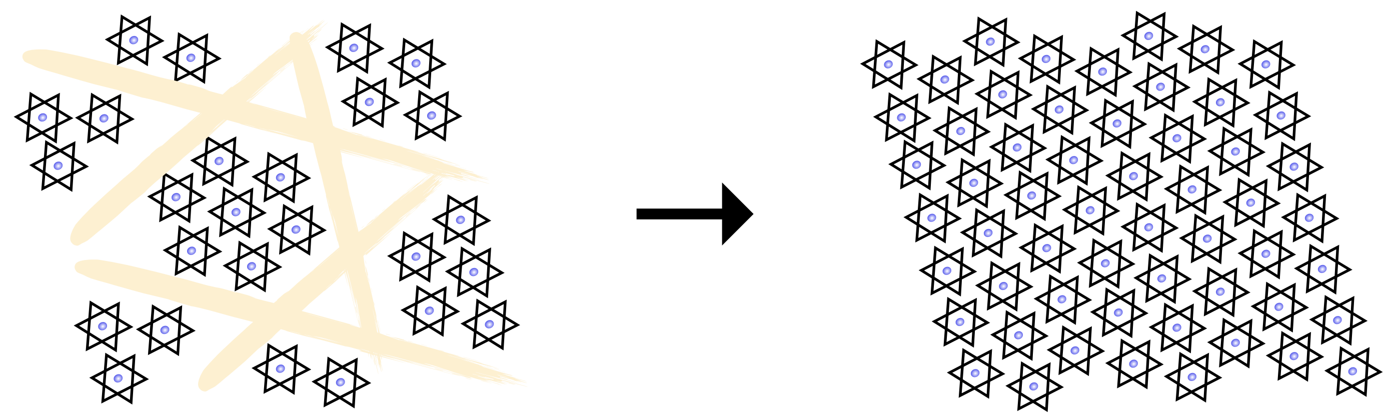
Ostwald ripening

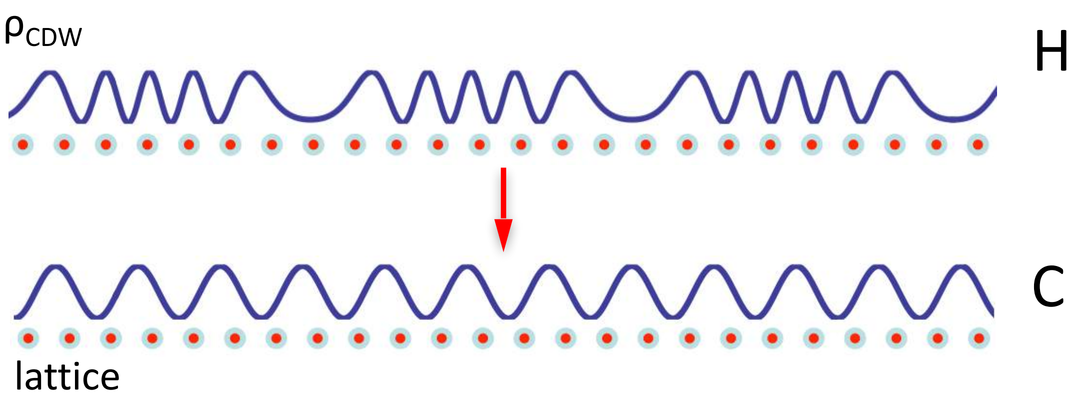




W. Ostwald. **1896**. *Lehrbuch der Allgemeinen Chemie*, vol. 2, part 1. Leipzig, Germany.

Incommensurate → commensurate
Relaxation (3)





Kontorova and Frenkel model (1938):

$$H = \int \left[\frac{1}{2} \left(\frac{d\varphi}{dn} - \delta \right)^2 + V(1 - \cos p\varphi) \right] dn$$

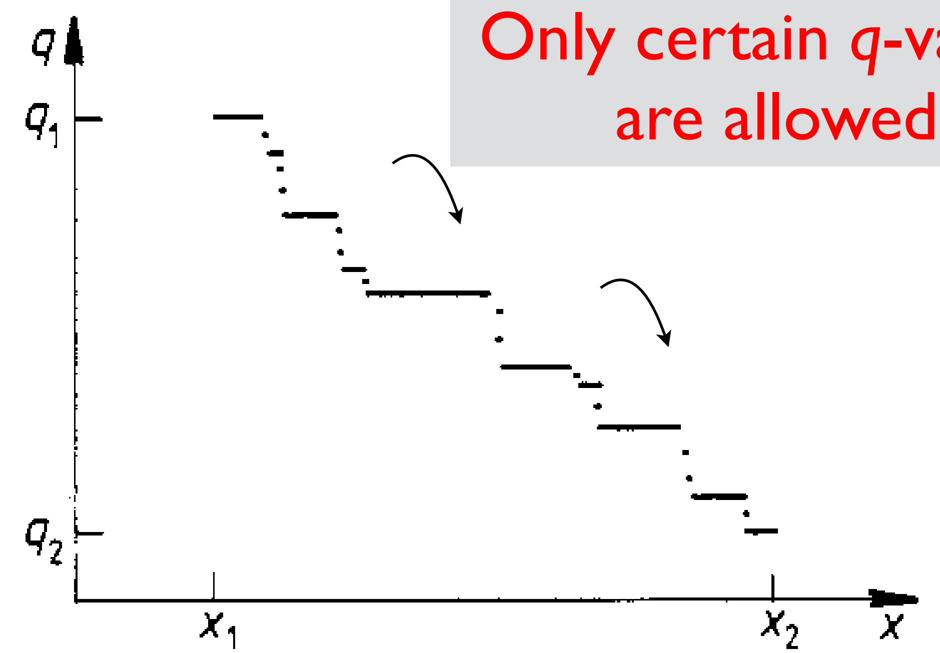
x_n is the position of the nth atom:

$$x_n = nb + \frac{b}{2\pi} \varphi_n$$

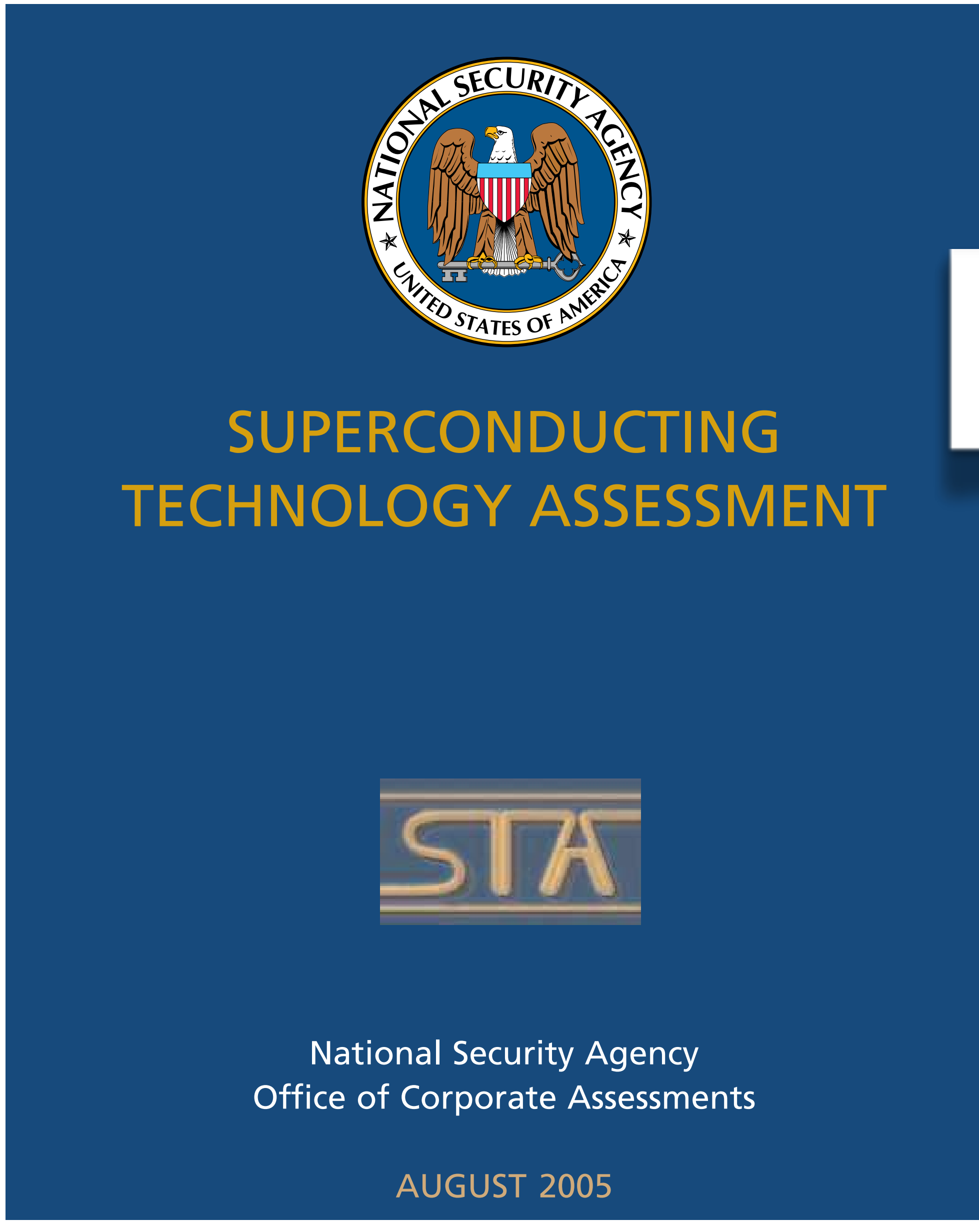
In the continuum limit: $\varphi_n - \varphi_{n-1} = d\varphi/dn$

A devil's staircase

Only certain q-values are allowed



time



Published online, 242 pages

Recent industry trends clearly establish that design tradeoffs between power, clock and metrology have brought CMOS to the limits of its scalability….

…Alternatives to CMOS must therefore be found.

…Cryogenic RAM has been **the most neglected superconductor technology** and therefore needs the most development…

Memory challenges:

High speed
Low energy dissipation
Low temperature operation

(2015)


IEEE TRANSACTIONS ON APPLIED SUPERCONDUCTIVITY, VOL. 23, NO. 3, JUNE 2013 1701610

Energy-Efficient Superconducting Computing—Power Budgets and Requirements

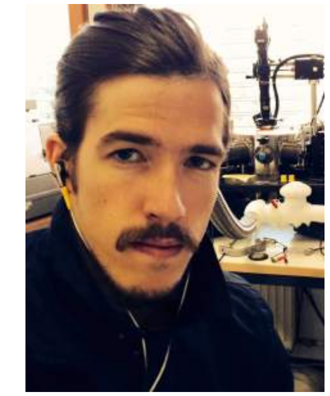
D. Scott Holmes, *Senior Member, IEEE*, Andrew L. Ripple, and Marc A. Manheimer

Is it possible to rapidly switch to a H state by *charge injection*?

(It could be useful)

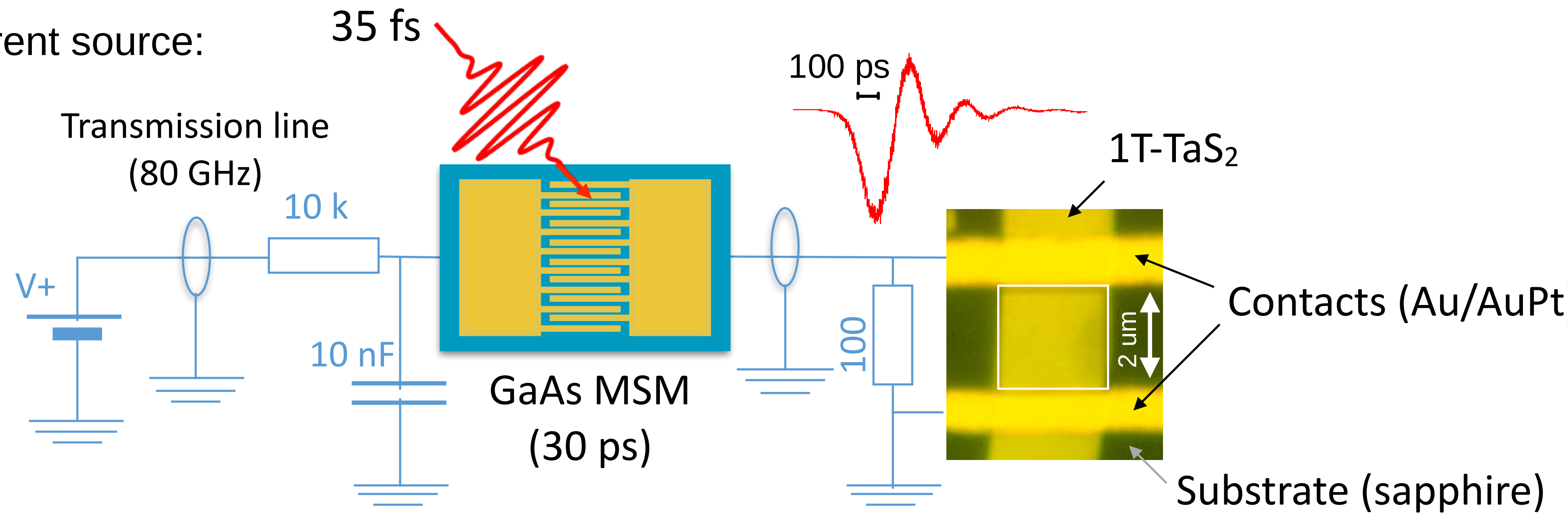
Vaskivskyi, I. *et al.* Fast non-thermal switching between macroscopic charge-ordered quantum states induced by charge injection. *arXiv* (2014).

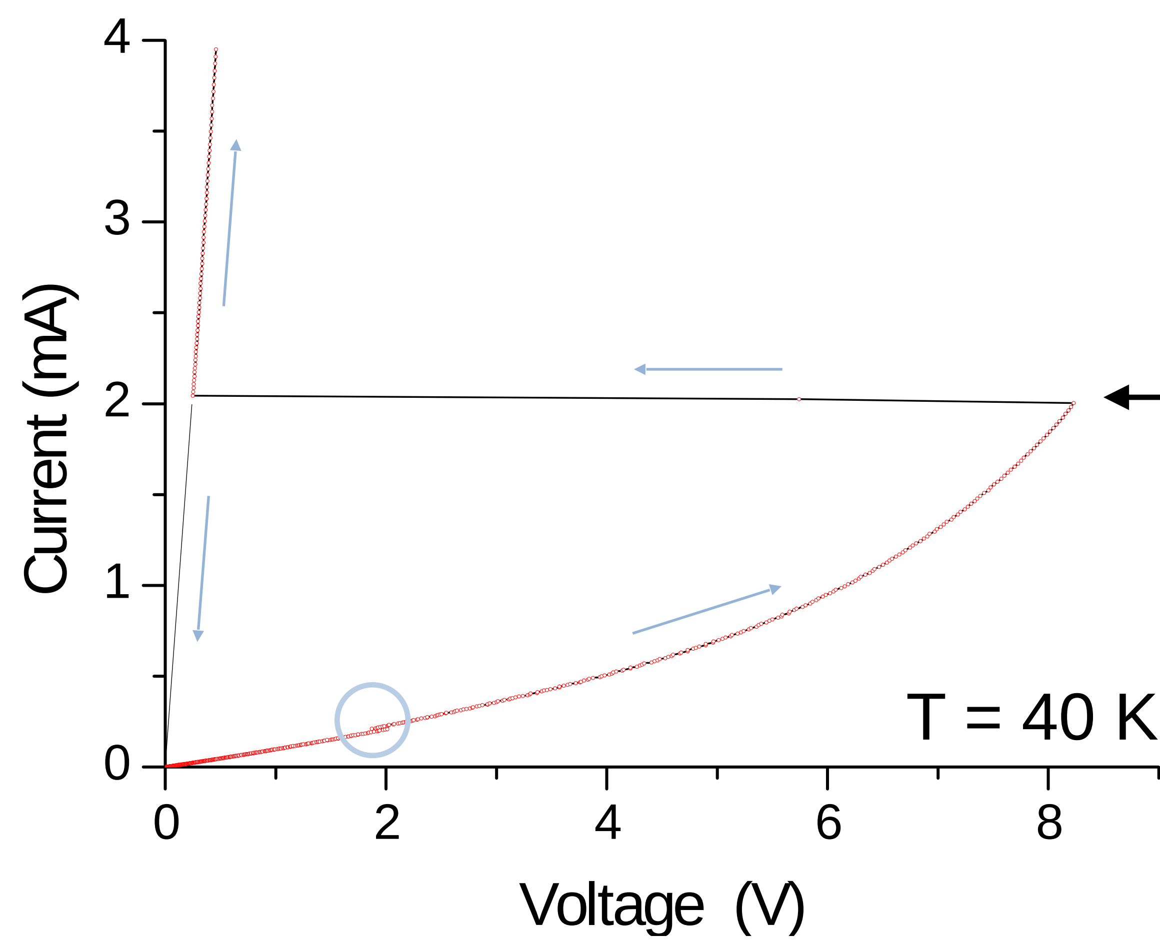
Switching to the H state using an ultrafast current source

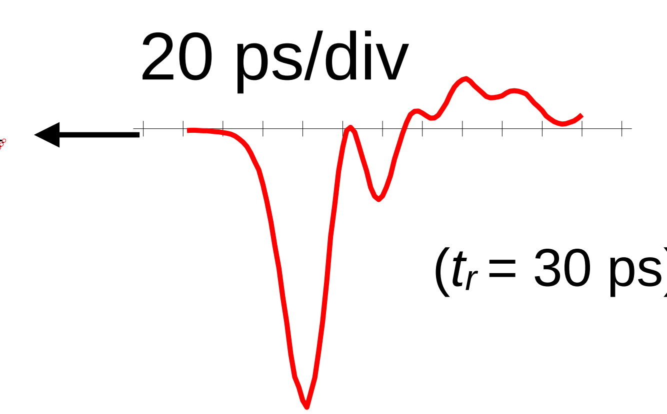


Ian Mihailovic

Pulsed current source:

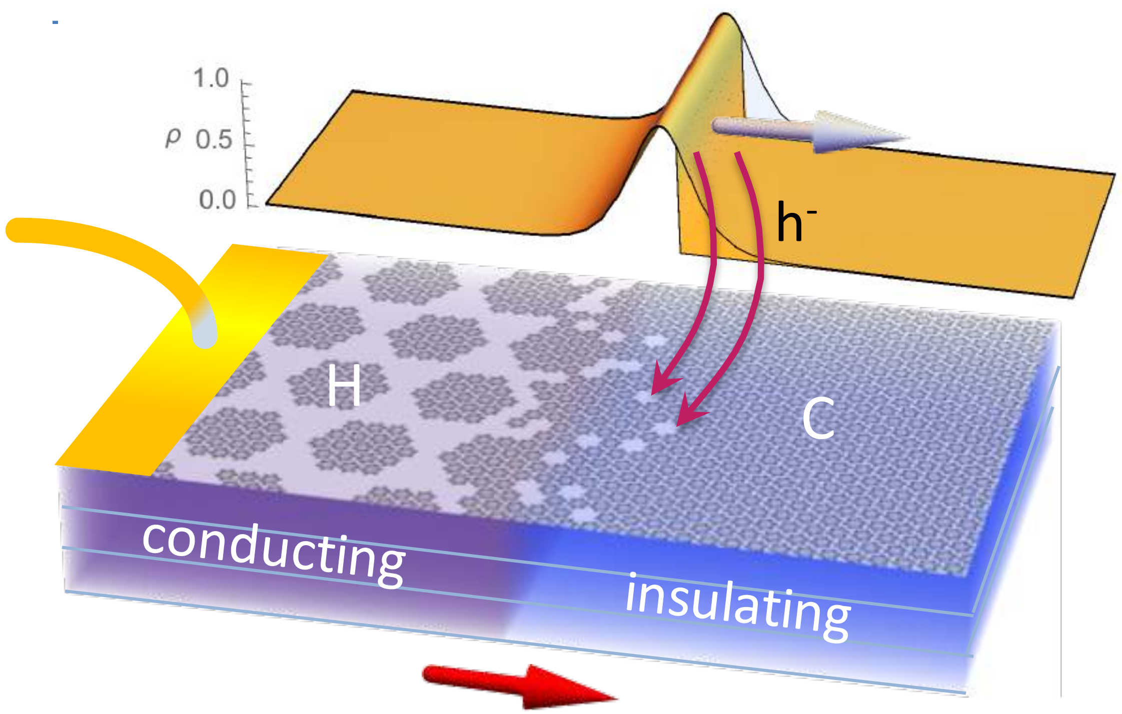






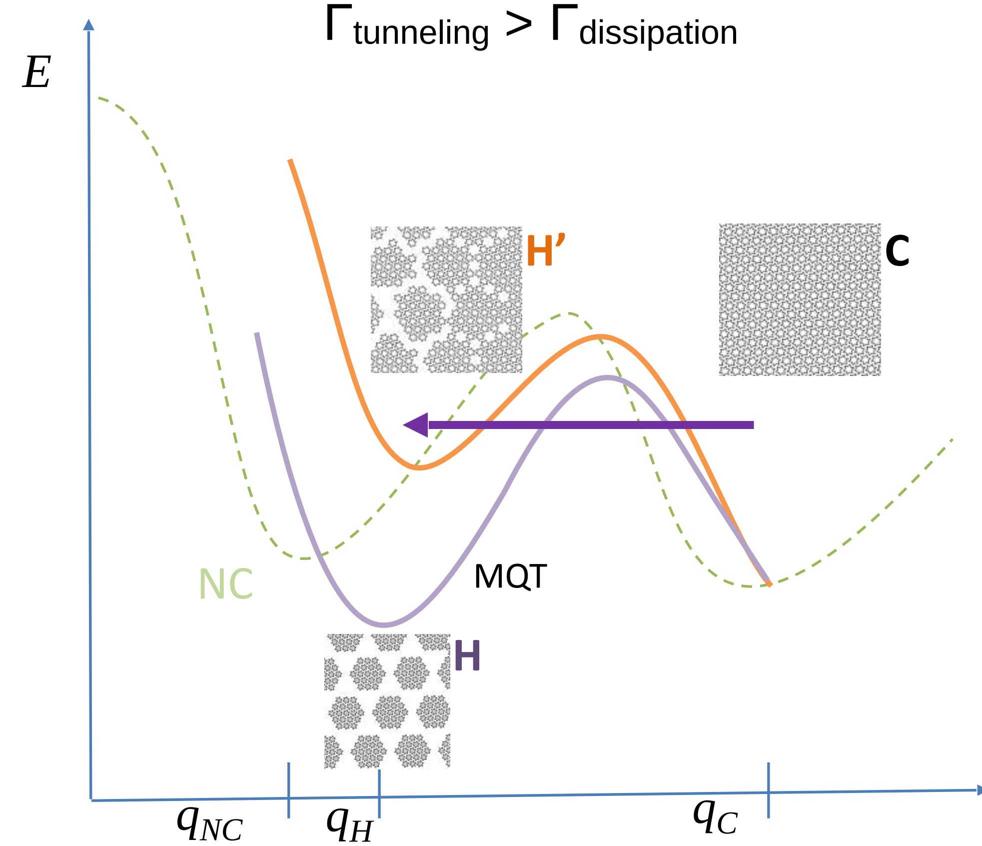
Vaskivskyi, I. *et al.* Fast non-thermal switching between macroscopic charge-ordered quantum states induced by charge injection. *arXiv* **cond-mat** (2014).

Macroscopic Quantum Tunneling with current injection?



Leggett's MQT criterion (1980):

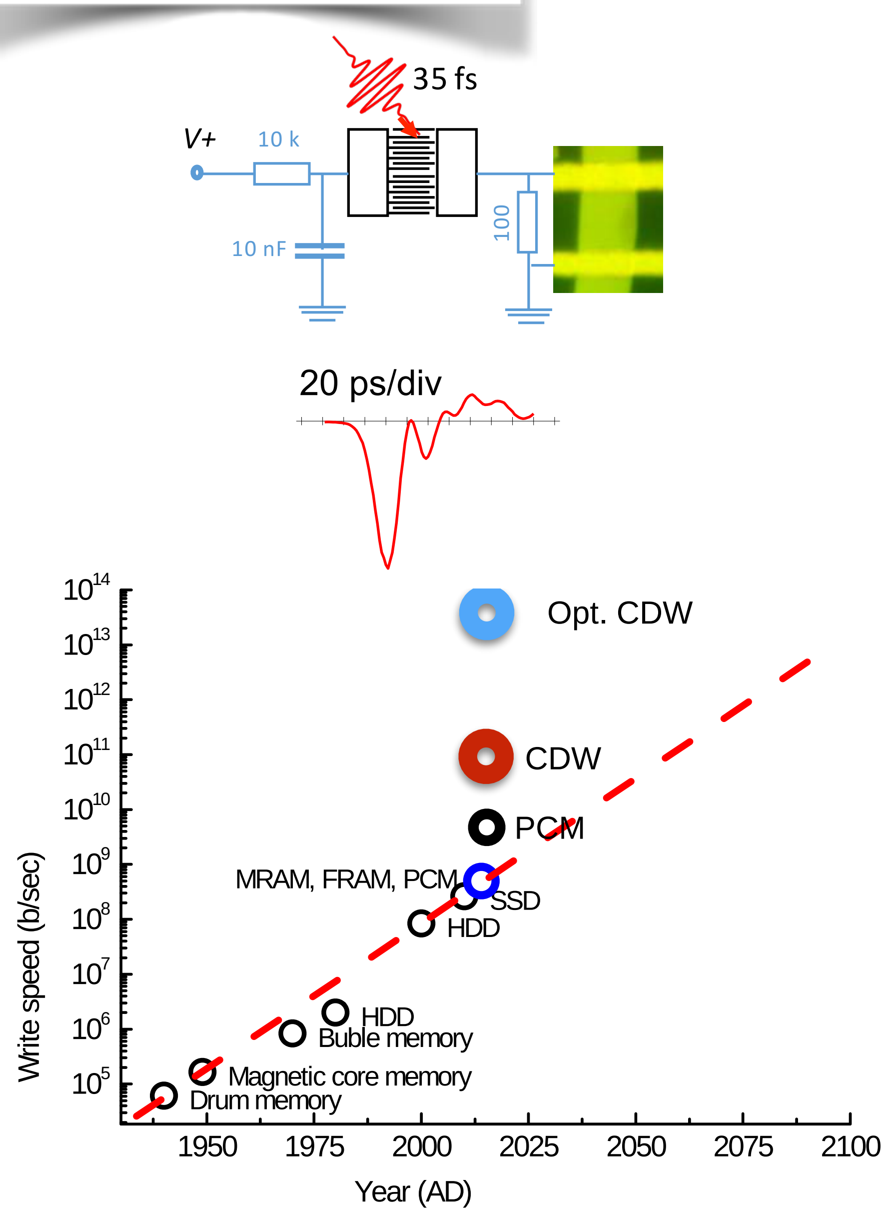
$\Gamma_{\text{tunneling}} > \Gamma_{\text{dissipation}}$



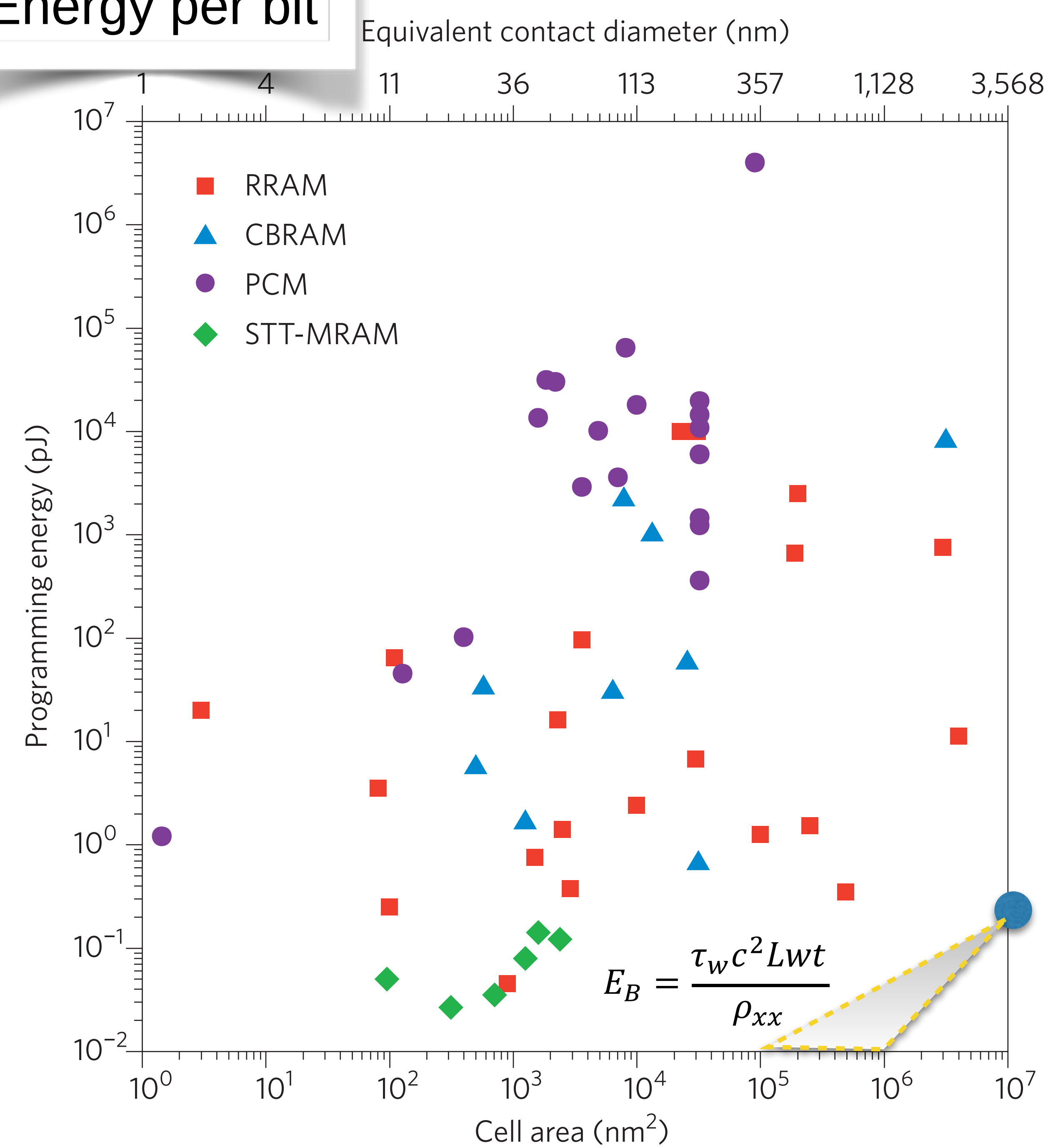
The state is spatially and temporally inhomogeneous

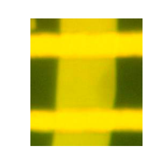
CDW memory performance

Switching speed



Energy per bit



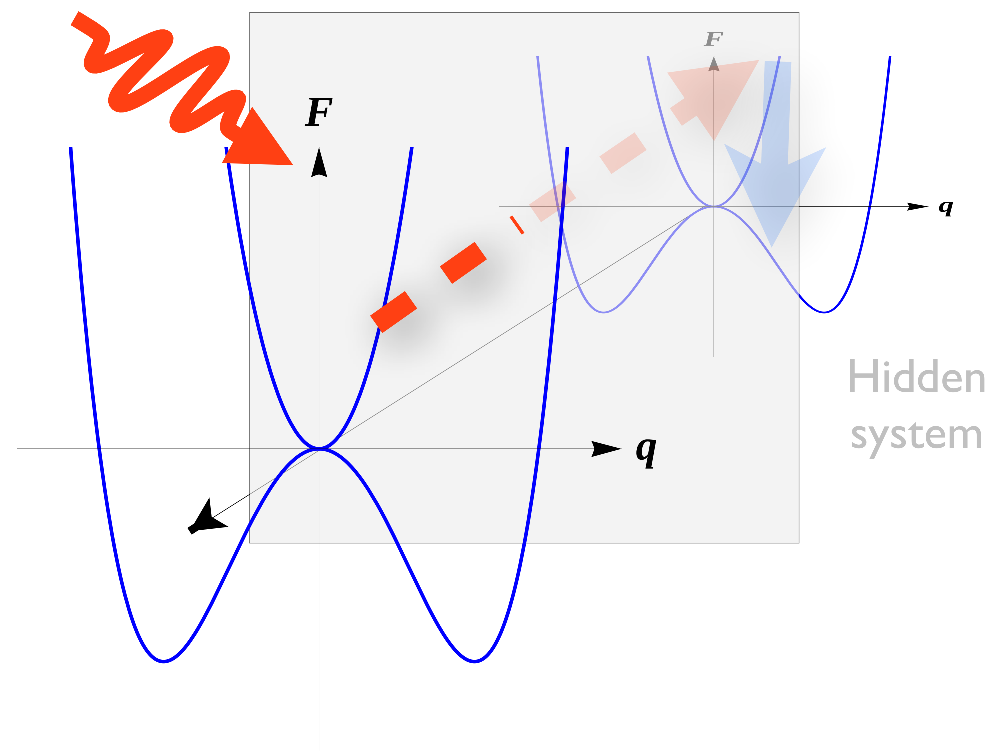


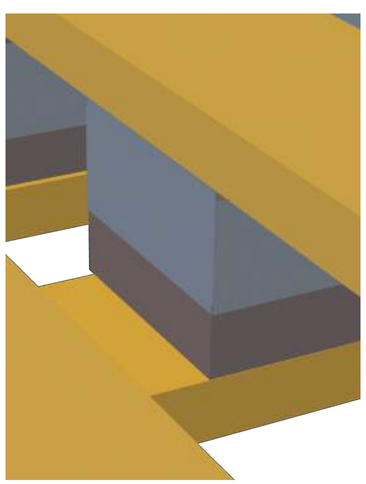
1T-TaS$_2$ device E_W~0.25 pJ

Yang et al, Nat. Nanot. (2010)

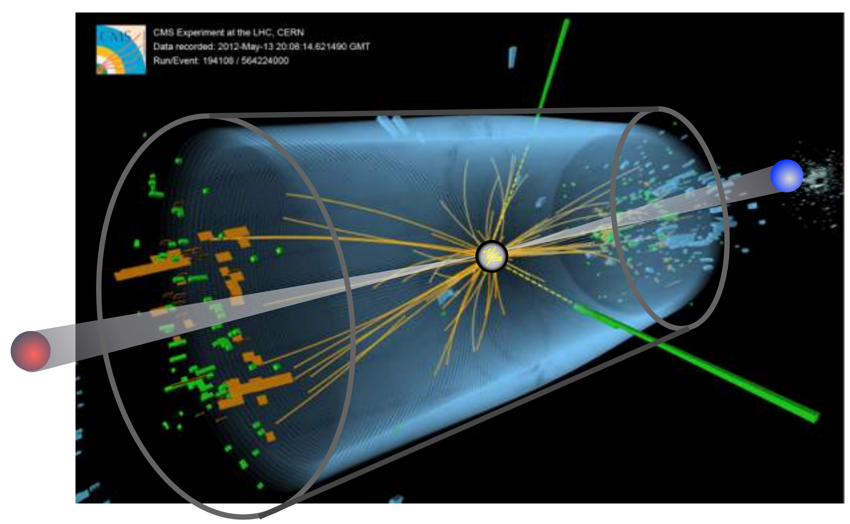
Vaskivskyi, I. *et al.* *arXiv* **cond-mat.mes-hall,** (2014).
I.V., I.A.M and D.M., UK *patent pend.*, 2014

The significance of hidden states of matter





Devices



New elementary particles under non-ergodic conditions
(different beam energies)

Hidden universes?

Mihailovic, D., et al, S. *J Phys-Condens Mat* **25,** 404206 (2013). (Invited paper by T.W.Kibble)