Trieste, Dec. 2014

A brisk walk through phase transitions in time: Oscillating order and topological states. in CDWs and superconductors

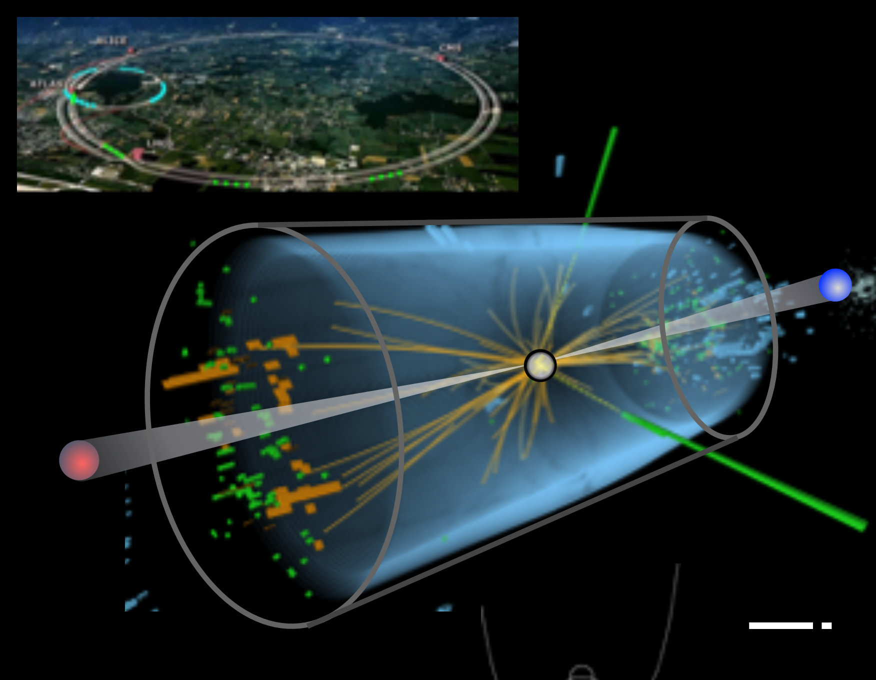
Dragan Mihailovic

Jozef Stefan Institute, Ljubljana, Slovenia
Nanocenter - Center of Excellence for Nanoscience and Nanotechnology
University of Ljubljana, Dept. of Physics
Jožef Stefan International Postgraduate School

European Research Council
Established by the European Commission

Transitions…in time

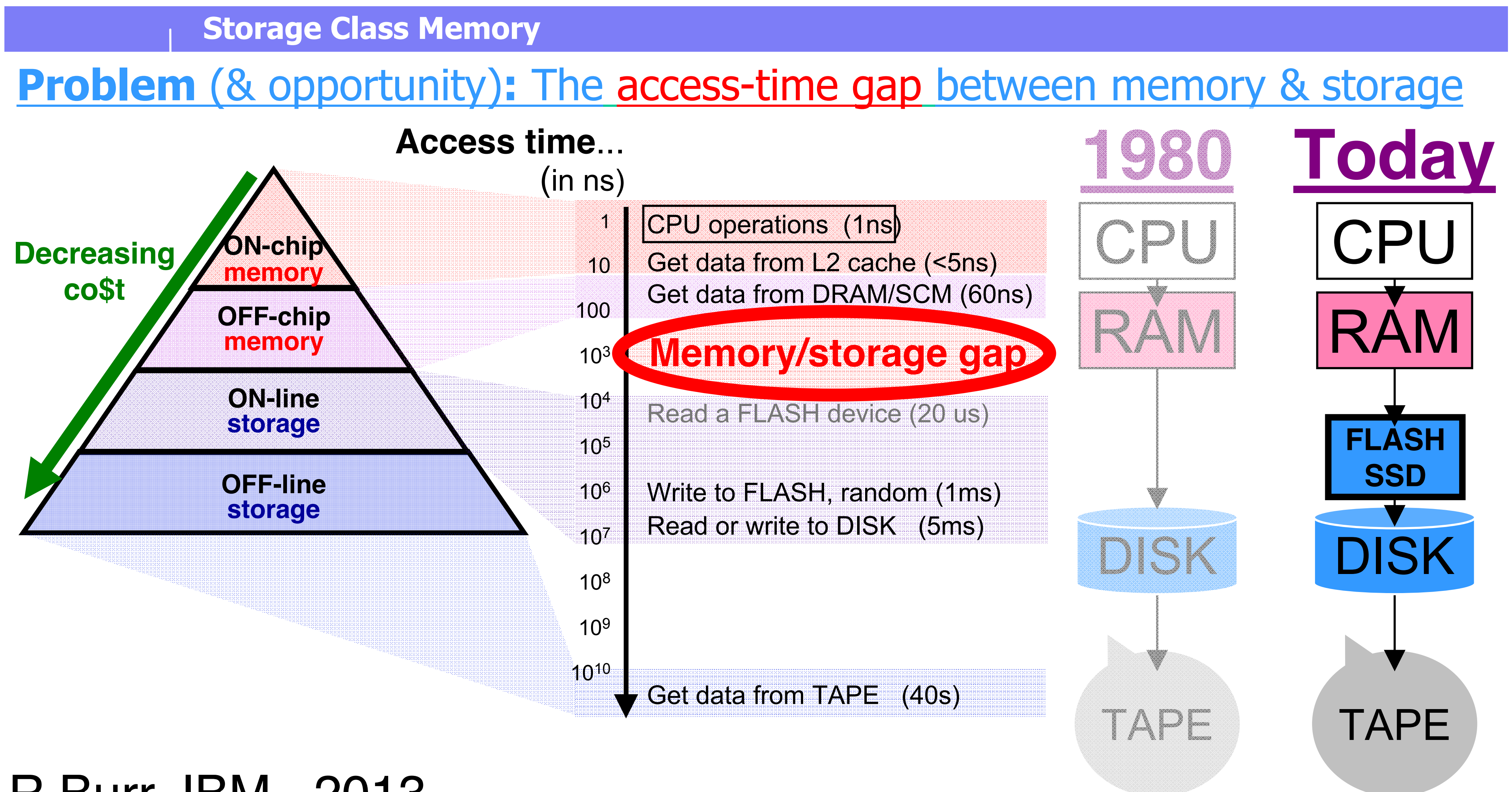
Stock market crashes



Elementary particle collisions

The Big Bang - hidden universes

The  challenge



R.Burr, IBM, 2013

The

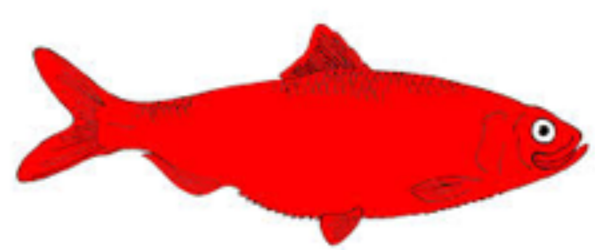
WHAT IS THE PSEUDOGAP? problem

pre-formed pairs? pre-formed SC? a vortex state?

stripes? CDWs or Friedel oscillation?

Dynamic or static inhomogeneity? Bulk or surface? Ionic or electronic origin? Magnetic order?

Or is it just a red herring? (FL theory applies.)



Optical experiments:
(at JSI)

Ljupka Stojchevska
Igor Vaskivskyi
Tomaz Mertelj
Primoz Kusar
Roman Yusupov

Current switching experiments

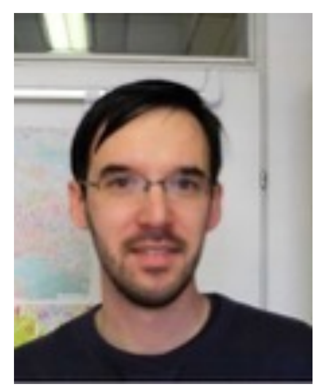
Ian Mihailovic

Samples+:

I. Fisher *(Stanford)*
P. Sutar *(JSI)*
H.Berger *(EPFL)*

Lithography:

D.Svetin *(JSI)*



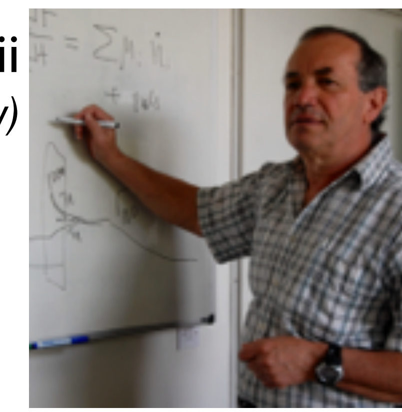
Special thanks to

L.Forro *(EPFL)*

Theory

Viktor Kabanov
(JSI)

Serguei Brazovskii
(Univ. Paris Sud Orsay)



TR ARPES

Patrick Kirchman + ZX Shen group
(Stanford)

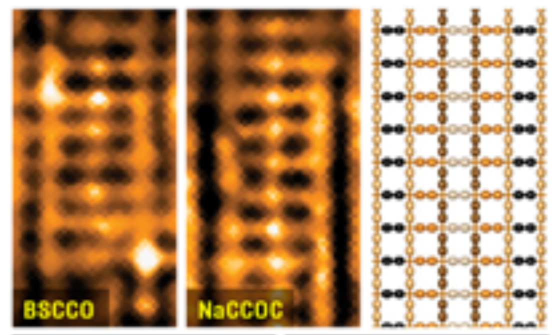
European Research Council
Established by the European Commission

Fermi surface instability, Mott physics (Coulomb repulsion), and strain

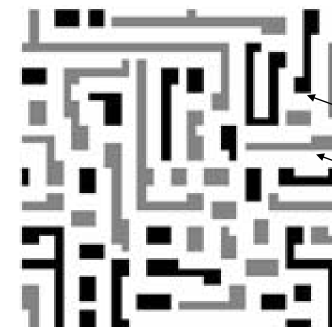
STM, REXS

Theory

Oxides



Fujita et al, PNAS (2014)



k≠0 e-p coupling

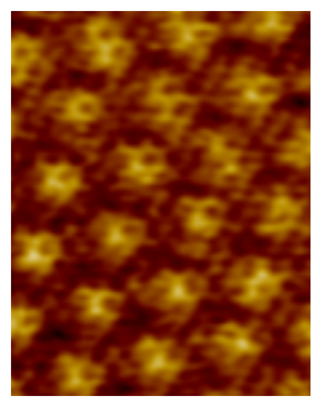
localised charge

uncharged

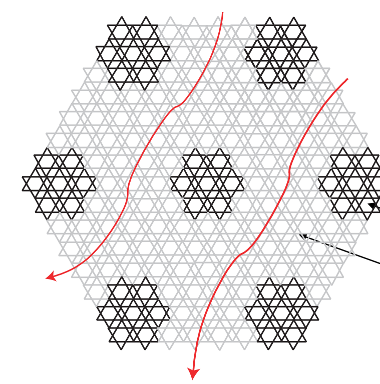
T. Mertelj et al., **PRL** , 147003 (2005), PRB **76**, 9 (2007).

1T-TaS_2

no AF order



I.Vaskivskyi (to be publ.)

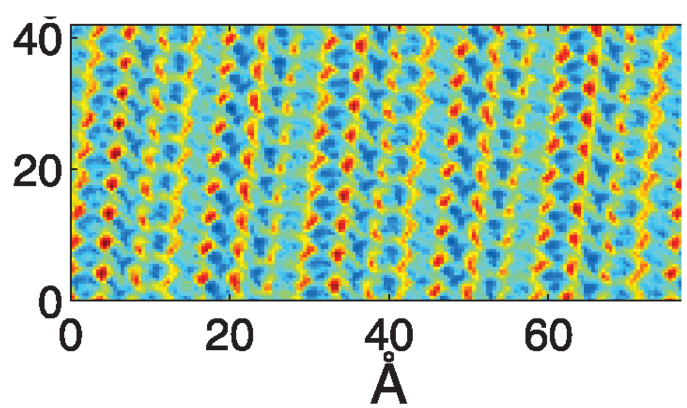


localised charge (polarons)

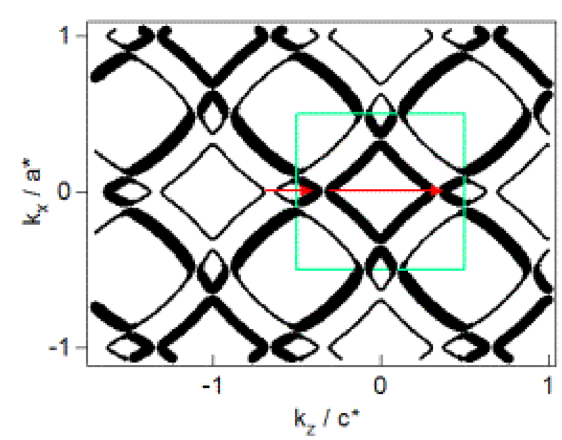
uncharged (delocalised)

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

$TbTe_3$



Fang, A.,PRL **99,** 046401 (2007).

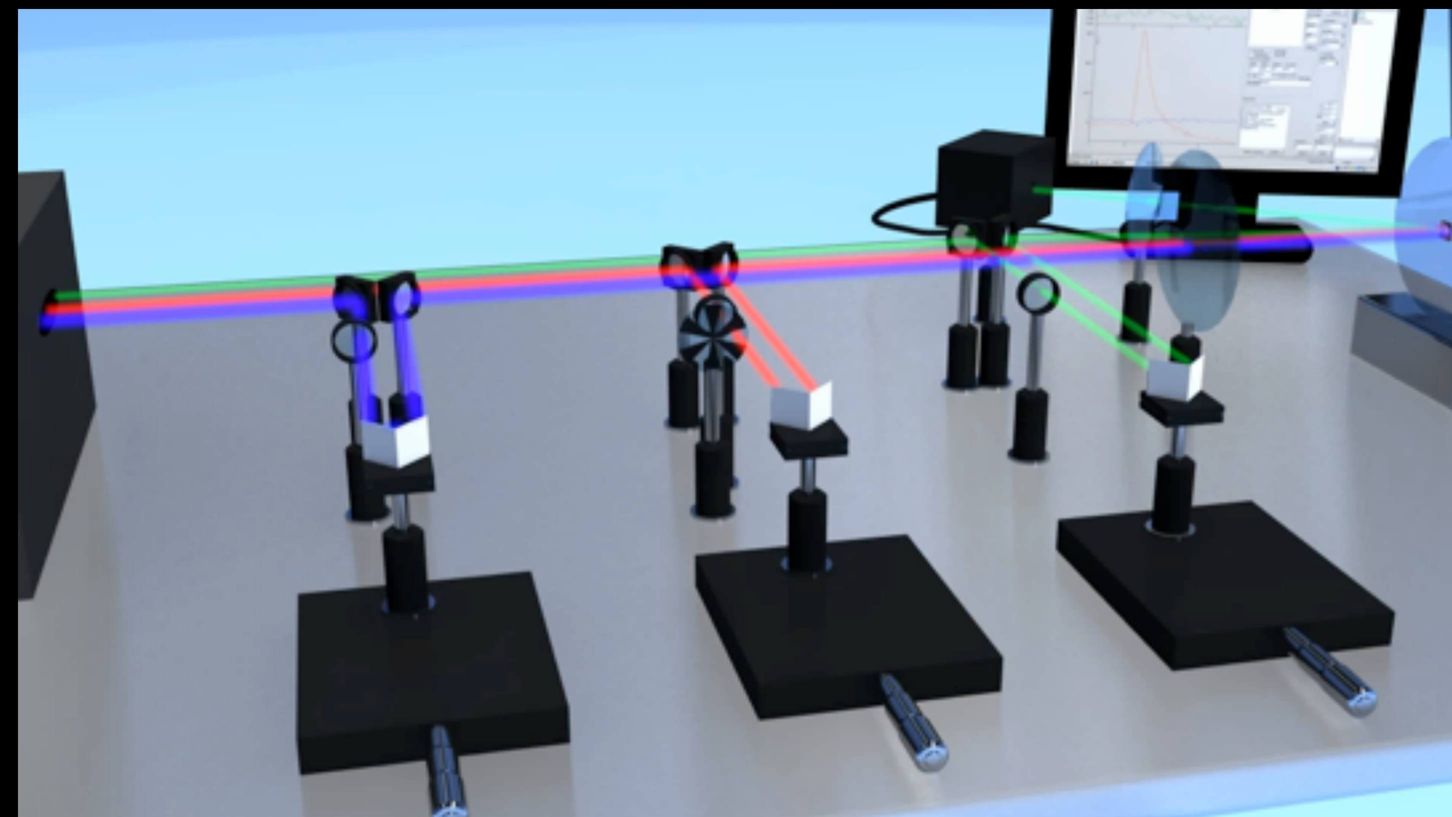


FS nesting?
or
k≠0 e-p coupling?

“Cosmic Quench” experiments

“Cosmology in L^{4}He”, Zurek (1985)

Optical experiments :

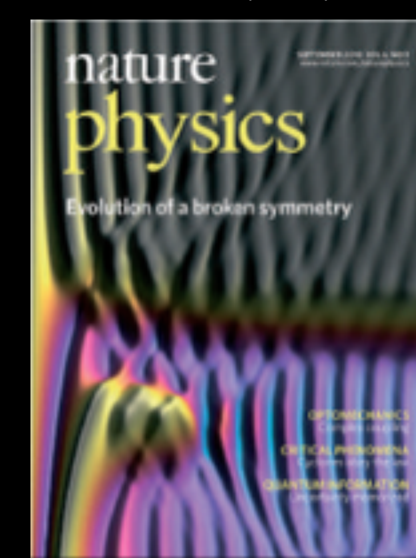


- offer high temporal resolution (easily to 7 fs)
- flexibility in probe wavelengths (THz - UV)
- we can probe the symmetry of different states

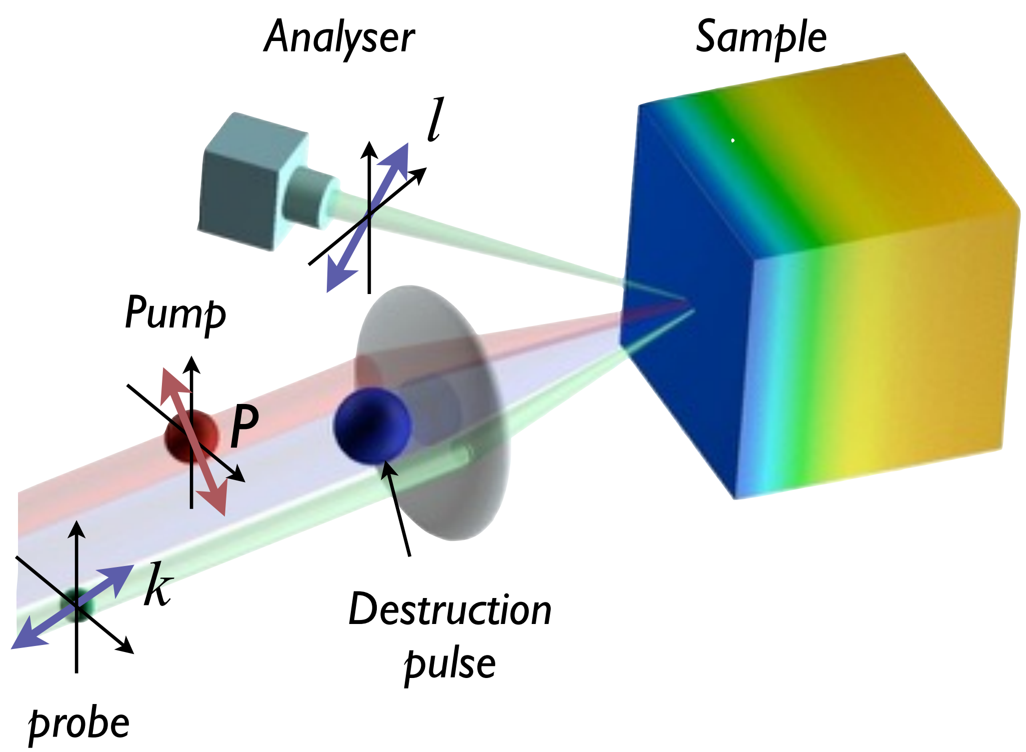
Other experiments :

- TR ARPES : Martin Wolf’s group, Alessandra Lanzara’s group, Z.X. Shen, Uwe Bovensiepen’s group, Luca Perfetti’s group…
- TRED : Jure Demsar &co., Dwayne Miller’s group
- TR XRD, TR XPS…

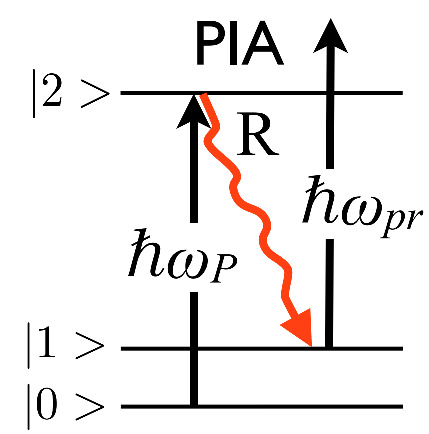
Yusupov, R. *et al. Nat Phys* **6,** 681–684 (2010).



The response of the probe in all-optical experiments



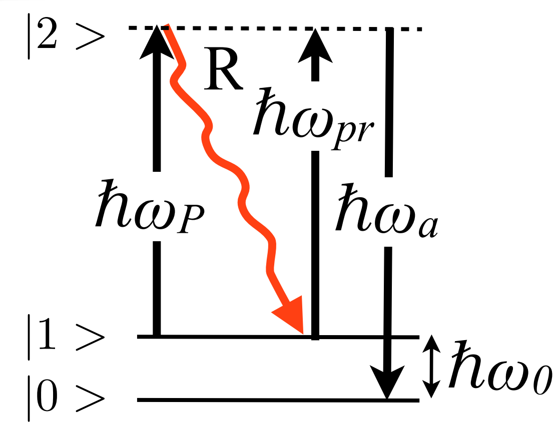
1. Photoinduced absorption (PIA):



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}

Toda, Y. *et al.* PRB **90**, 094513 (2014)

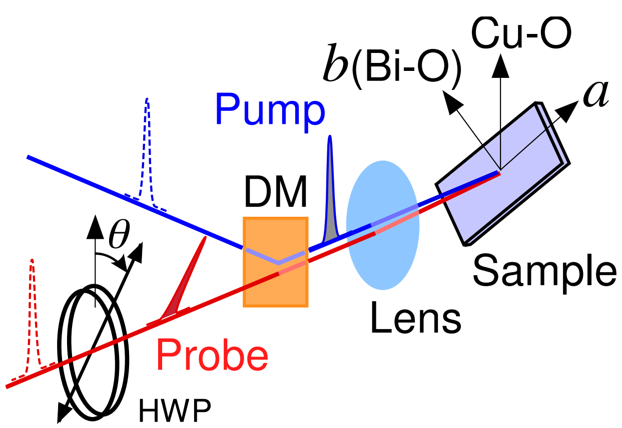
1. Garrett, G., Albrecht, T., WHITAKER, J. & Merlin, R. *Phys Rev Lett* **77,** 3661–3664 (1996).
2. Stevens, T. E., Kuhl, J. & Merlin, R. *Phys Rev B* **65,** 144304 (2002).

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

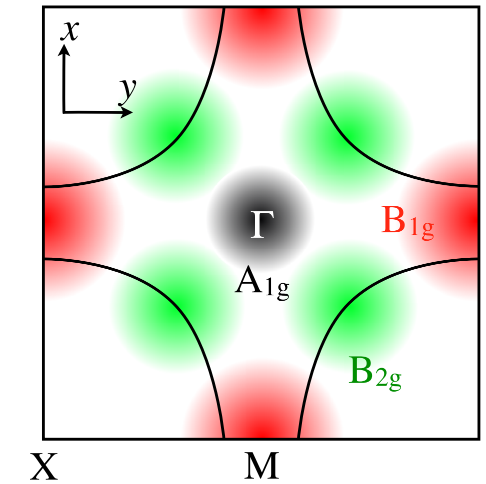
PHYSICAL REVIEW B **90**, 094513 (2014)

Rotational symmetry breaking in $Bi_2Sr_2CaCu_2O_{8+\delta}$ probed by polarized femtosecond spectroscopy

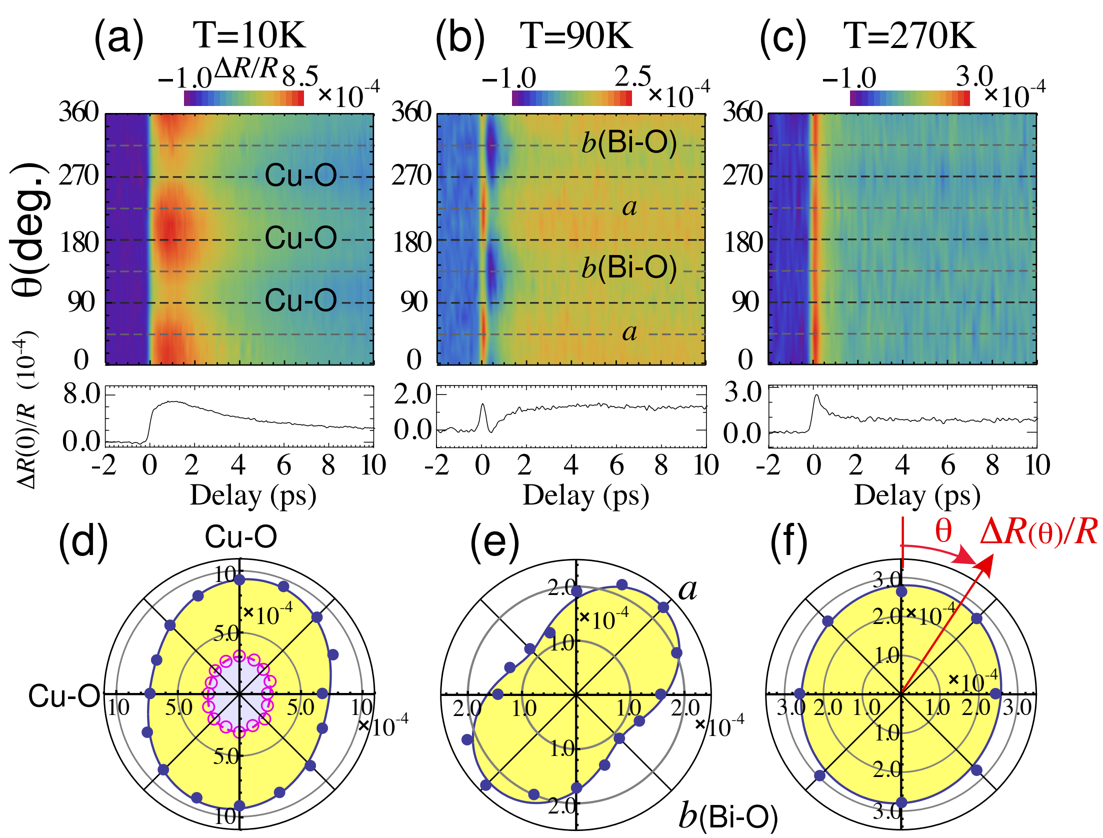
Y. Toda,[1] F. Kawanokami,[1] T. Kurosawa,[2] M. Oda,[2] I. Madan,[3] T. Mertelj,[3] V. V. Kabanov,[3] and D. Mihailovic[3]

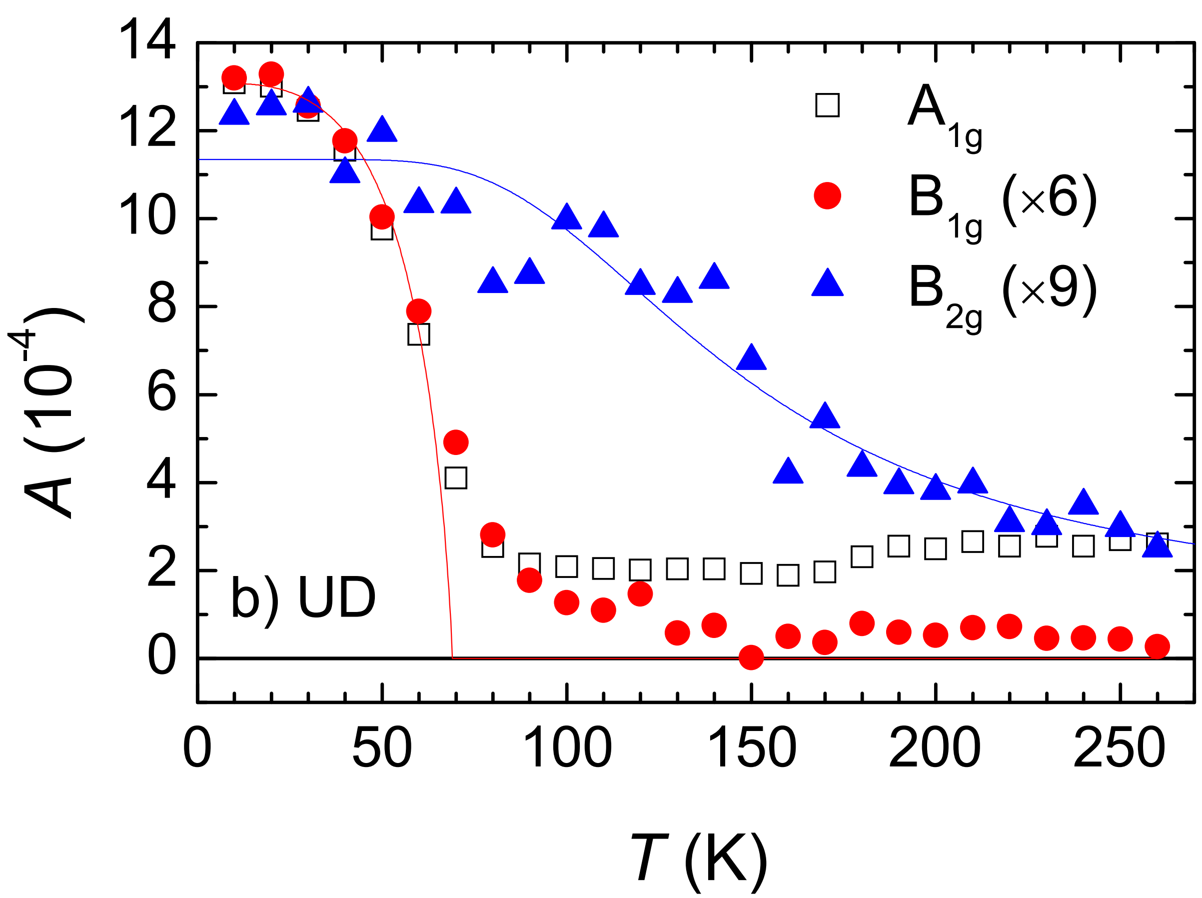


$$\Delta R(\theta) = \frac{\partial R}{\partial \epsilon_1}\left[\Delta\epsilon_1^{A_{1g}} + \Delta\epsilon_1^{B_{1g}}\cos(2\theta) + \Delta\epsilon_1^{B_{2g}}\sin(2\theta)\right] + \frac{\partial R}{\partial \epsilon_2}\left[\Delta\epsilon_2^{A_{1g}} + \Delta\epsilon_2^{B_{1g}}\cos(2\theta) + \Delta\epsilon_2^{B_{2g}}\sin(2\theta)\right] \quad (2)$$



Temperature dependence of different symmetry components, A_{1g}, B_{1g} and B_{2g}:

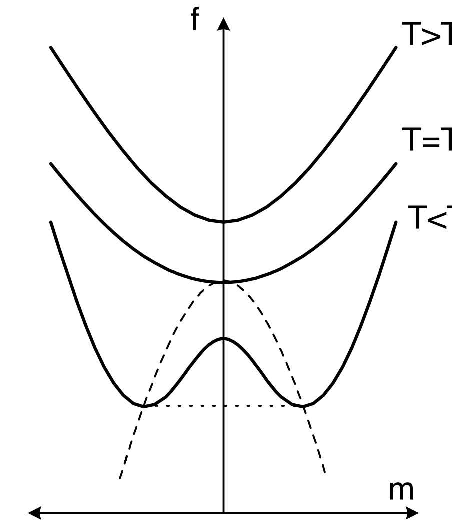




The system trajectory in GL theory

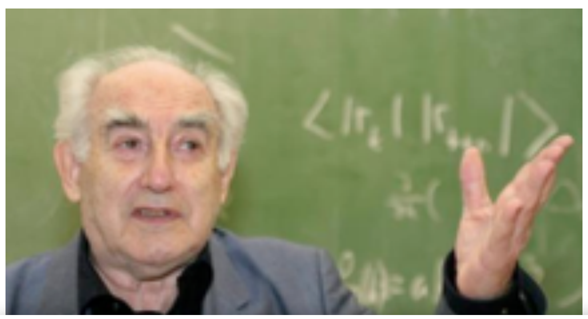
The Landau non-linear energy functional originally written to describe a **structural phase transition**:

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



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

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



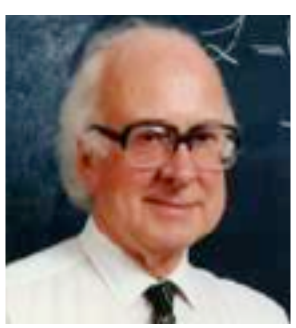
Complex order parameter

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

LUME 13, NUMBER 16 PHYSICAL REVIEW LETTERS 19 OCTOB

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. term

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

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

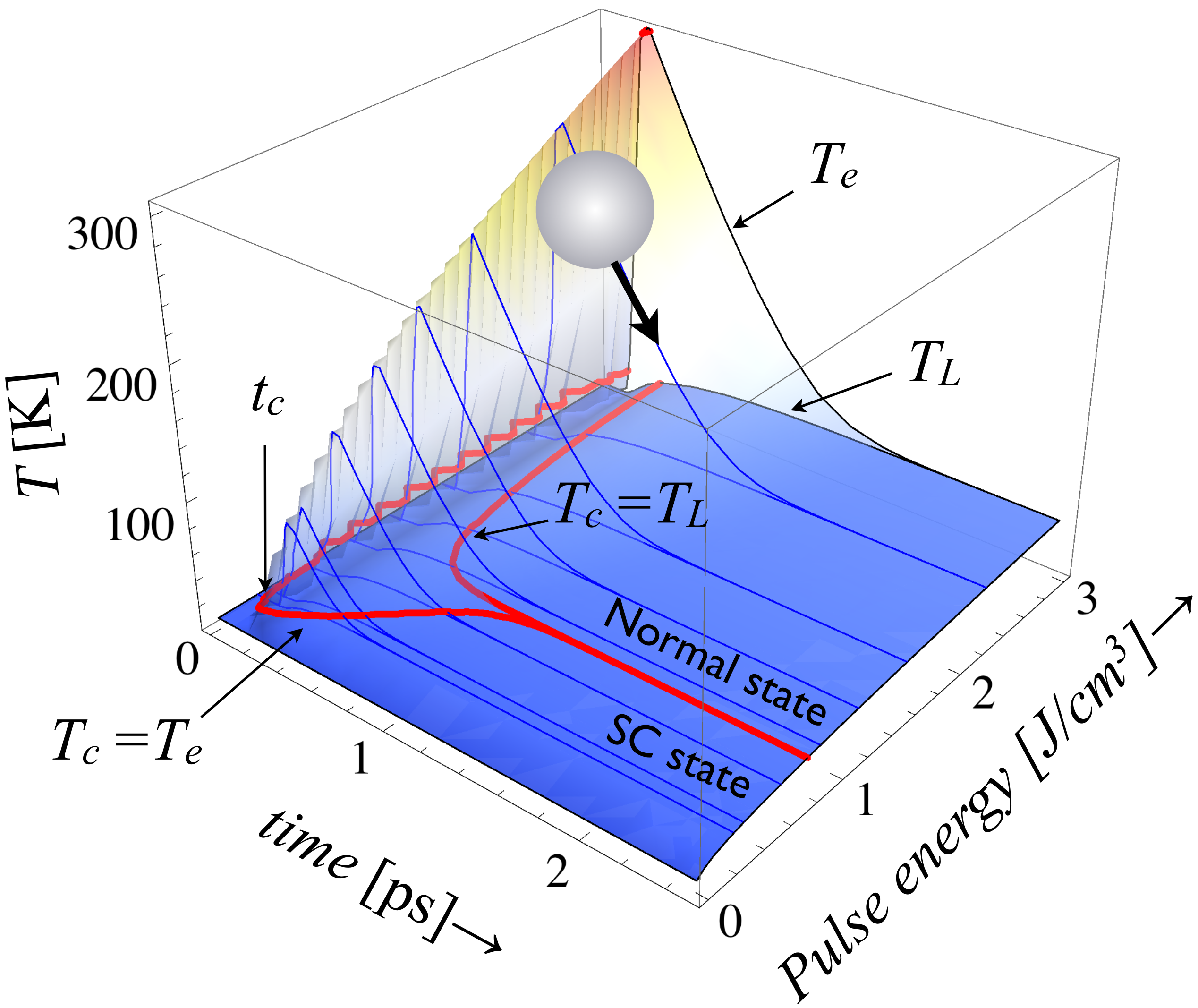
Although our discussion will be quite general, for illustrative purposes it is convenient to have a specific example in mind. Let us consider an N-component real scalar field ϕ with a Lagrangian invariant under the orthogonal group $O(N)$, and coupled in the usual way to $\frac{1}{2}N(N-1)$ vector fields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

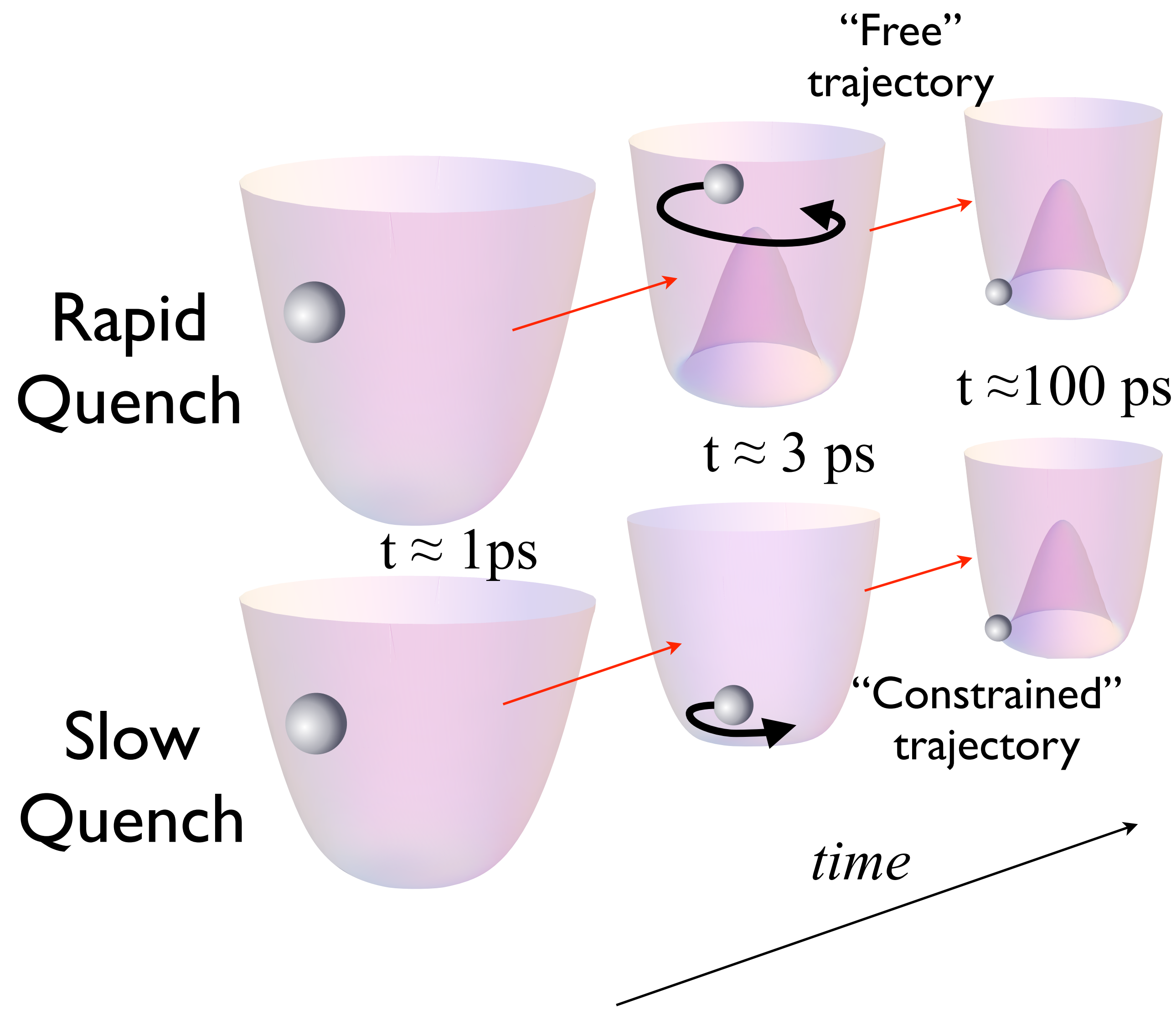
$$L = \tfrac{1}{2}(D_\mu\phi)^2 - \tfrac{1}{8}g^2(\phi^2 - \eta^2)^2 + \tfrac{1}{8}\mathrm{Tr}(B_{\mu\nu}B^{\mu\nu}) \quad (1)$$

with

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

The quench through T_c



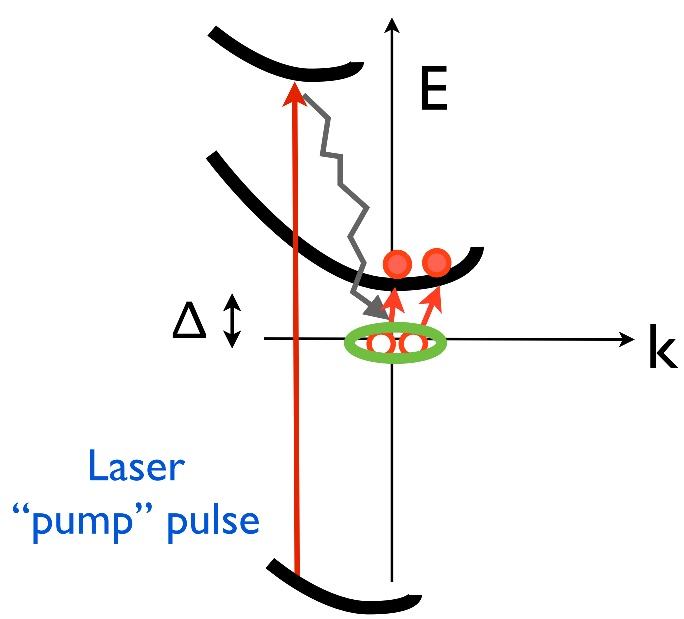


Higgs mode osc. period: $2\pi/\omega_H \sim 0.1$ ps

Quench time: $\tau_Q = 0.1 \sim 30$ ps

Ginzburg-Landau time: $\tau_{GL} = 1/\Delta_0 = 0.1 \simeq 5\text{ps}$

Laser vaporisation of the superconducting condensate

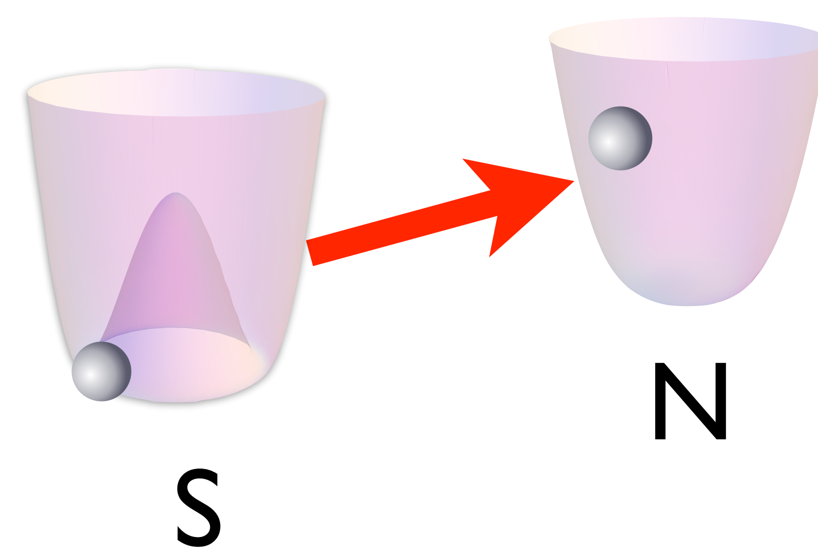


Single particle (electron) energy relaxation via boson (phonon) emission

↓ $t_D \sim 1$ ps

Bosons destroy pairs, creating QPs

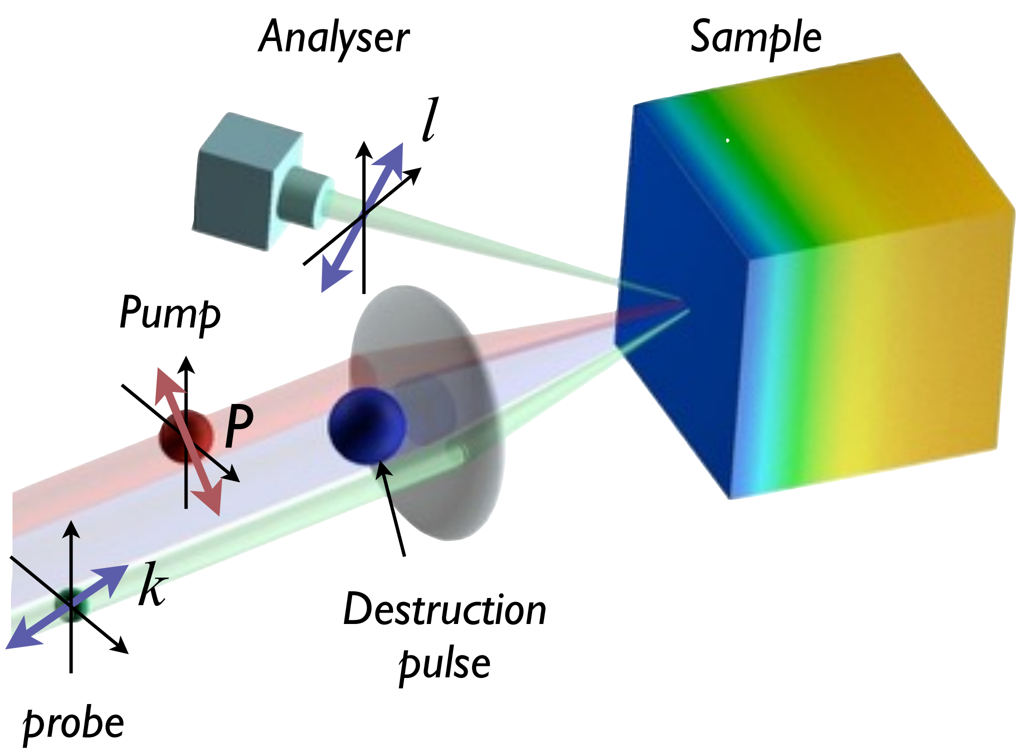
The SC condensate is vaporised in less than 1 ps

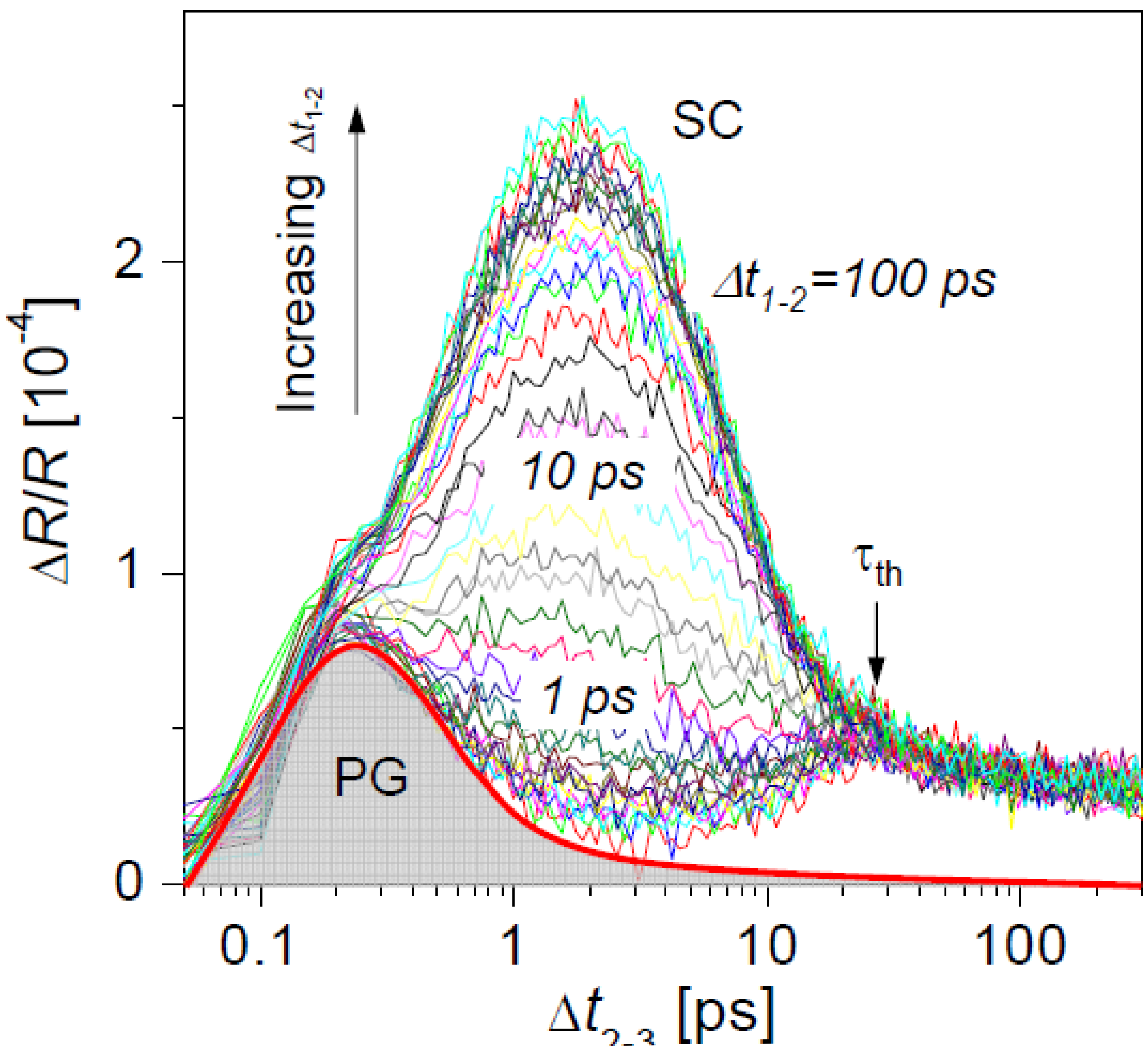


$t_D \sim 1$ ps

1. Kusar, P., Kabanov, V., Demsar, J. & Mertelj, T. Controlled Vaporization of the Superconducting Condensate in Cuprate Superconductors by Femtosecond Photoexcitation. *Phys Rev Lett* **101**, 227001 (2008).
2. Stojchevska, L. *et al.* Mechanisms of nonthermal destruction of the superconducting state and melting of the charge-density-wave state by femtosecond laser pulses. *Phys Rev B* **84**, 180507(R) (2011).

Transient reflectivity

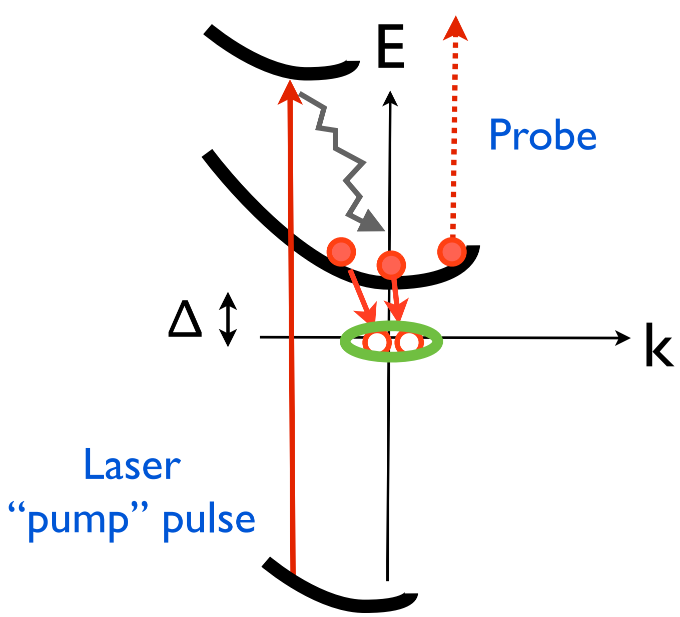


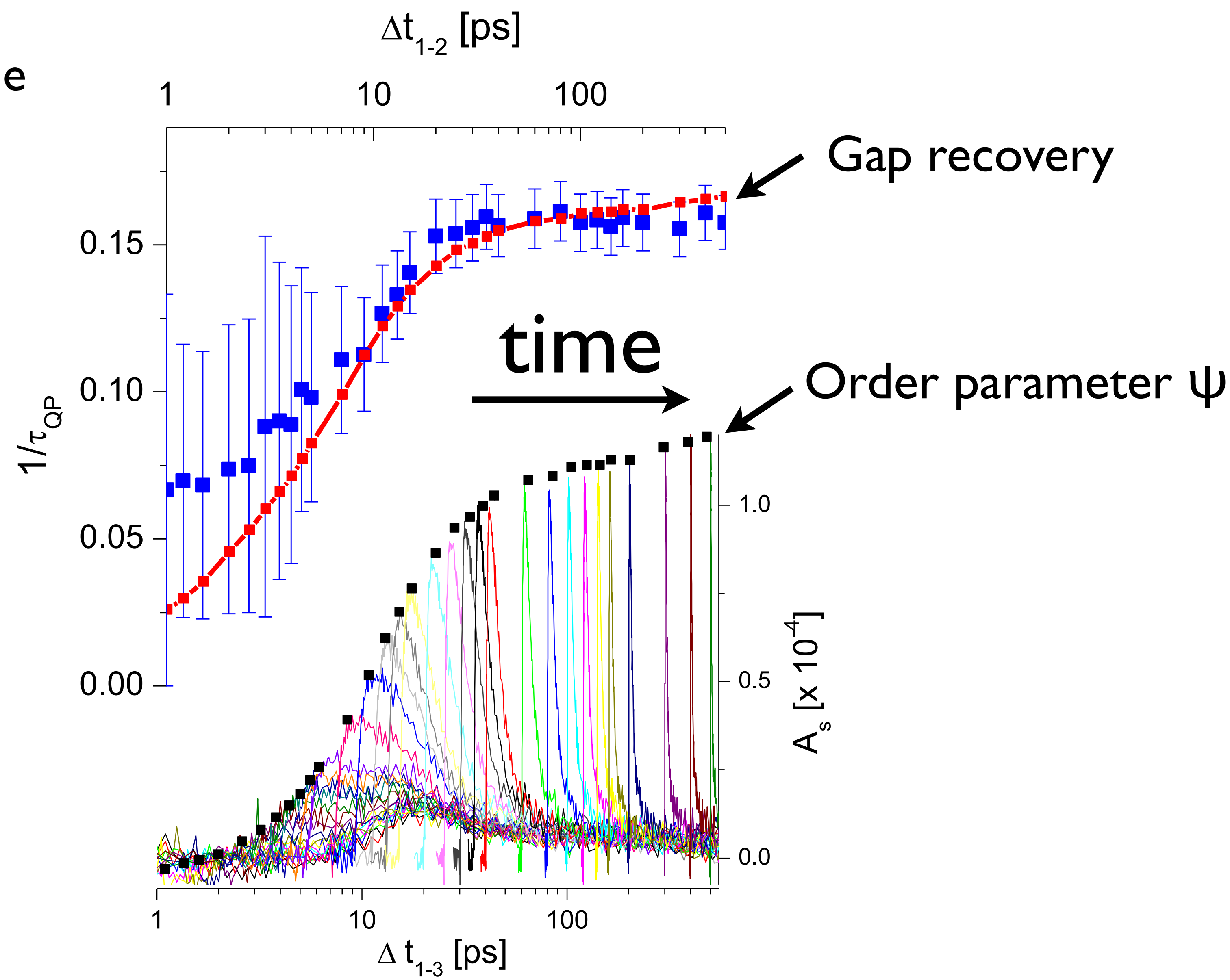


Superconducting gap recovery: QP recombination time compared with reflectivity amplitude

Quasiparticle recombination time

$$1/\tau_{QP} \sim \Delta(t)$$





No Higgs oscillations observed in $A_s(t)$!

Modelling of superconducting state recovery:

TDGL:

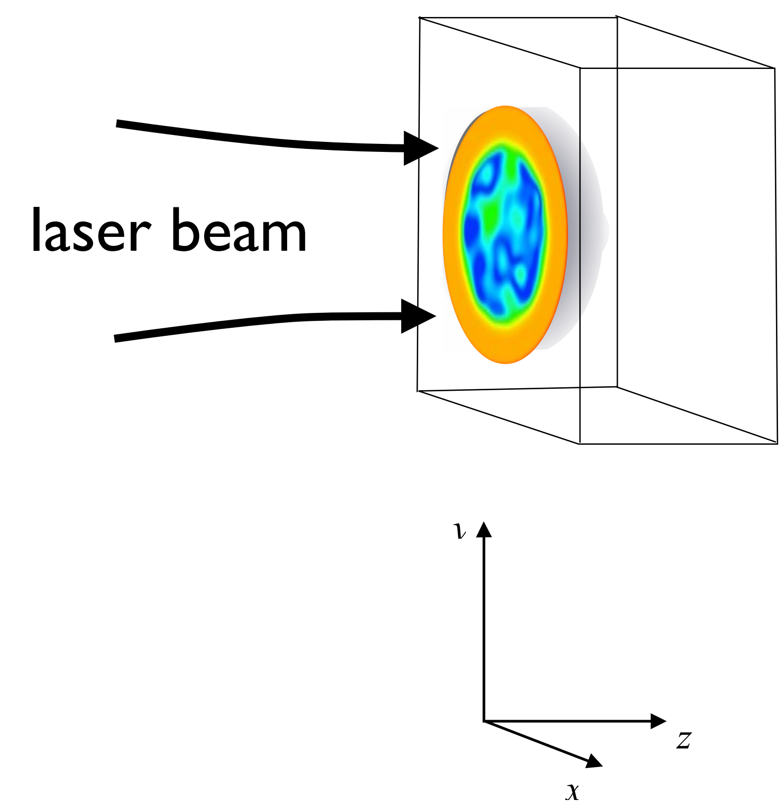
$$u\left(\frac{\partial\psi}{\partial t}\cancel{+i\Phi\psi}\right) = -\alpha_r(t,z)\psi - \psi|\psi|^2 - (i\nabla \cancel{+\mathbf{a}})^2\psi + \eta$$

$$\cancel{\nabla^2\Phi = -\nabla\left[\frac{i}{2}(\psi^*\nabla\psi - \psi\nabla\psi^*) + \mathbf{a}|\psi|^2\right]}$$

no external field

Diffusion:

$$\frac{\partial T_D(t,\mathbf{s})}{\partial t} = D\nabla^2 T_D$$



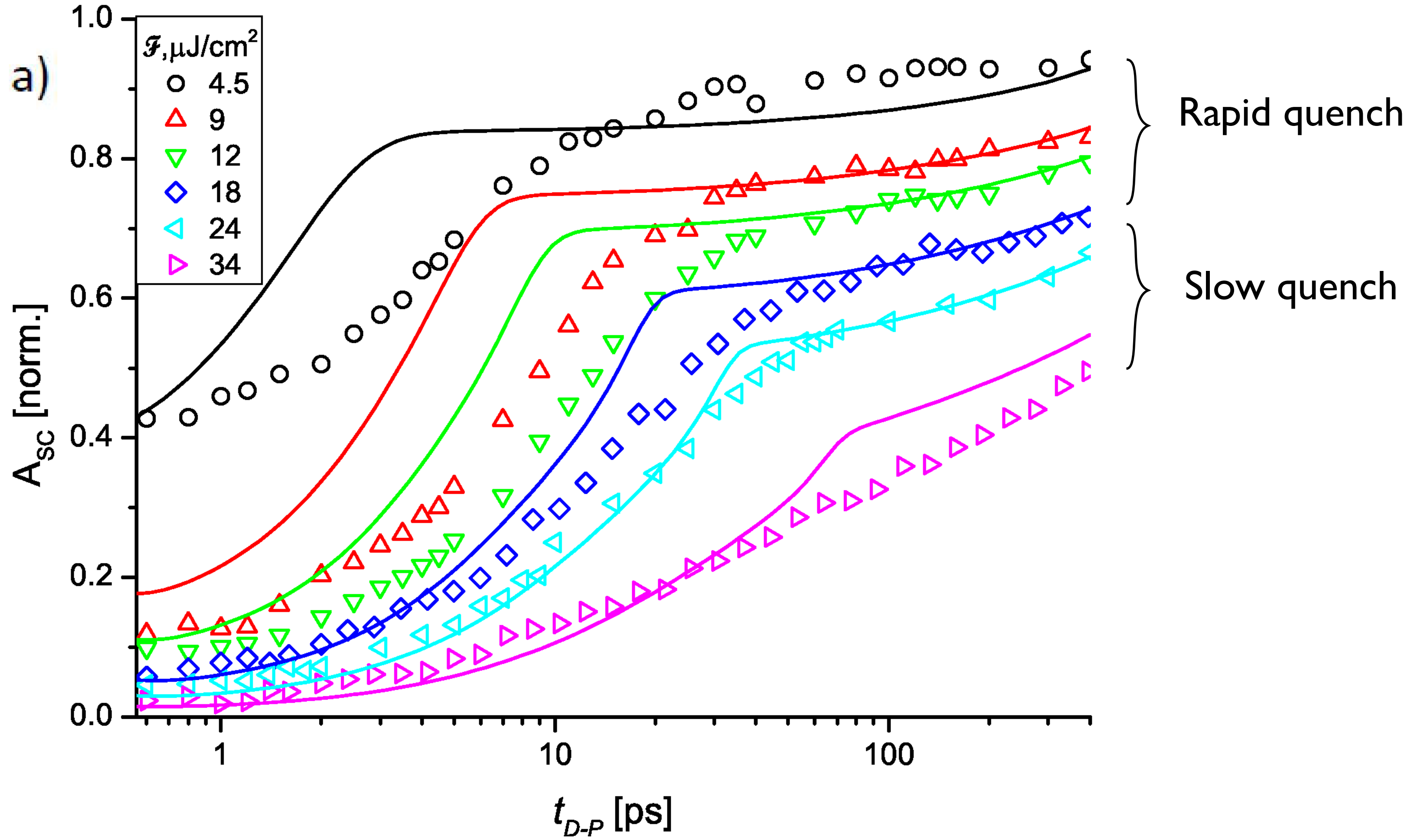
Photoexcited electron energy loss:

$$\gamma_e T_e \frac{dT_e}{dt} = -\gamma_L(T_e - T_L) + P(t)$$

$$C_L(T)\frac{dT_L}{dt} = \gamma_L(T_e - T_L)$$

Basic TDGL equation: $$\frac{\partial \psi}{\partial t} = \alpha_r(t,z)\psi - \psi|\psi|^2 + \nabla^2\psi + \eta$$

Boundary conditions: $$\psi(0,z) = \begin{cases} 0 & ,\mathcal{F}(z) > \mathcal{F}_T; \\ (1 - \frac{\mathcal{F}}{\mathcal{F}_T} e^{-z/\lambda})\sqrt{1 - T(0,z)/T_c} & ,\mathcal{F}(z) < \mathcal{F}_T. \end{cases}$$

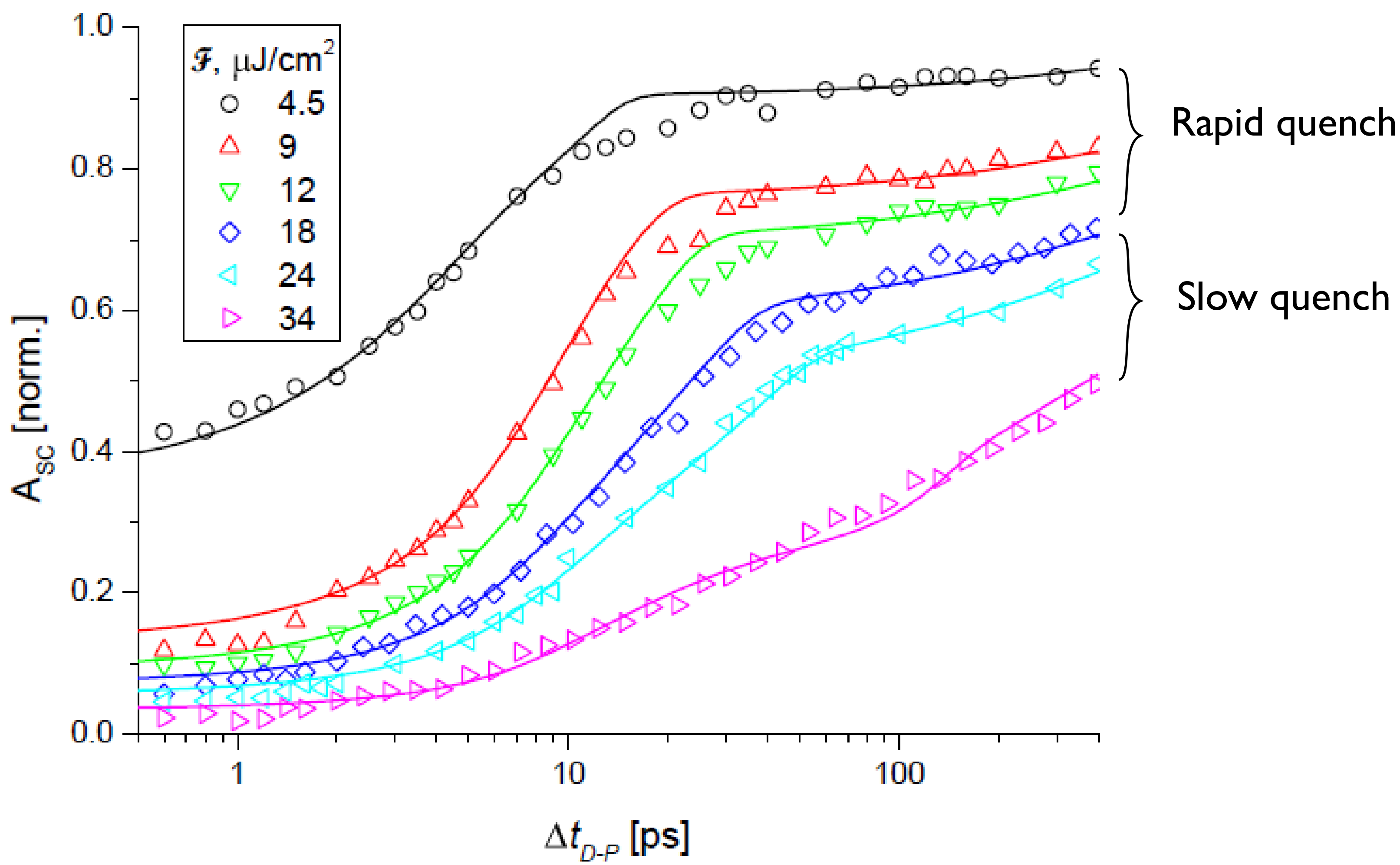


Doesn’t work very well!

Basic TDGL equation: $$\frac{\partial \psi}{\partial t} = \alpha_r(t,z)\psi - \psi|\psi|^2 + \nabla^2\psi + \eta$$

Boundary conditions: $$\psi(0,z) = \begin{cases} 0 & ,\mathcal{F}(z) > \mathcal{F}_T; \\ (1 - \frac{\mathcal{F}}{\mathcal{F}_T} e^{-z/\lambda})\sqrt{1 - T(0,z)/T_c} & ,\mathcal{F}(z) < \mathcal{F}_T. \end{cases}$$

Introducing fluctuations for t < 0 (Volovik, 2000)†:
$$\psi(t=0) = \kappa z$$



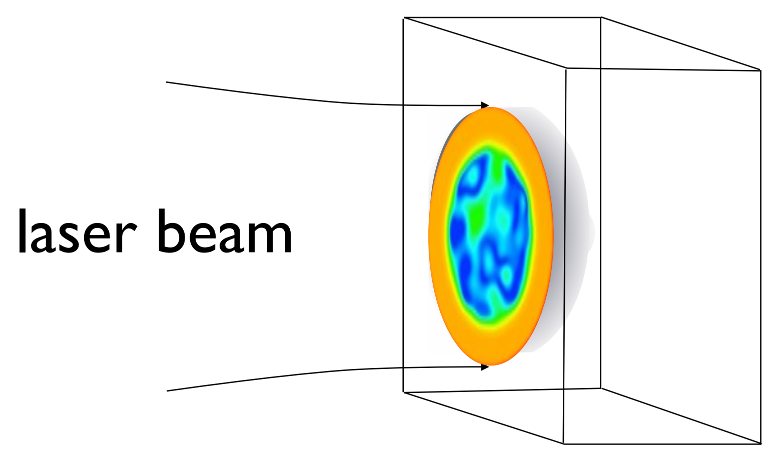
Pre-existing order: a much better fit

† G. E. Volovik, *Physica B* **280** 122-127 (2000).

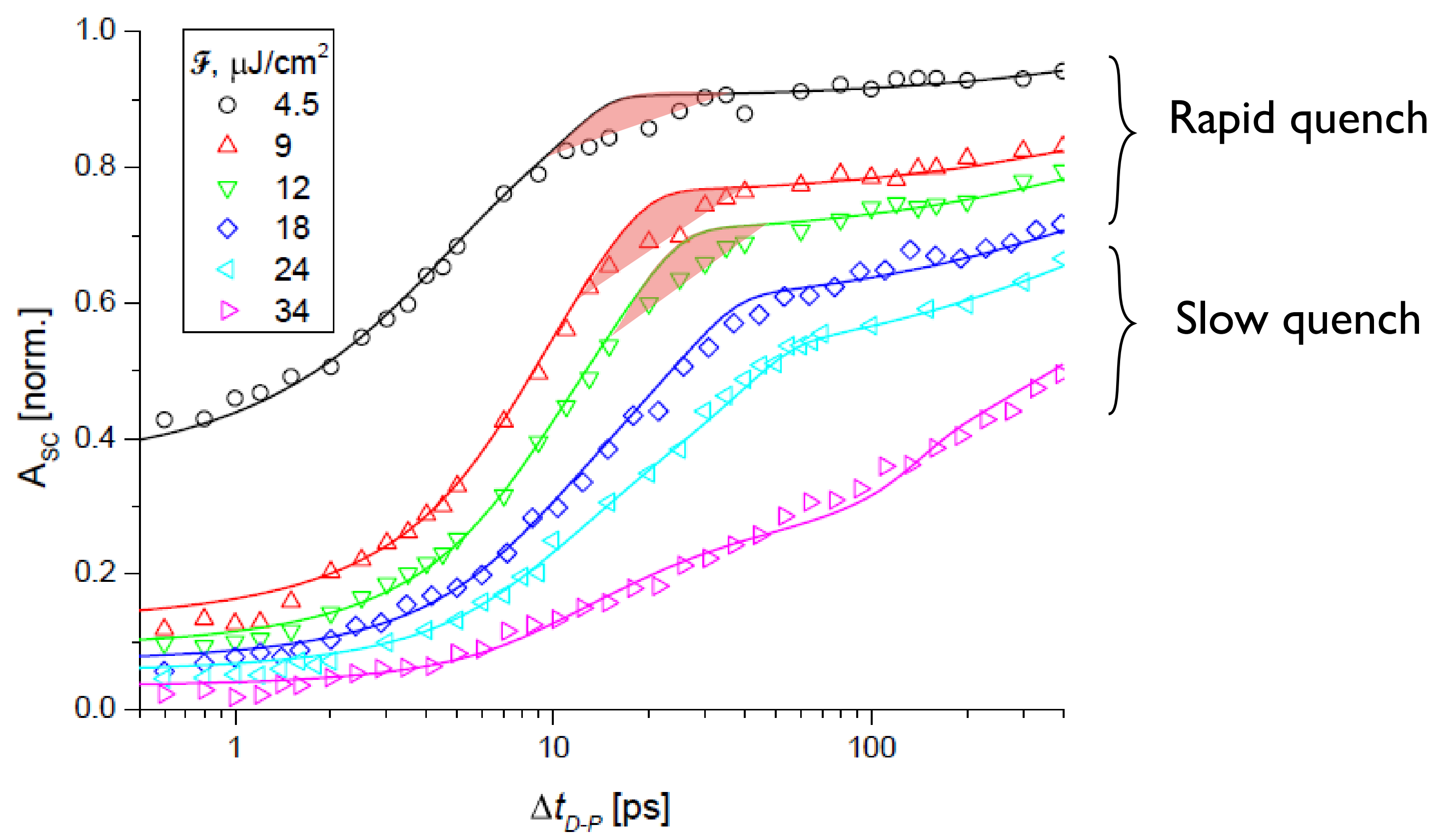
Kibble-Zurek mechanism: Evidence for vortex formation and annihilation on 10 ps timescale

Laser spot size: $d = 60\mu\mathrm{m}$

Coherence length: $\xi_{||} \simeq 2\mathrm{nm}$



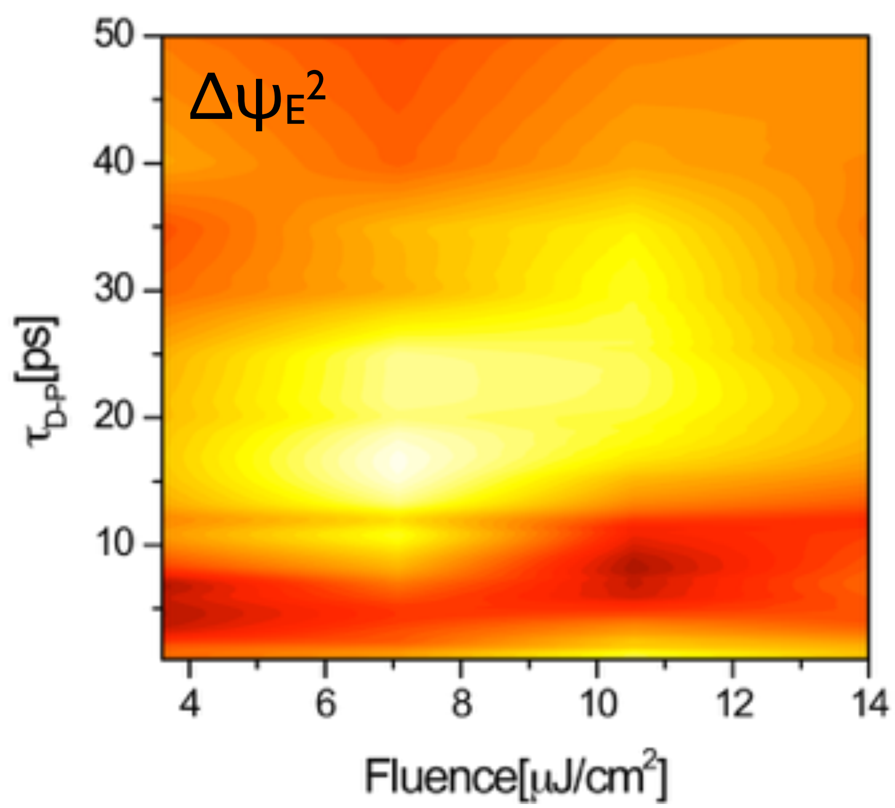
Regions are causally unconnected and evolve independently after the quench which causes the **formation of topological defects**.



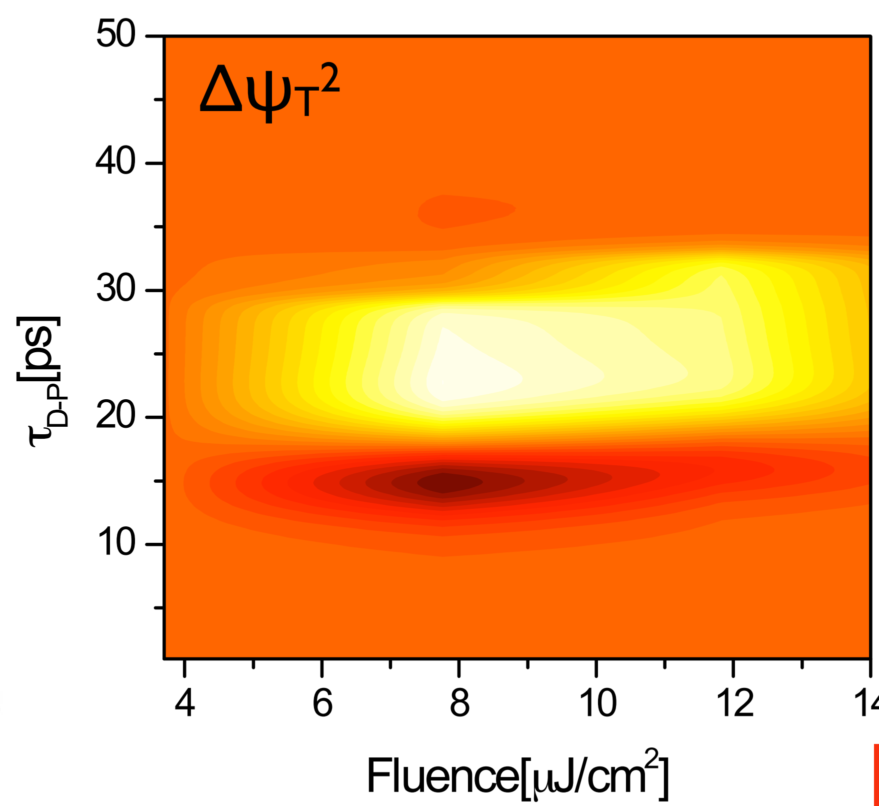
Mihailovic et al., J Phys-Condens Mat 25, 404206 (2013).

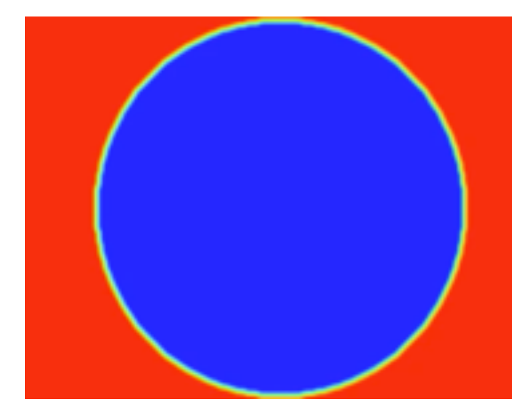
Vortices created in the quench annihilate on a timescale of 10-30 ps

Experiment



Theory calculation



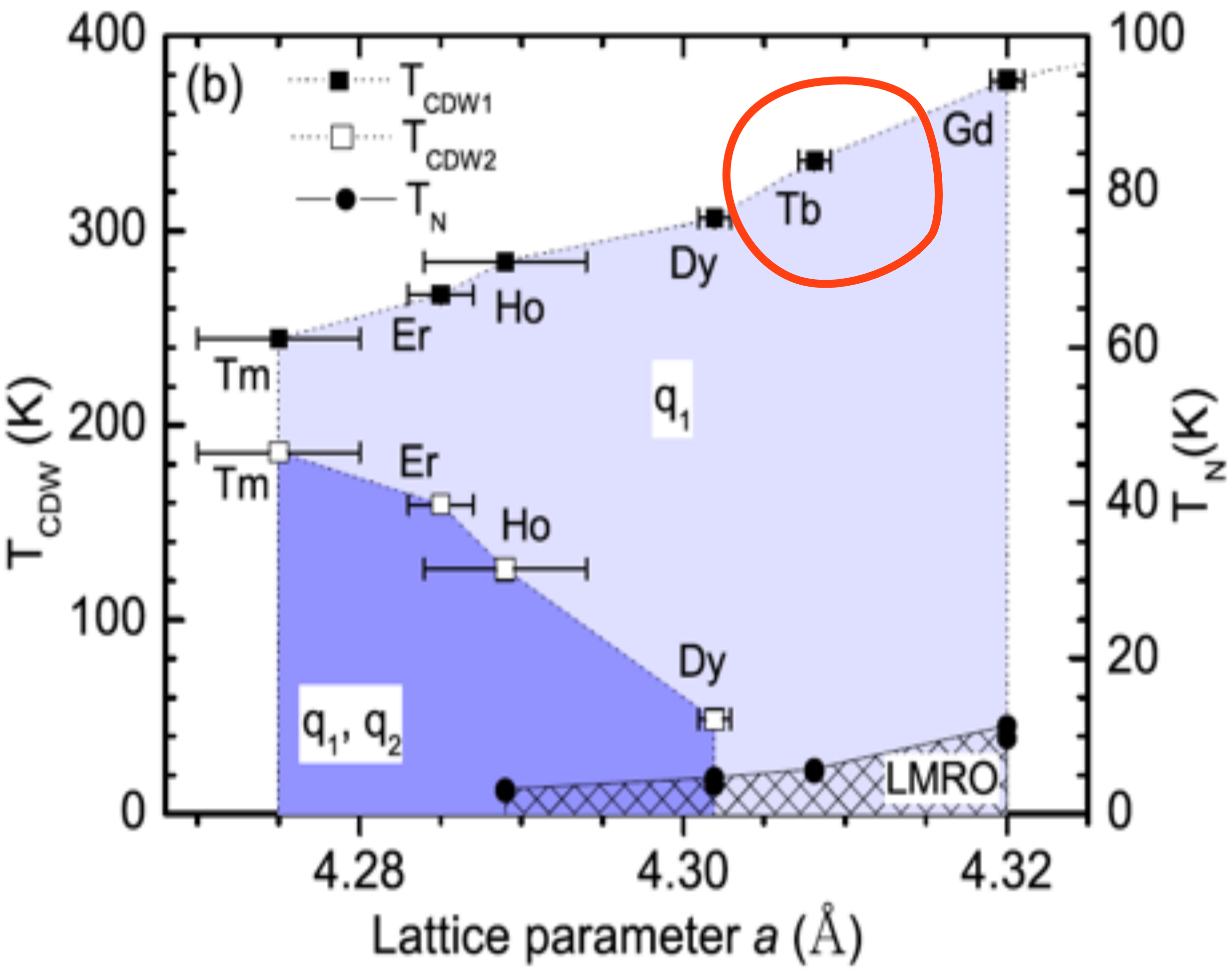


Laser spot

D. Mihailovic, T. Mertelj, V.V. Kabanov, and S. Brazovskii, J Phys-Condens Mat 25, 404206 (2013).

$TbTe_3$

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



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, strongly 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.

The system trajectory: TDGL theory for a CDW

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:

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

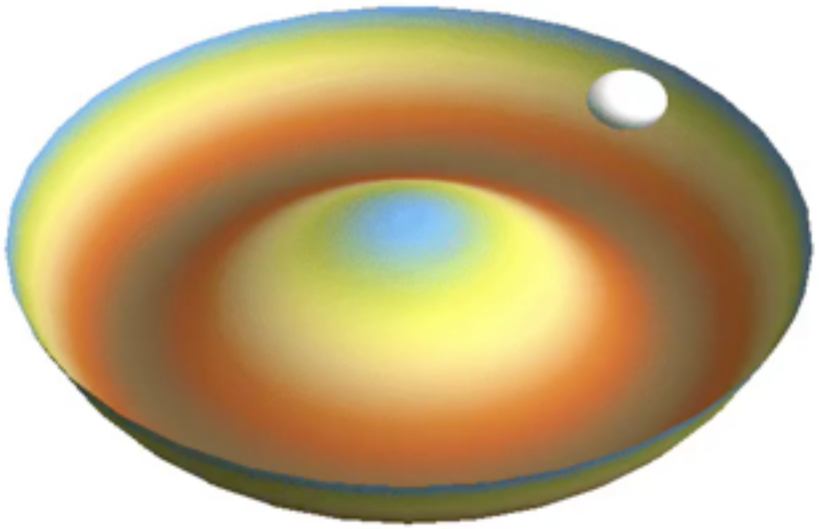
"The quench process"

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

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

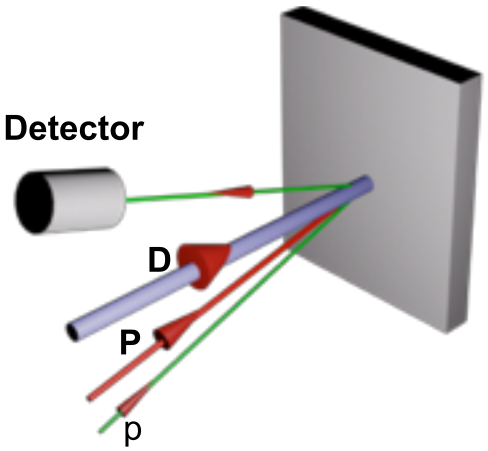
The order parameter, $\psi(t) = A(t)e^{i\phi(t)}$

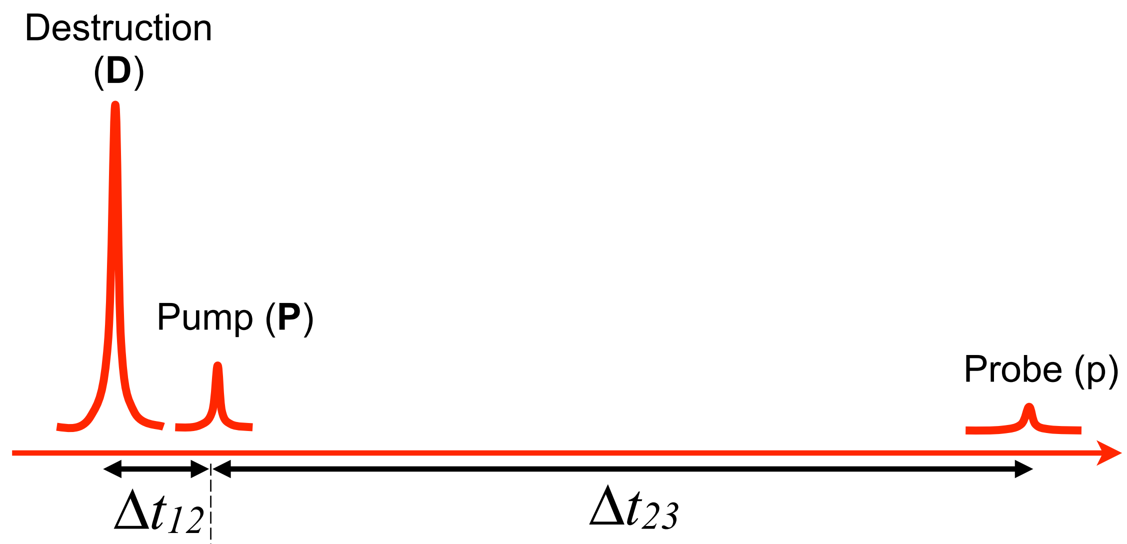
† Phase fluctuations are assumed to be slow.

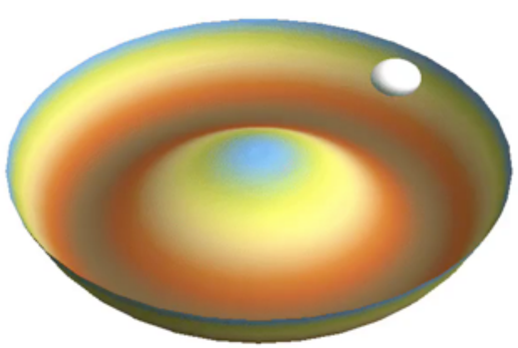


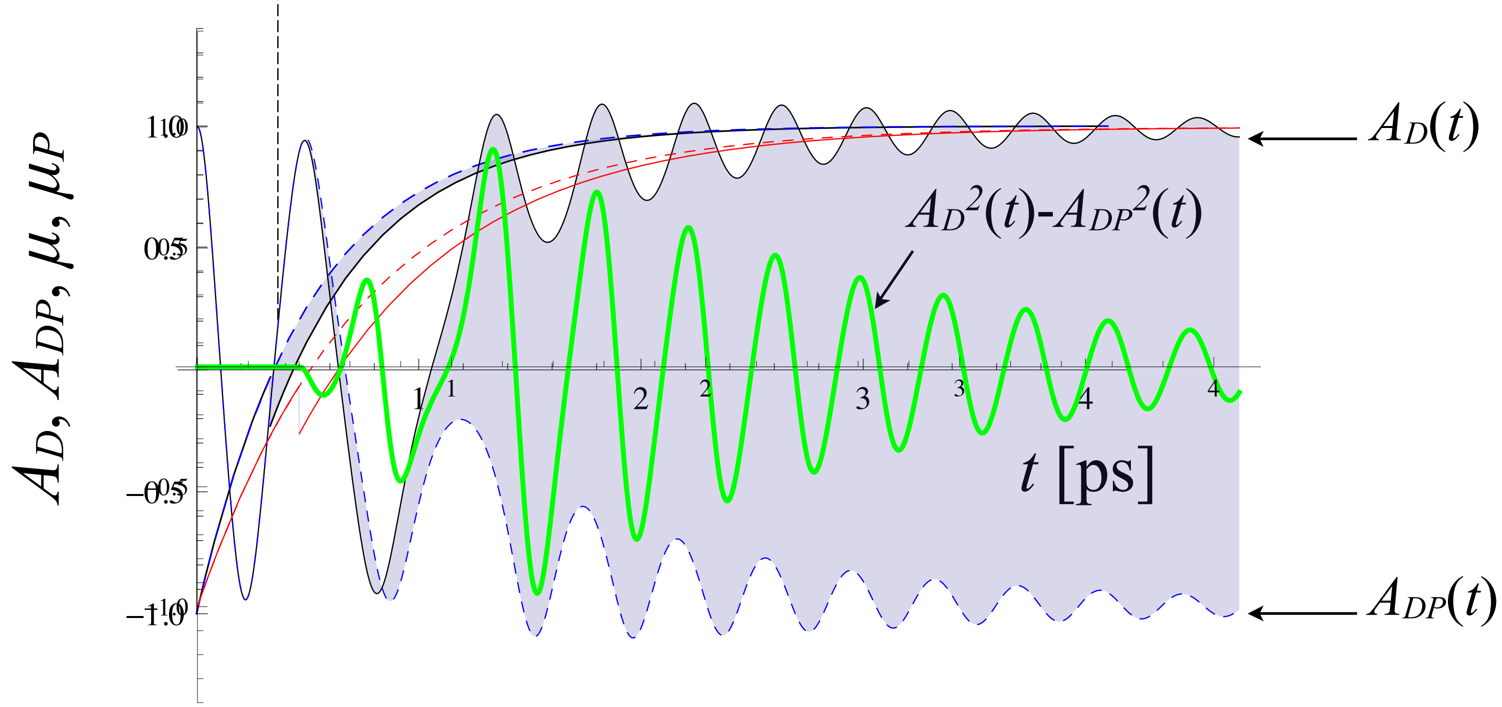
Yusupov, R. *et al.* Coherent dynamics of macroscopic electronic order through a symmetry breaking transition. *Nat Phys* **6,** 681–684 (2010).

The predicted optical response of the collective mode



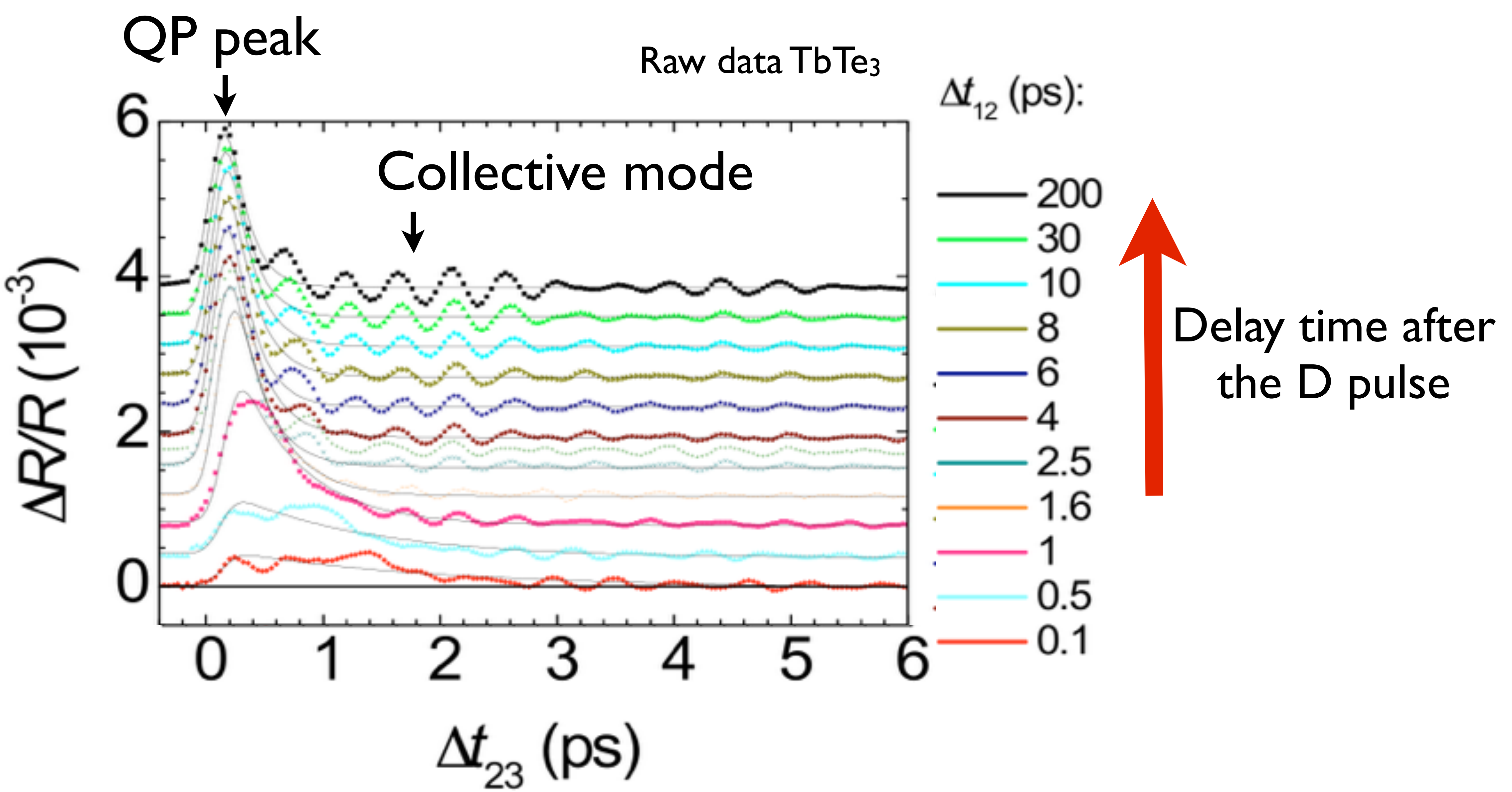






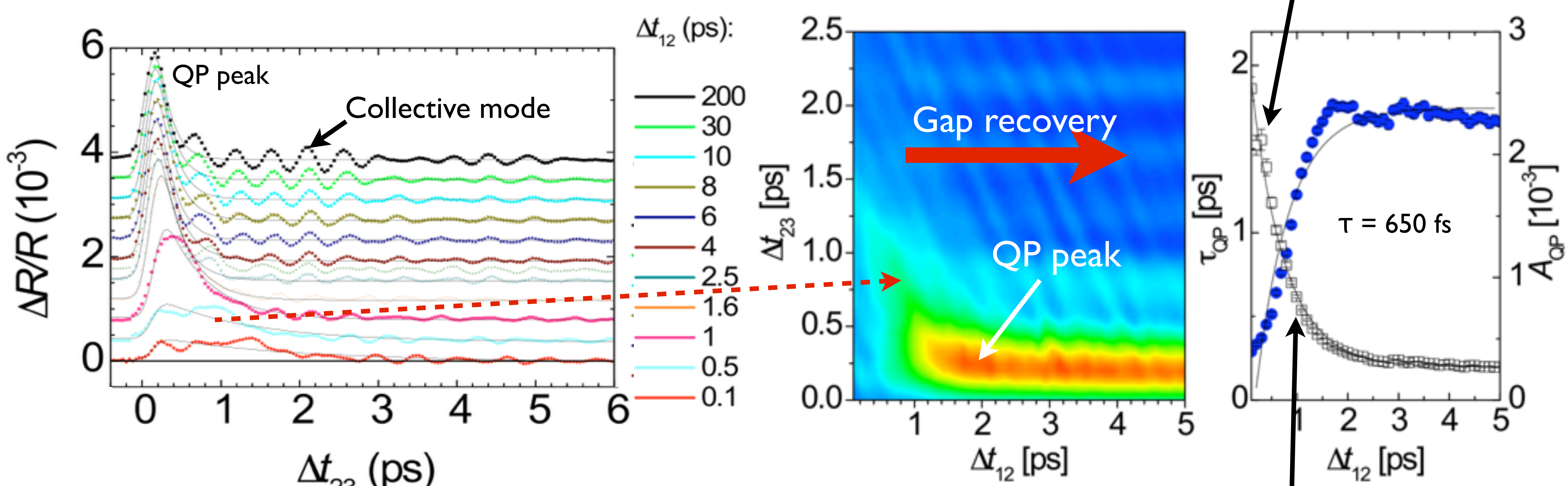
The reflectivity, $\Delta R(t) \propto \left(\frac{\partial R}{\partial \epsilon}\right) \Delta\epsilon \propto \int [A_{DP}^2(t, \mathbf{r}, \Delta t_{12}) - A_D^2(t, \mathbf{r})] e^{-z/\lambda} d^3\mathbf{r}.$

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



Quasi-particle (Fermion) kinetics: gap recovery

$$\delta R = -\frac{2A\Delta_0^2}{\omega^2} n_p.$$



time after quench→

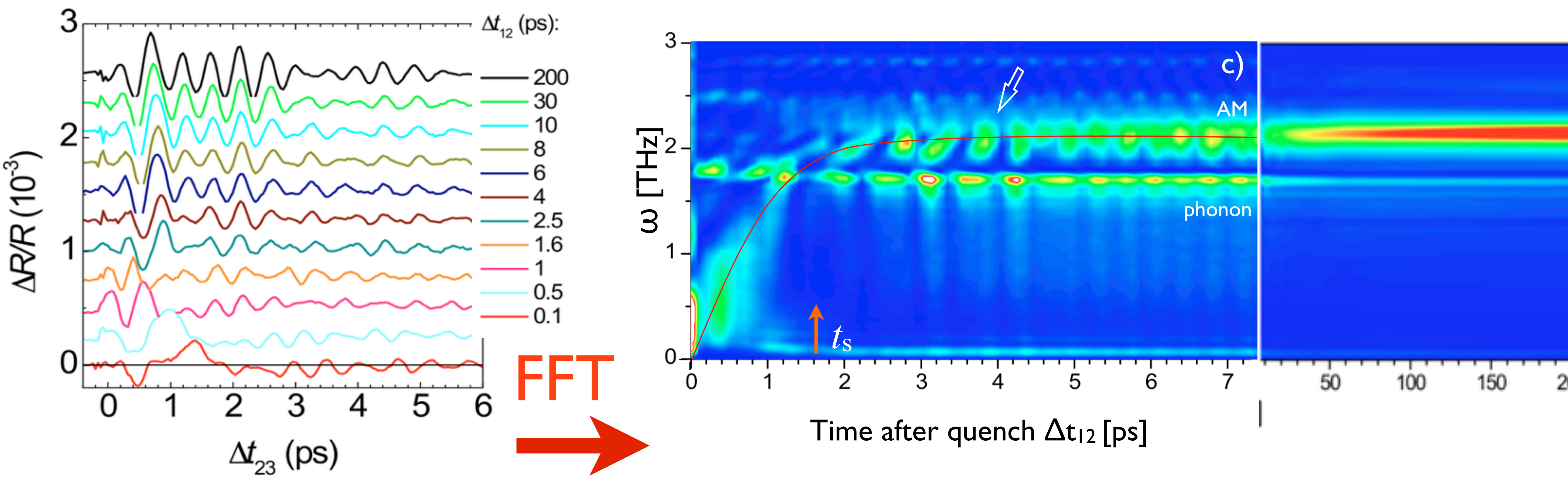
Relaxation
time recovery

$$\tau \propto \frac{1}{\Delta}$$

The time-dependence of ΔR/R during the recovery of ψ:

$$\delta R/R \propto \frac{1/(|\psi(t)| + T(t)/2)}{1 + B\sqrt{2T(t)/|\psi(t)|}\exp[-|\psi(t)|/T(t)]}$$

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



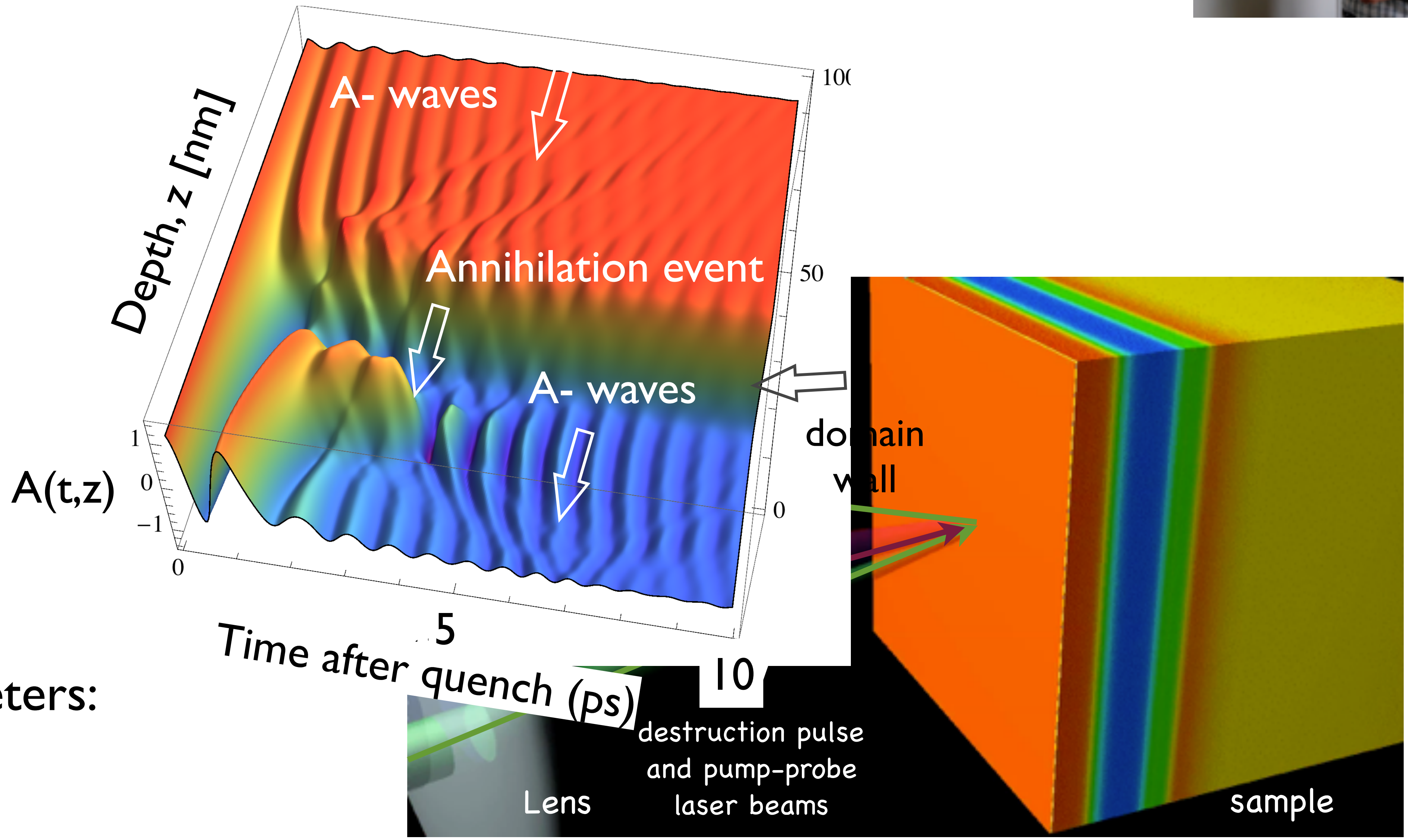
The most obvious feature:
oscillations of intensity of the collective mode

Order parameter calculation

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:

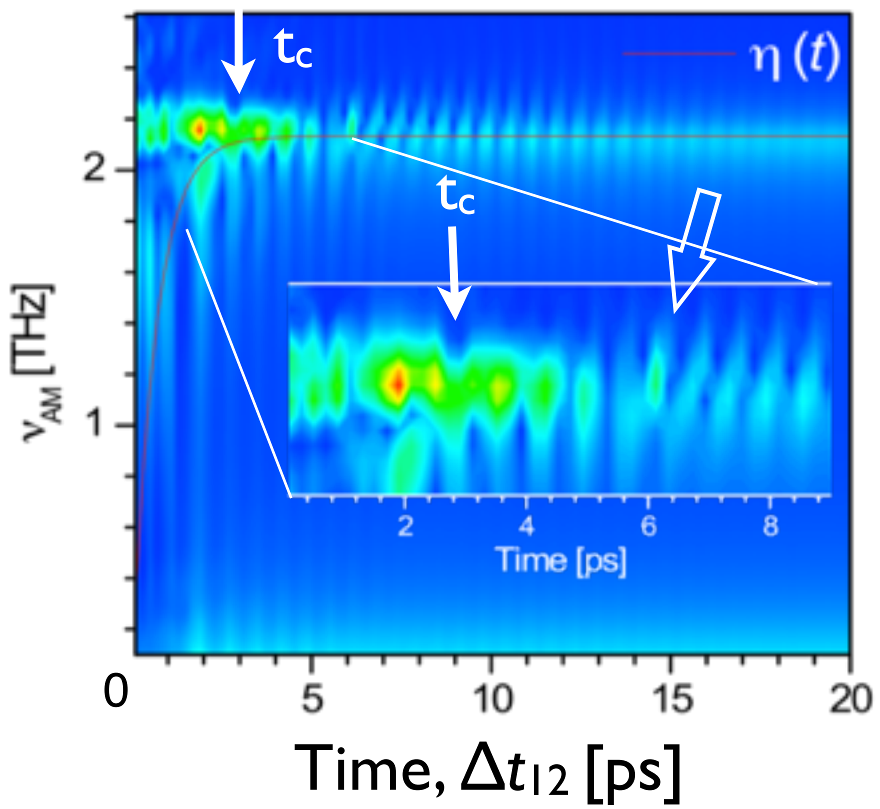


Experimental parameters:

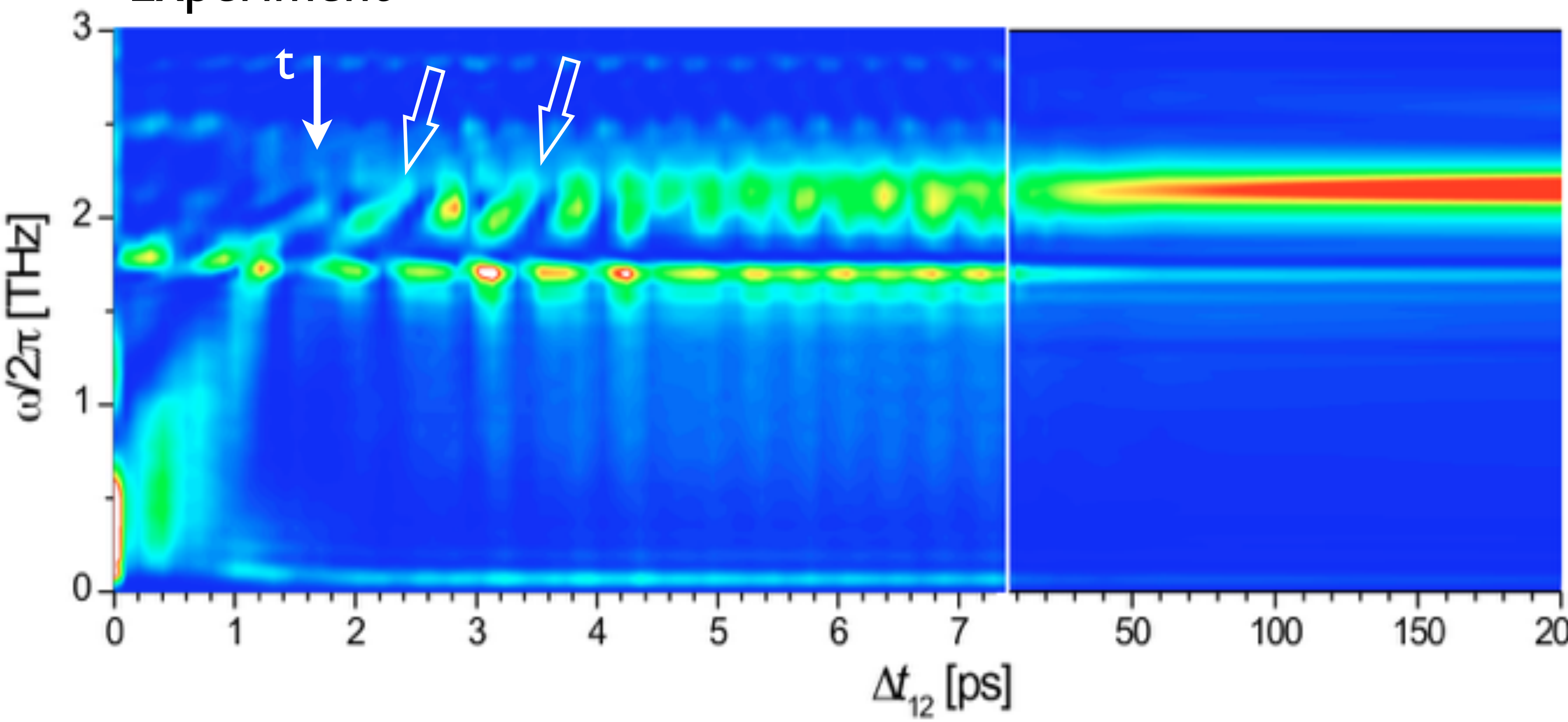
$\tau_{QP} = 650$ fs
$\omega_0/2\pi = 2.18$ THz
$\eta = 2$
$\alpha = 0.1$

Order parameter dynamics: TDGL theory vs. experiment

Theory

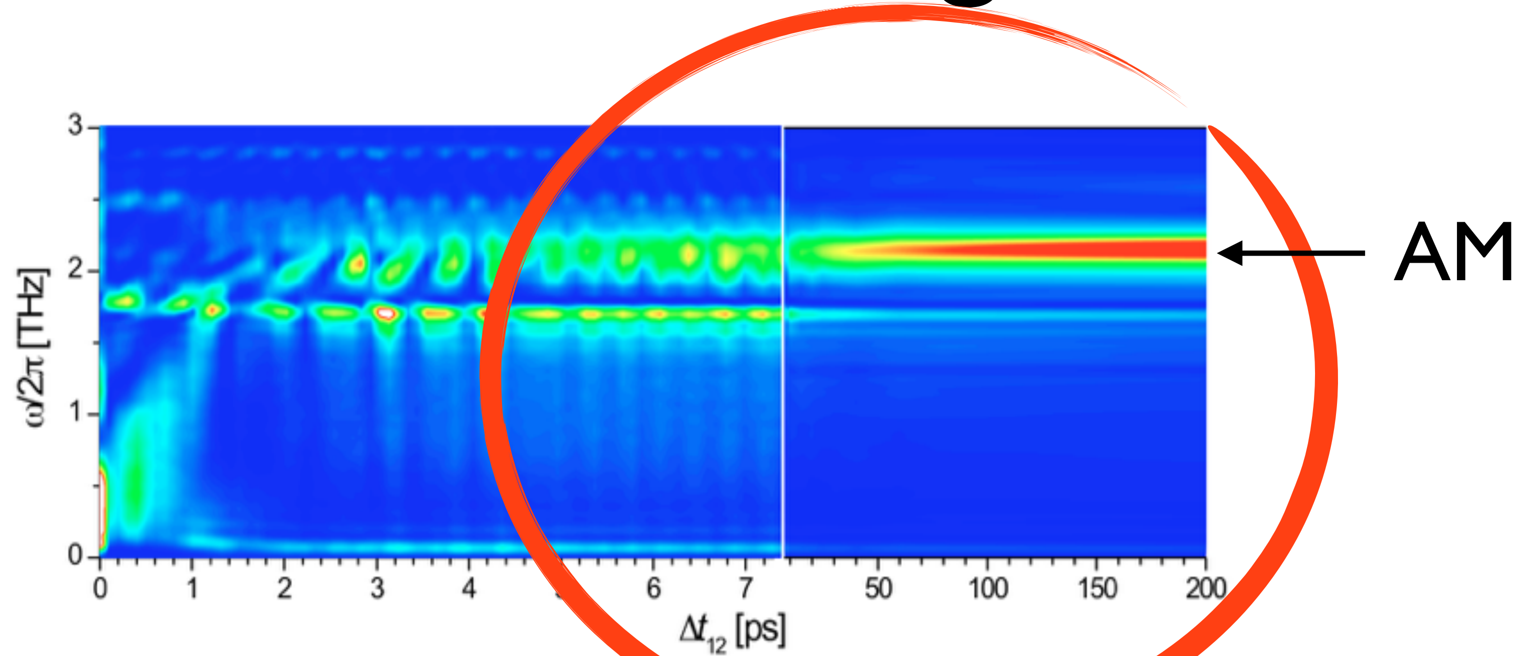


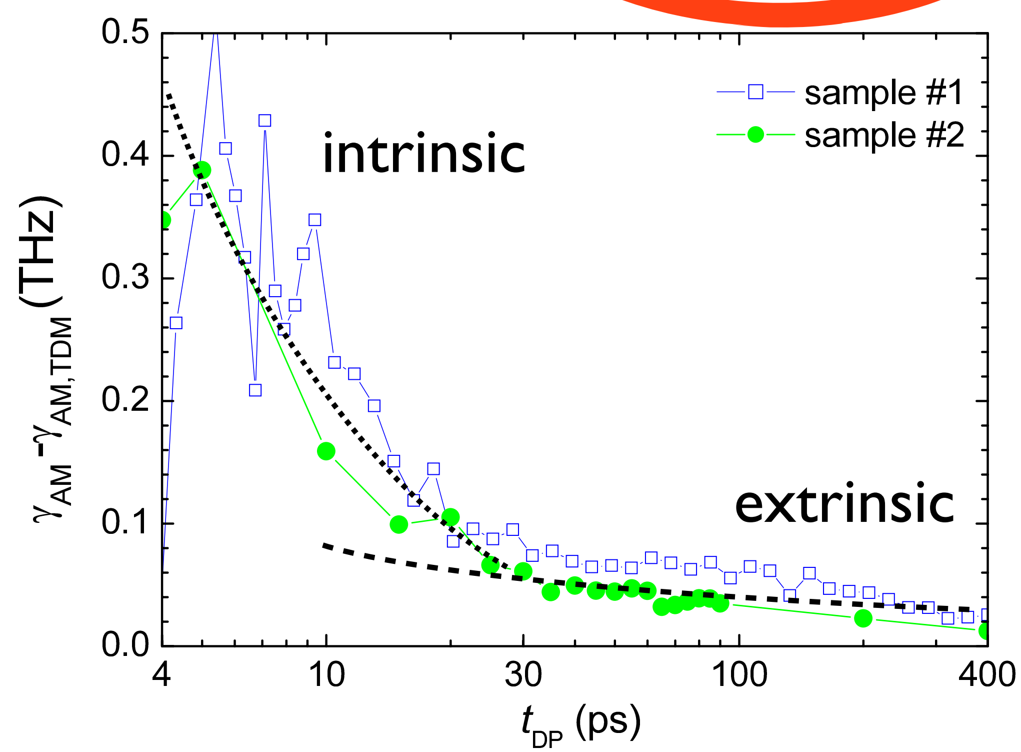
Experiment



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

Incoherent topological defect dynamics: collective mode broadening for Δt_{12} > 7 ps

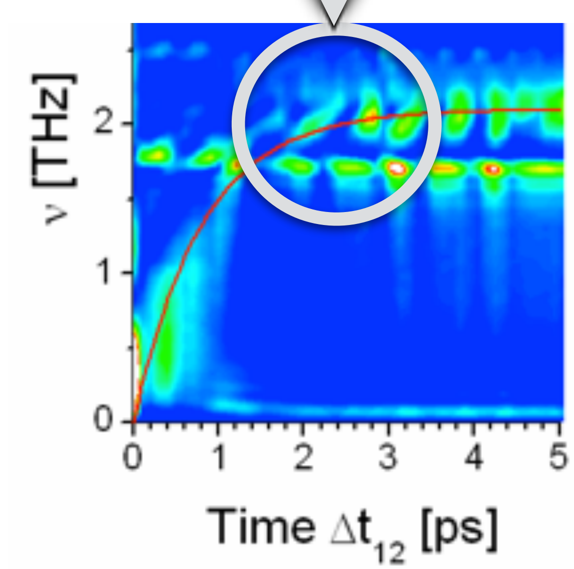




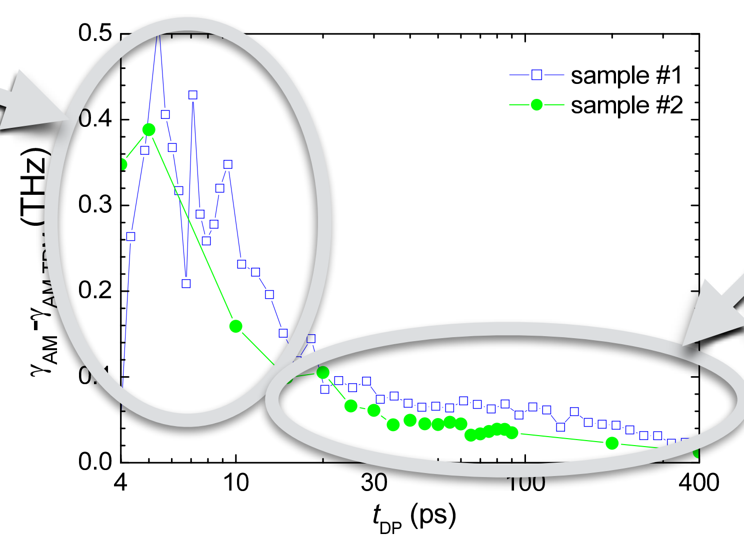
Mertelj, T. *et al.* Incoherent Topological Defect Recombination Dynamics in TbTe_{3}. *Phys Rev Lett* **110,** 156401 (2013).

Topological defect dynamics: timescales

1 ~ 5 ps: Coherent defect annihilation

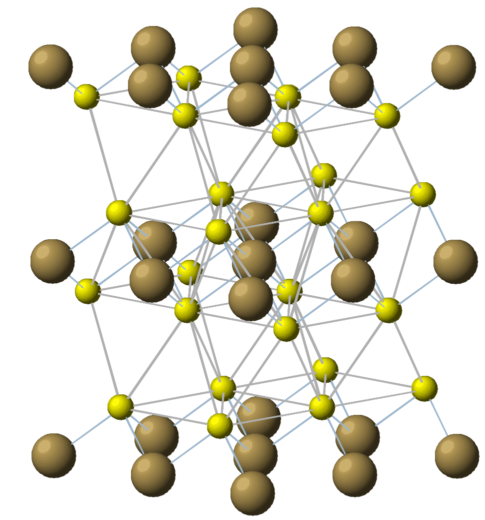


~5-30 ps: incoherent (intrinsic) dynamics:



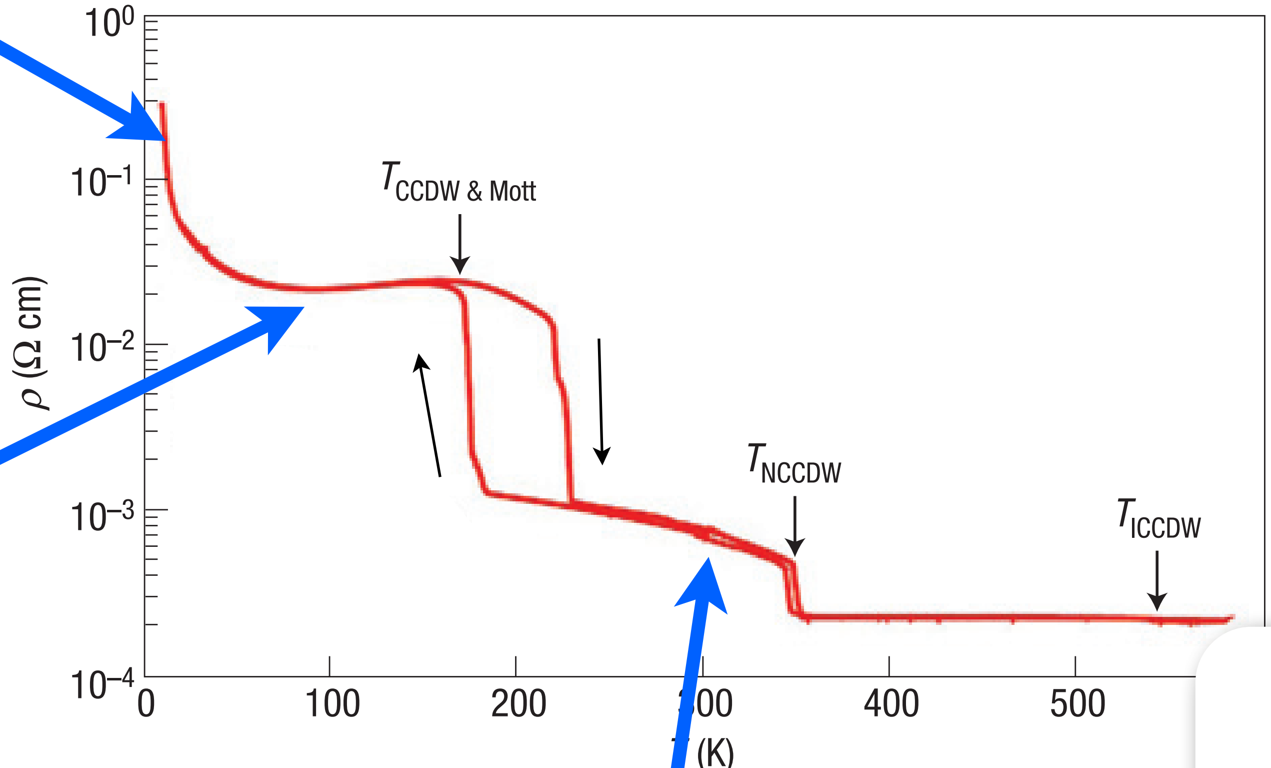
~ 50 ps to > 1 μs: extrinsic defect-related dynamics

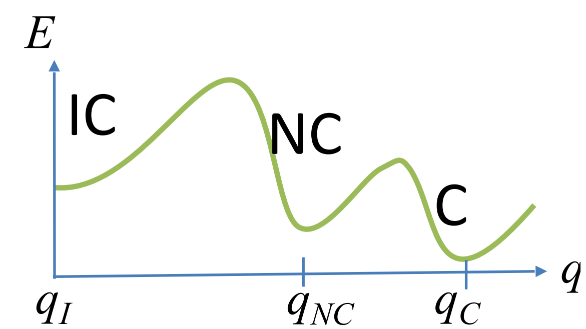
The competing states of $1T$-TaS_2 under equilibrium conditions



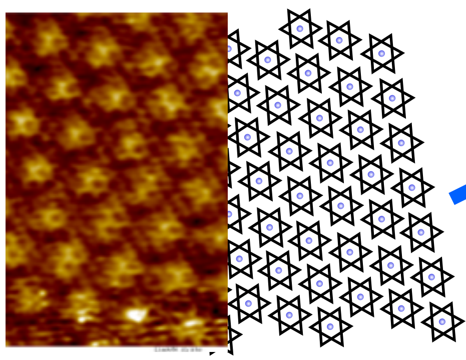
Resistivity of $1T$-TaS_2

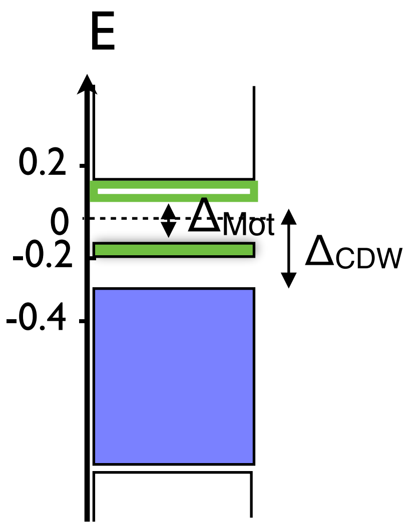
Mott state



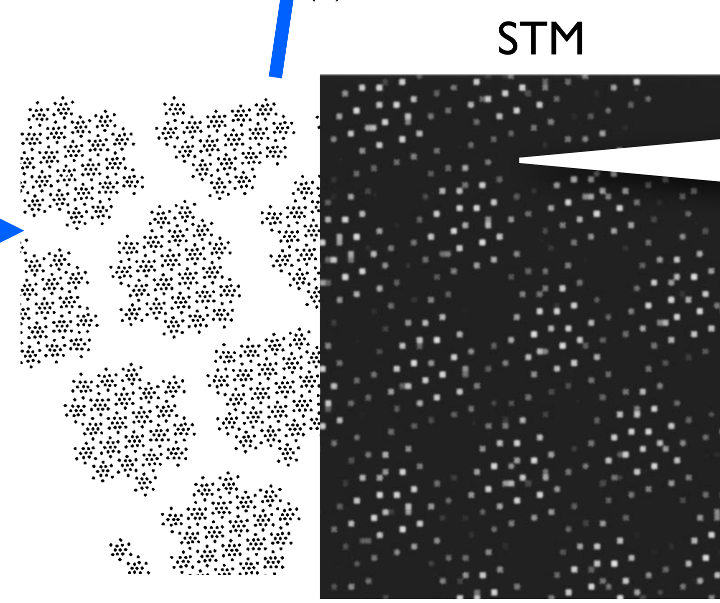


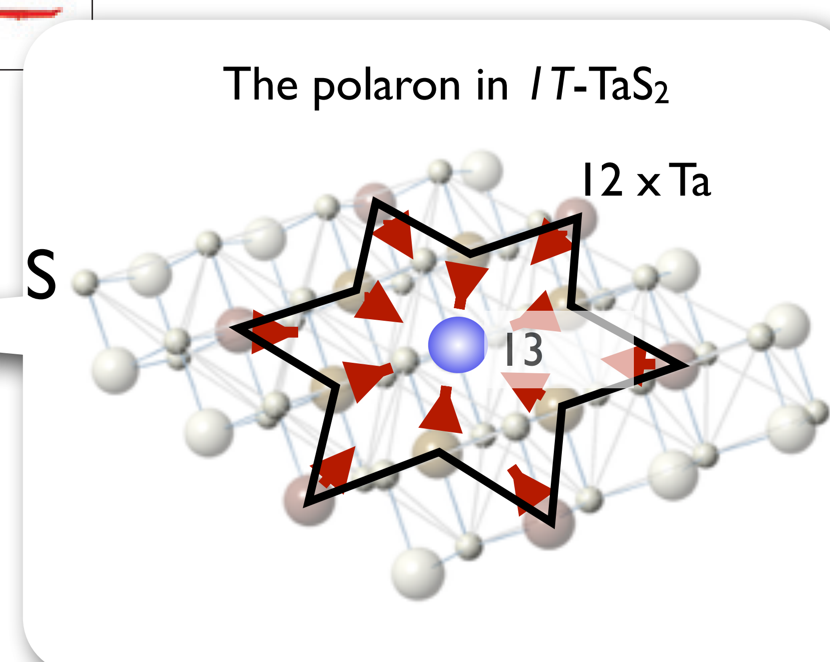
Honeycomb structure
(Commensurate CDW phase)
Tossati and Fazekas (1974)





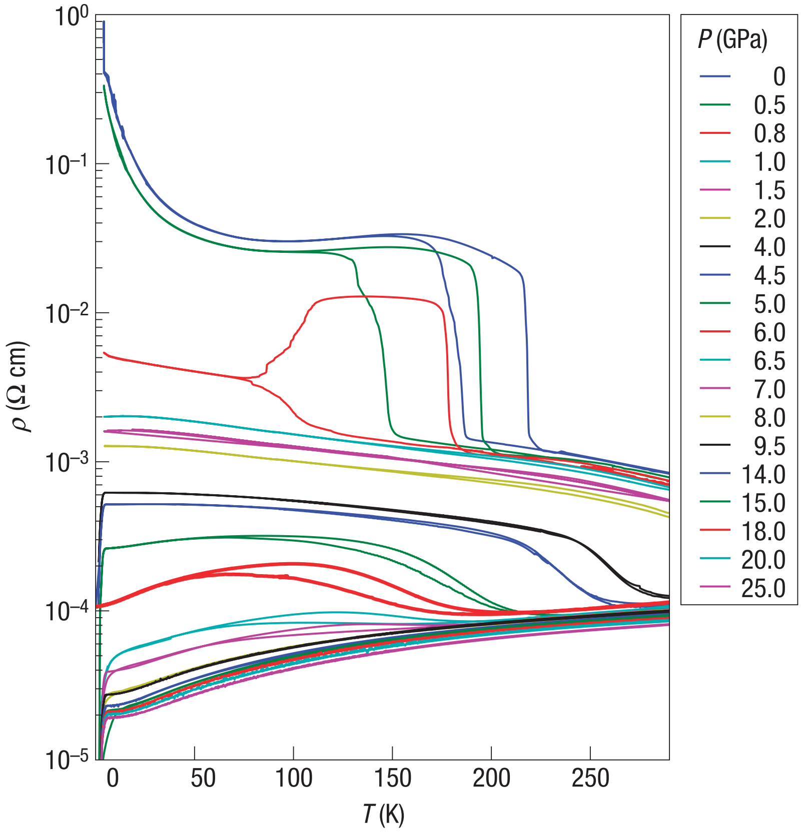
domain walls

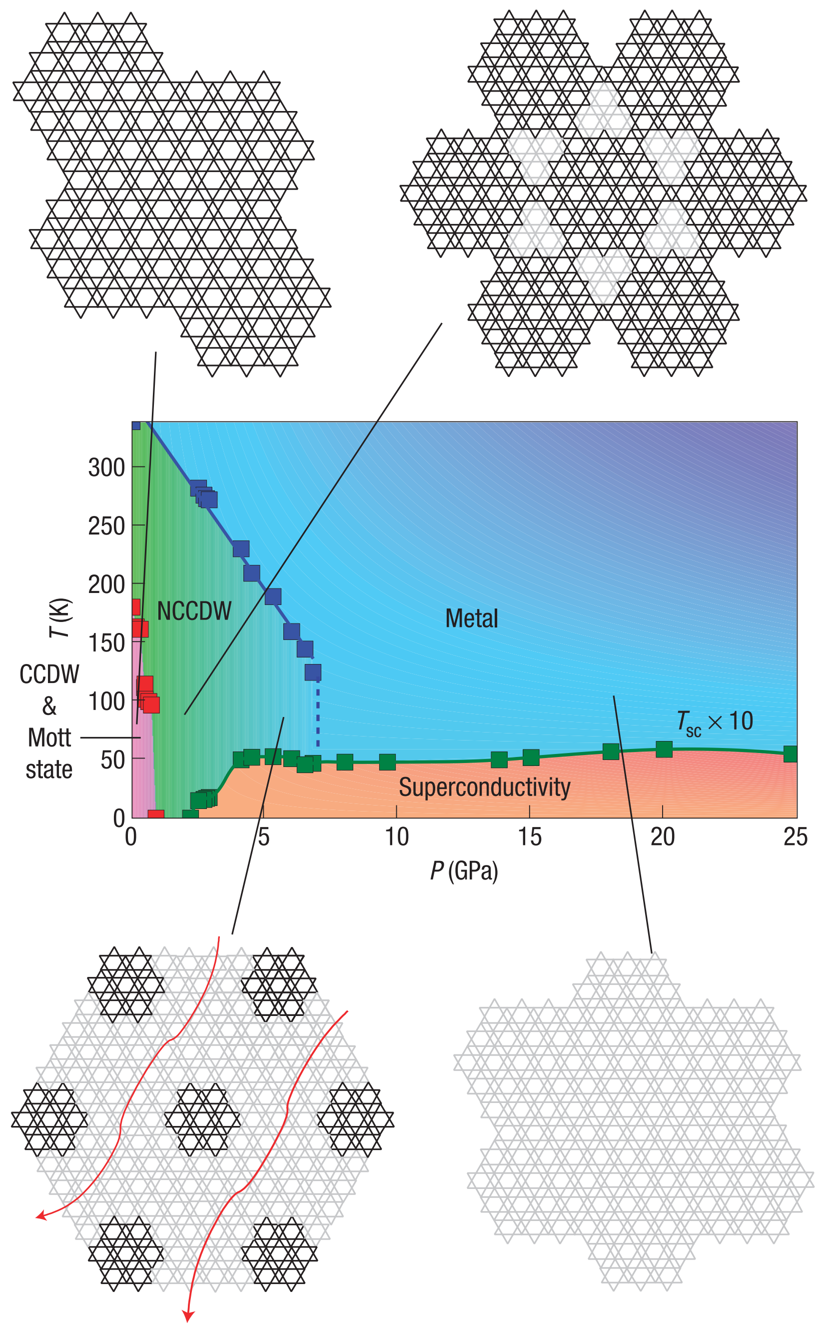




Other nearby states in 1T-TaS_2: Superconductivity under pressure, or Fe, or Se doping etc.

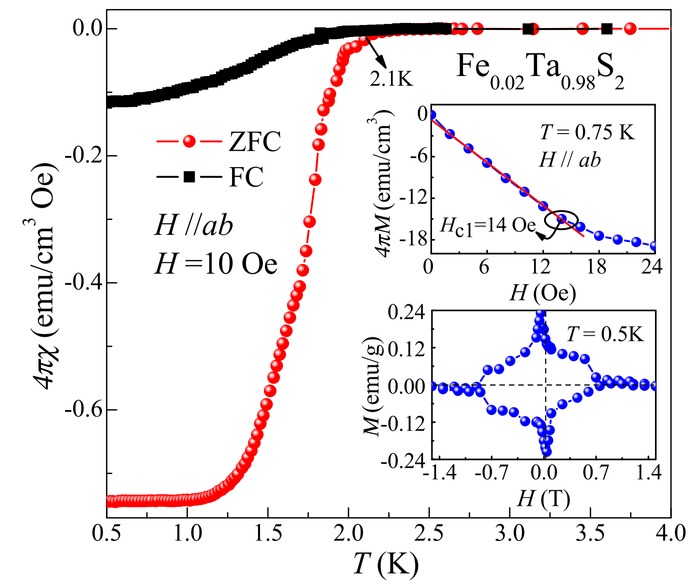
Pressure:

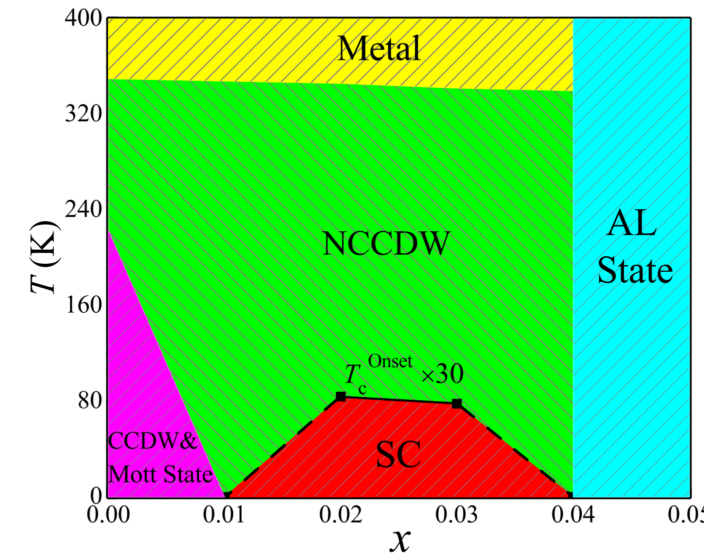




Sipos et al (Nat.Mat. 2008)

Fe doping:



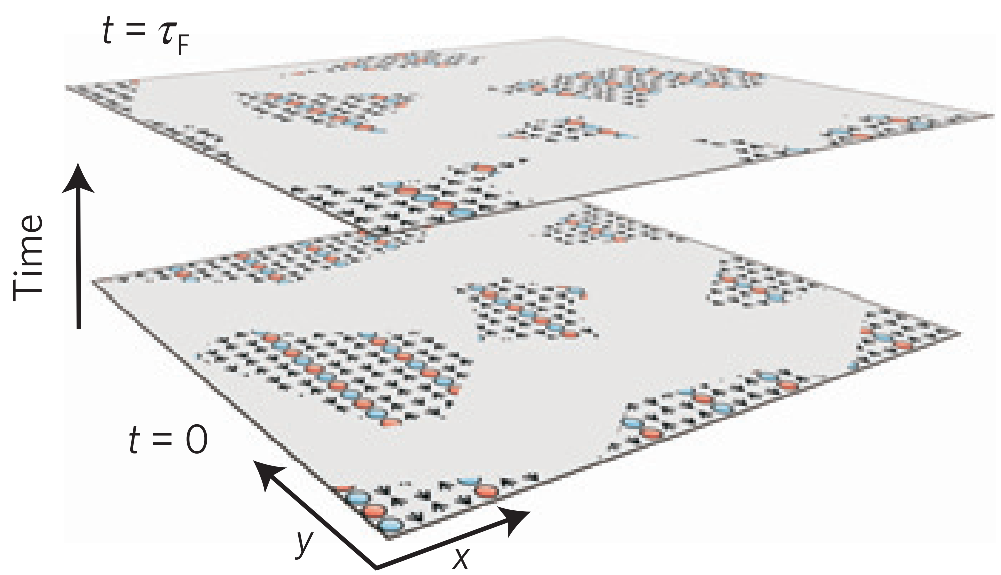


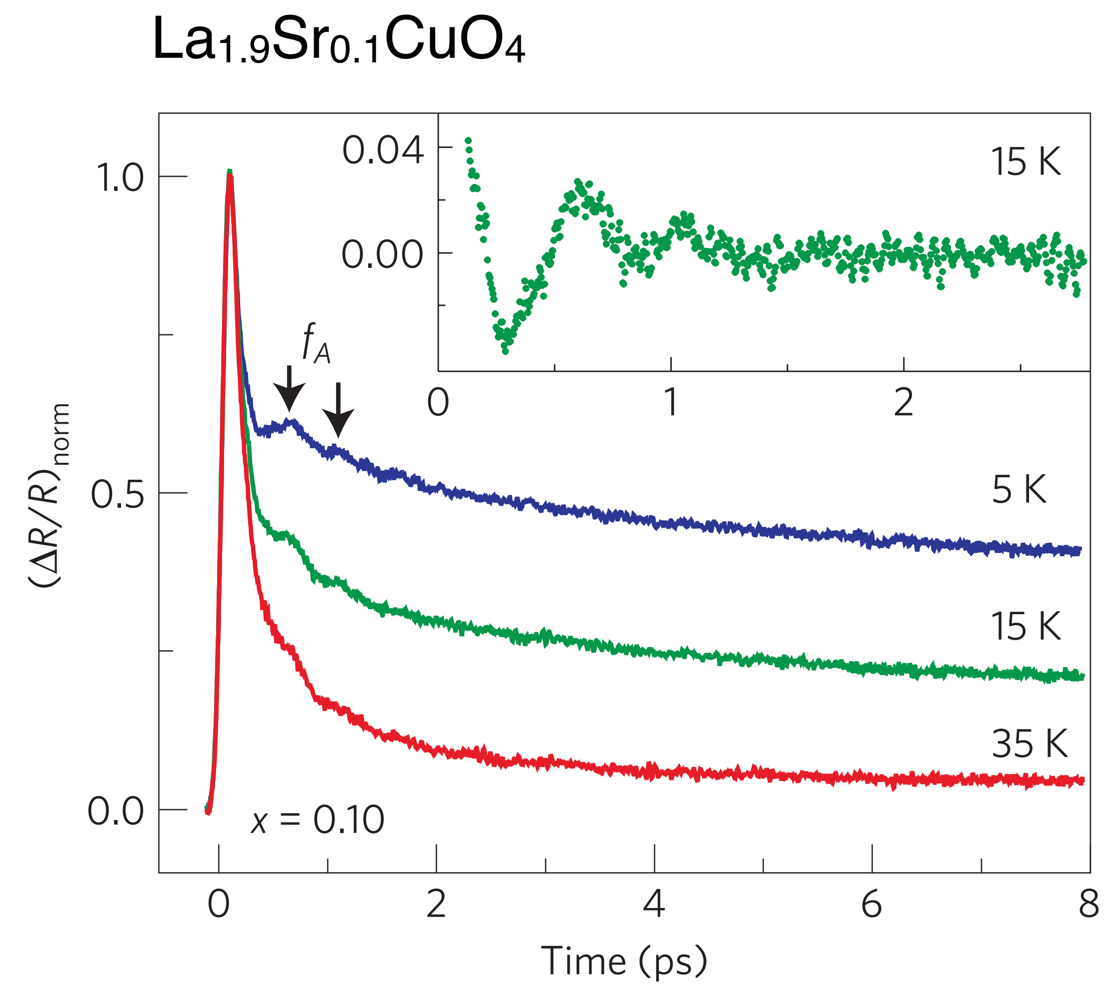
Li et al. EPL 2012

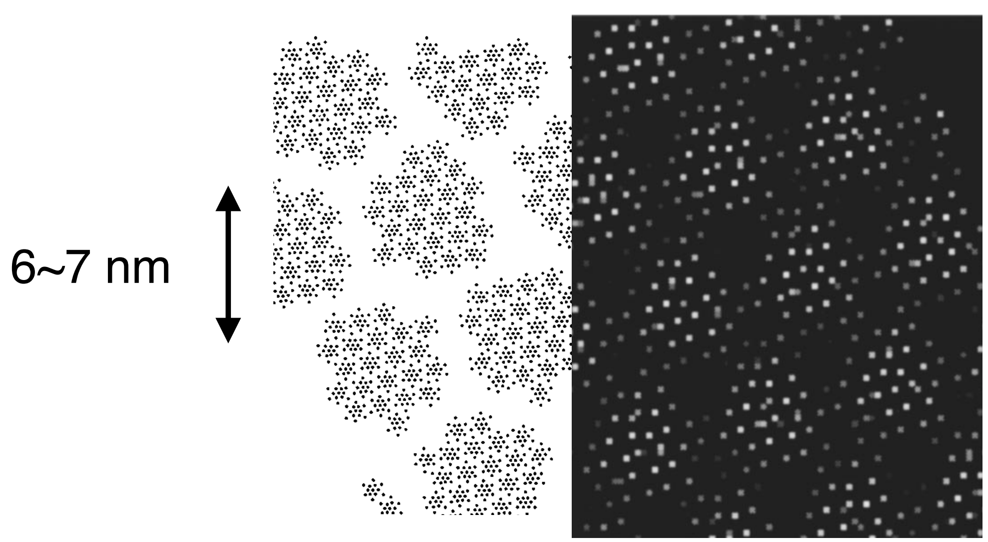
nature materials
LETTERS
PUBLISHED ONLINE: 24 FEBRUARY 2013 | DOI: 10.1038/NMAT3571

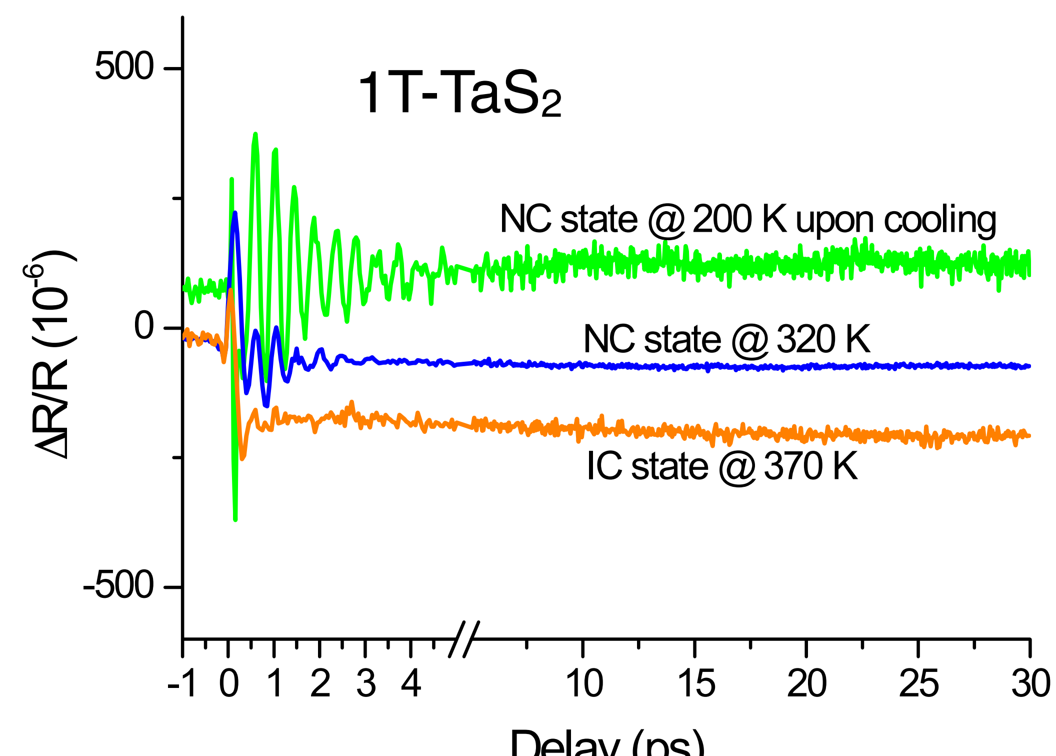
Fluctuating charge-density waves in a cuprate superconductor

Darius H. Torchinsky[1†], Fahad Mahmood[1†], Anthony T. Bollinger[2], Ivan Božović[2] and Nuh Gedik[1★]

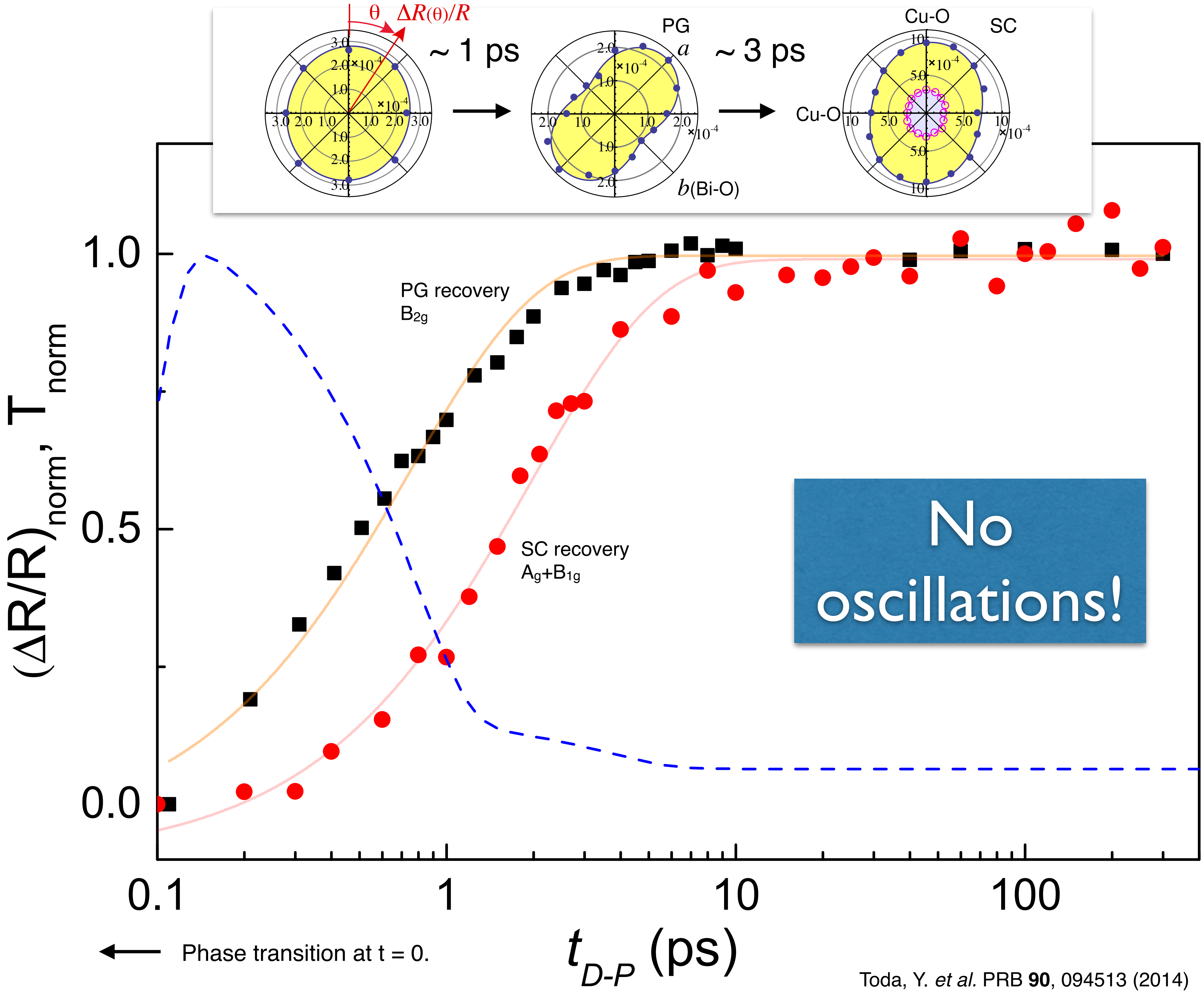








Emergence of broken symmetry in real time BiSCO-2212-OP

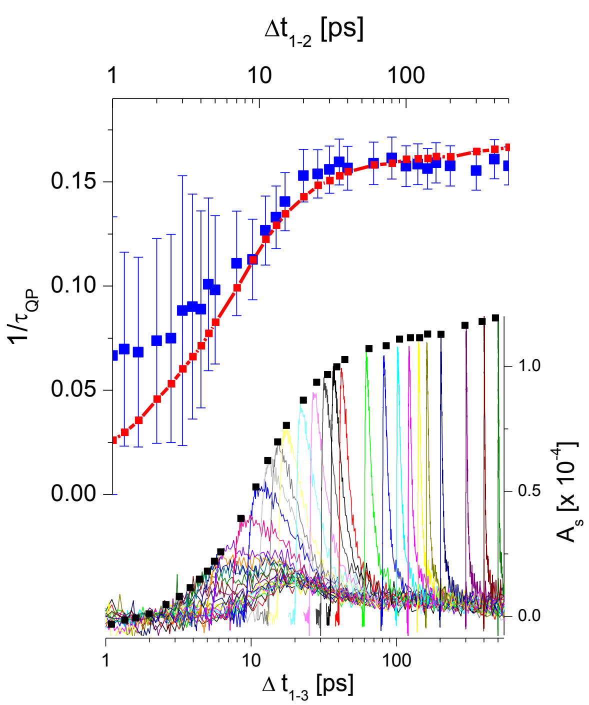


← Phase transition at t = 0.

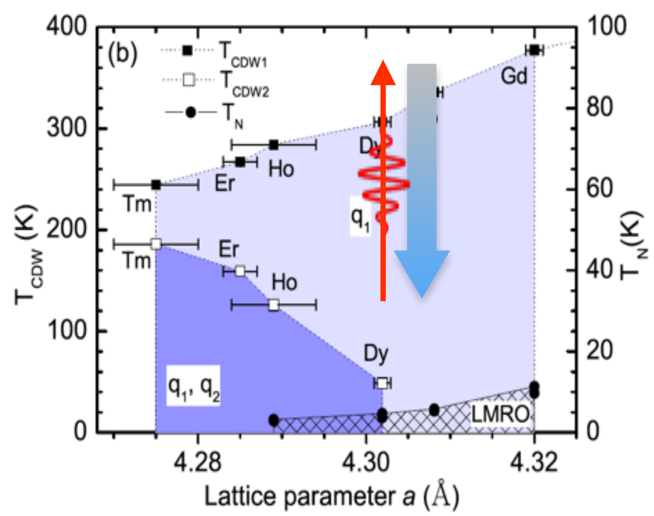
Toda, Y. *et al.* PRB **90**, 094513 (2014)

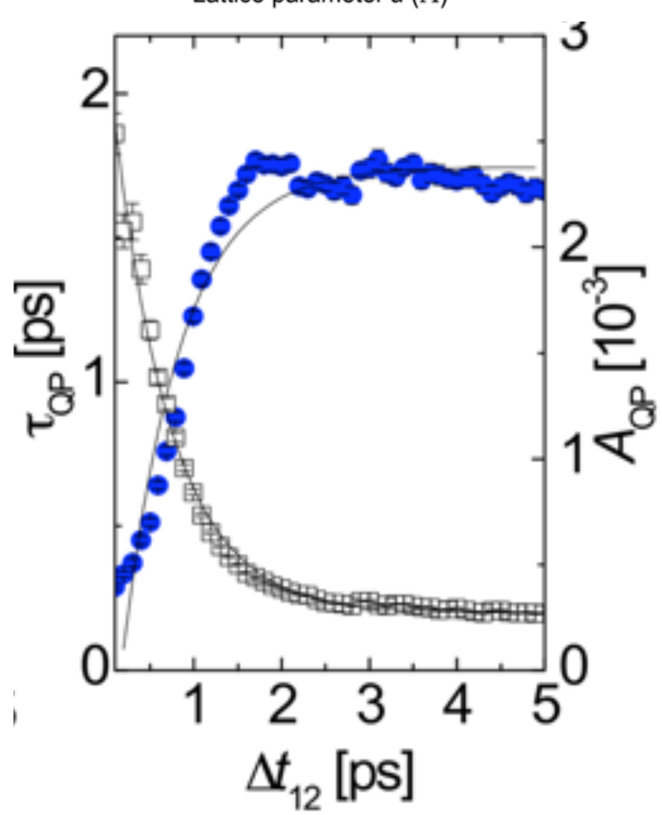
Toda, Y. *et al.* PRB **84**, 174516 (2011).

The evolution of SP lifetimes through the “transition”

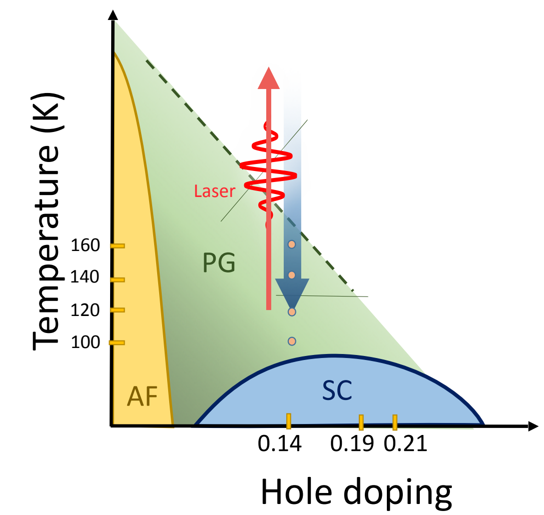


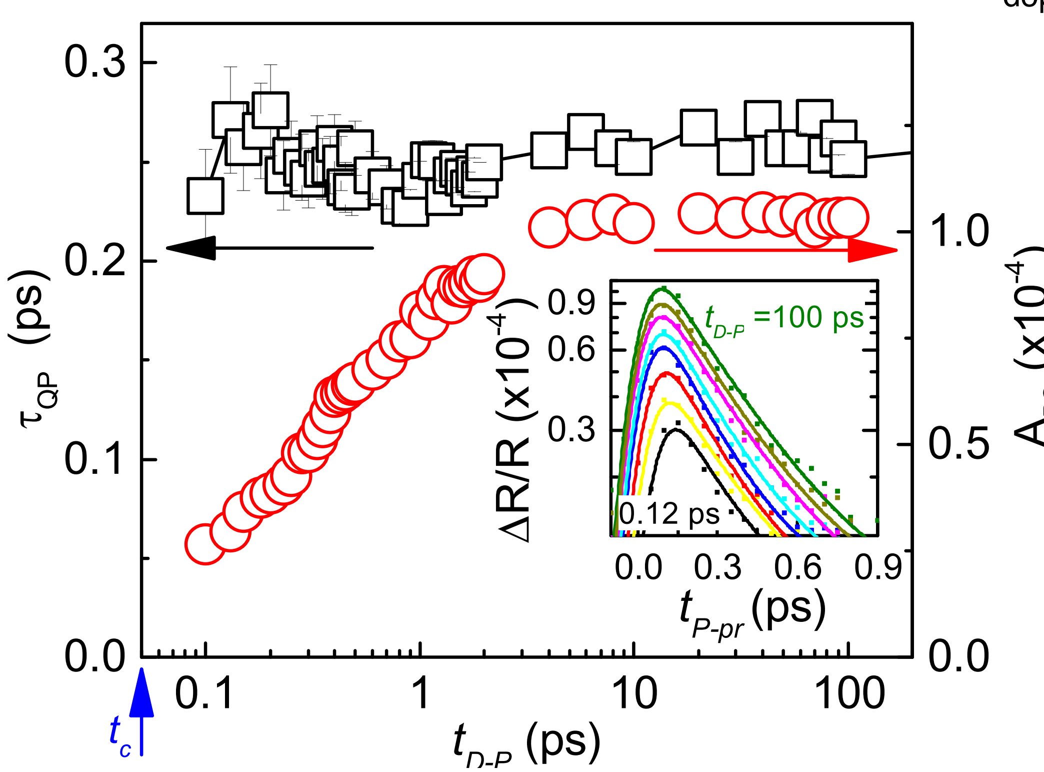
CDW transition in TbTe3





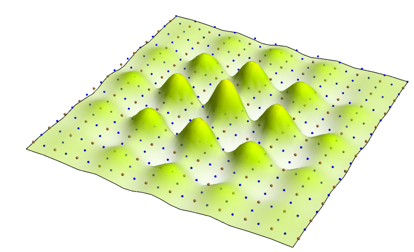
PG state in Bi2212 UD





$$\xi = \sqrt{\frac{\hbar^2}{2m|\alpha|}},$$

<3~5 nm



D_{2h} (or C_{2h})

Friedel oscillation around dopant ion sites?

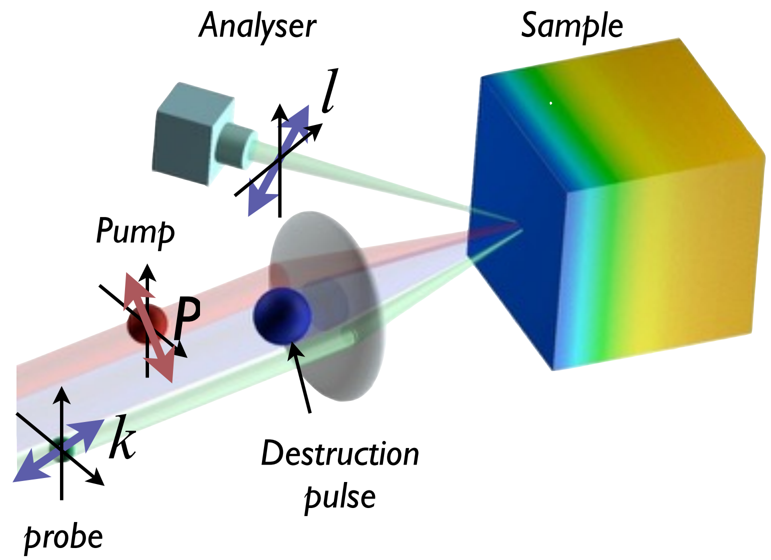
arXiv **cond-mat.str-el,** (2014).

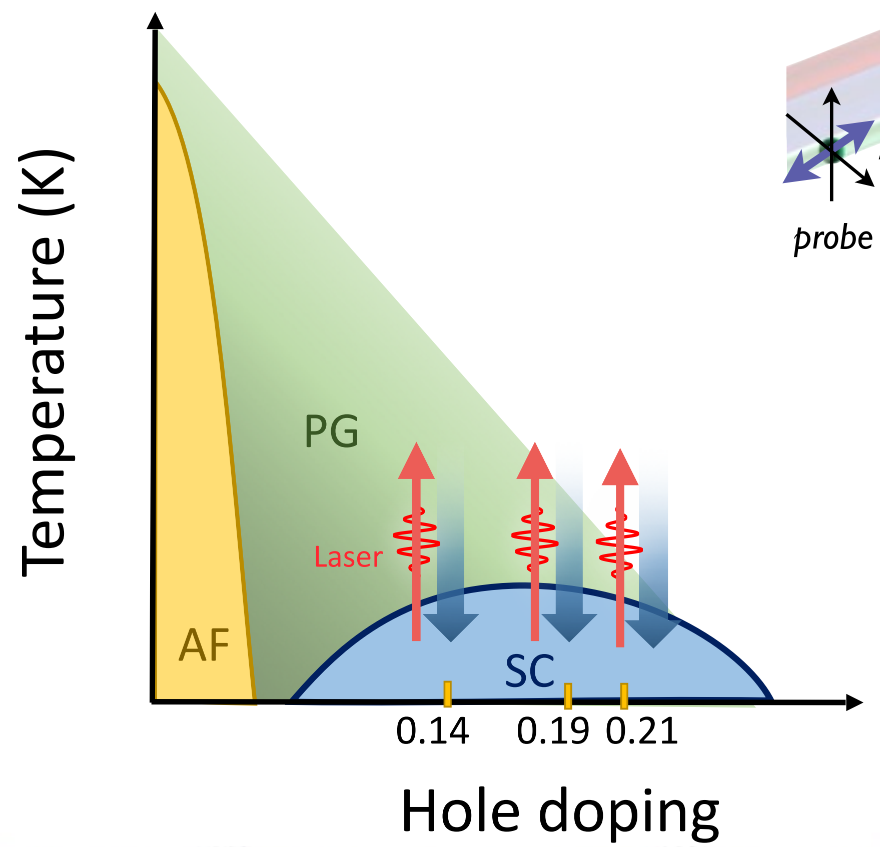
Torchinsky, D. H., Mahmood, F., Bollinger, A. T., Božović, I. & Gedik, N. Fluctuating charge-density waves in a cuprate superconductor. *Nat. Mater.* **12**, 387–91 (2013).

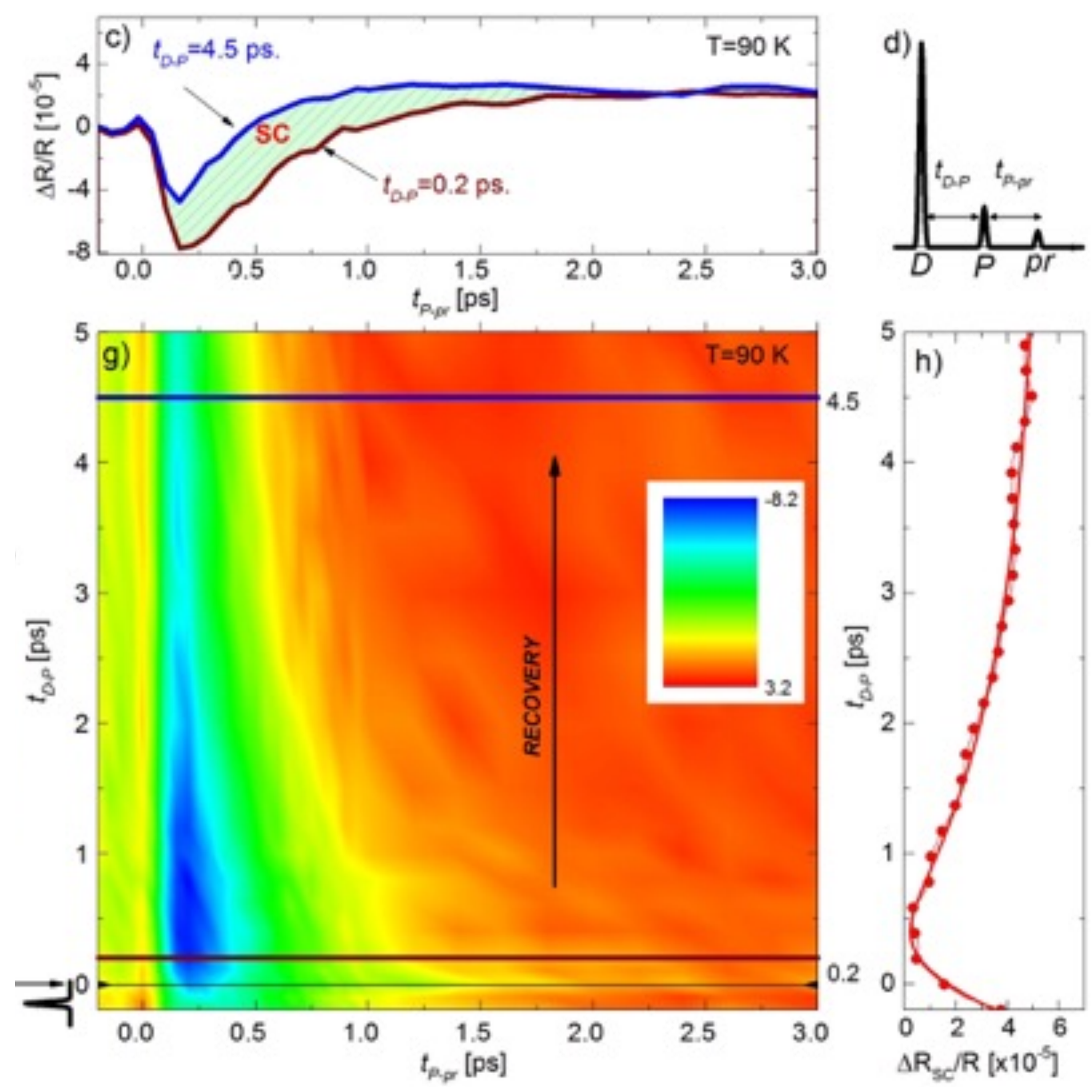
Separating pairing from quantum phase coherence dynamics above the superconducting transition by femtosecond spectroscopy

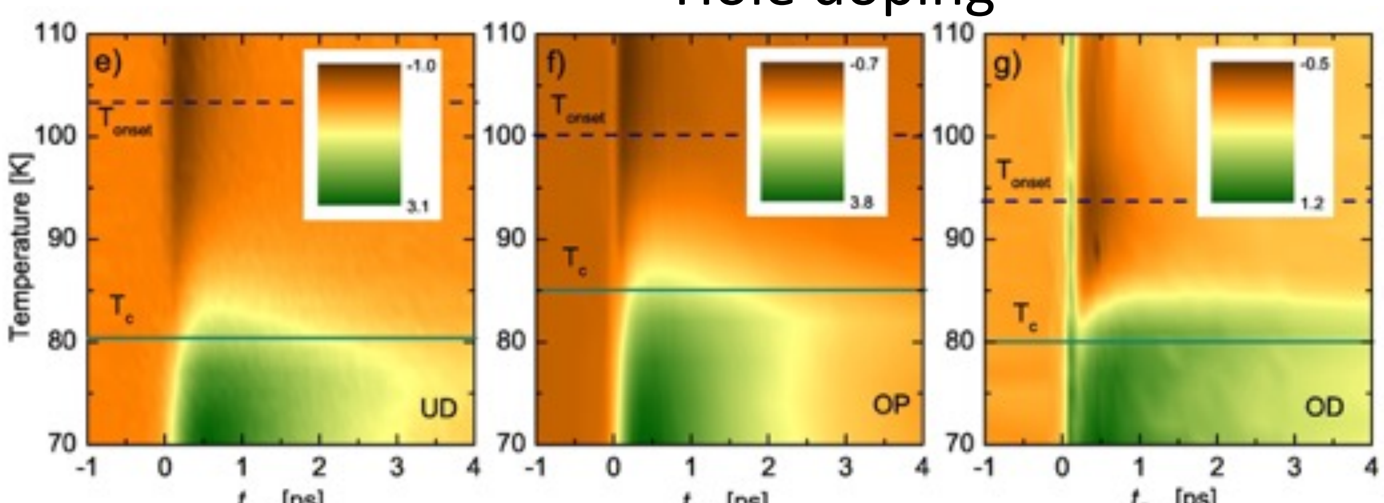
I. Madan[1], T. Kurosawa[2], Y. Toda[3], M. Oda[2], T. Mertelj[1], P. Kusar[1] & D. Mihailovic[1]

Madan et al., *Scientific Reports* **4**, 5656 (2014)



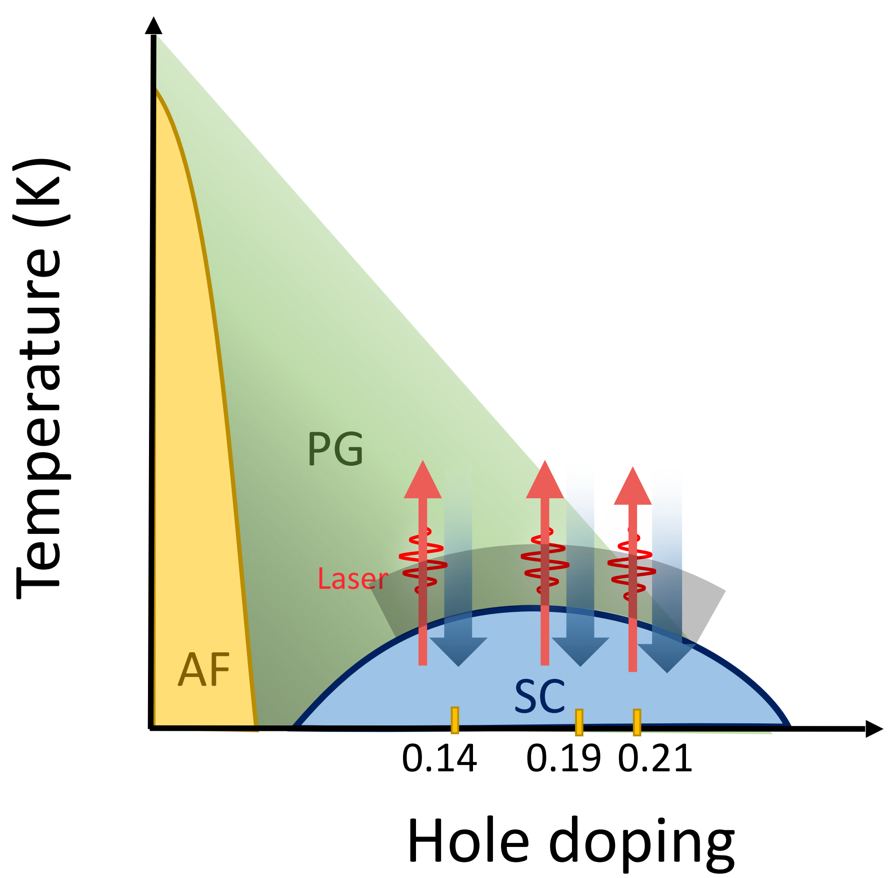


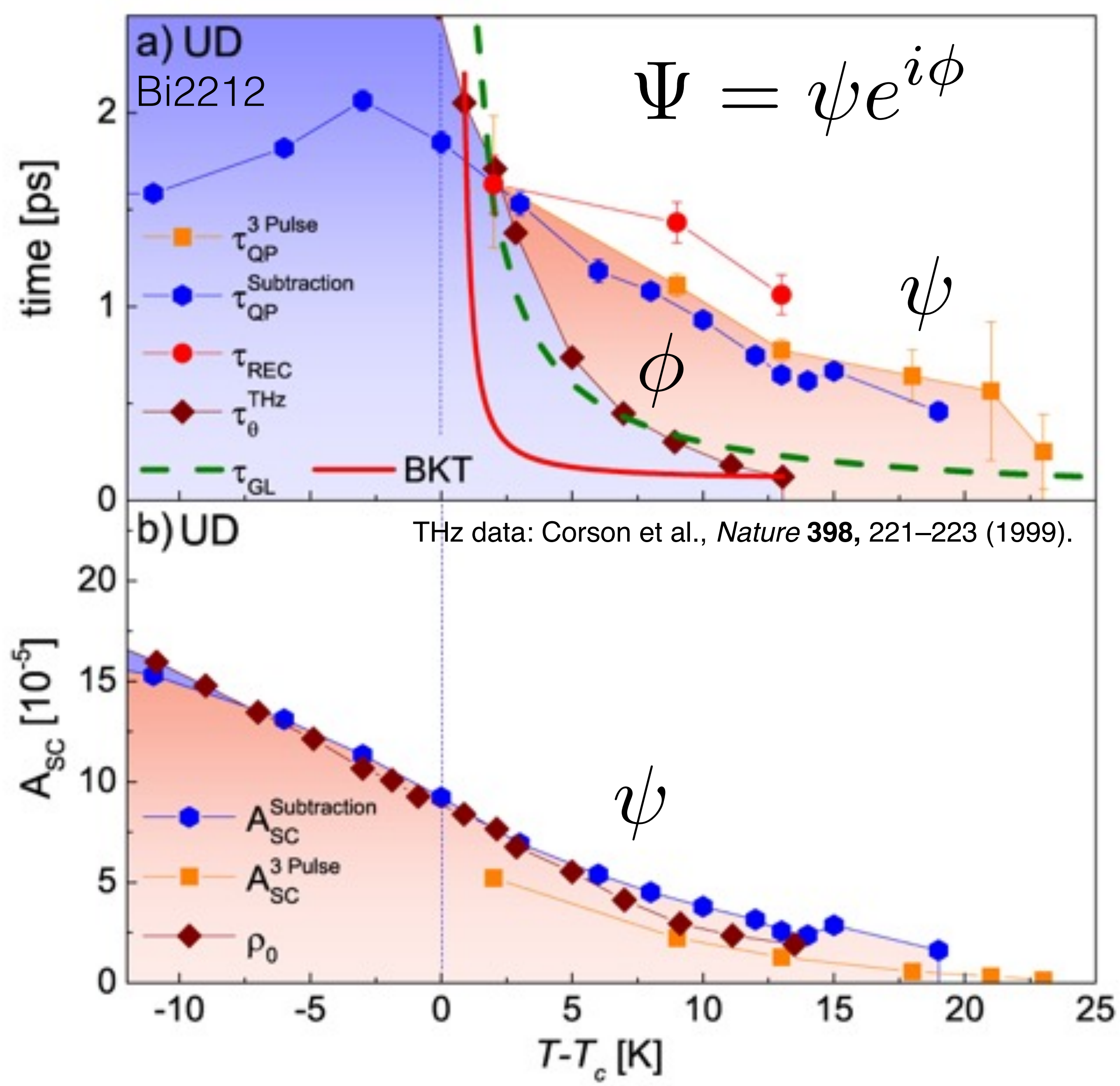




Phase coherence and SC gap formation are distinct processes which occur on different timescales.

Madan et al., *Scientific Reports* **4**, 5656 (2014)

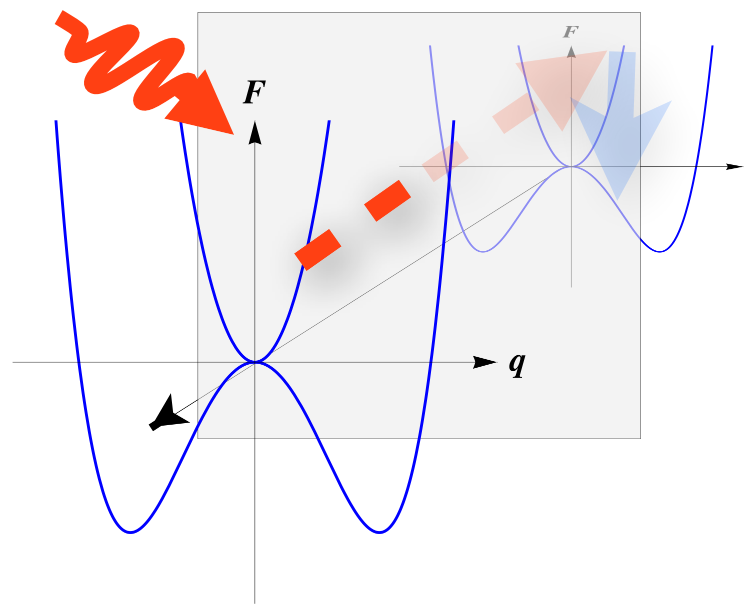




The SC gap persists to ~1.25 T_c on a femtosecond timescale

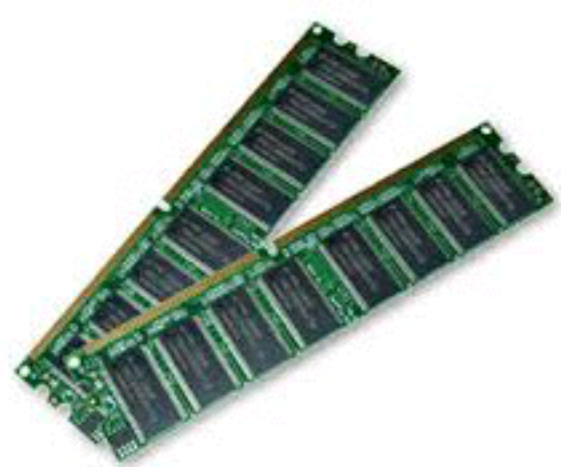
Conclusions

- nanoscale physics unavoidably involves topological defects in cuprates and dichalcogenides
- the PG state is a result of carrier localisation, not a proper CDW phase.
- phase coherence and pairing gap are separable in time-domain (distinct from the PG)



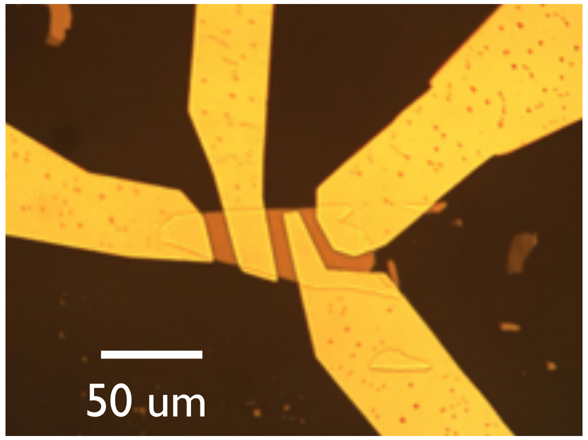
Ultrafast memory

Switching to a hidden topologically protected state

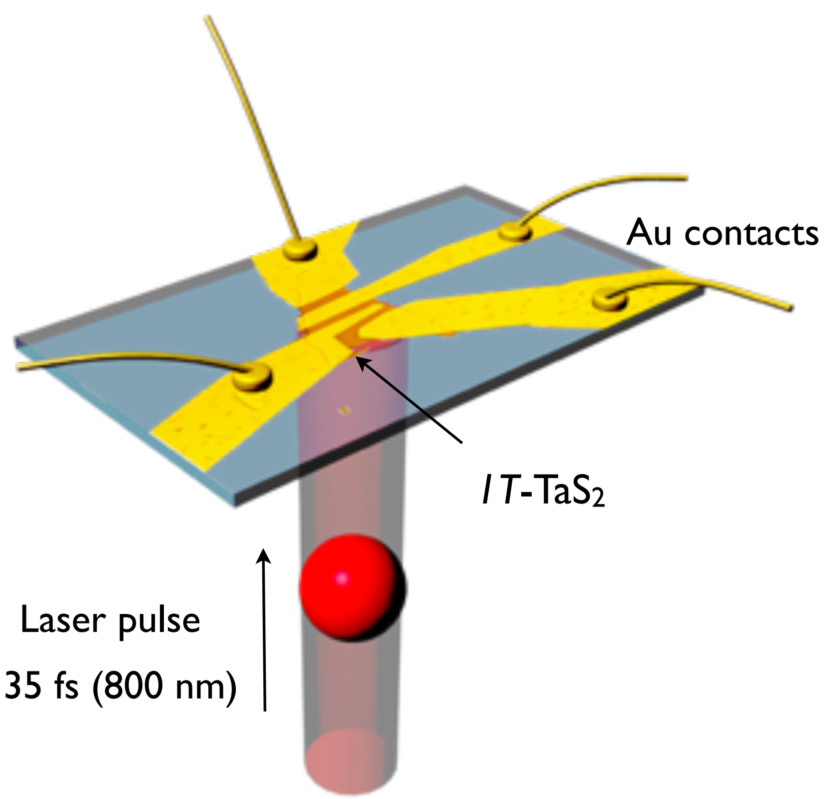


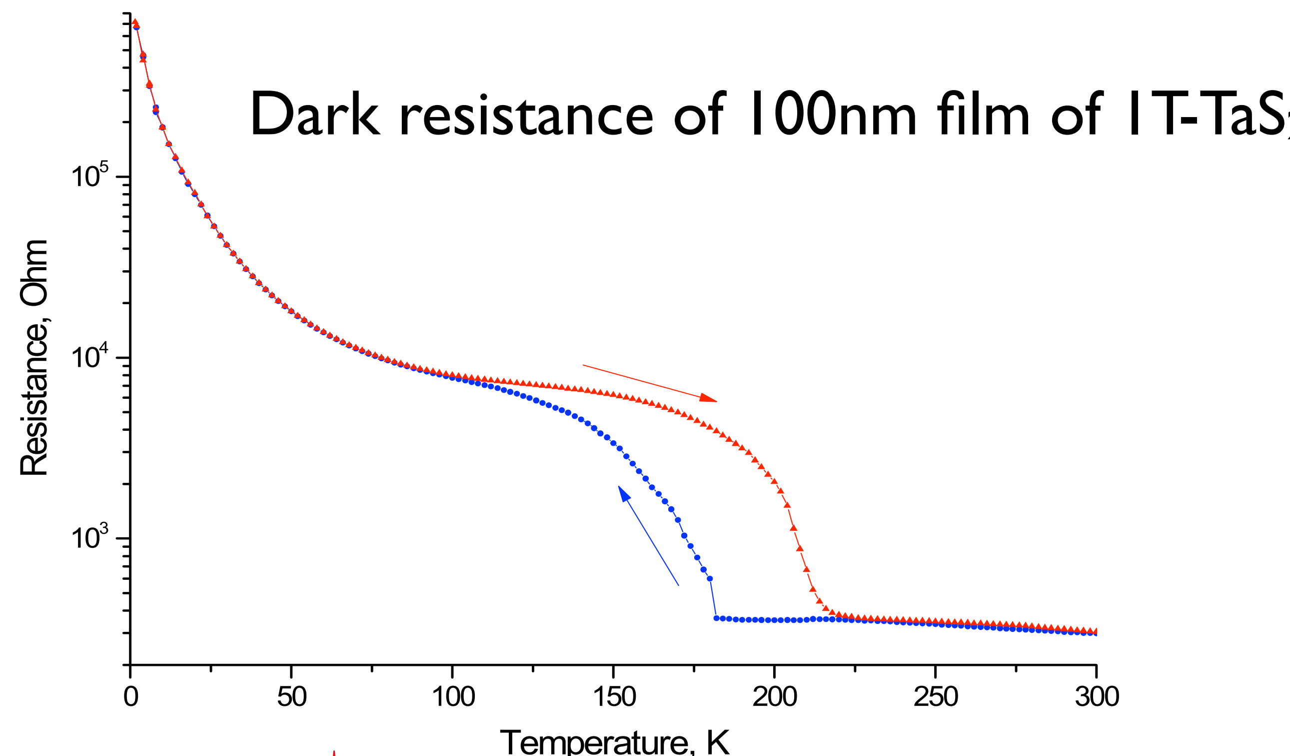
Switching to a hidden state in $1T$-TaS_2 : Resistance change after a (single) 35 fs pulse

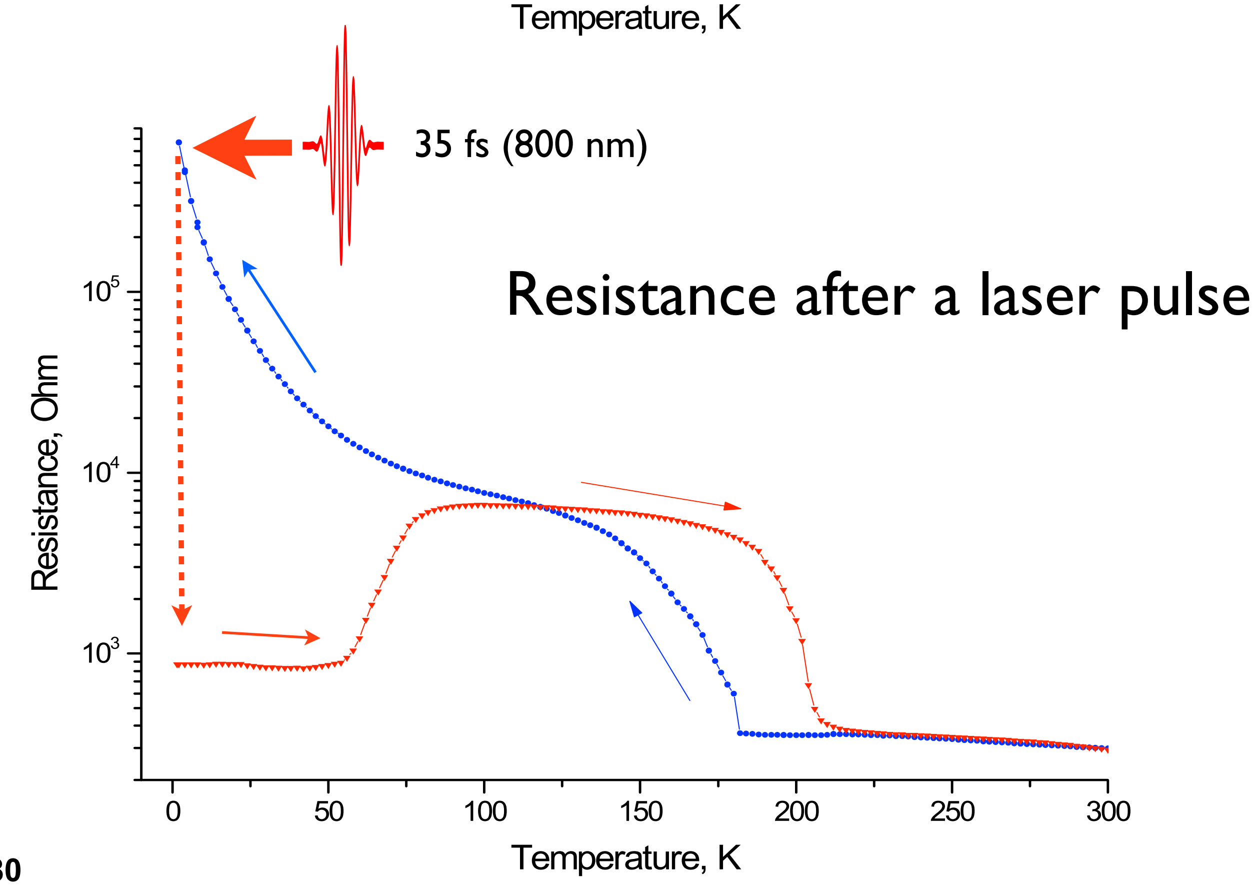
Igor Vaskivskyi



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

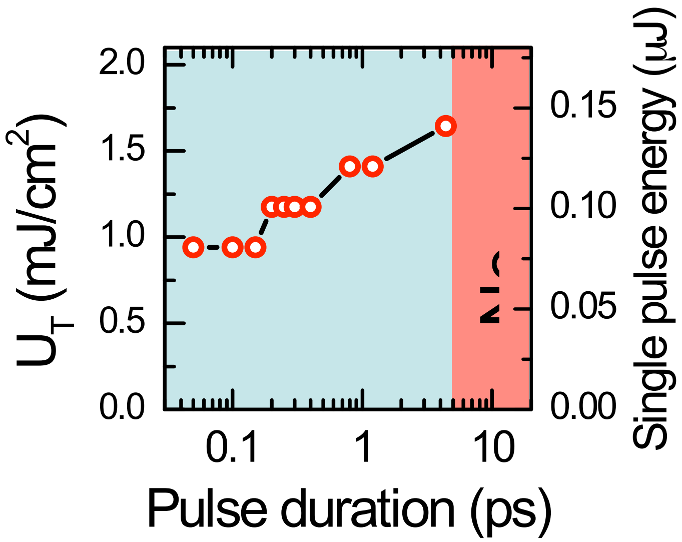


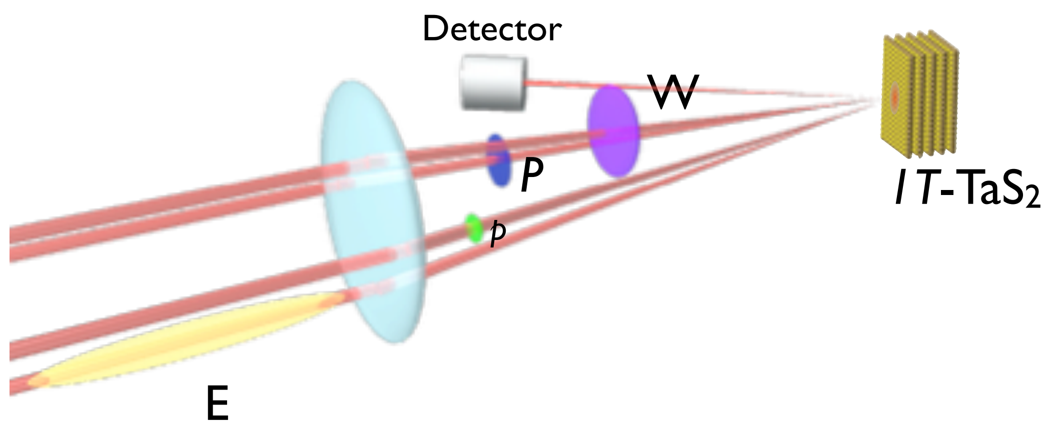




L Stojchevska et al. Science 2014;344:177-180

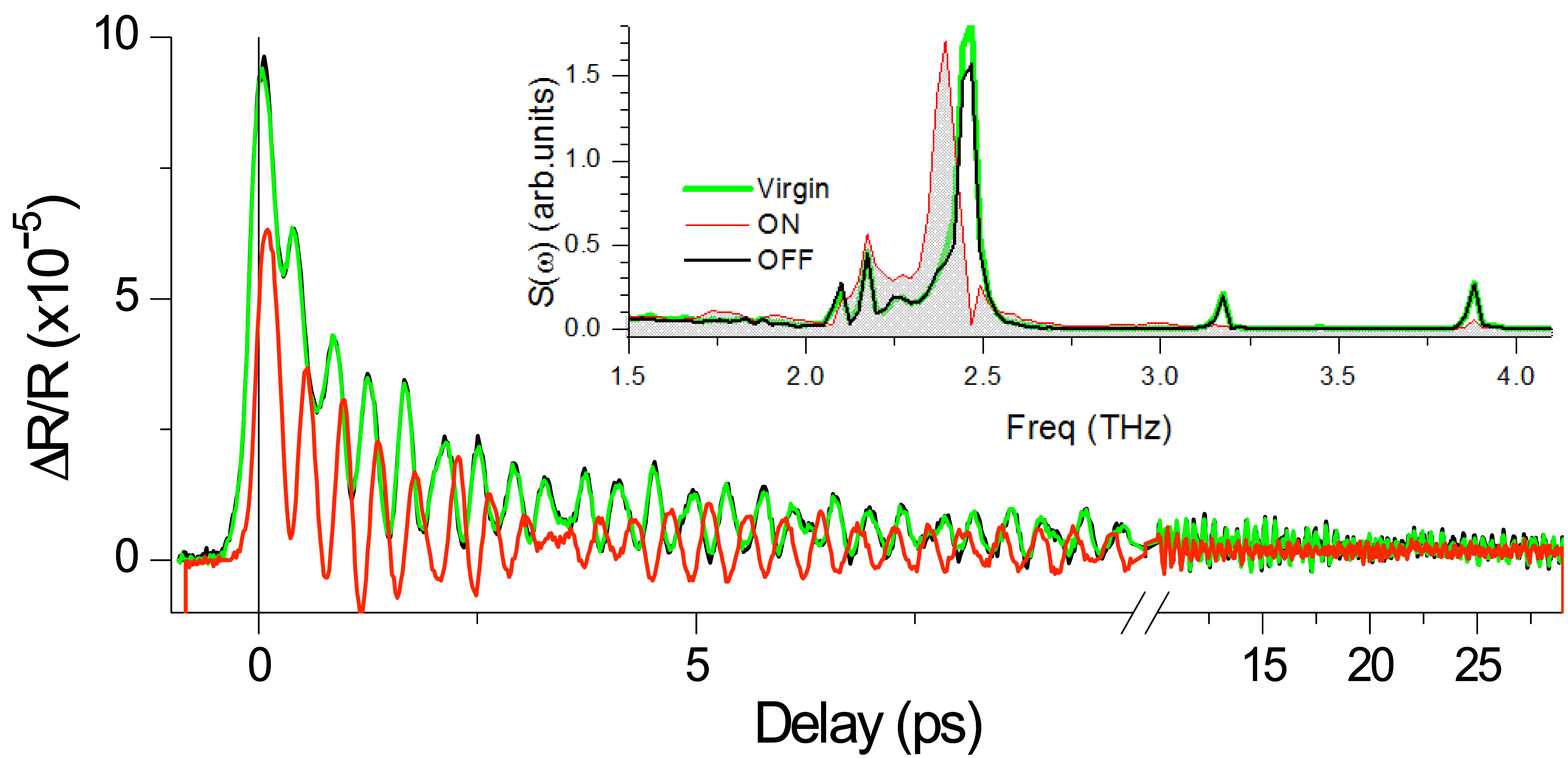
1 T-TaS_2: Collective mode switching





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

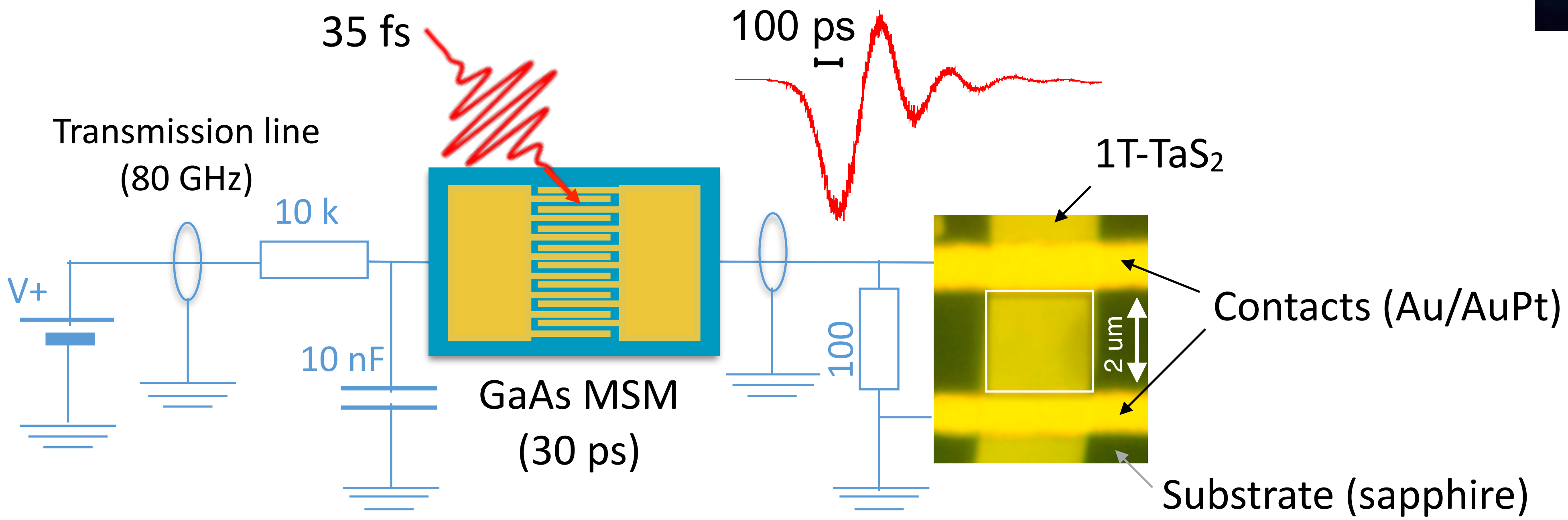
Ljupka Stojchevska

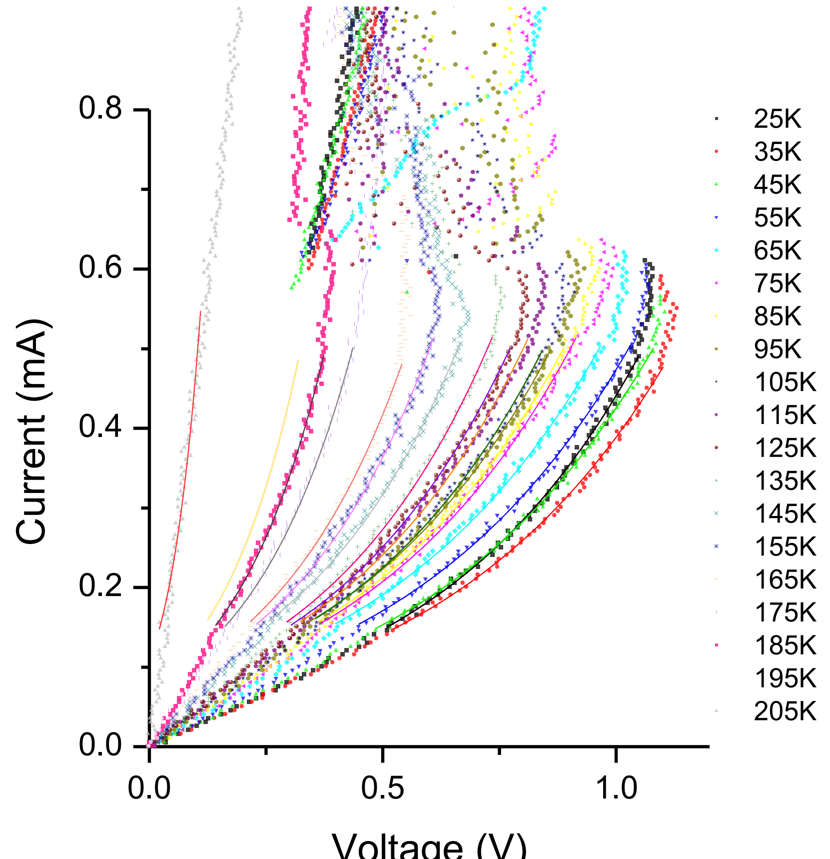


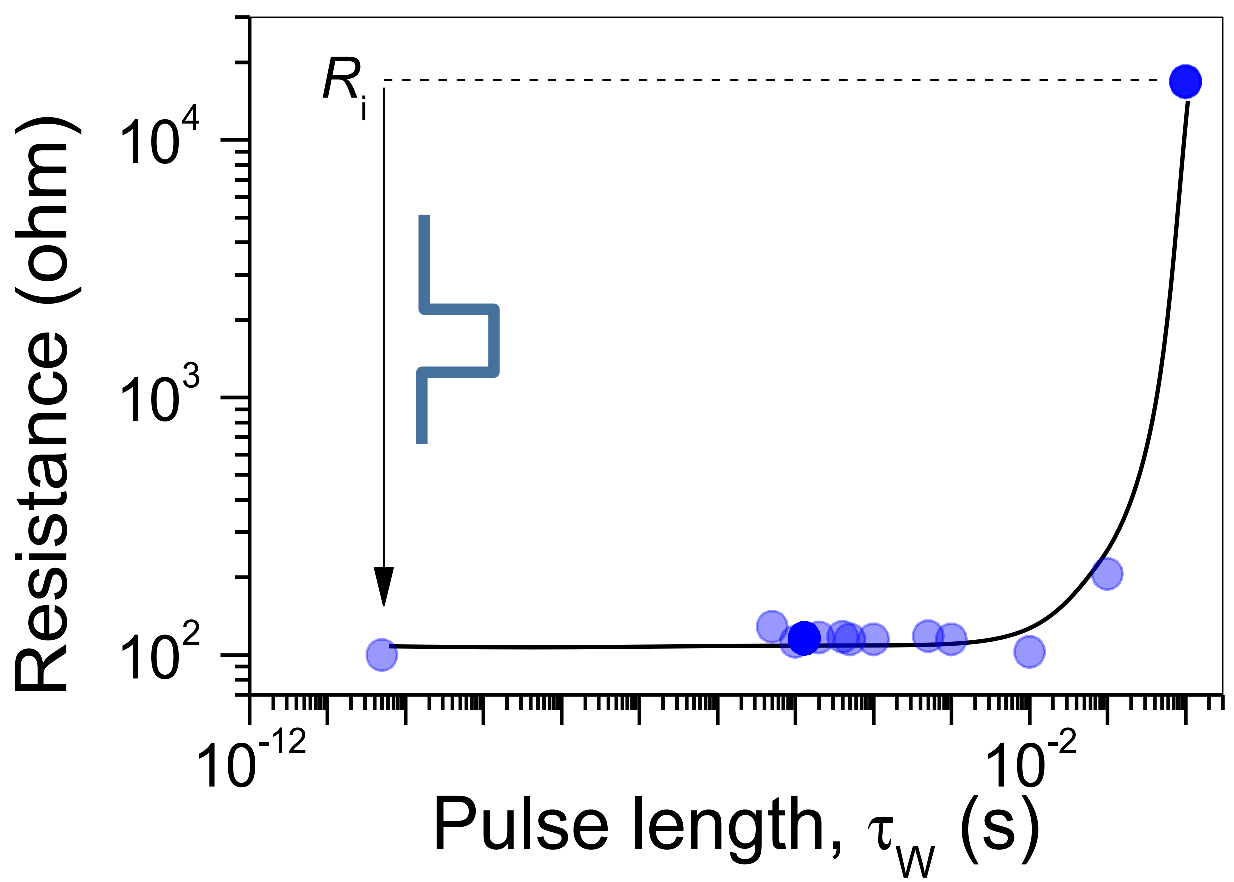
Science AAAS
L Stojchevska et al. Science 2014;344:177-180

Switching by an ultrafast current source


Ian Mihailovic

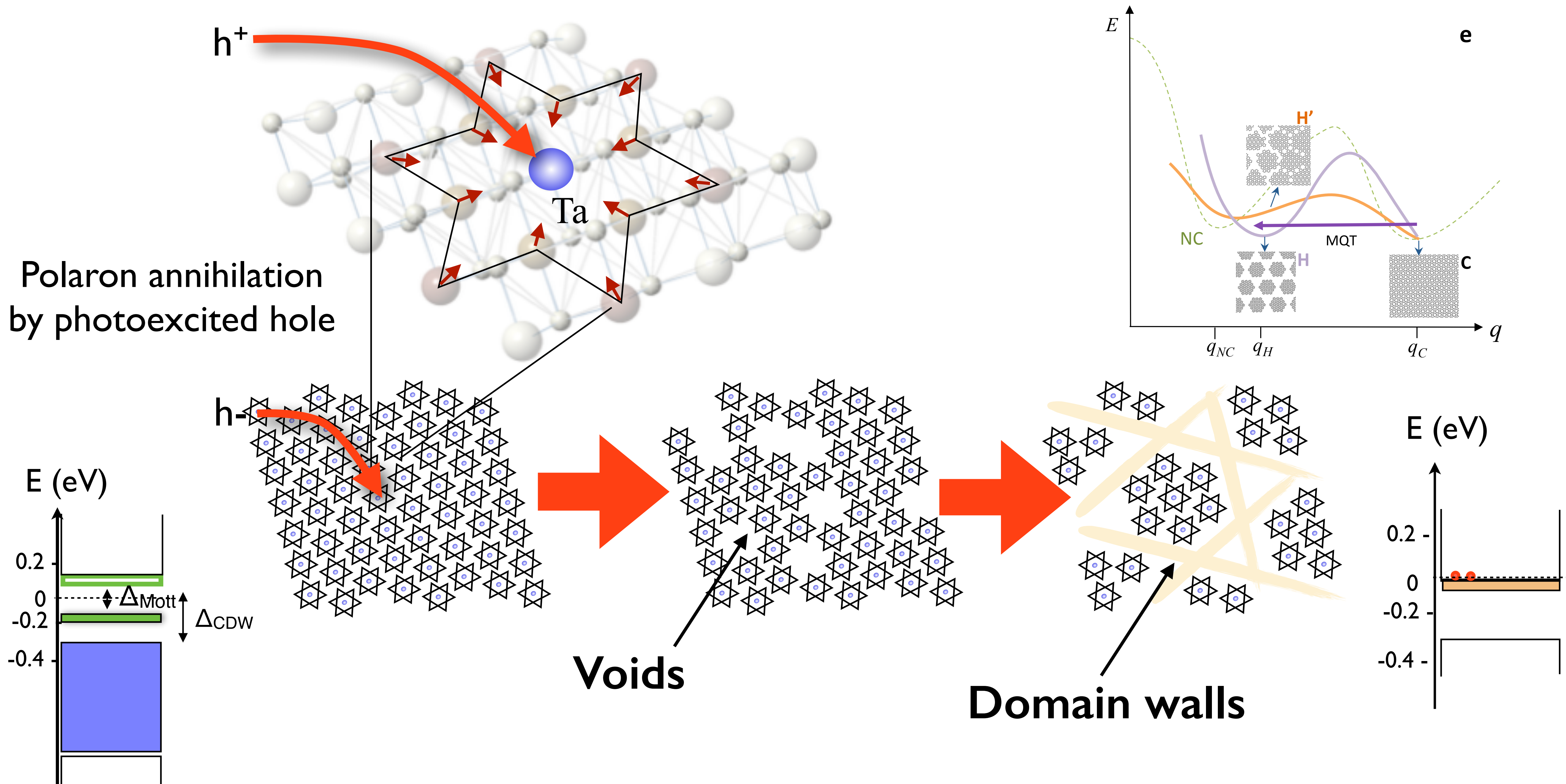




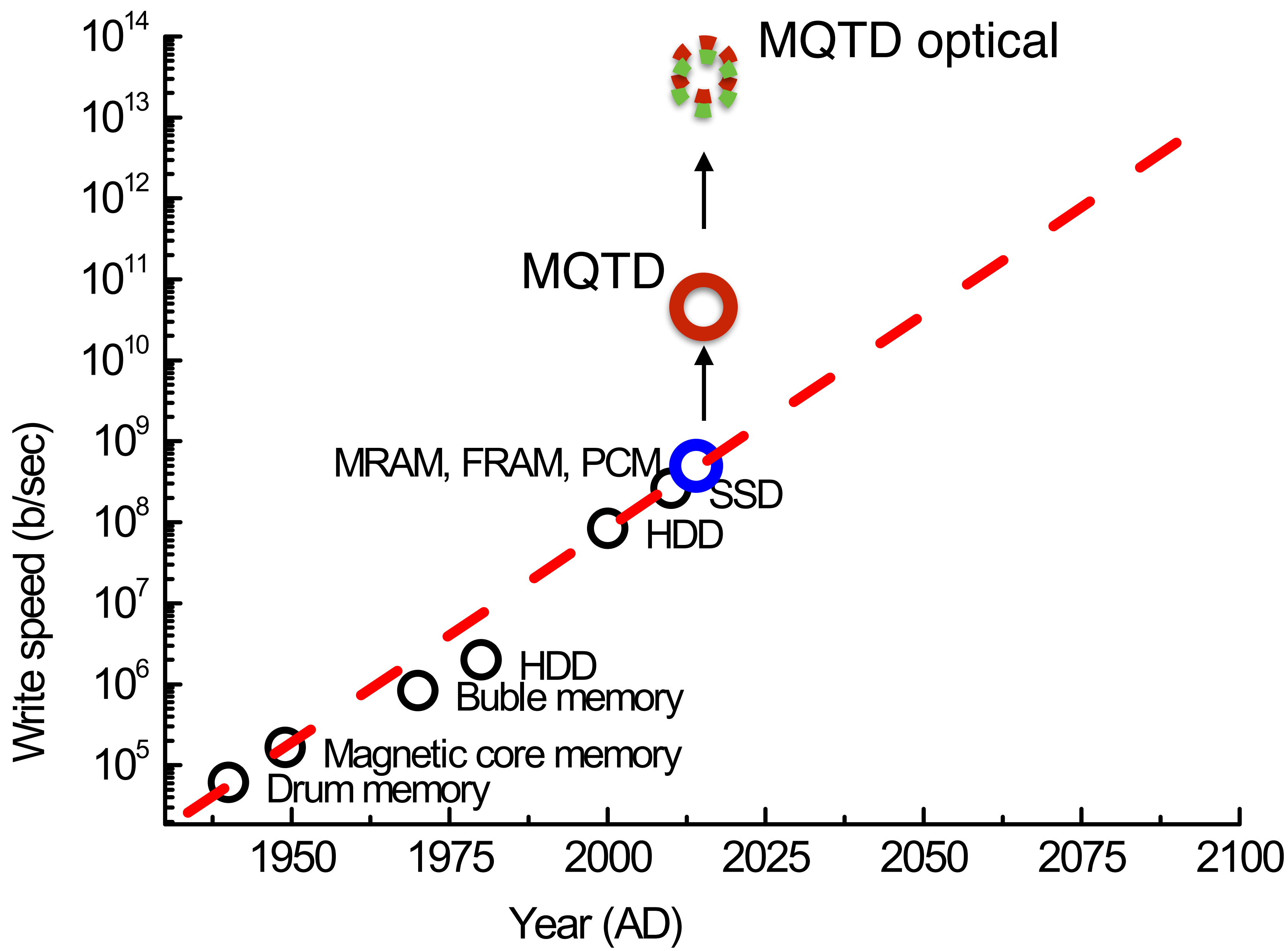


Photo"doping" and ordering of voids

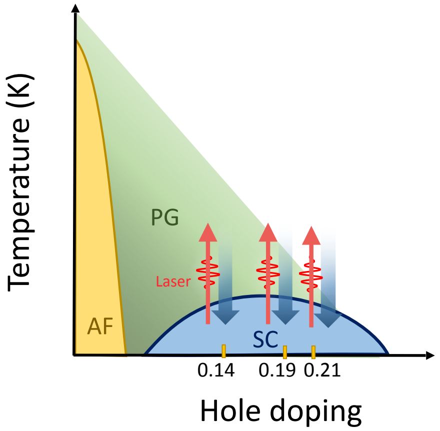
The addition of a h+ to the C structure annihilates a polaron, creating a void.



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

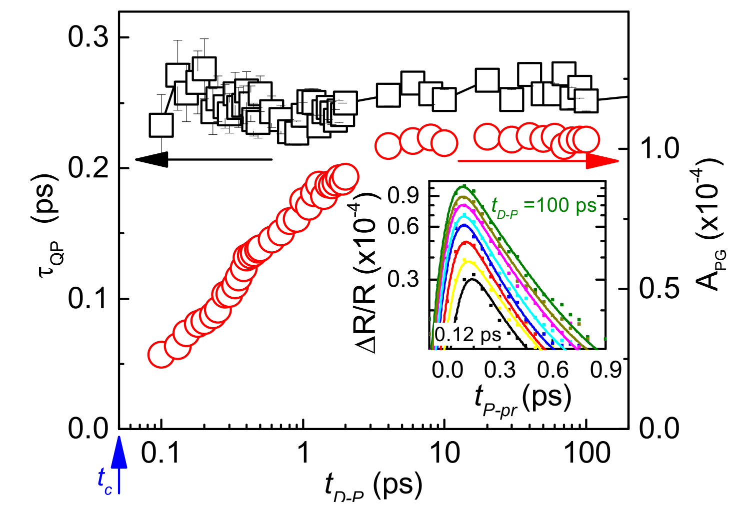


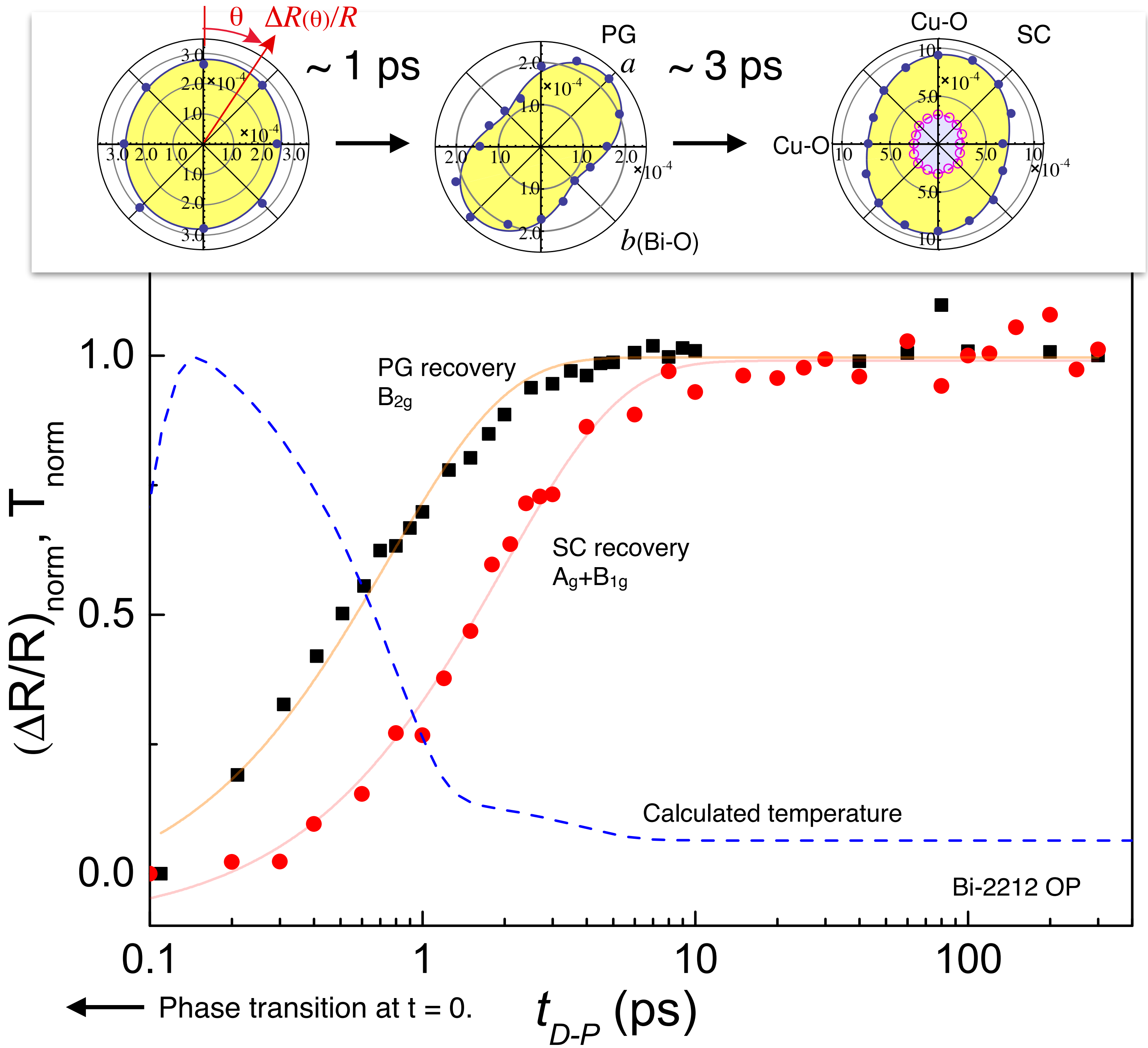
Yang et al, Nat. Nanot. (2010)



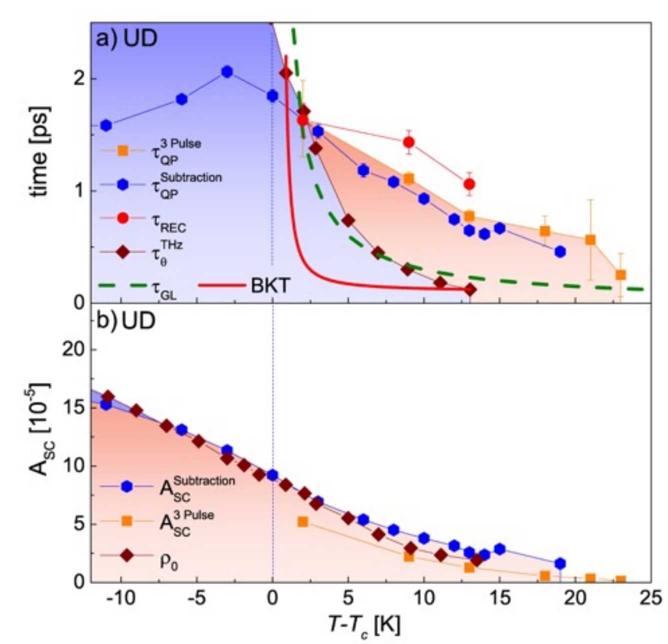
Emergence of broken symmetry in real time BiSCO-2212 (UD-OP-OD)

Ultrafast carrier localisation in the PG state not proper CDW





Phase coherence and pairing timescales vs. T



Toda, Y. *et al.* PRB **90**, 094513 (2014)

Toda, Y. *et al.* PRB **84,** 174516 (2011).

Madan, I. *et al. Sci Rep* **4,** 5656 (2014).

Madan, I. *et al. to be published* (2014)

L Stojchevska et al. Science (2014);344:177