Kyoto, Feb. 12, 2014

Peculiar phenomena in the time-dynamics of condensed matter systems undergoing symmetry-breaking transitions.

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European Research Council
Established by the European Commission




Experiments:

(at JSI)

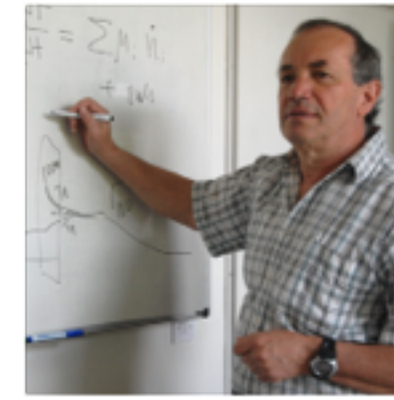
L. Stojchevska
I.Vaskivskyi
T. Mertelj
P. Kusar
R.Yusupov

Samples+:

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

Lithography:

D.Svetin *(JSI)*

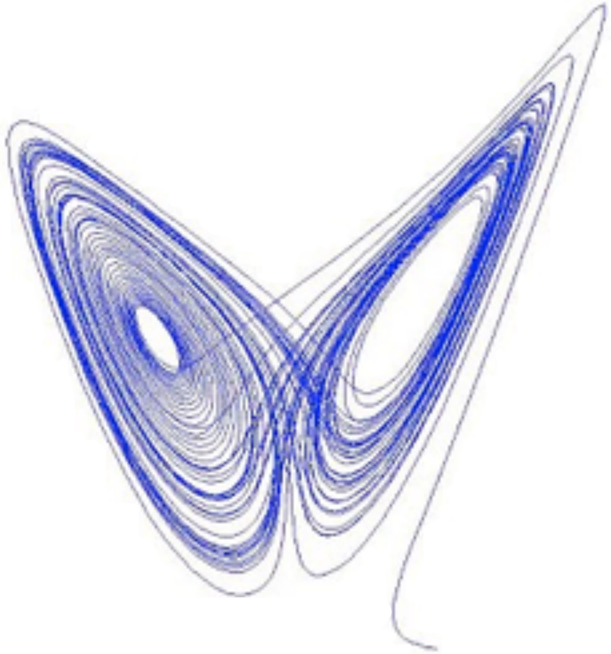


Theory

Serguei Brazovskii
(Univ. Paris Sud Orsay)

Viktor Kabanov
(JSI)

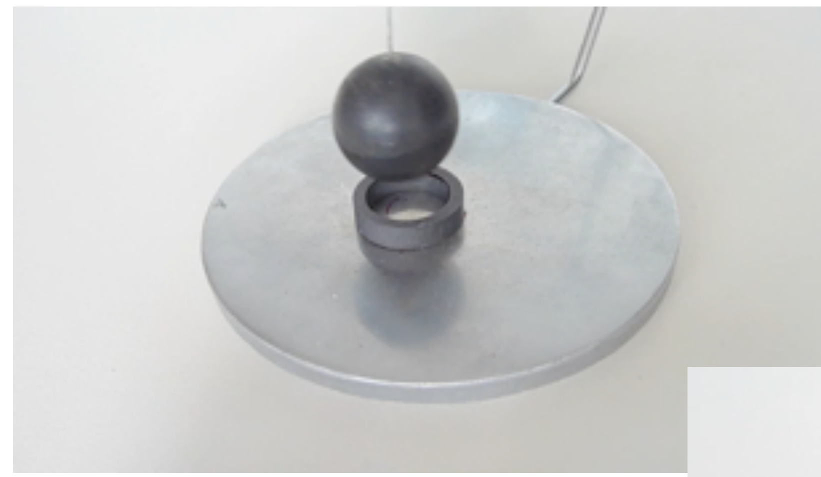
European Research Council
Established by the European Commission



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

Examples:

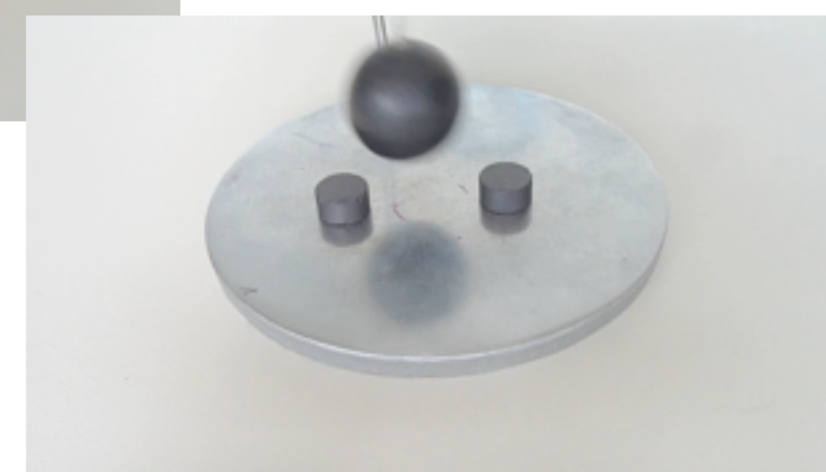
- HT Superconductors
- CDW transitions



$$\psi(t) = A(t)e^{i\phi(t)}$$

(τ_1 and τ_2)

$$\psi(t) \simeq A(t)$$ (τ_1)

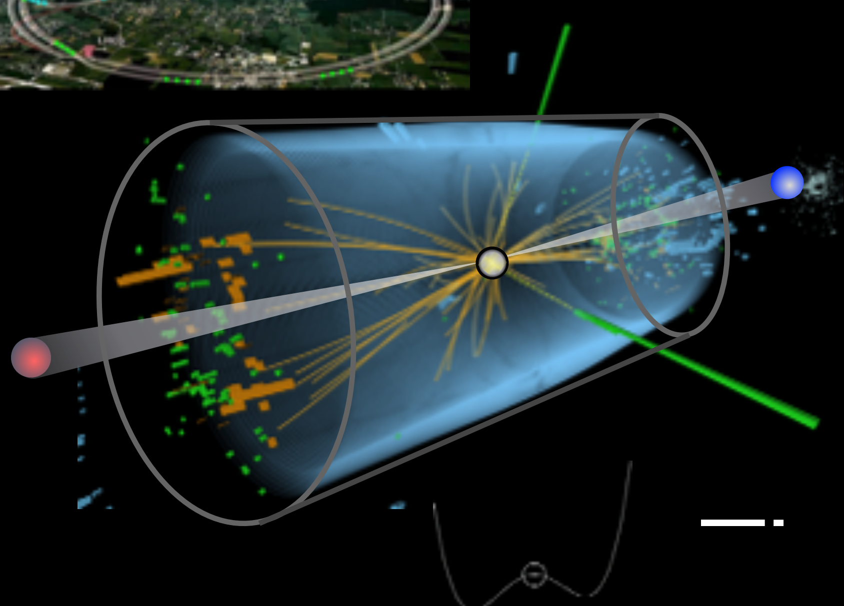


Peculiar phenomena:

- Amplitude mode decay and creation from defect annihilation
- Coupling to collective modes from previous eons
- A transition to a **hidden** state: *1T*-TaS_2

Transitions…

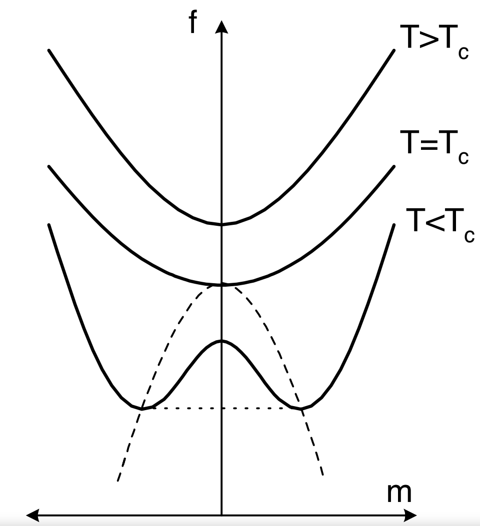
in time



The non-linear energy functional

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

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



leads to the Ginzburg-Landau equation for a **superconductor**:

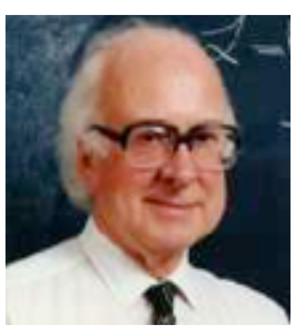
$$F = F_0 + \alpha|\psi|^2 + \frac{\beta}{2}|\psi|^4 + \frac{1}{2m}|(-i\hbar\nabla - 2e\mathbf{A})\psi|^2 + \frac{|\mathbf{B}|^2}{2\mu_0}$$

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. 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, Lon

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

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

with

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

The time-dependent GLT

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 <u>time-dependent</u>:

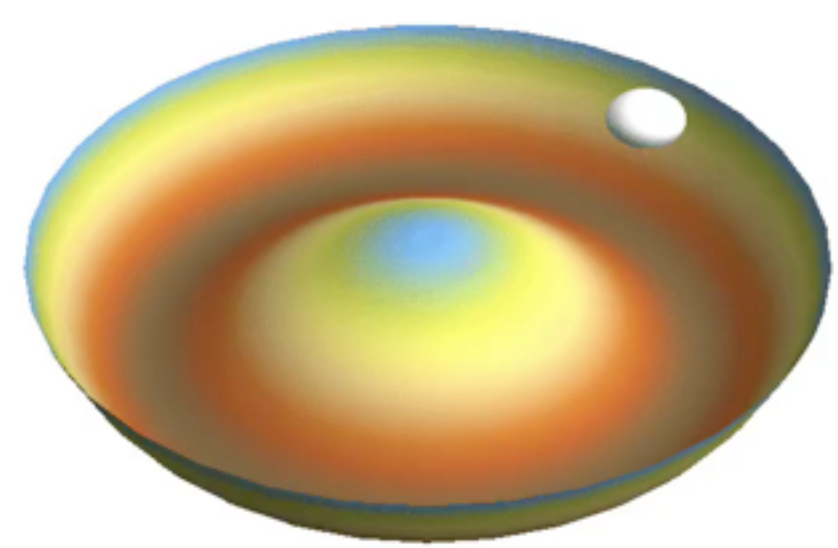
$$\alpha = [1 - \frac{T_e(t, \mathbf{r})}{T_c}]$$

"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, $\psi(t) = A(t)e^{i\phi(t)}$



Yusupov et al, Nat Phys. (2010)

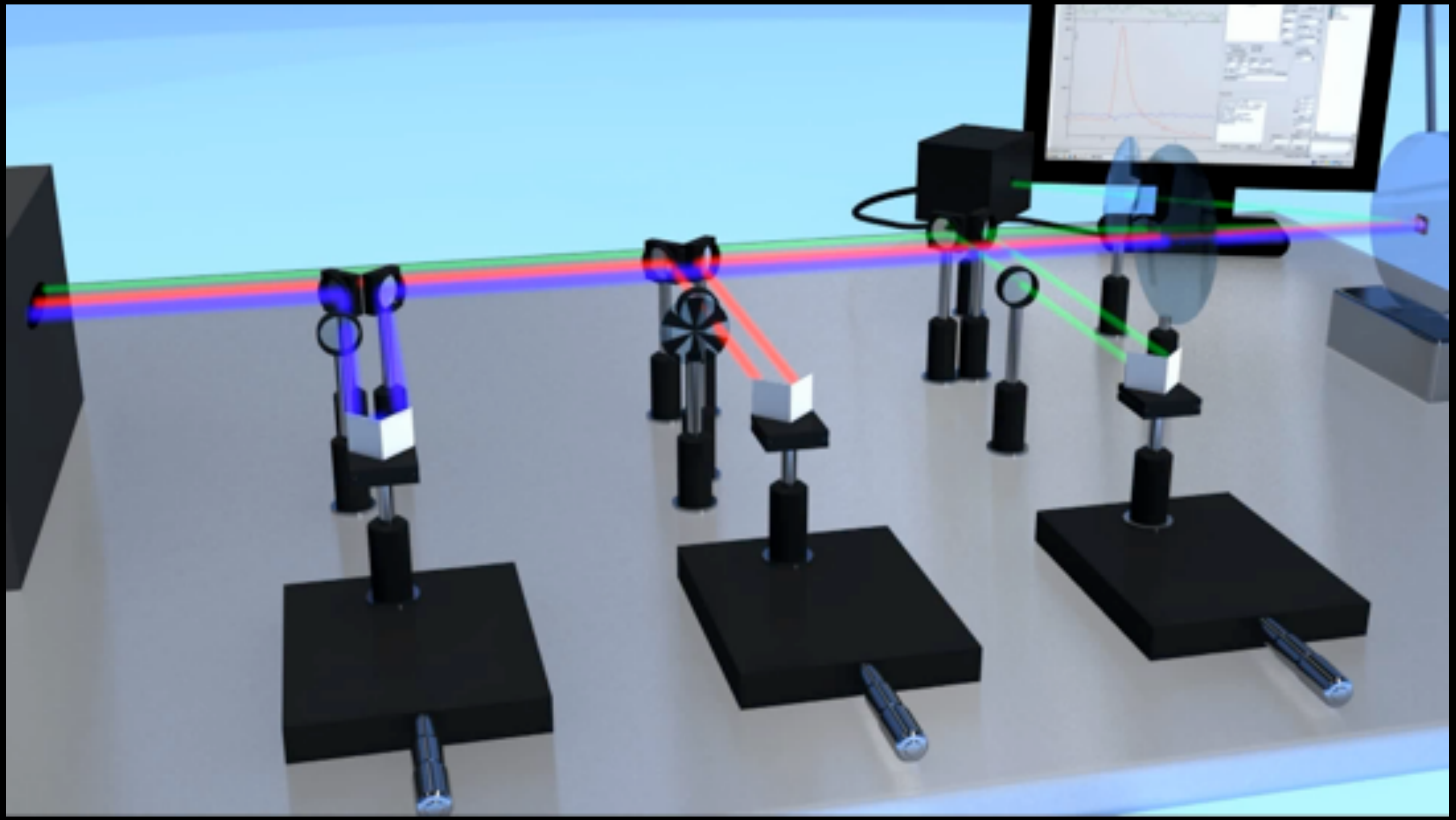
[†] Phase fluctuations are assumed to be slow.

“Cosmic Quench” experiments

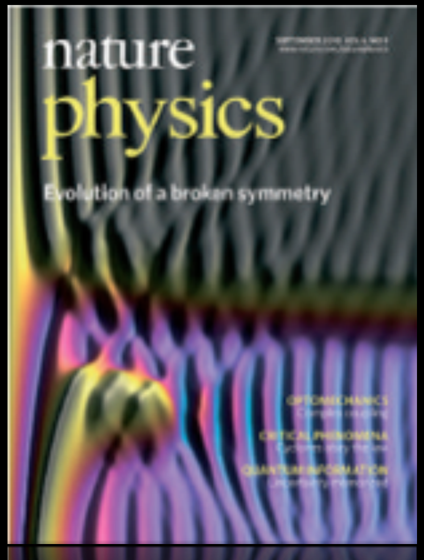
“Cosmology in L^4He”, Zurek (1985)

Optical experiments :

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

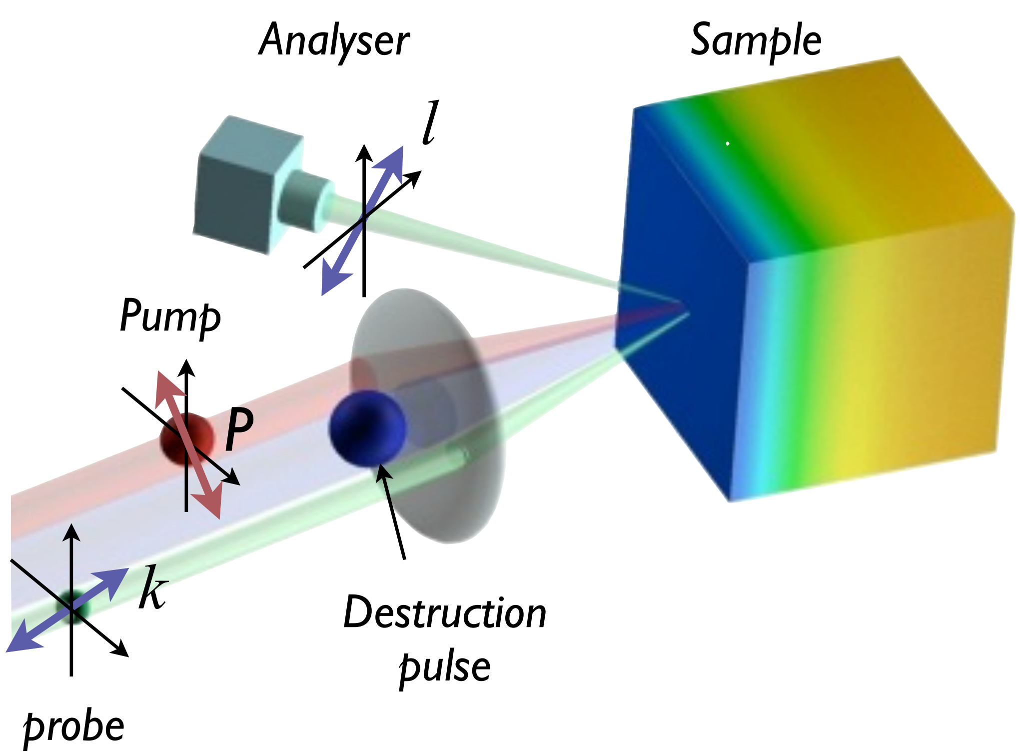


Yusupov, R. *et al. Nat Phys* **6,** 681–684 (2010).

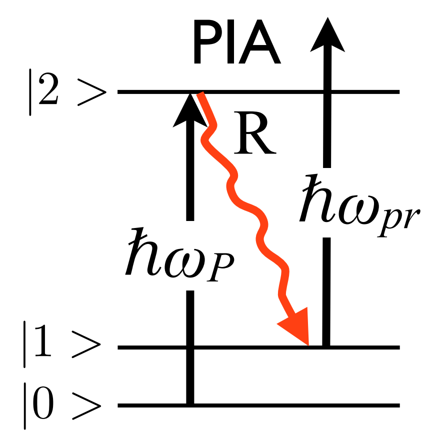


The response function is related to Raman-like processes

Toda et al., arXiv:1311.4719



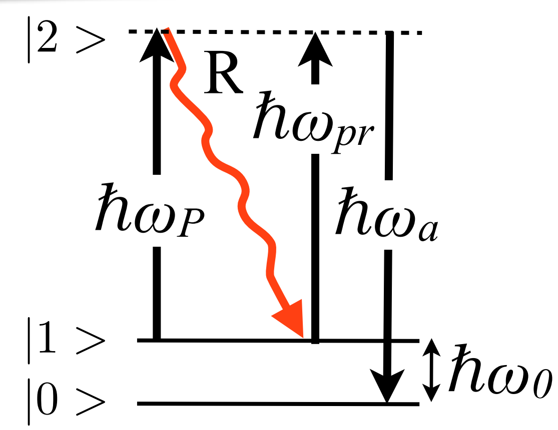
1. Photoinduced absorption (PIA):



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).

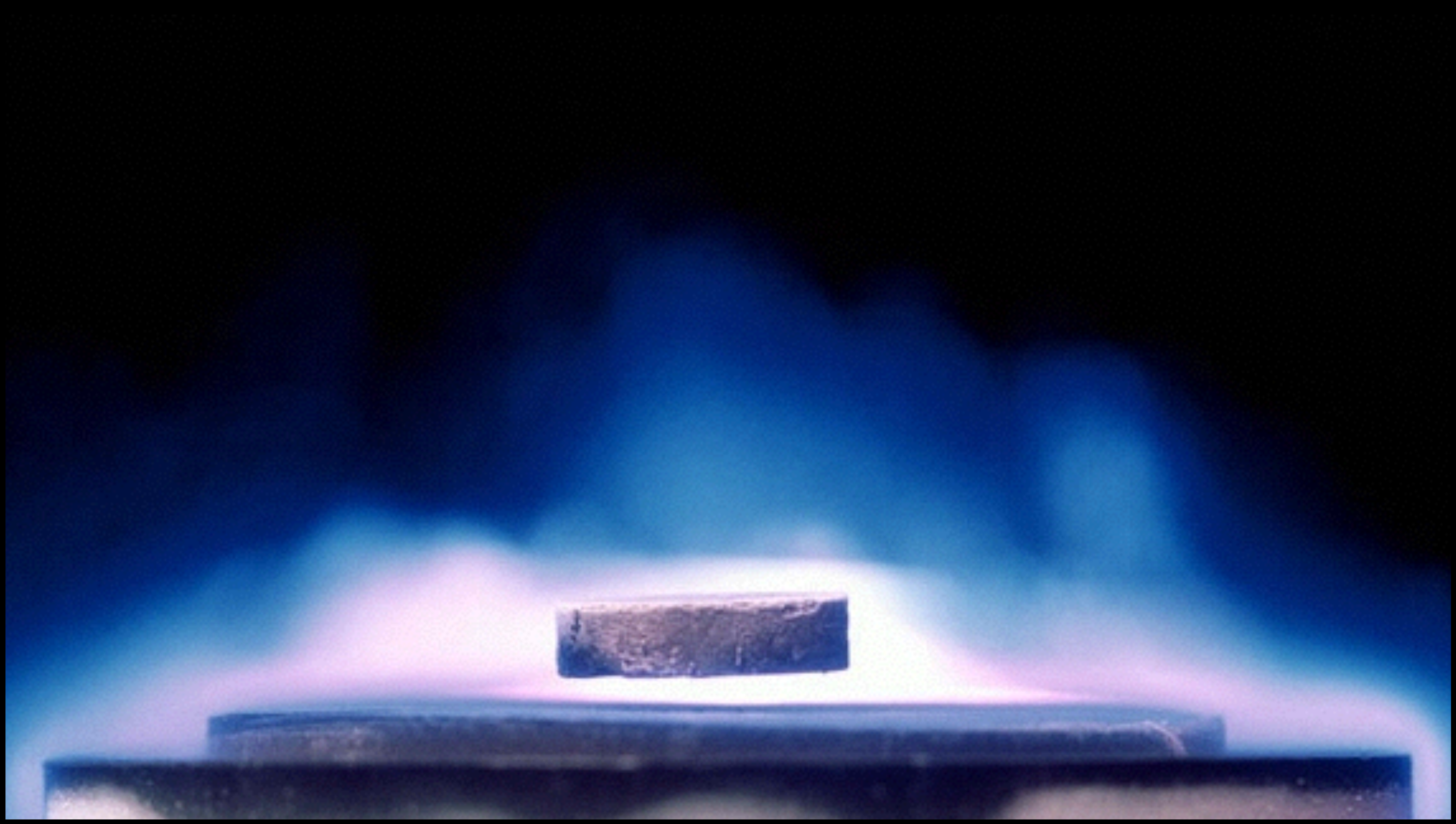
2. Coherent Raman-like (CRS) process:



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



Dynamics of broken symmetry nodal and anti-nodal excitations in $Bi_2Sr_2CaCu_2O_{8+\delta}$ probed by polarized femtosecond spectroscopy

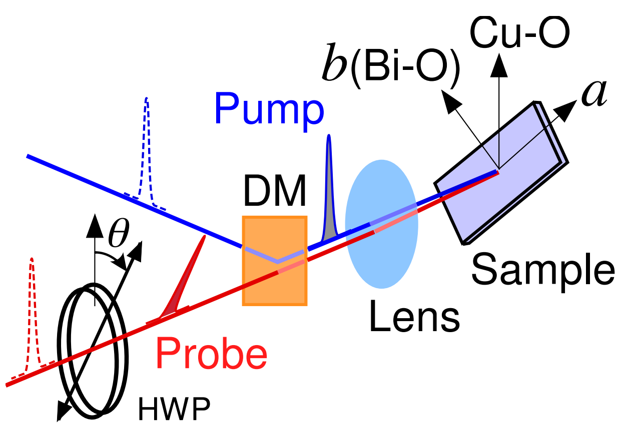
Y. Toda and F. Kawanokami
Department of Applied Physics, Hokkaido University, Sapporo 060-8628, Japan.

T. Kurosawa and M. Oda
Department of Physics, Hokkaido University, Sapporo 060-0810, Japan.

I. Madan, T. Mertelj, V. V. Kabanov, and D. Mihailovic
Complex Matter Dept., Jozef Stefan Institute, Jamova 39, Ljubljana, SI-1000, Slovenia

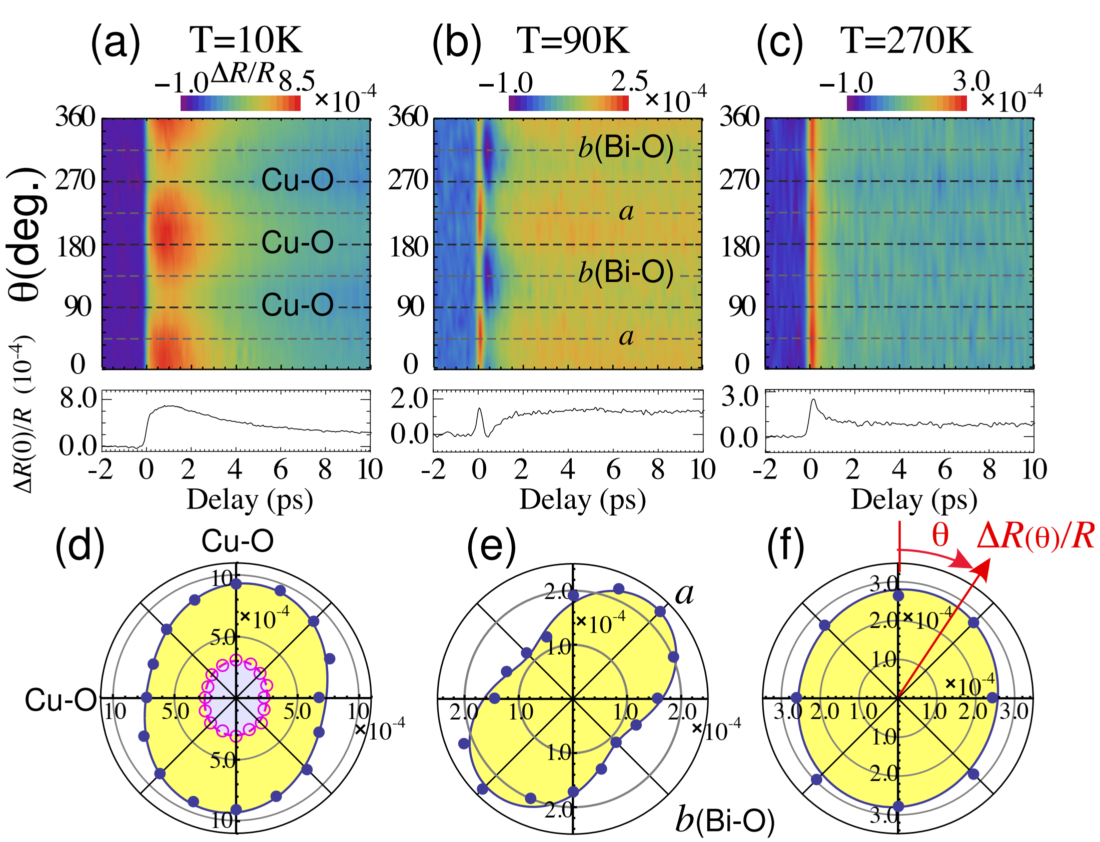
(Dated: January 29, 2014)

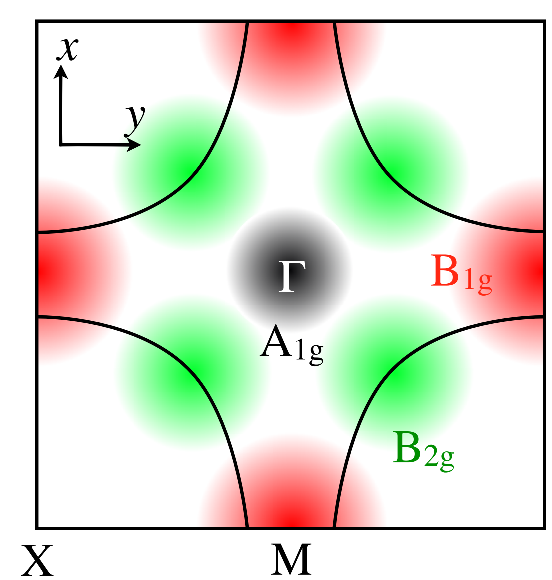
arXiv: Toda et al., (2013)

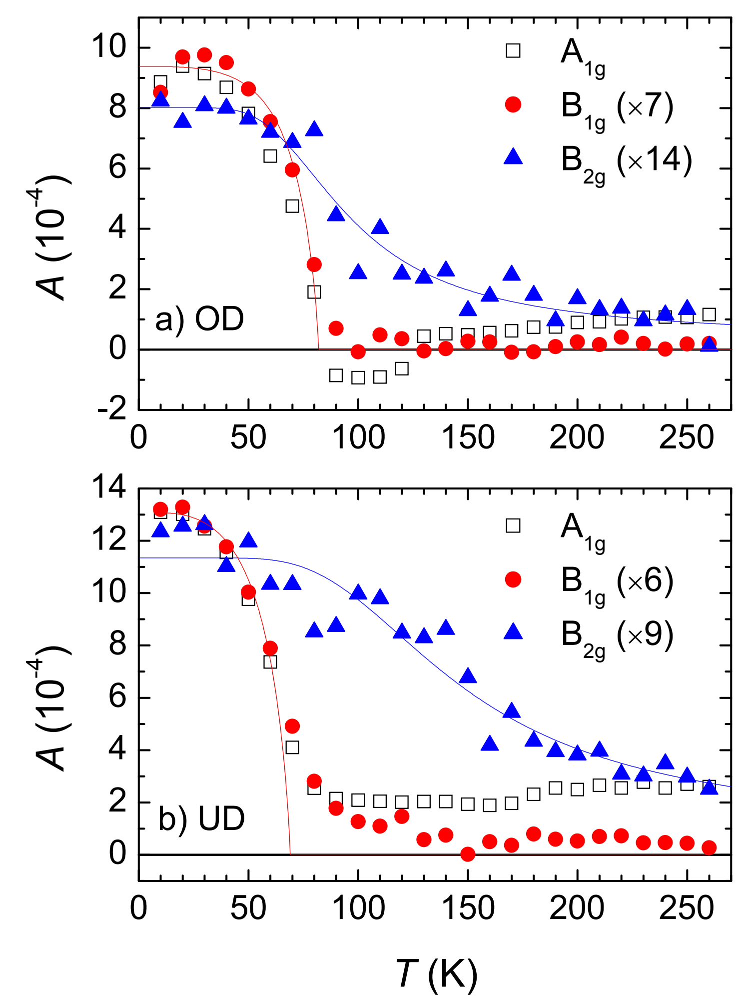


Temperature dependence of different symmetry components, A_{1g}, B_{1g} and B_{2g}:

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

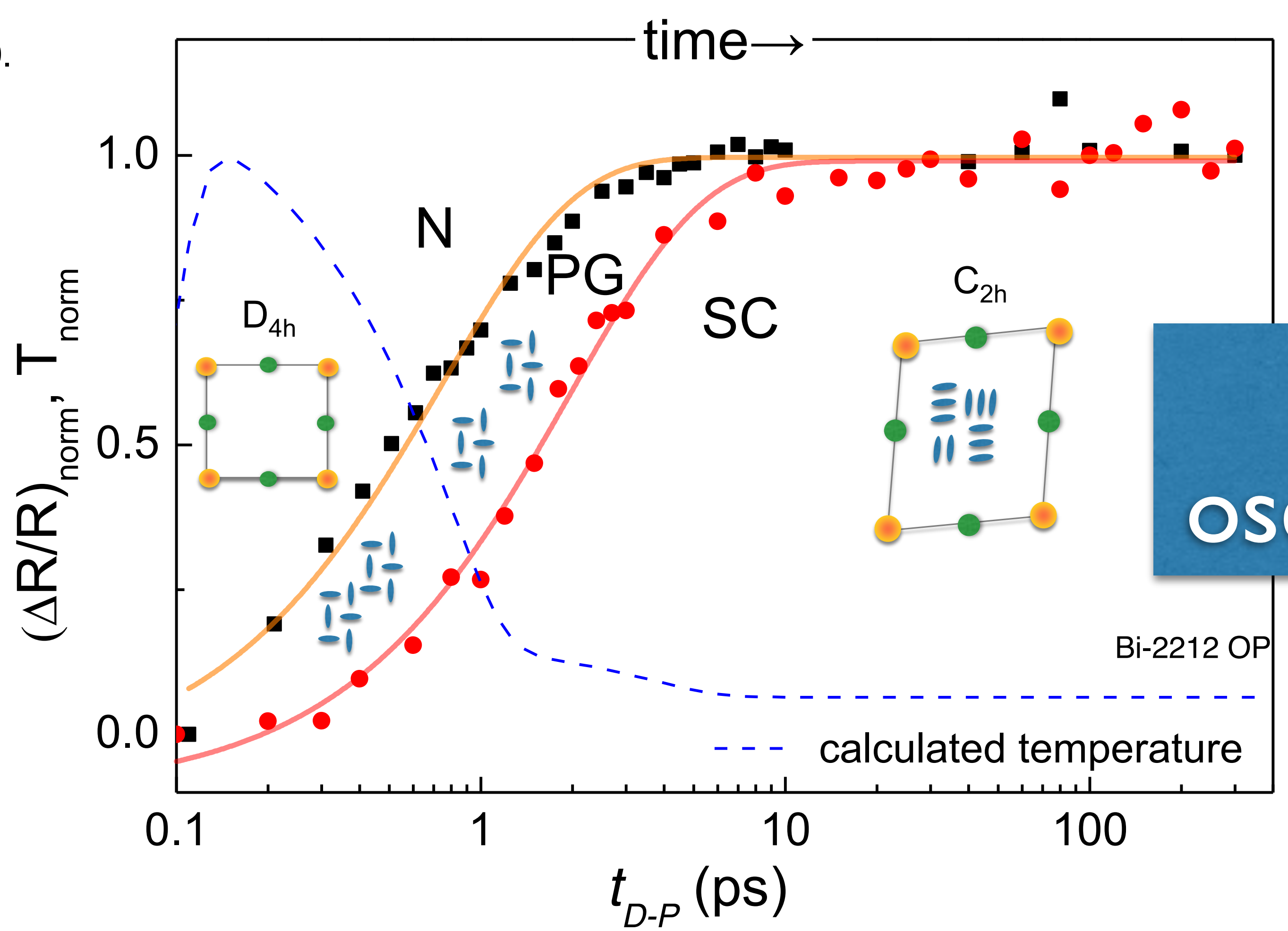






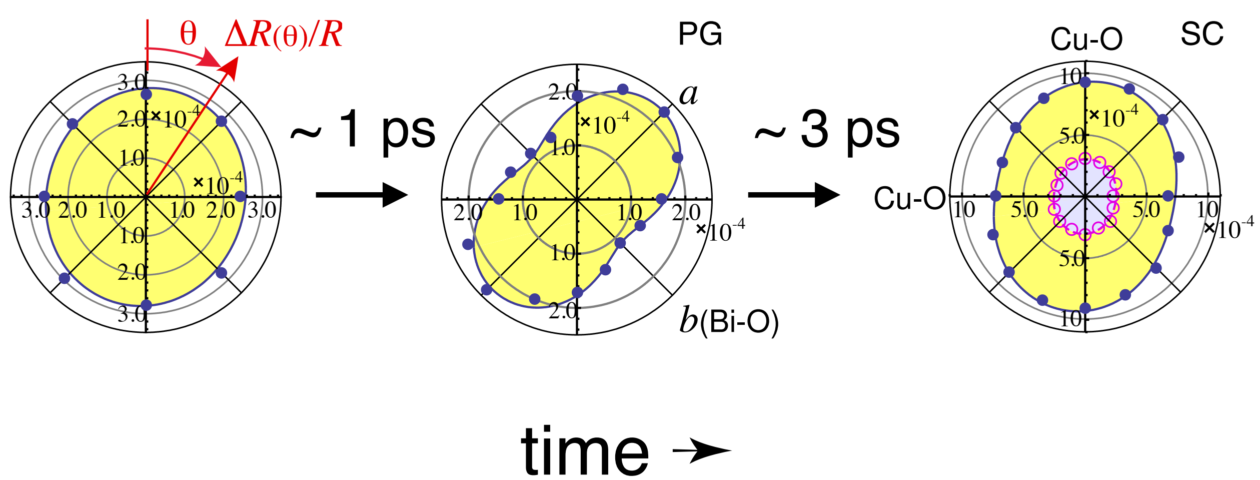
Time-dynamics of broken (rotational) symmetry in a high-Tc superconductor (Bi-2212-OP)

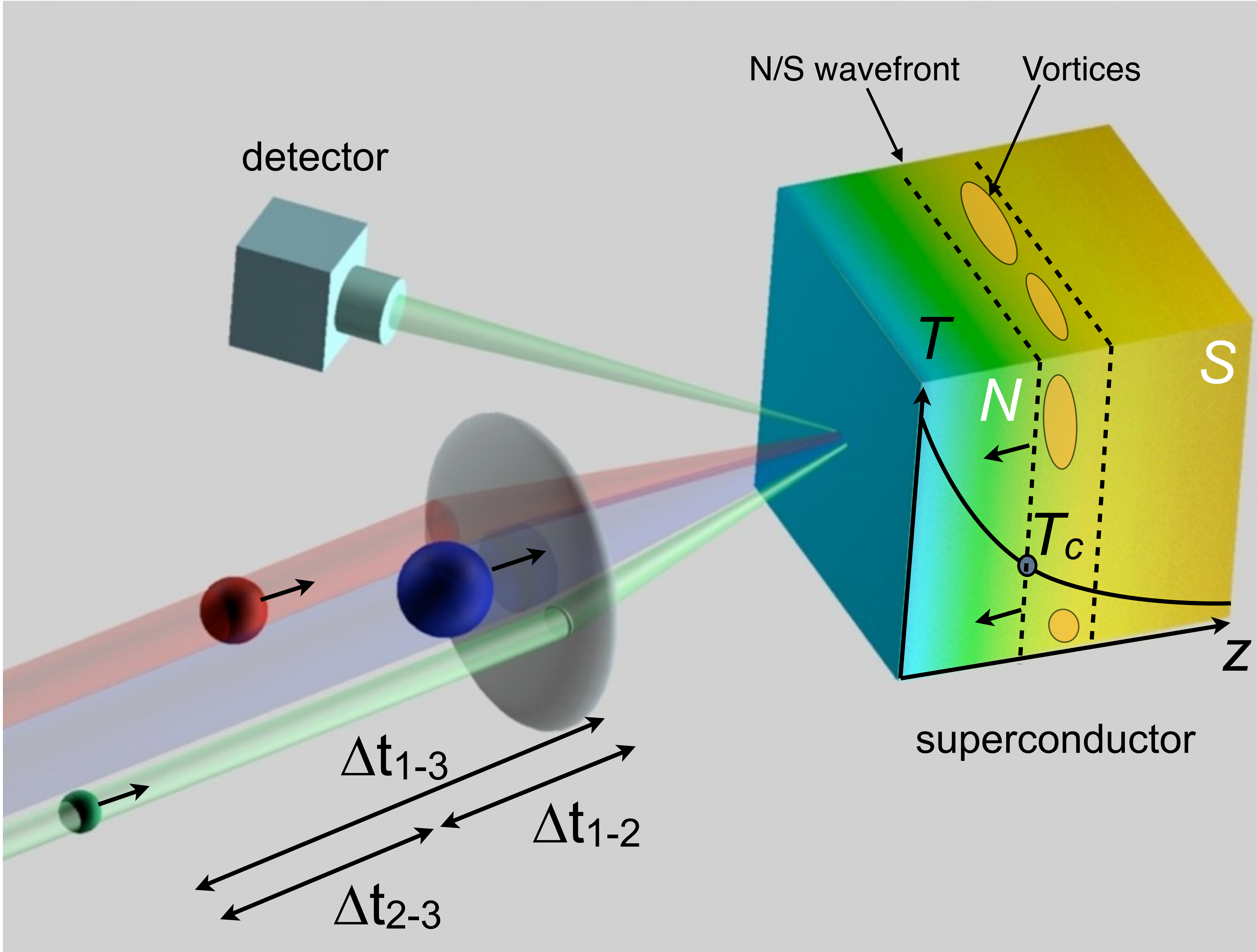
The D laser pulse is incident at t = 0.



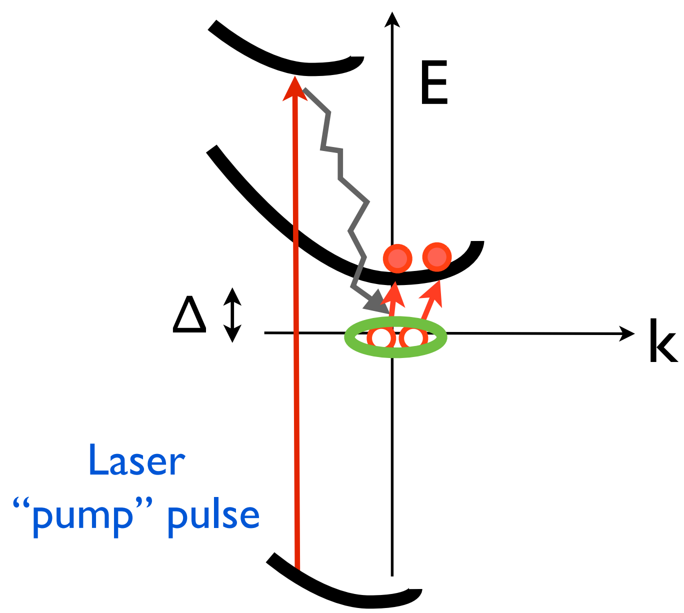
No oscillations!

Polarisation anisotropy changes with time




N/S wavefront
Vortices
detector
T
S
N
T_c
z
superconductor
Δt_{1-3}
Δt_{1-2}
Δt_{2-3}

Laser vaporisation of the superconducting condensate

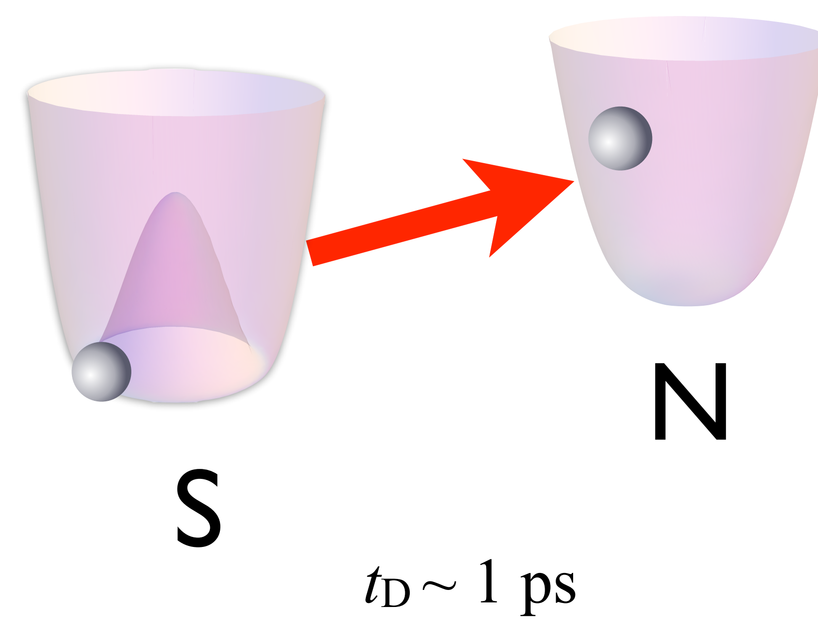


Single particle (electron) energy relaxation via boson (phonon) emission

↓ $t_D \sim 1$ ps

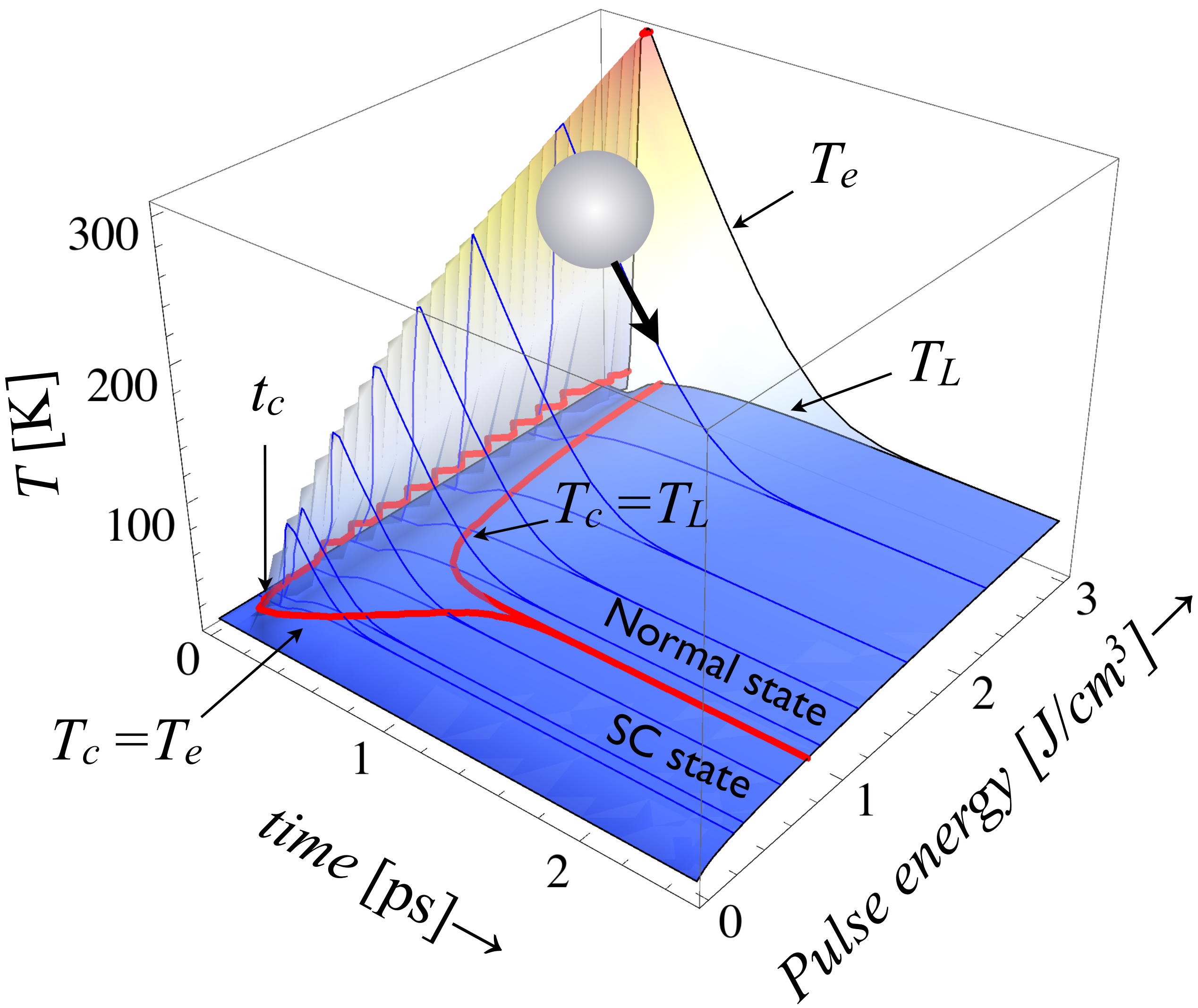
Bosons destroy pairs, creating QPs

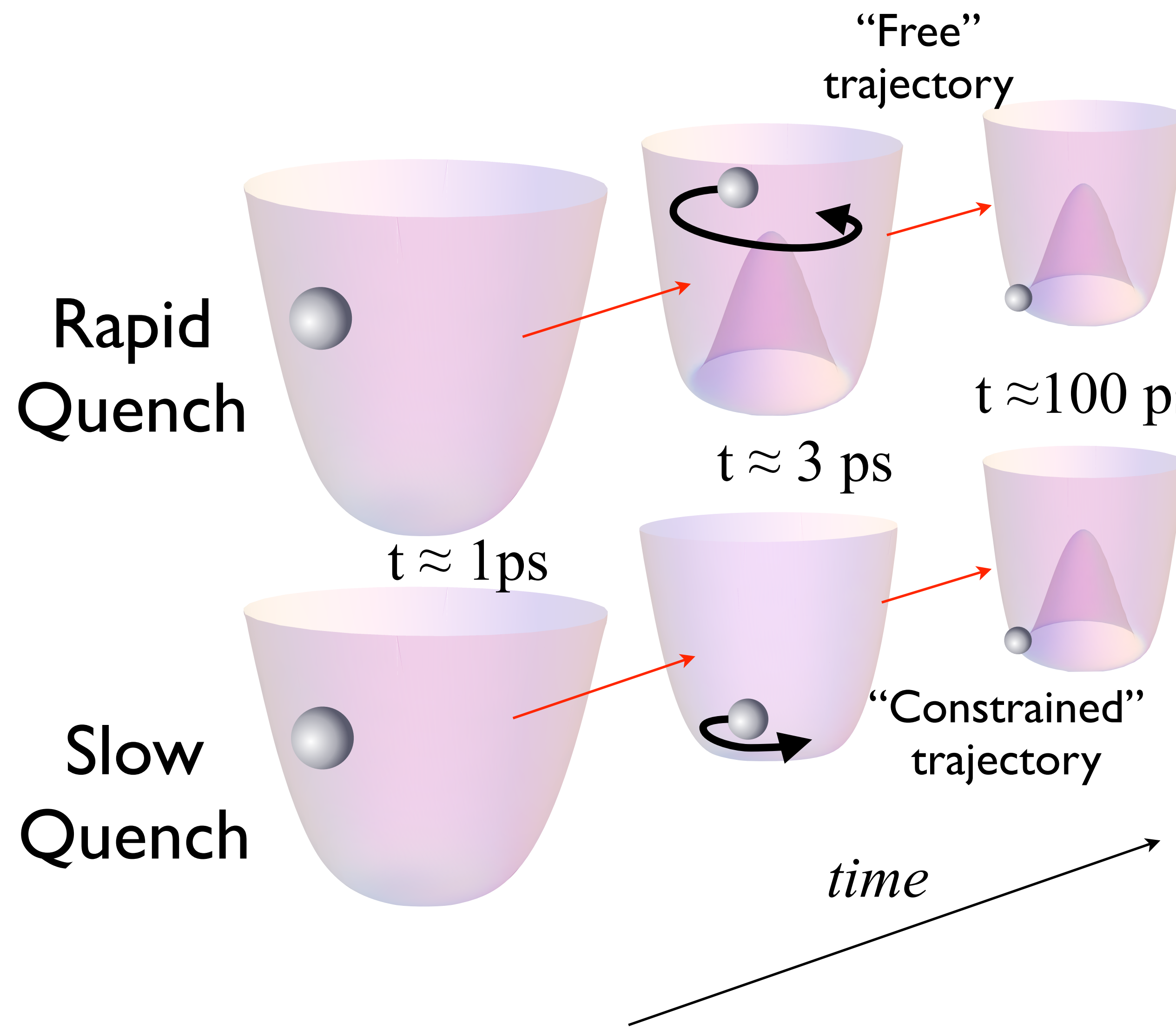
The SC condensate is vaporised in less than 1 ps



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).

The quench through T_c



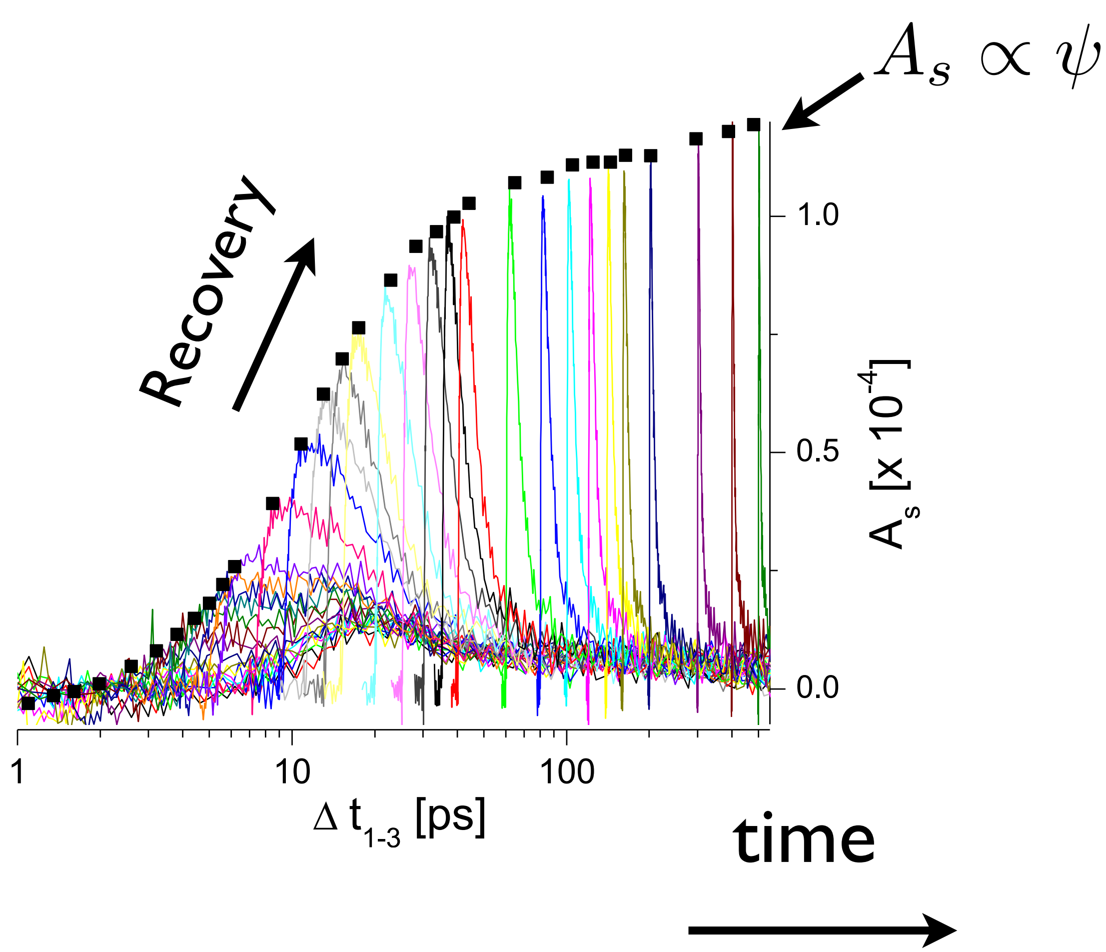


Higgs mode osc. period: $2\pi/\omega_H \sim 0.1$ ps

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

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

Recovery of the superconducting state measured by the amplitude of the transient reflectivity A_s(t)

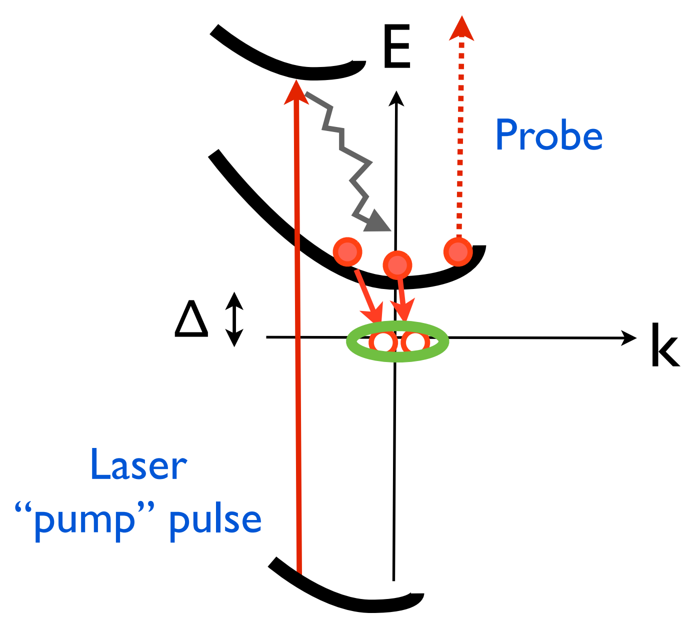


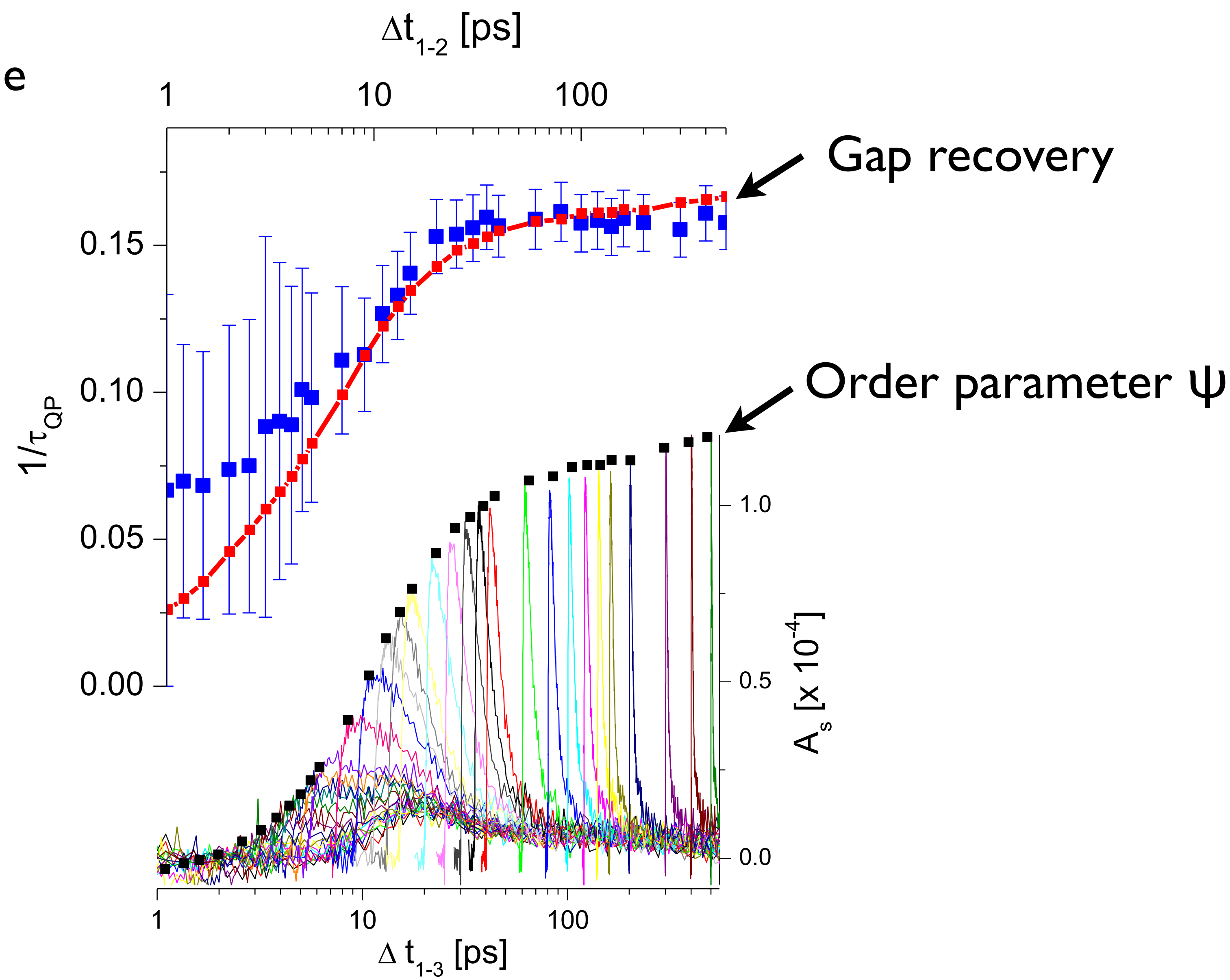
No Higgs oscillations observed in A_s(t)!

Superconducting gap recovery: QP recombination time compared with reflectivity amplitude

Quasiparticle recombination time

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





time

Modelling of superconducting state recovery:

TDGL:

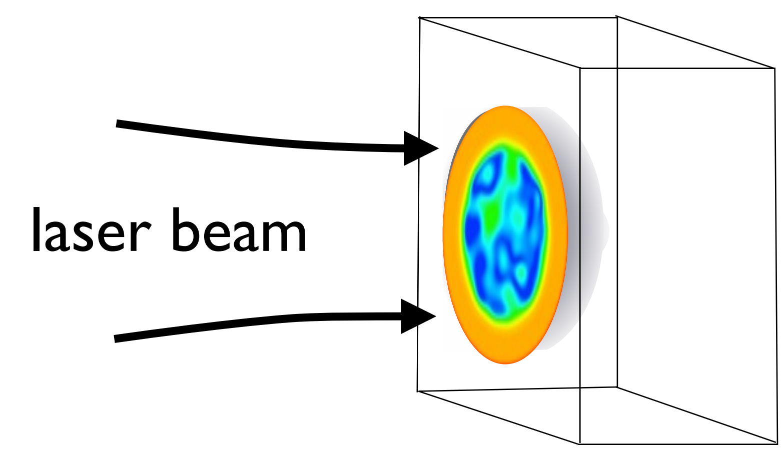
$$u\left(\frac{\partial\psi}{\partial t}\cancel{+i\Phi\psi}\right) = -\alpha_r(t,z)\psi - \psi|\psi|^2 - (i\nabla \cancel{+\mathbf{a}})^2\psi + \eta$$

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

no external field

Diffusion:

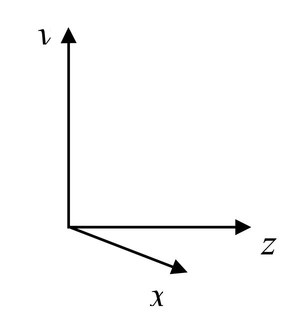
$$\frac{\partial T_D(t,\mathbf{s})}{\partial t} = D\nabla^2 T_D$$



Photoexcited electron energy loss:

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

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

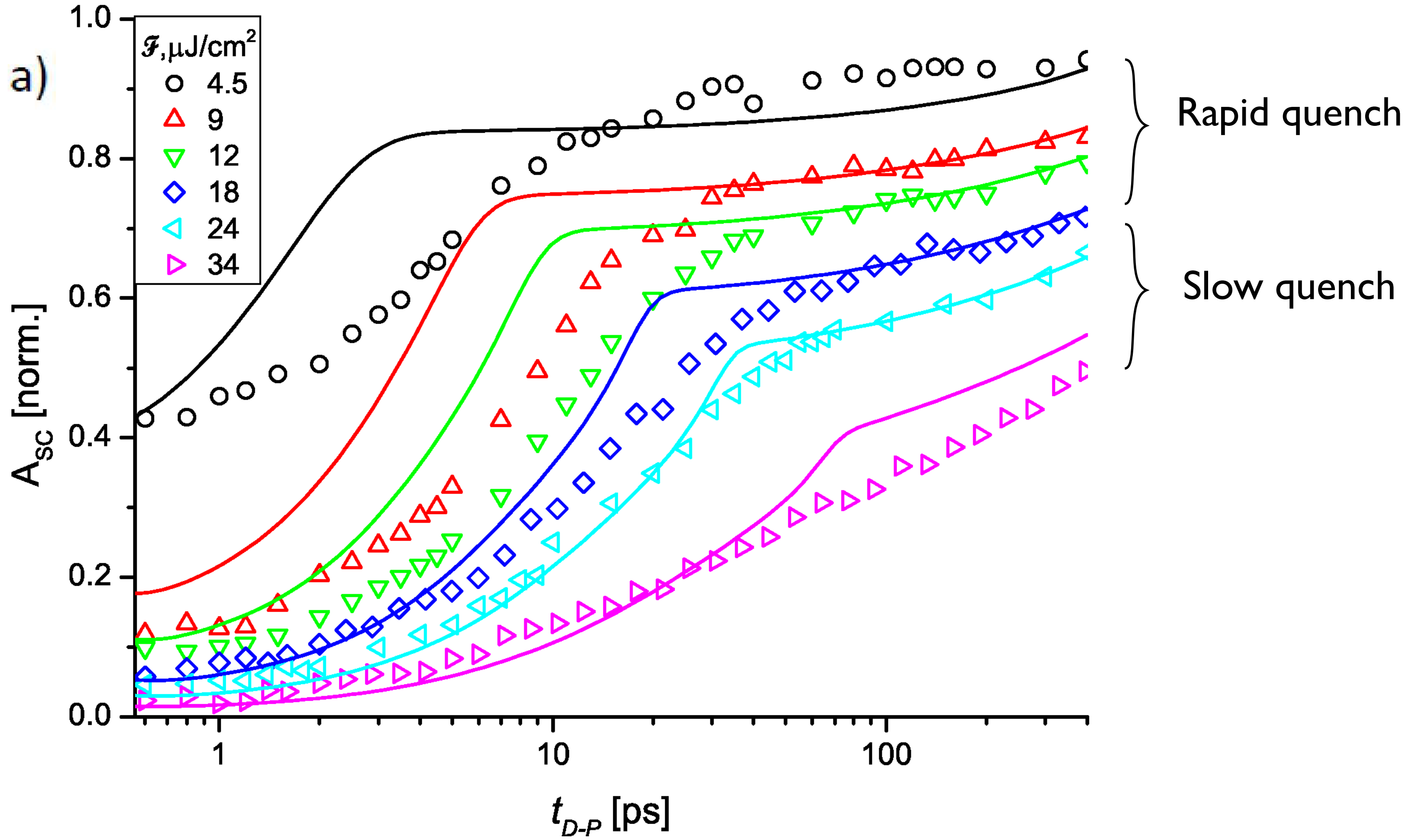


Basic TDGL equation:

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

Boundary conditions:

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

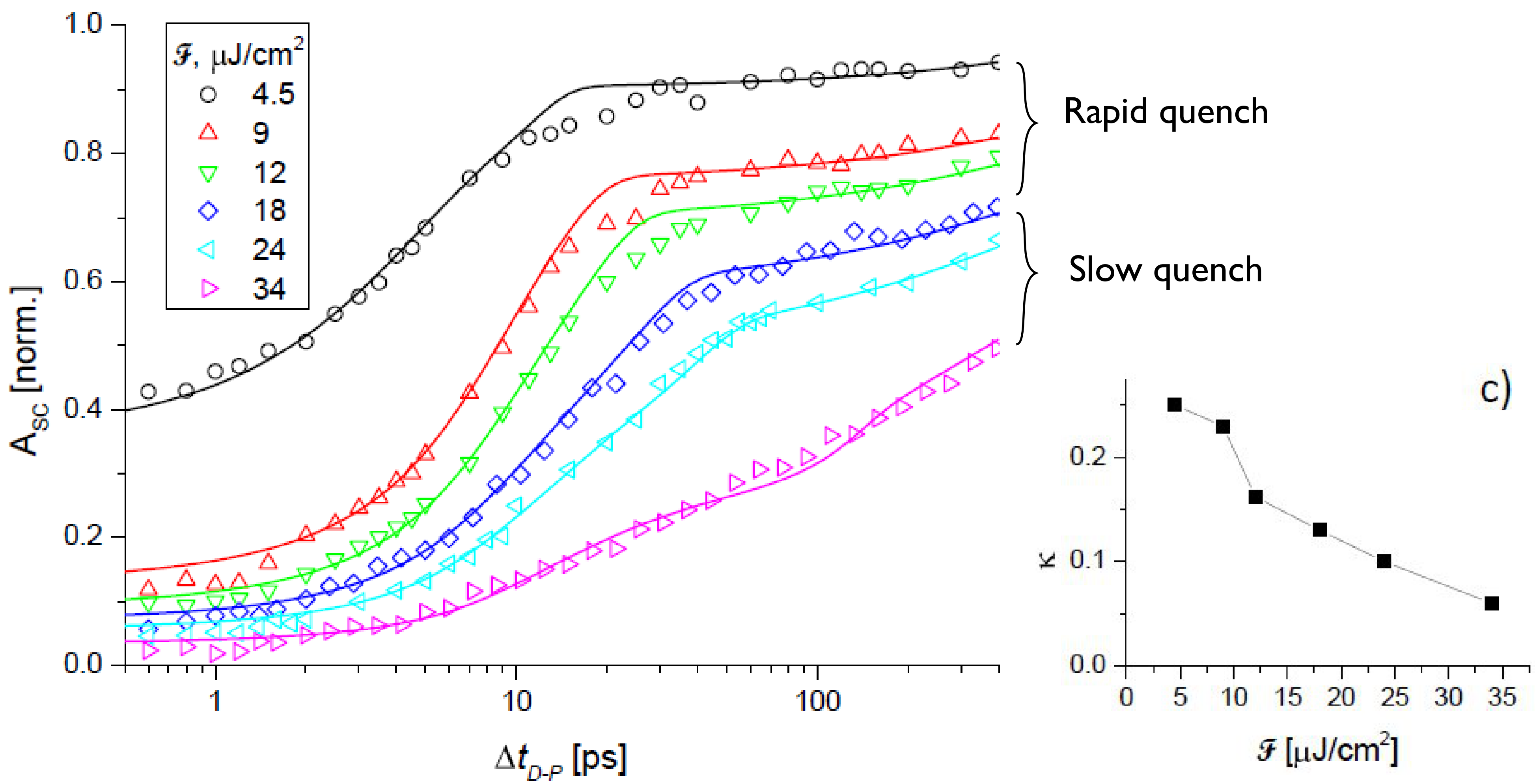


Doesn’t work very well!

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 at *t* = 0 (Volovik, 2000)†:
$$\psi(t=0) = \kappa z$$



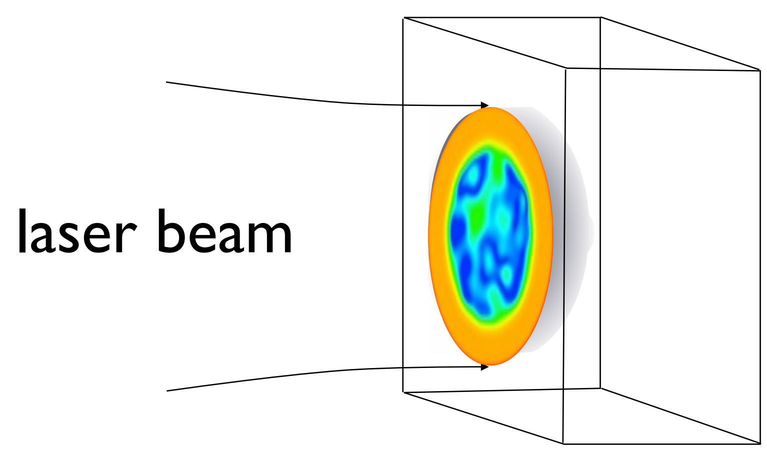
Much better

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

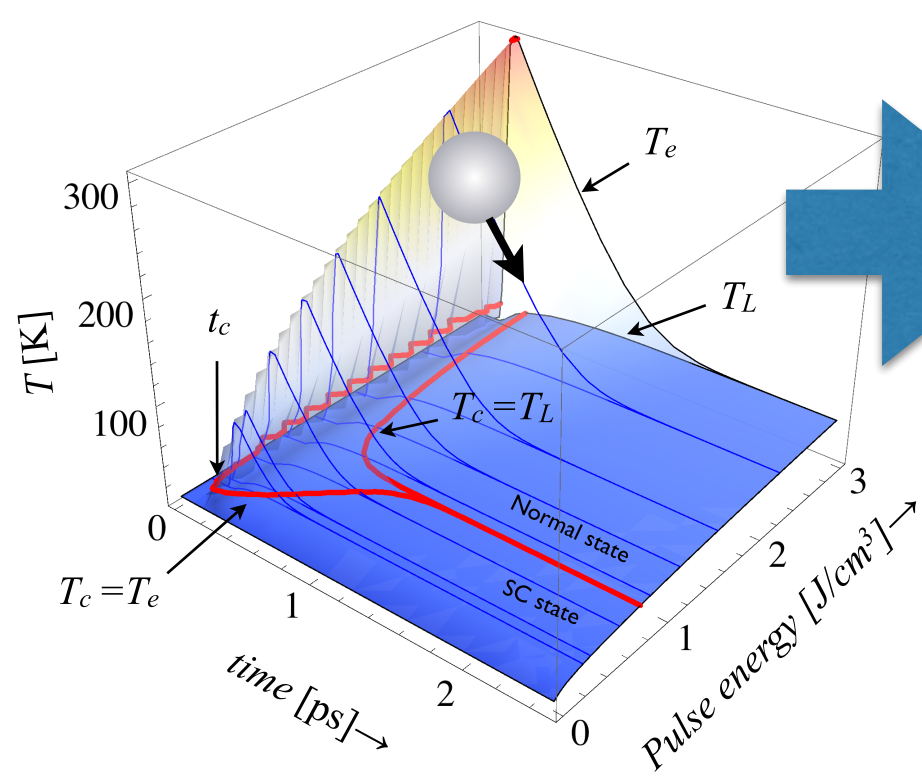
Kibble-Zurek mechanism: Evidence for vortex formation and annihilation on 10 ps timescale

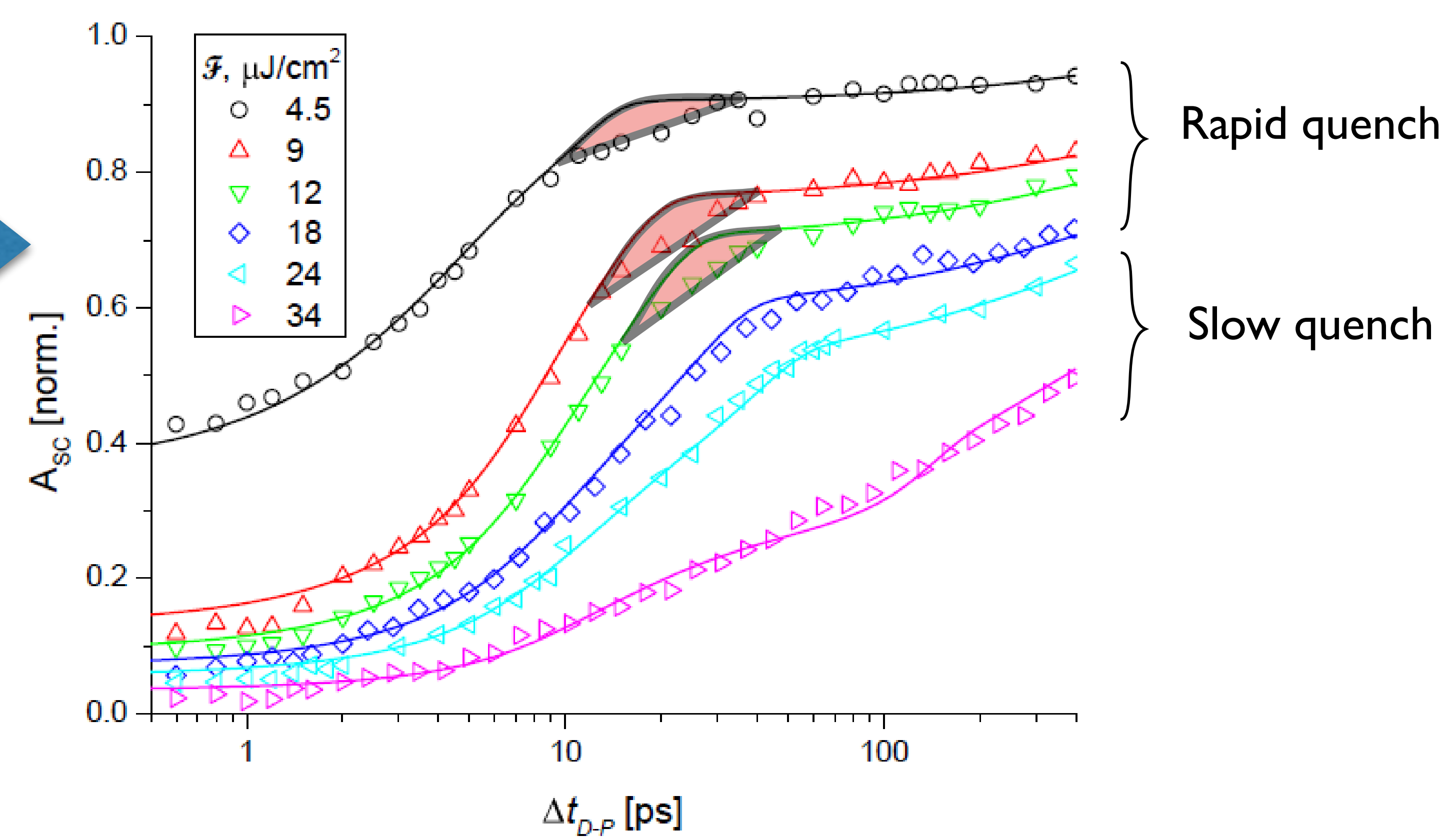
Laser spot size: $d = 60\mu\mathrm{m}$

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



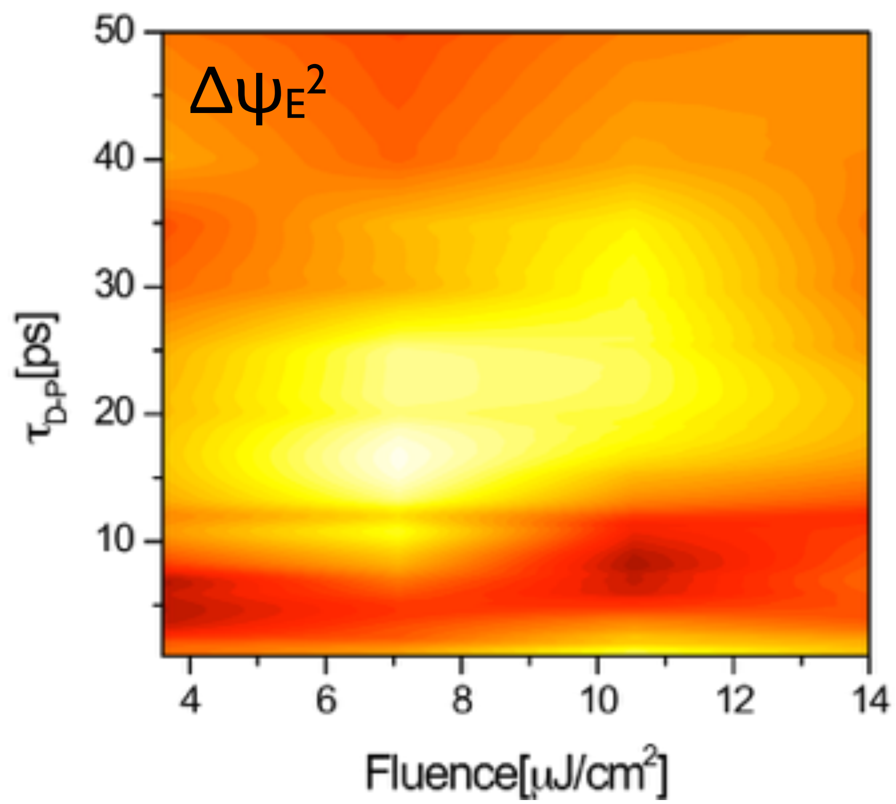
Regions are causally unconnected and evolve independently after the quench which causes the **formation of topological defects**.



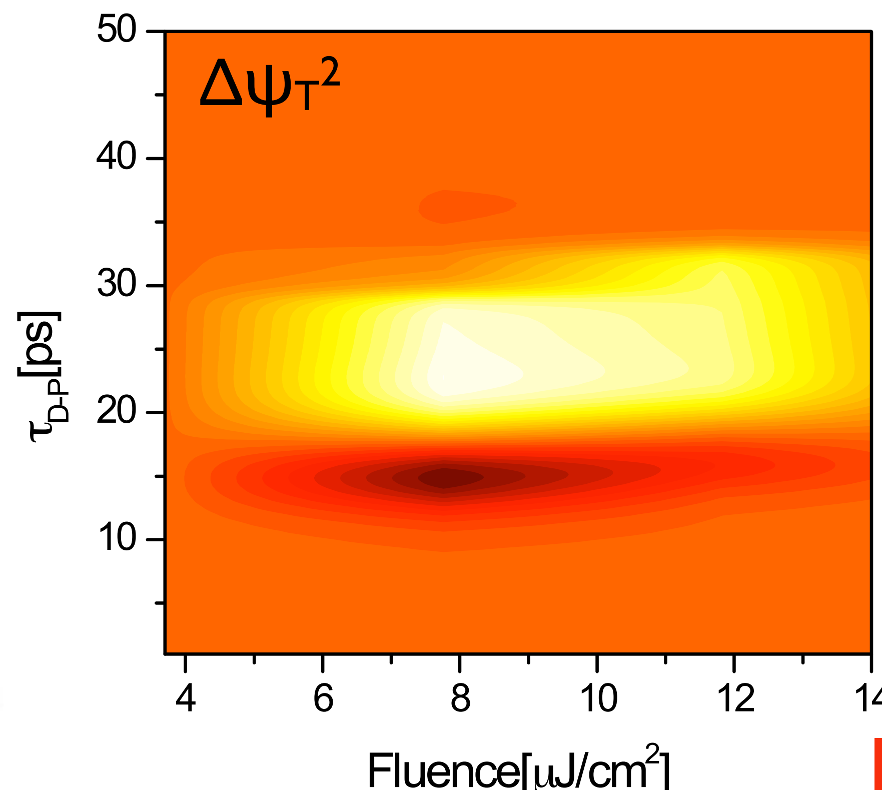


Mihailovic et al., J Phys-Condens Mat 25, 404206 (2013).

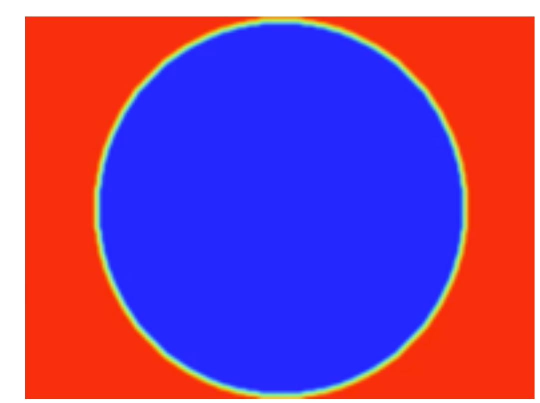
Vortices created in the quench annihilate on a timescale of 10-30 ps



Experiment



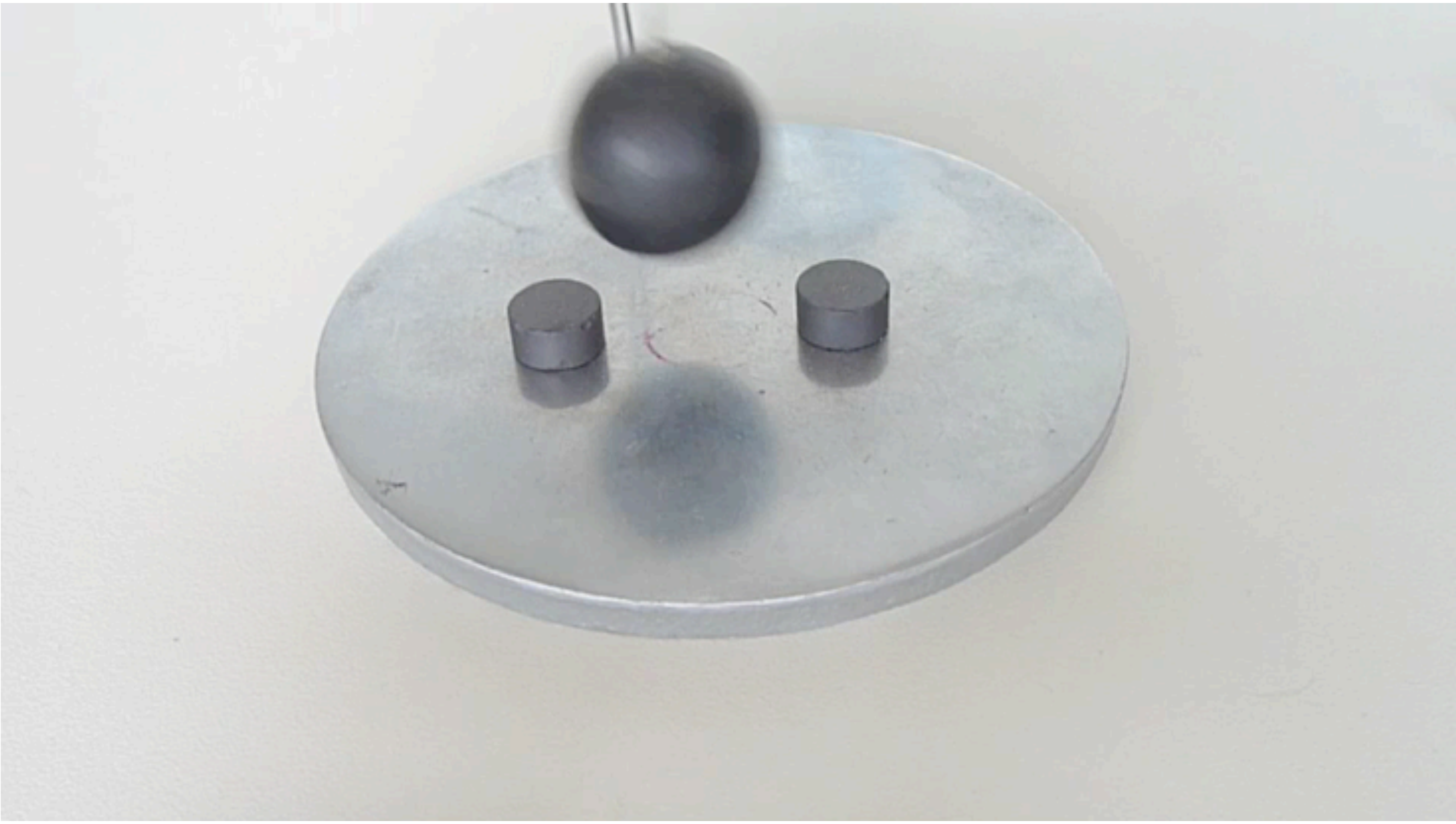
Theory calculation



Laser spot

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

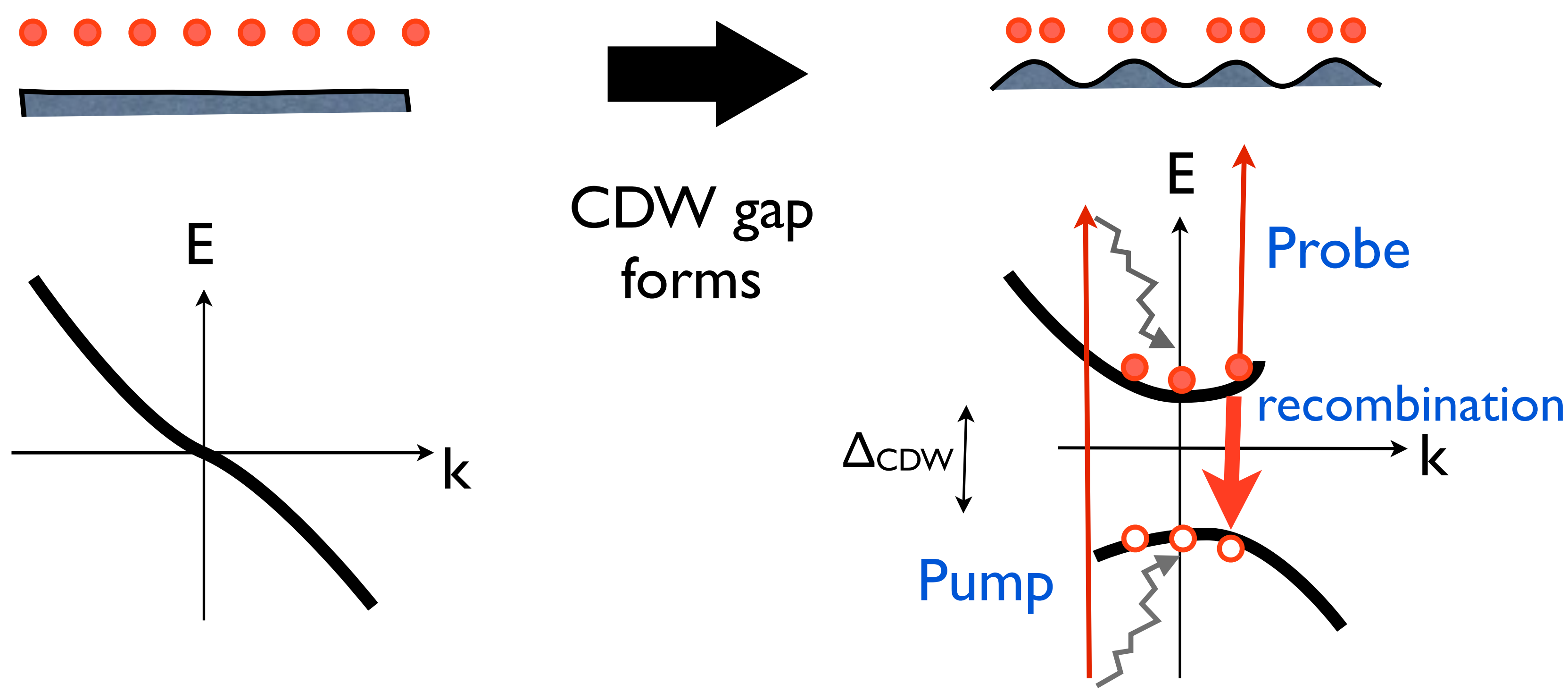
CDWs

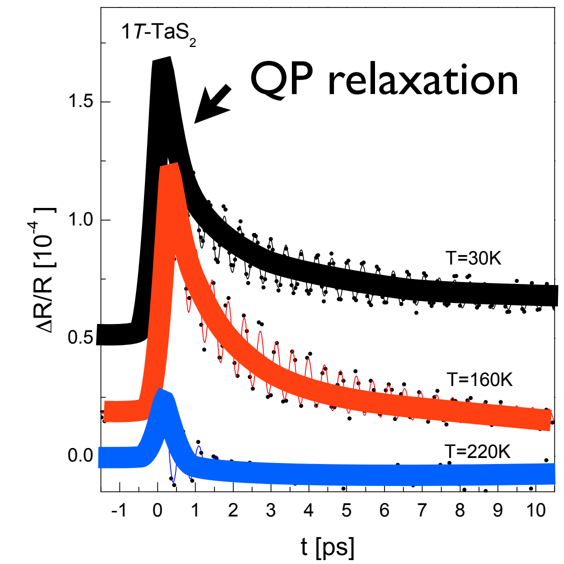


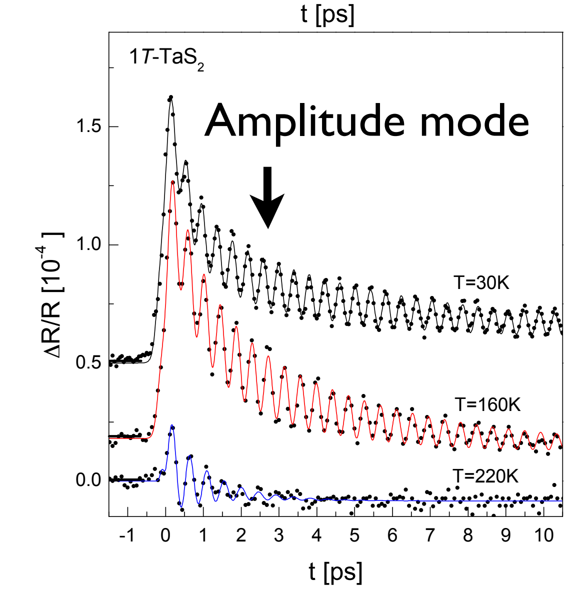
For $\phi \ll 2\pi/\omega_{AM},\ \psi(t) \simeq A(t)$

Detection of the onset of order in CDW systems: The elementary excitations

1. Detection of the gap through quasiparticle (fermionic) excitations



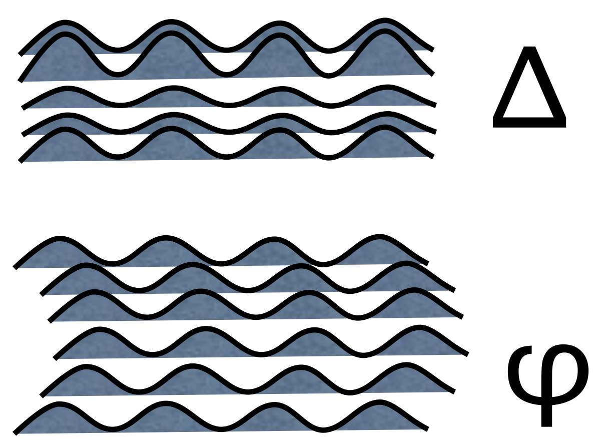




2. Collective mode (bosonic) excitations

The amplitude and phase modes

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



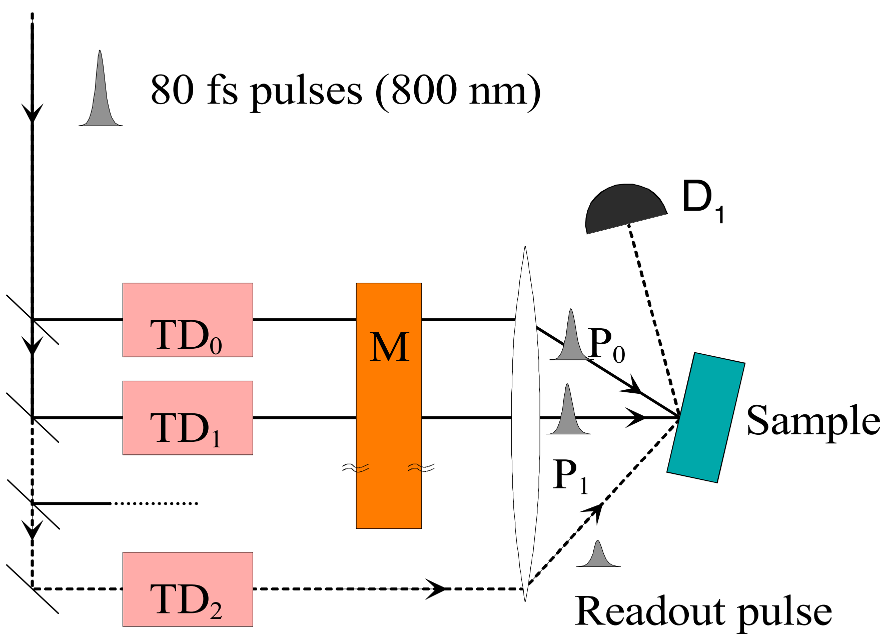
Demsar et al., PRL **83**, 800 (1999).
Demsar, J.,et al, PRB **66**, (2002).
Demsar, et al., PRB **82**, 4918 (1999).
Kusar et al, PRL 101, 227001 (2008)
Yusupov et al.,PRL **101**, 246402 (2008).

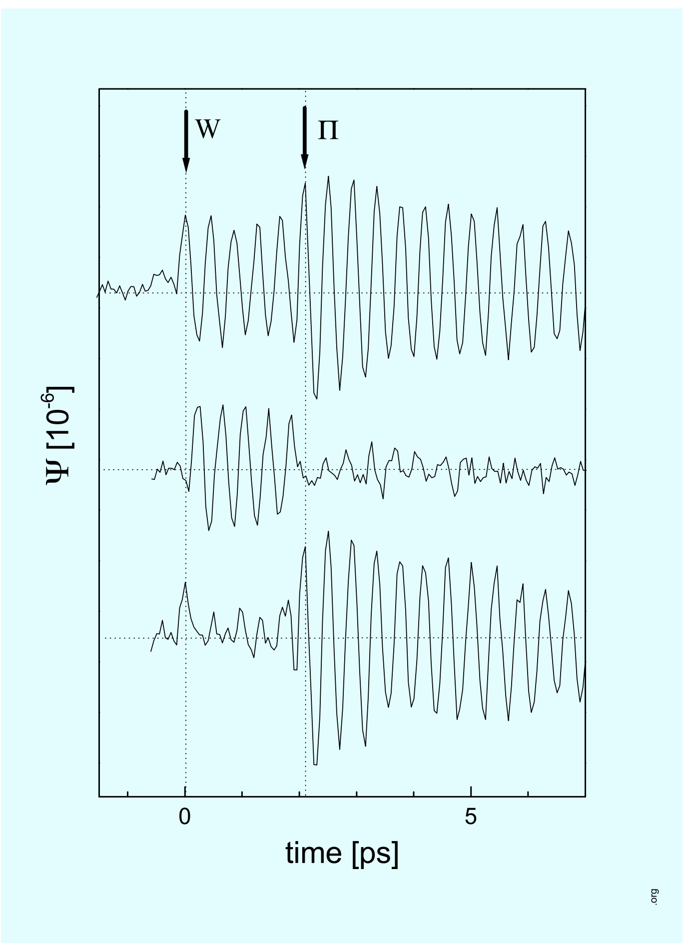
Coherent control and data processing using the Amplitude mode and Grover's superpositional search algorithm

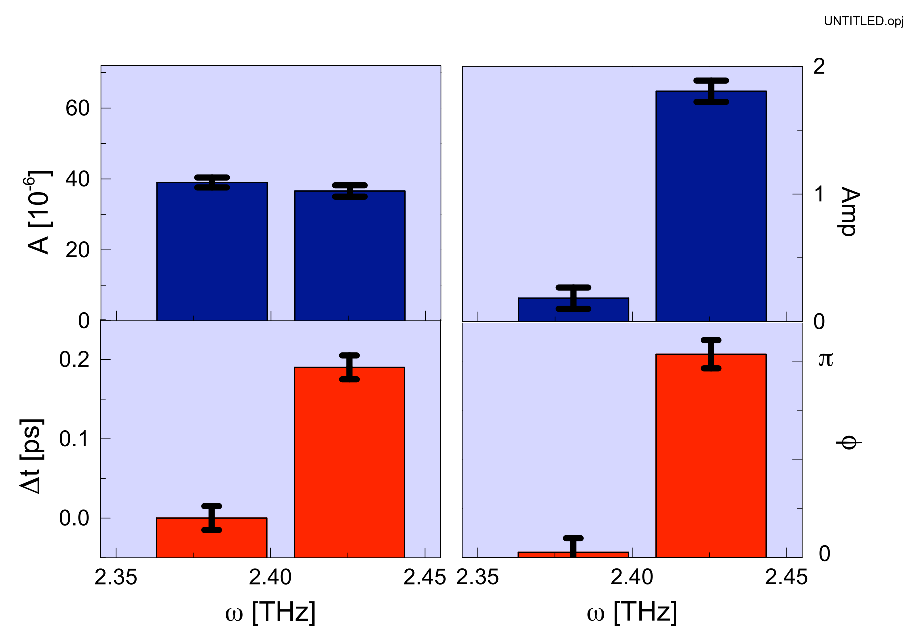
APPLIED PHYSICS LETTERS VOLUME 80, NUMBER 5 4 FEBRUARY 2002

Femtosecond data storage, processing, and search using collective excitations of a macroscopic quantum state

D. Mihailovic,[a)] D. Dvorsek, V. V. Kabanov, and J. Demsar
Jozef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia



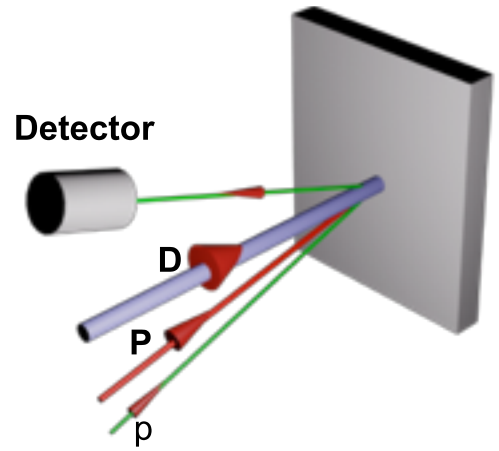


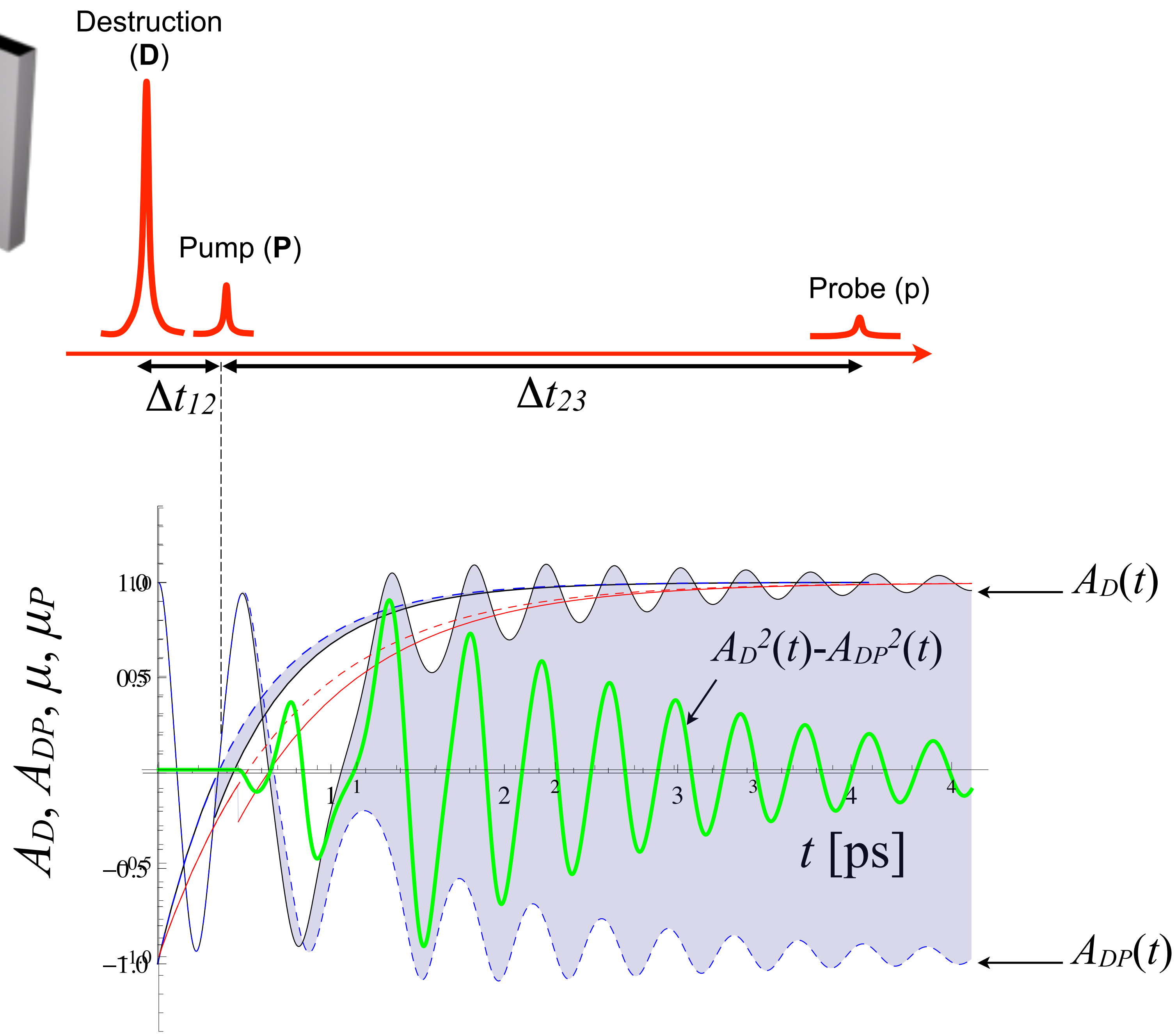


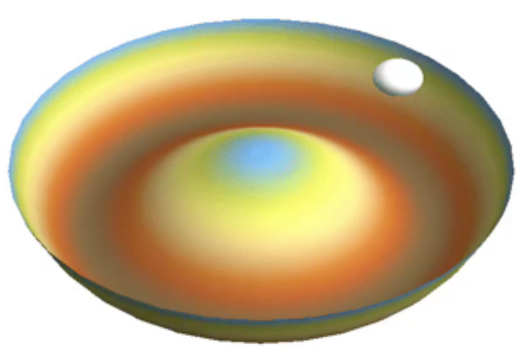
Mihailovic, D. *et al. Appl Phys Lett* **80,** 871–873 (2002).

Ackn. Seth Lloyd (2011)

The calculated optical response of the **collective mode after a quench**

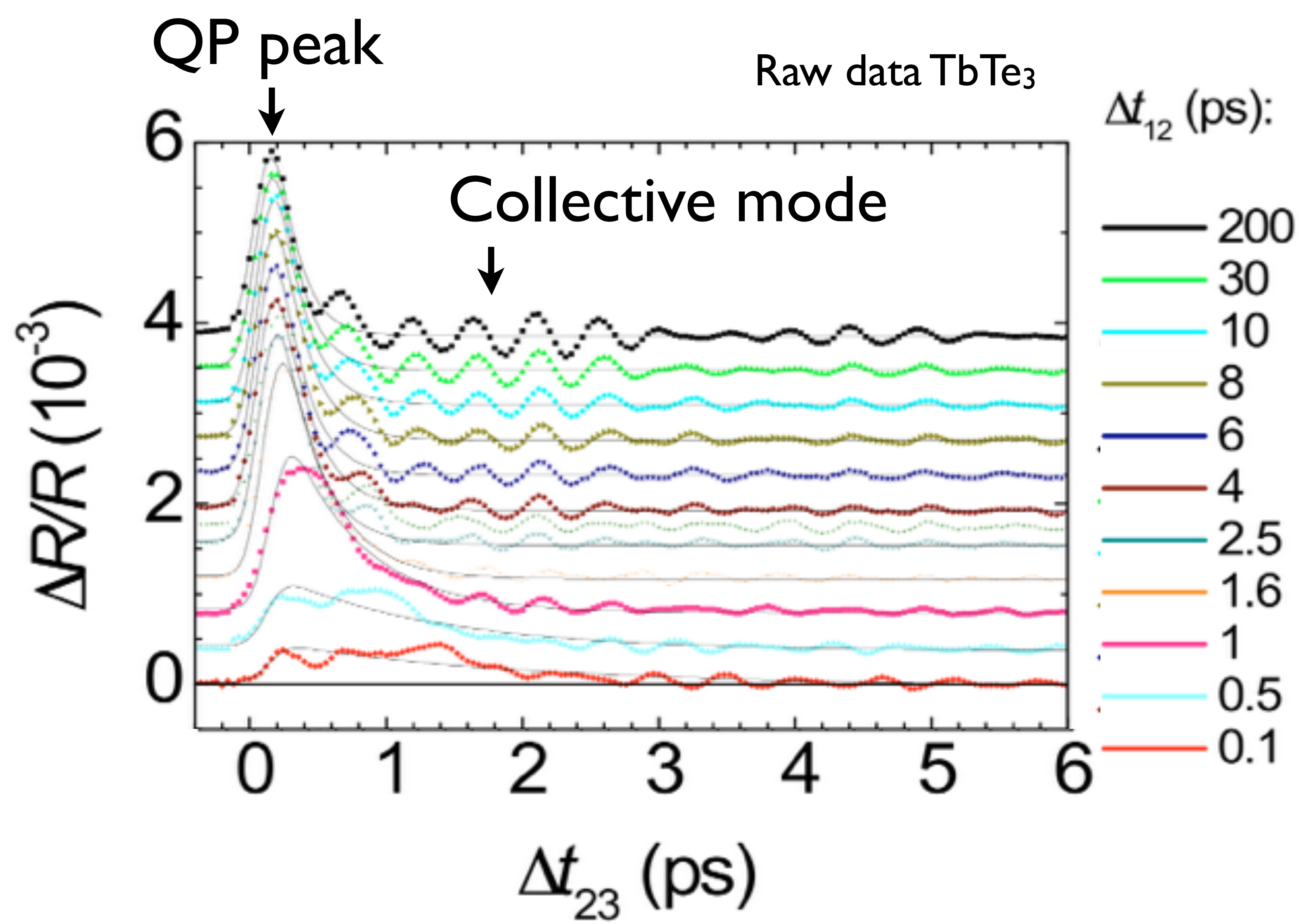






The reflectivity, $\Delta R(t) \propto \left(\frac{\partial R}{\partial \epsilon}\right) \Delta\epsilon \propto \int [A_{DP}^2(t, \mathbf{r}, \Delta t_{12}) - A_D^2(t, \mathbf{r})] e^{-z/\lambda} d^3\mathbf{r}.$

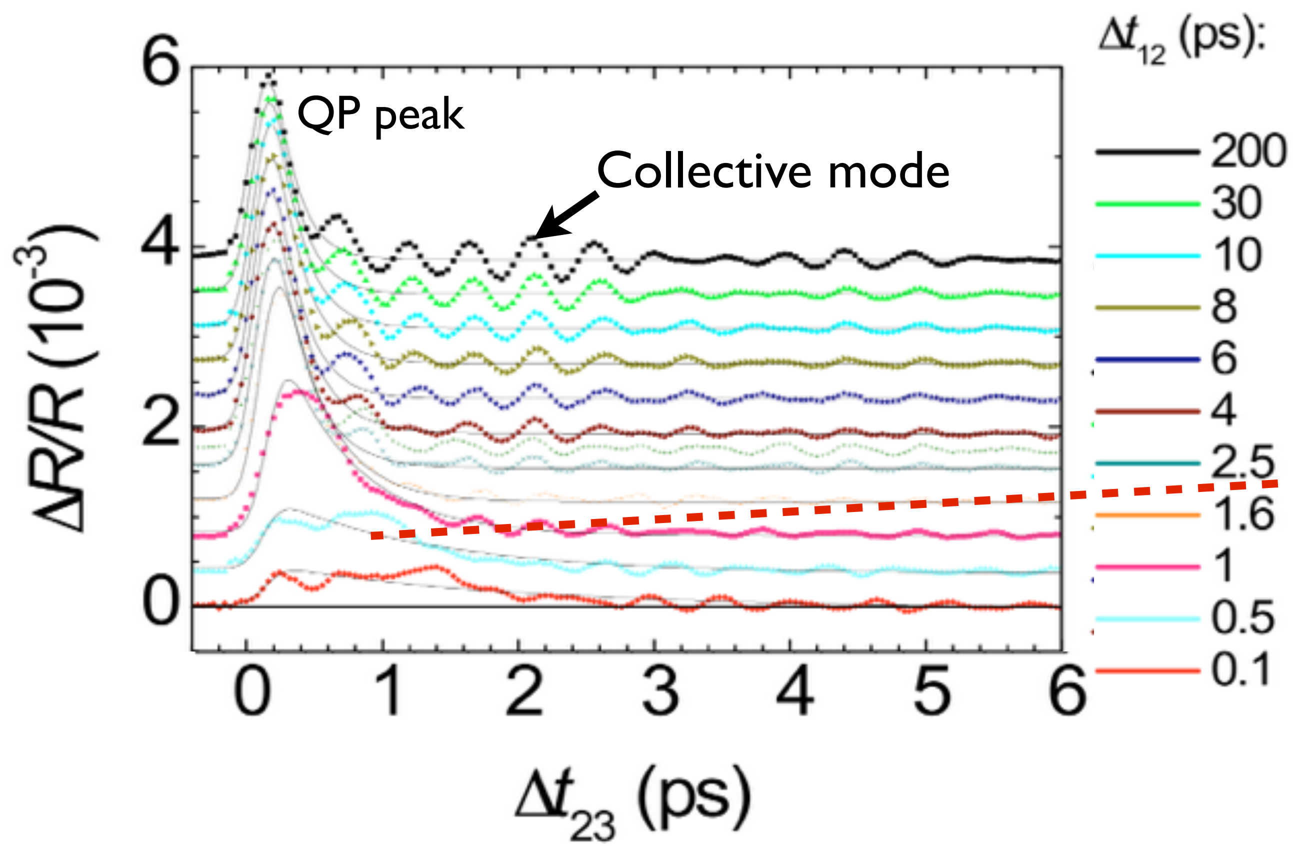
The transient reflectivity $\Delta R/R$ after a quench at $\Delta t_{12}=0$

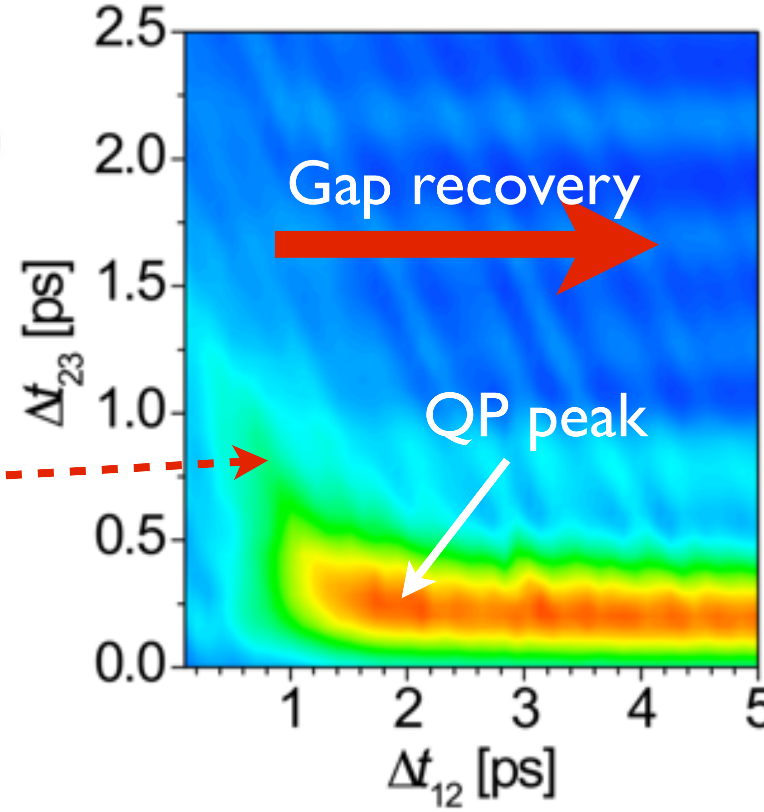


Delay time after the D pulse

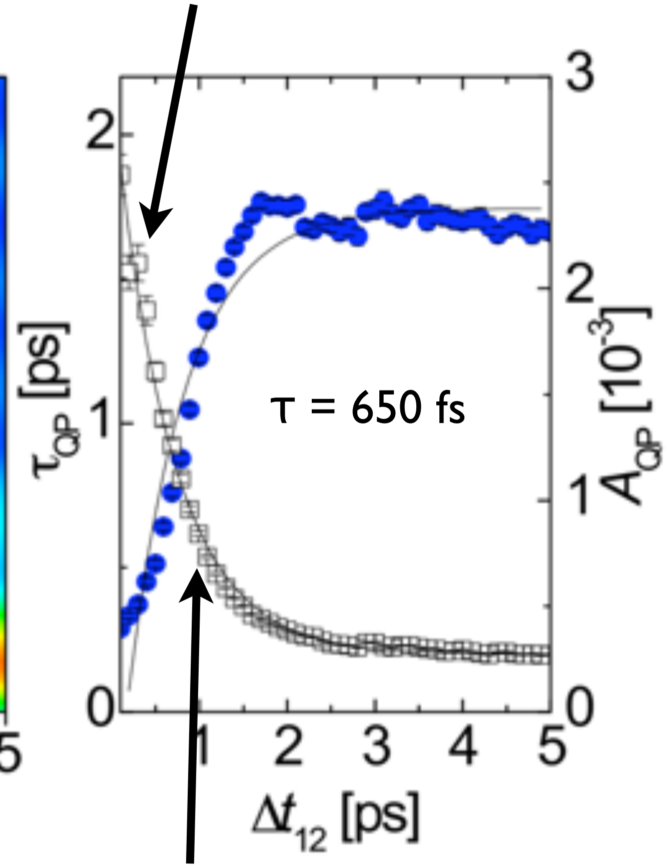
Quasi-particle (Fermion) kinetics: gap recovery

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





time after quench→



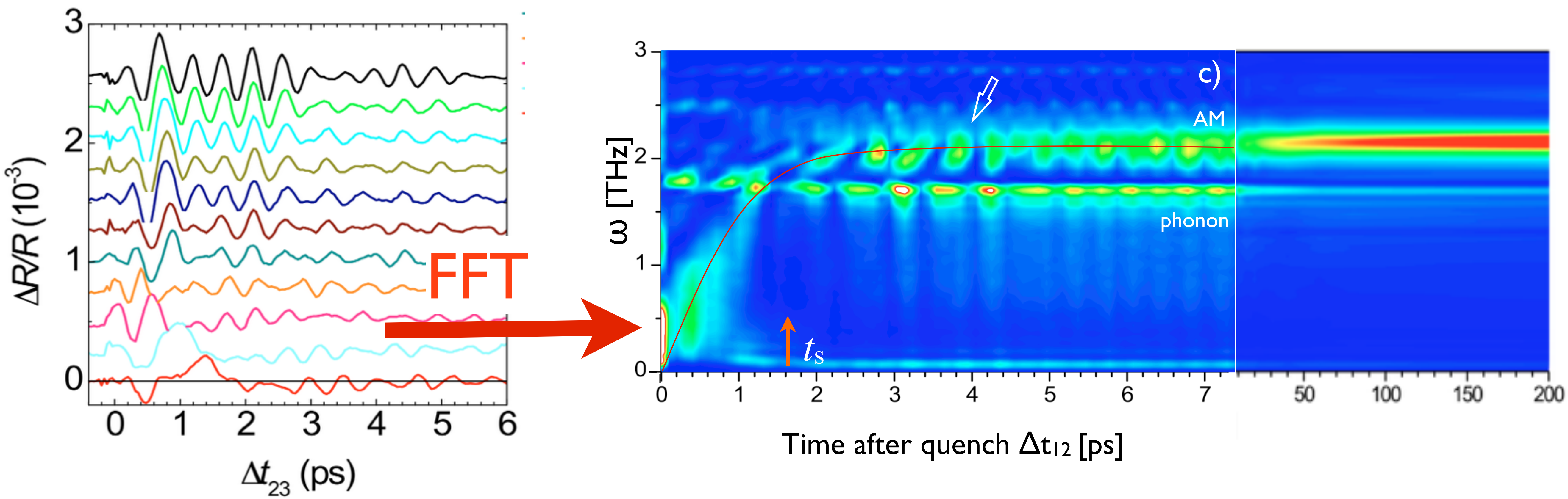
Relaxation time recovery

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

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 spectrum as a function of time after quench



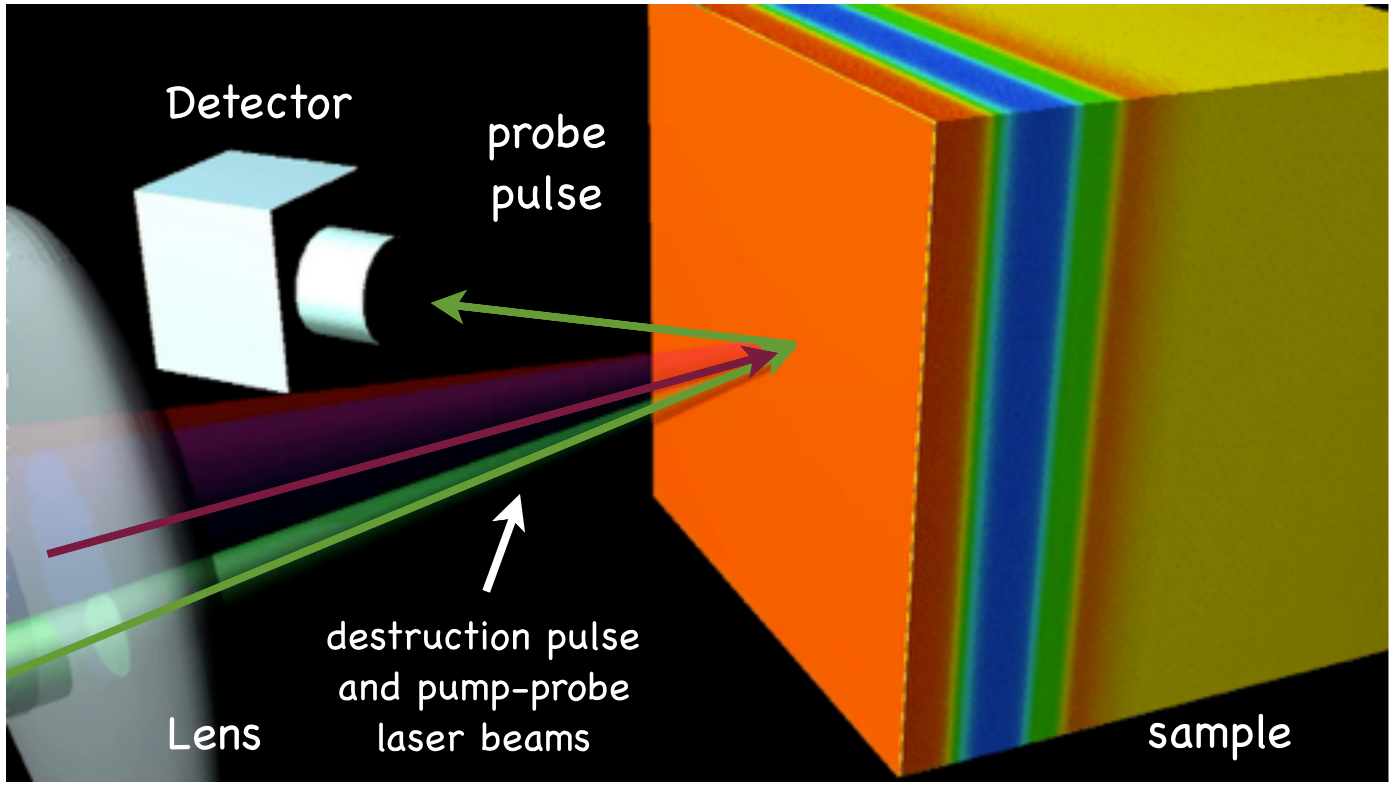
The most obvious feature:
oscillations of intensity of the collective mode

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:

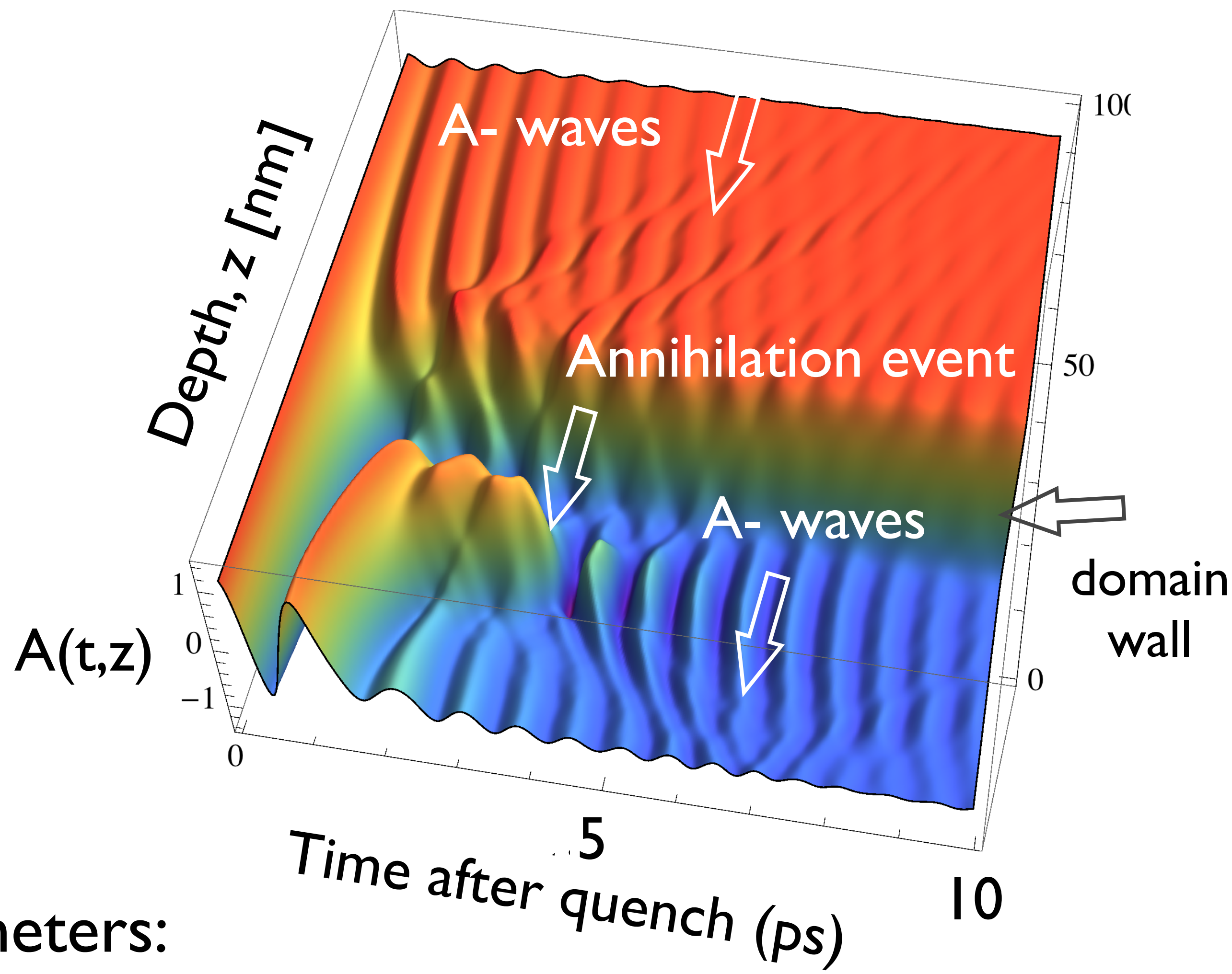
$\tau_{QP} = 650$ fs
$\omega_0/2\pi = 2.18$ THz
$\eta = 2$
$\alpha = 0.1$

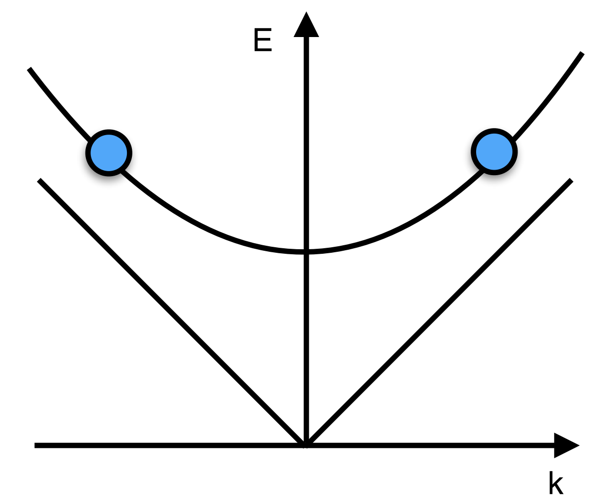
Order parameter calculation

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:

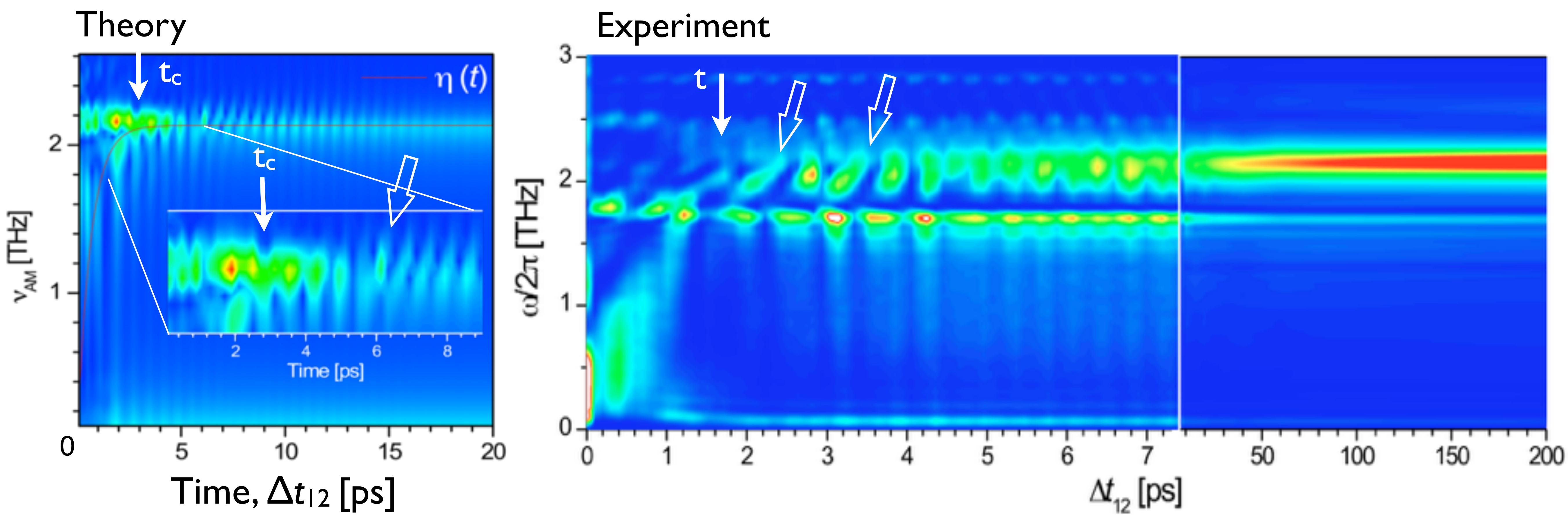




Experimental parameters:

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

Order parameter dynamics: theory vs. expt.



Theory predictions:

Oscillations of Δ or |Ψ|

Critical slowing down

Collective mode softening

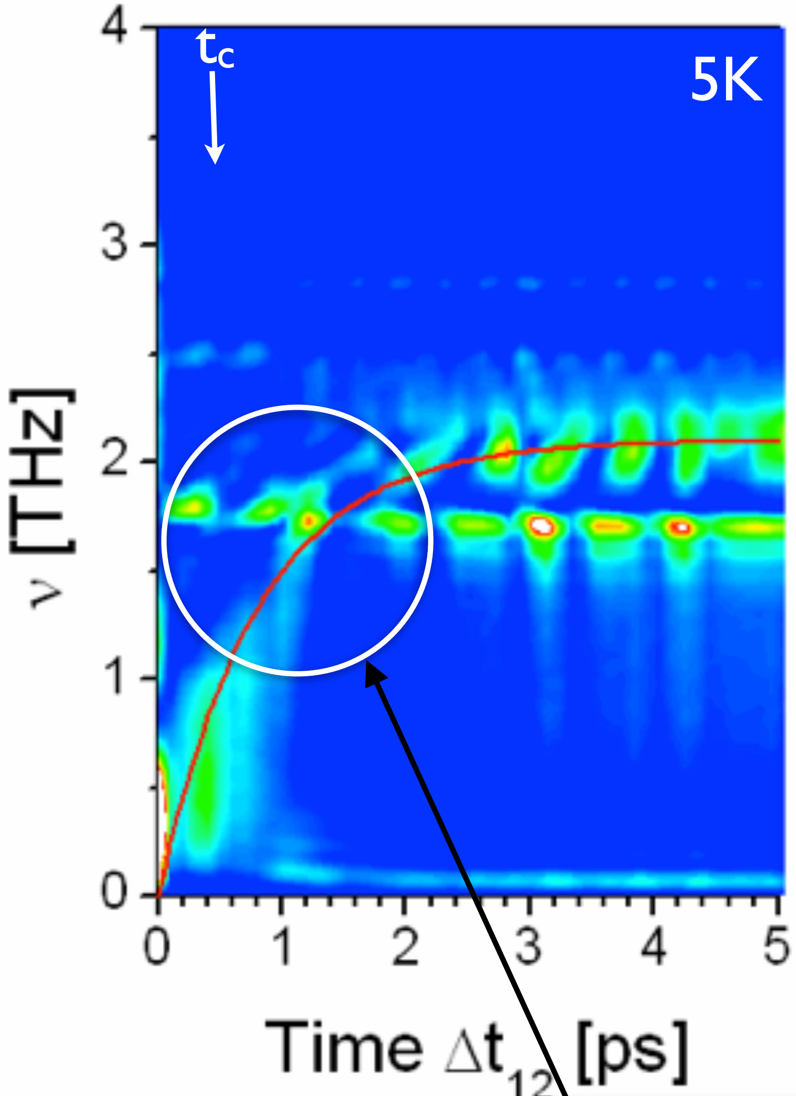
Domain formation

Ψ Higgs waves +

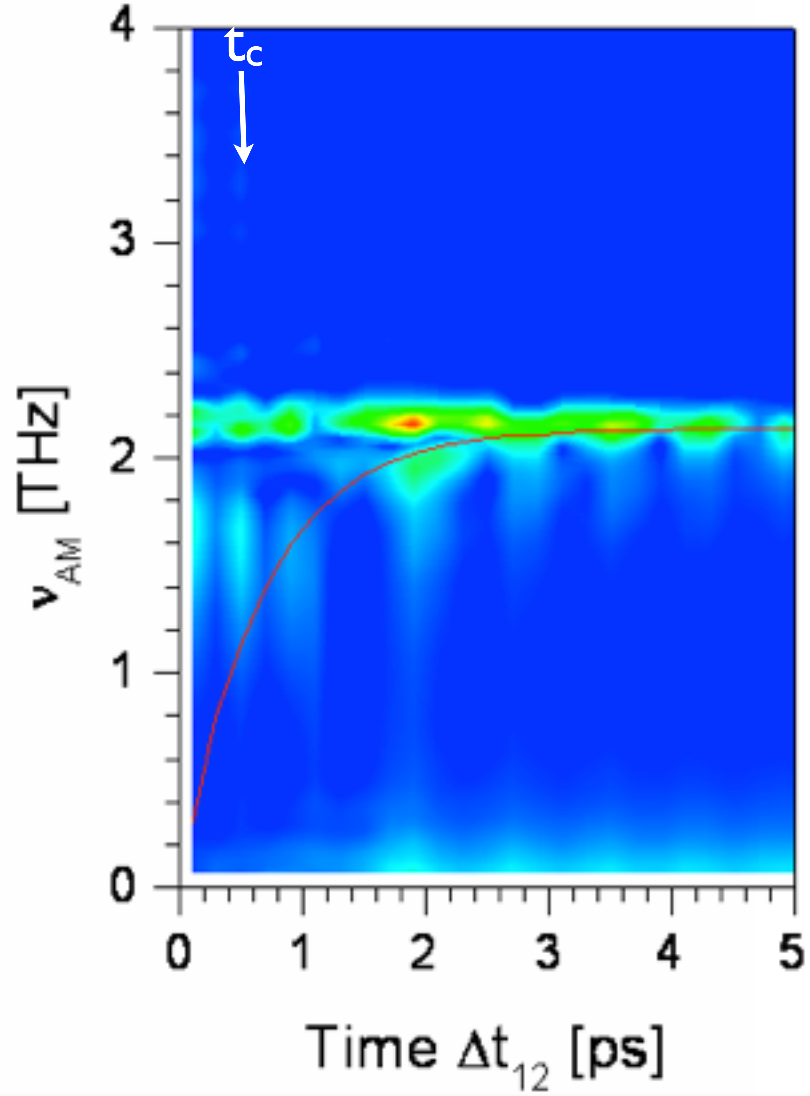
The first three picoseconds:

Critical dynamics as $t \to t_c$

Experimental data



Theory

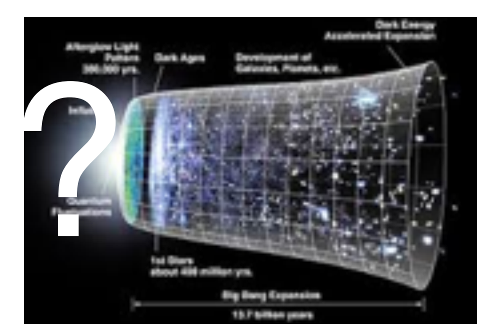


<u>ISSUES NOT ADDRESSED:</u>

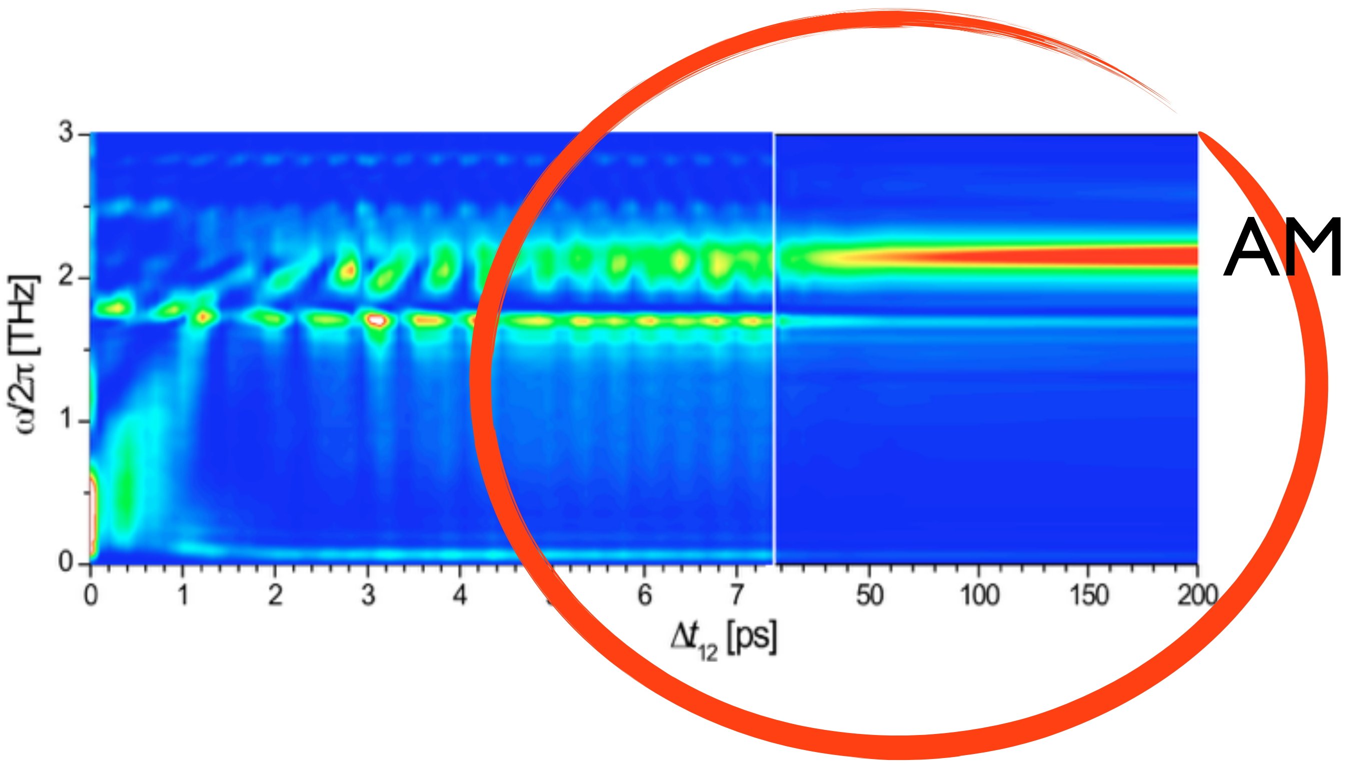
- Response selection rules: AM has A symmetry only $t > t_c$
- coupling to other pre-existing collective modes (phonons)
- initial energy relaxation
- fluctuation phenomena

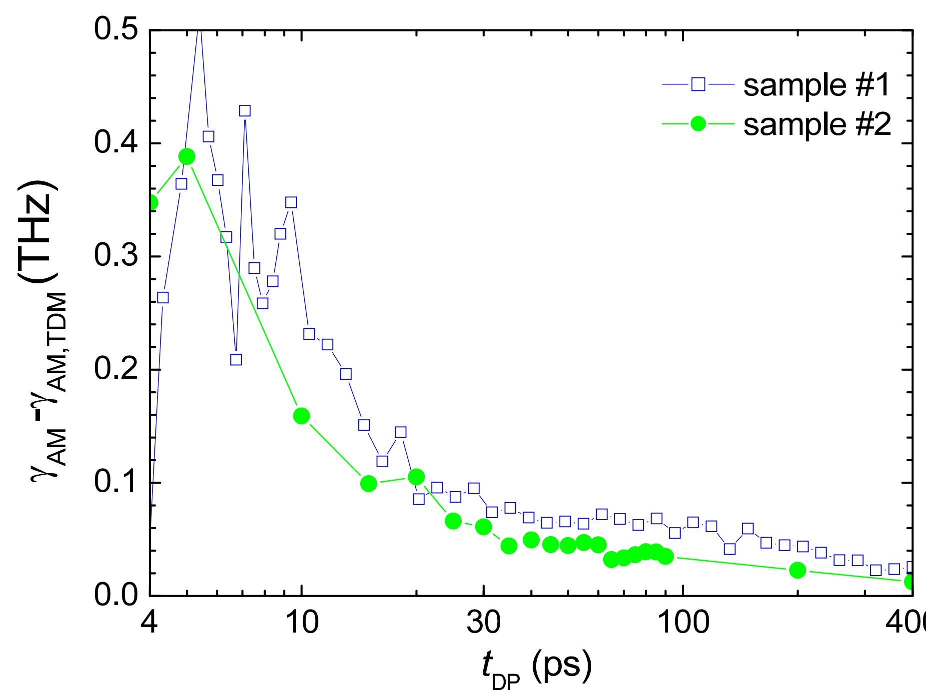
Coupling of AM to a collective mode from a previous eon!

(Different from Penrose's eons. More like Littlewood-Varma-Klein NbSe2 resonance)



Incoherent topological defect dynamics: collective mode broadening for $\Delta t_{12} > 7$ ps



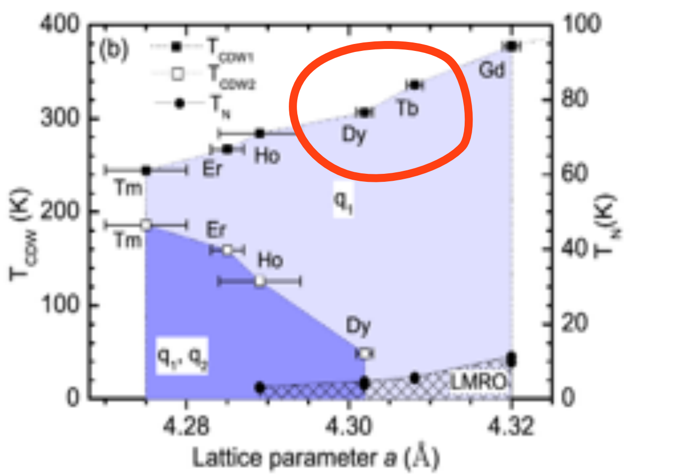


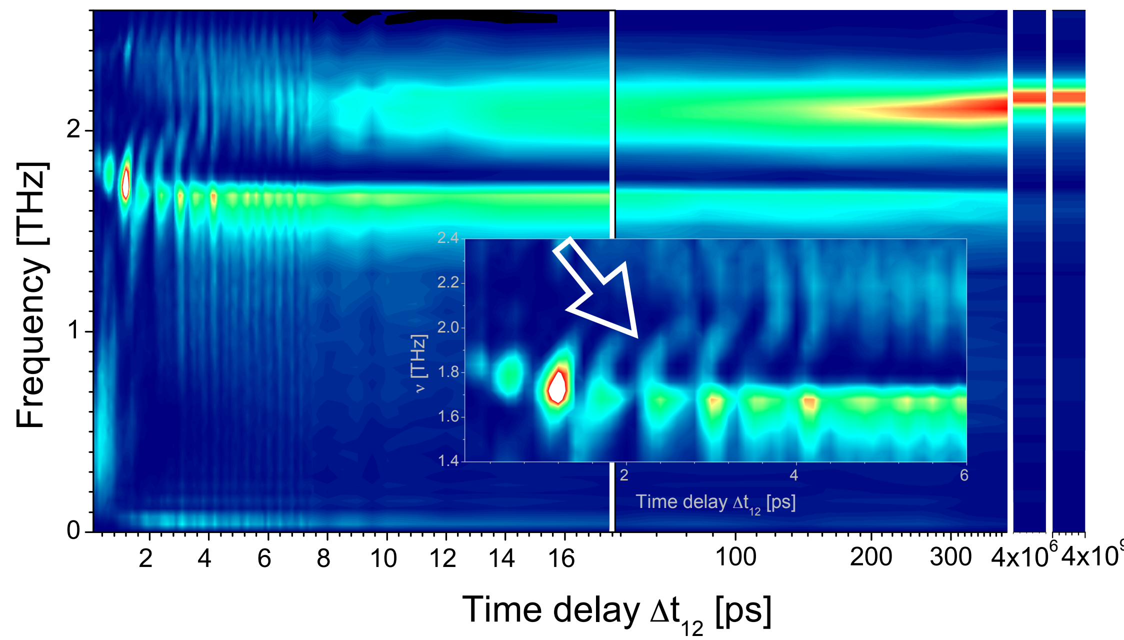
The collective mode linewidth reflects the presence of domain walls

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

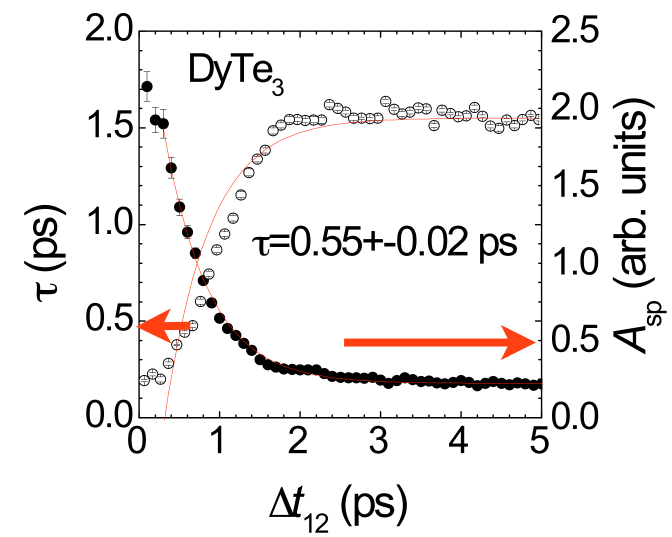
Related systems show (not so subtle) differences

$DyTe_3$ <u>Very</u> similar to $TbTe_3$



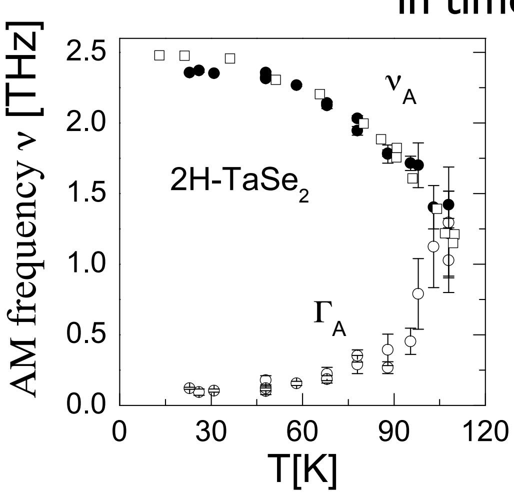


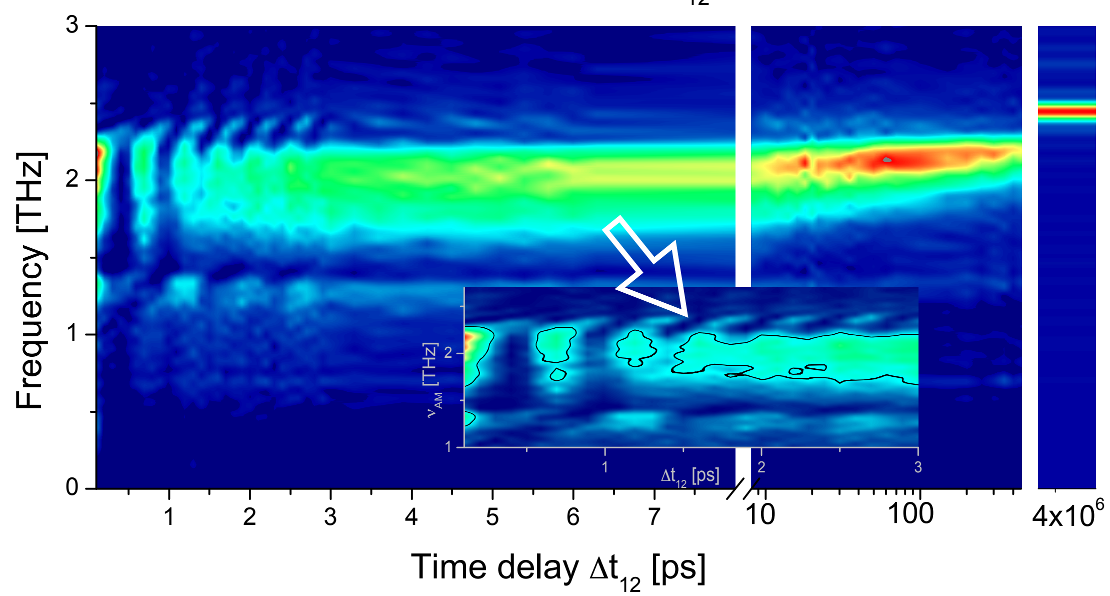
Gap closure (QP dynamics)

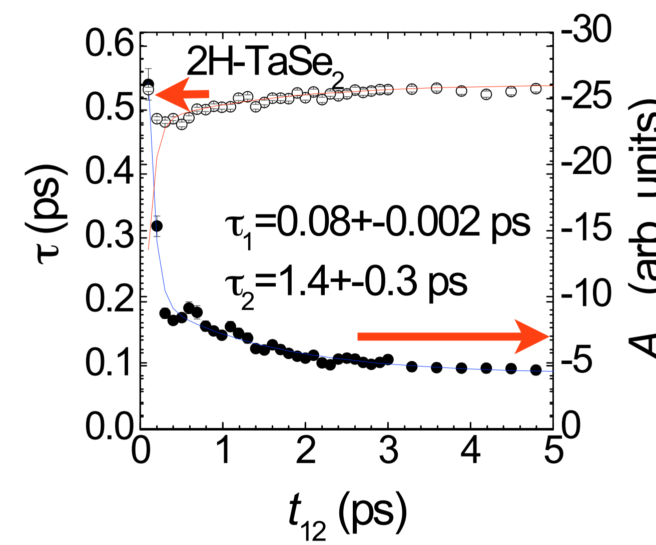


2H-$TaSe_2$

2D system
No soft mode
in time

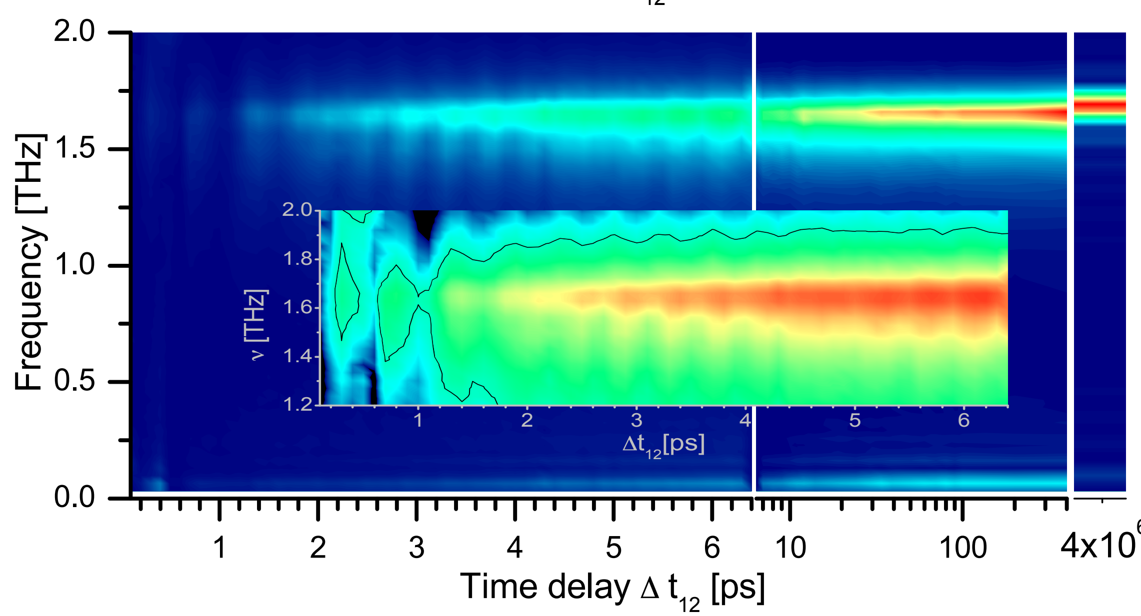


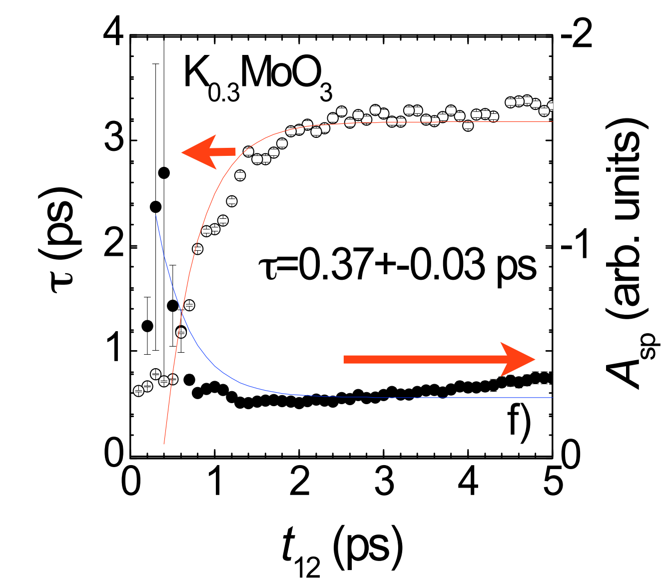




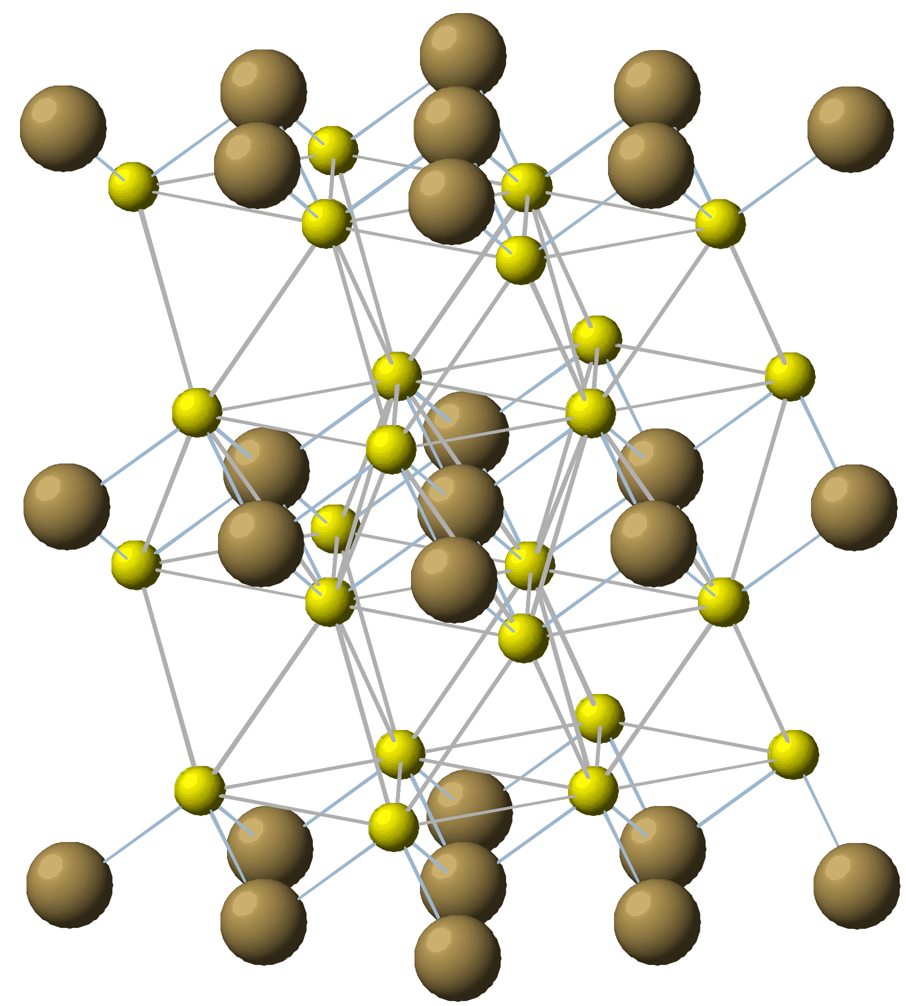
$K_{0.3}MoO_3$

1D chains
only 10% softening of the AM





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



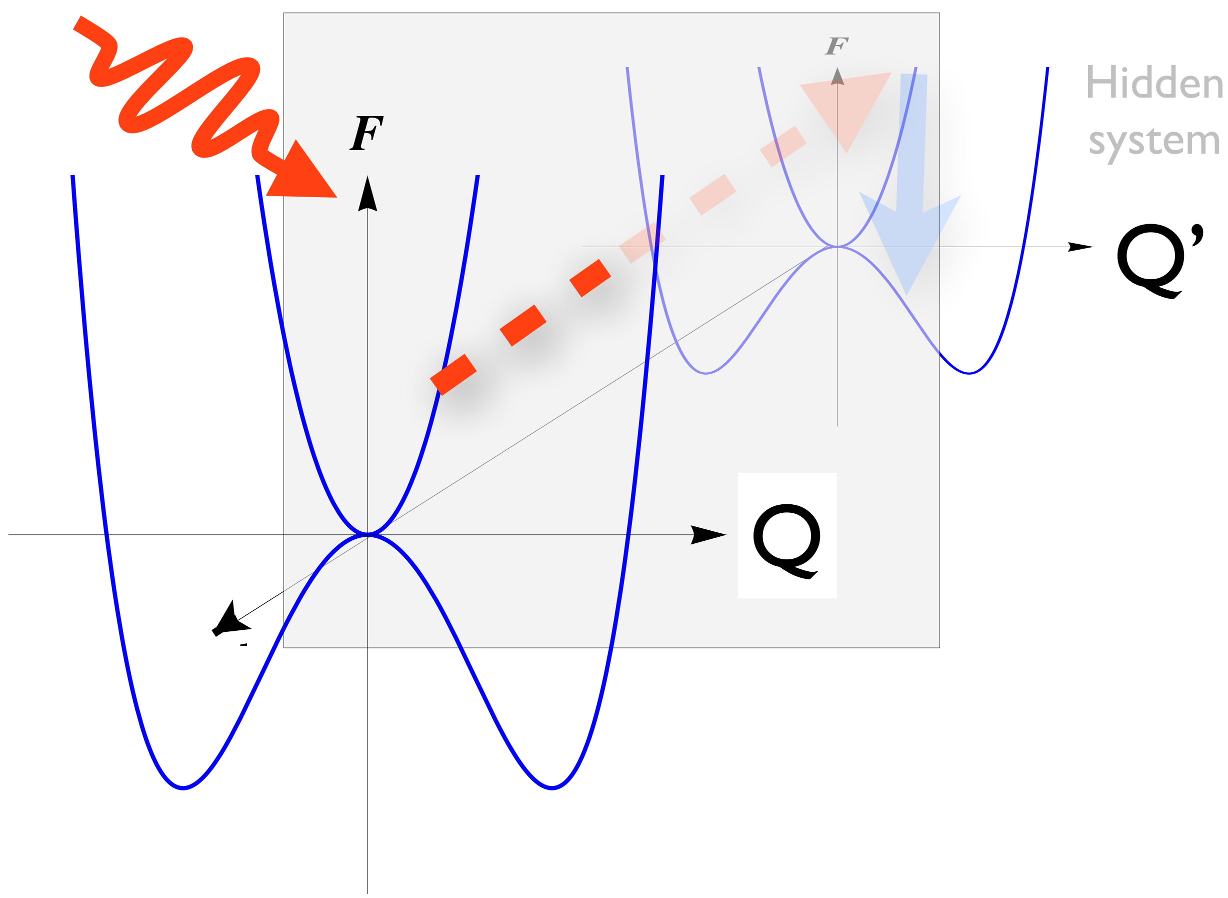
1T-TaS_2

Stojchevska et al, Science **344**, 177 (2014).

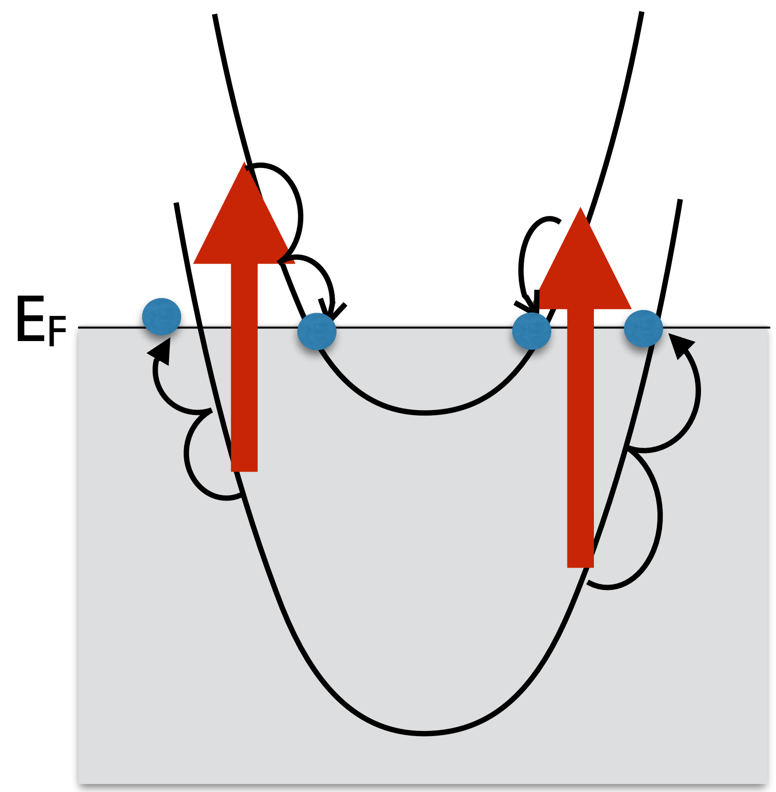
What is a hidden state?

It is a state of matter which cannot be reached under ergodic conditions, by slowly changing *T*, *P*, *B*-field, etc.

Switching to a hidden state can be achieved by a **non-thermal process** which occurs under highly non-equilibrium conditions of the underlying vacuum

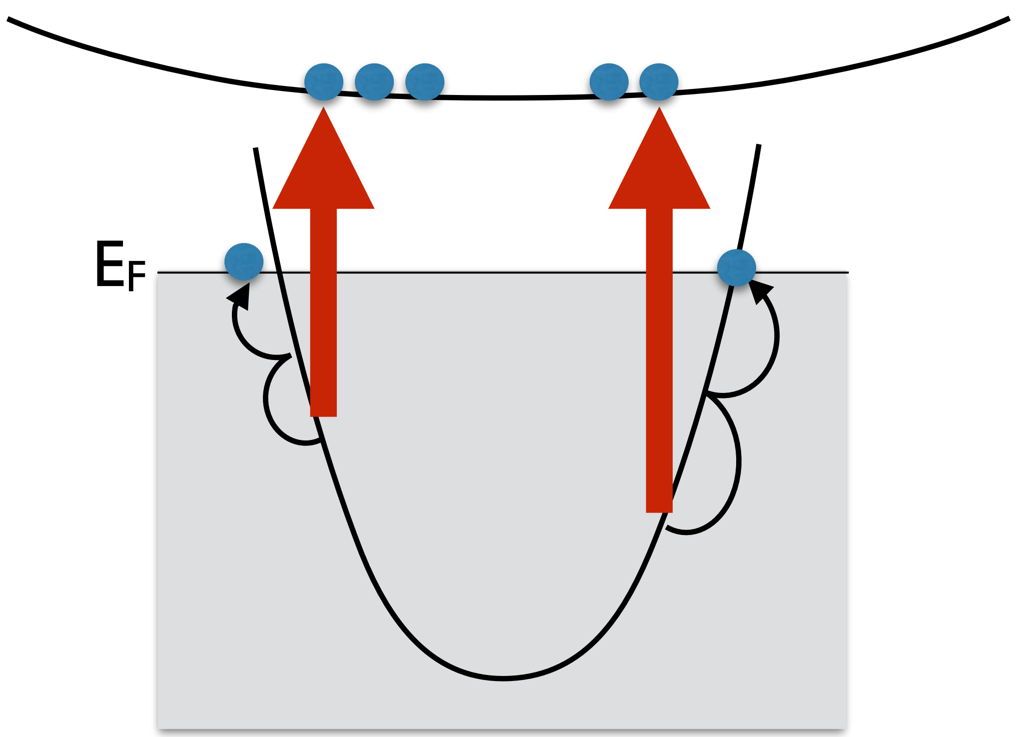


The importance of e-h (a)symmetry for creating photoinduced states

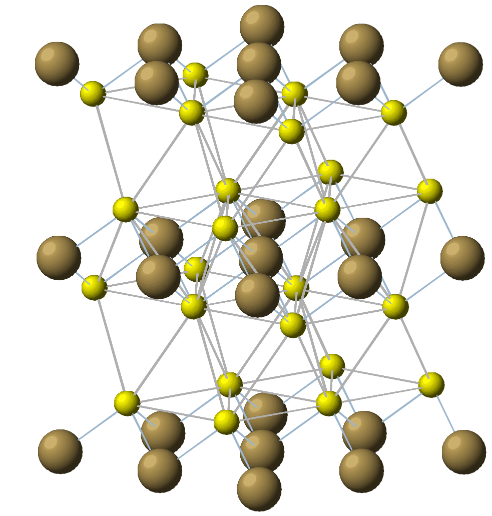


Just heating ($T_e^* = T_L^* = \ldots$).
No doping.

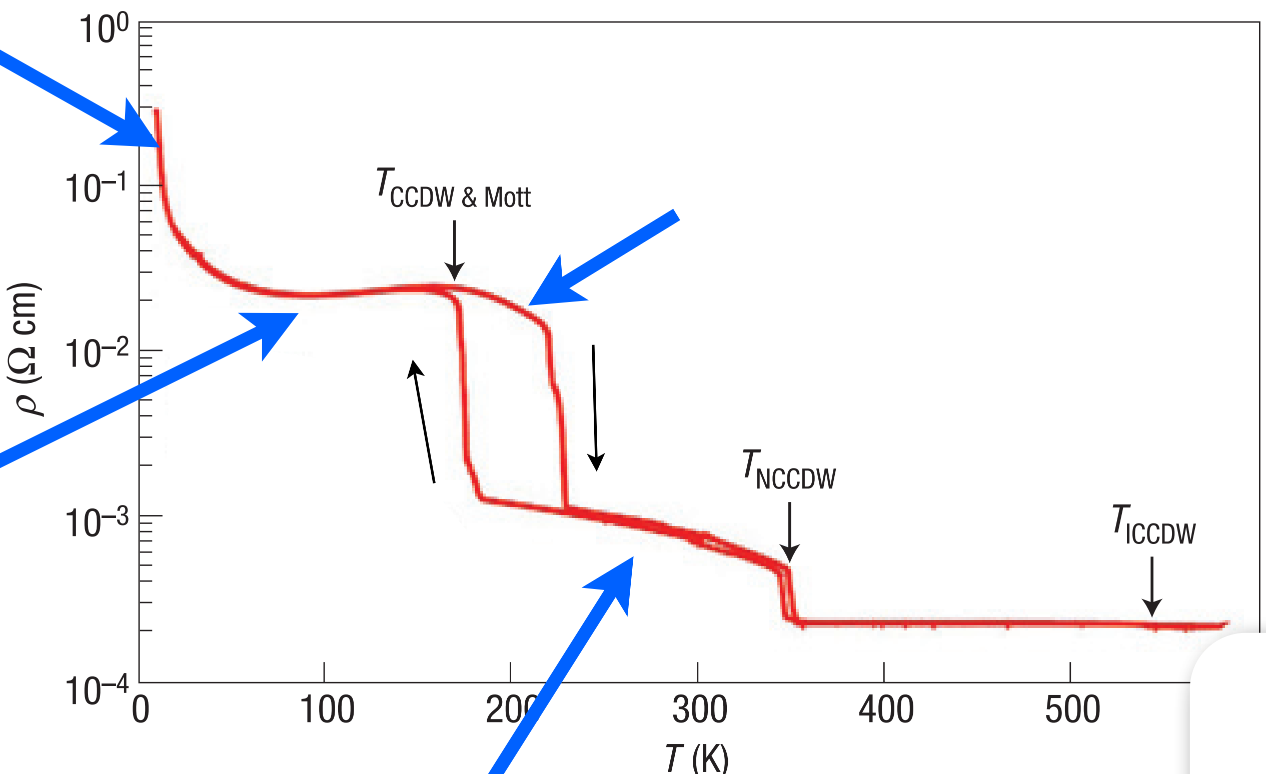
The importance of e-h (a)symmetry for creating photoinduced states



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

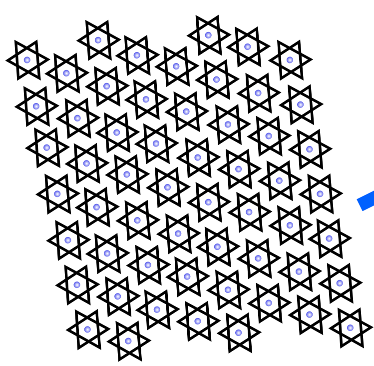


Resistivity of $1T$-TaS_2

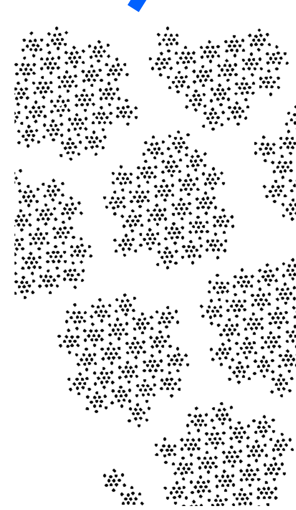


Mott state

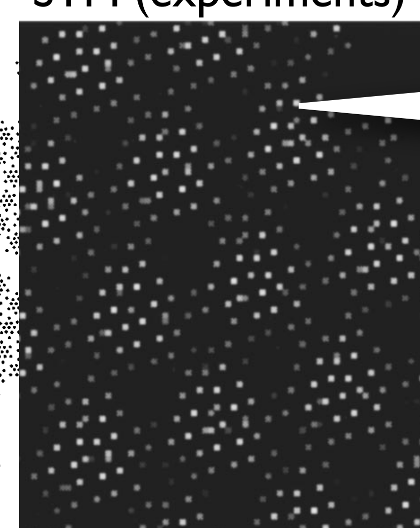
Honeycomb structure
(Commensurate CDW phase)
Tossati and Fazekas (1974)



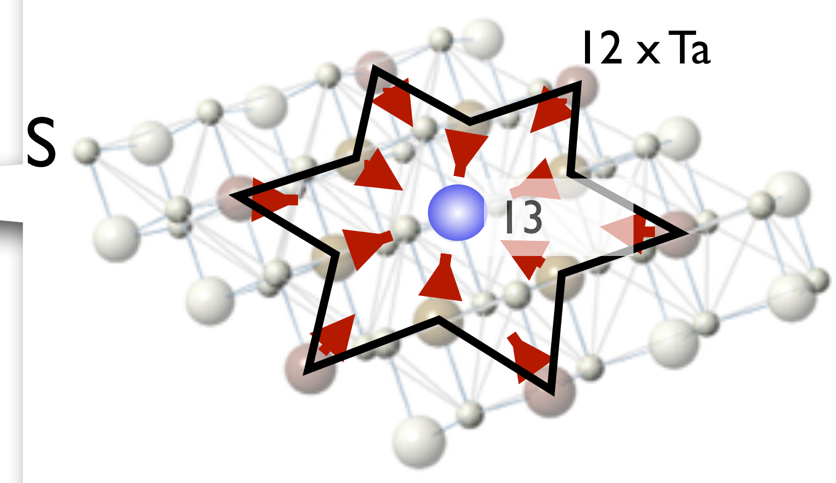
Stretched honeycomb structure of C phase
with domain walls in between
(Nearly Commensurate CDW phase)
Wilson et al



STM (experiments)

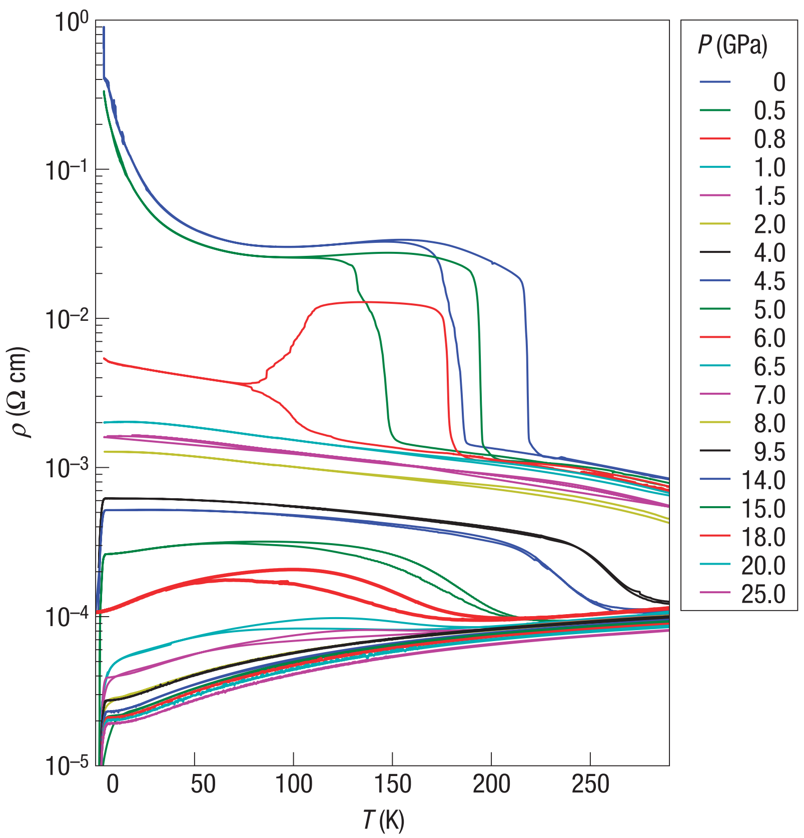


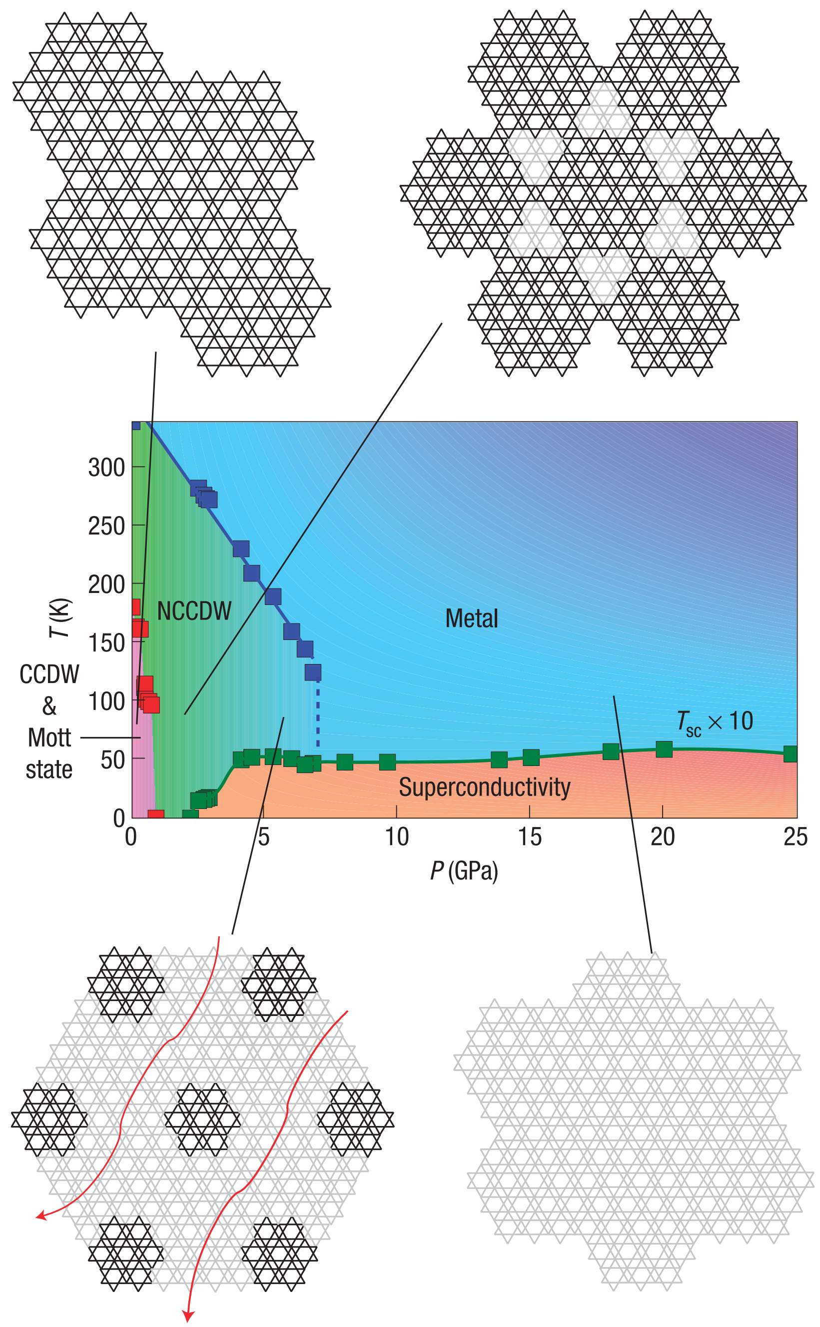
The polaron in $1T$-TaS_2



Other nearby states in $1T\text{-}TaS_2$: Superconductivity under pressure, or Fe doping

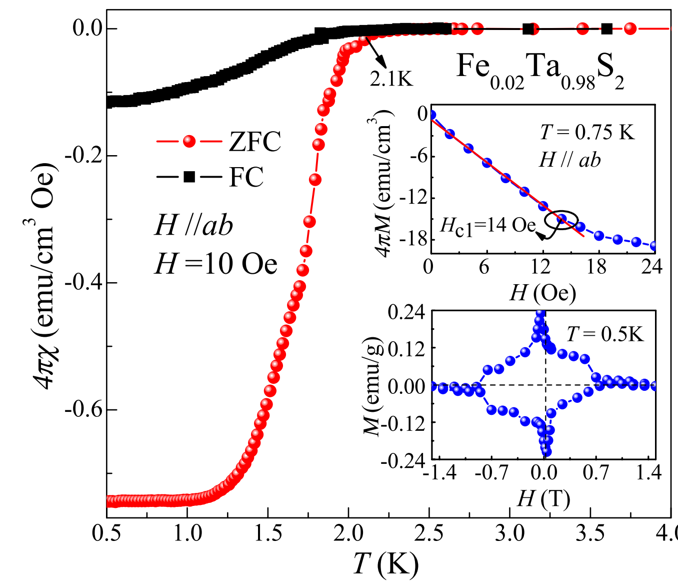
Pressure:

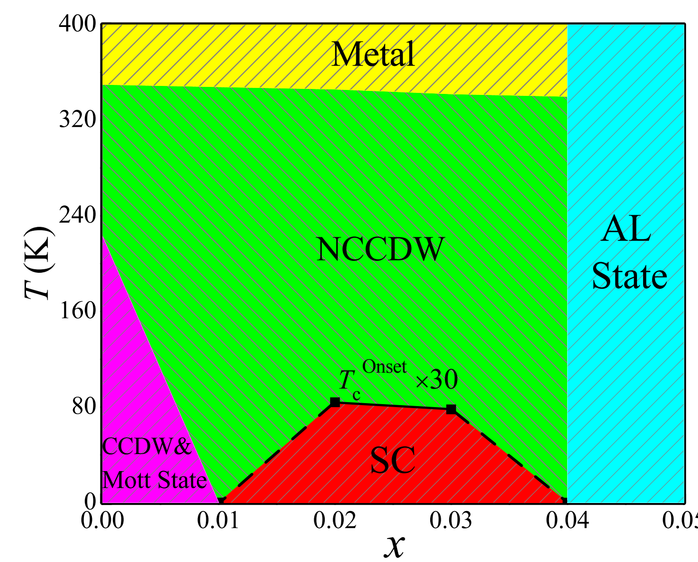




Sipos et al (Nat.Mat. 2008)

Fe doping:

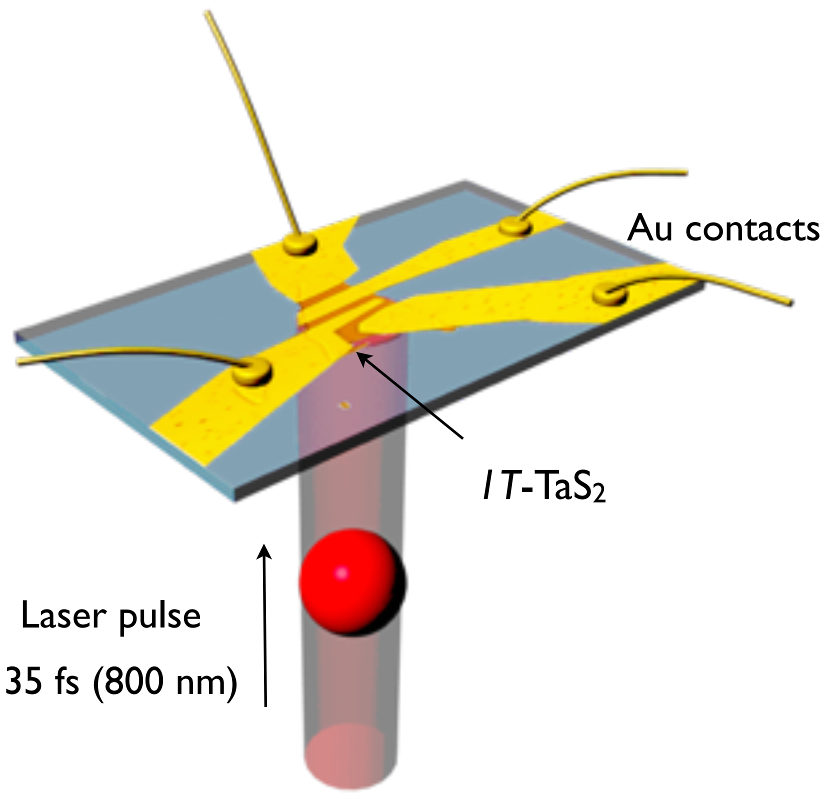


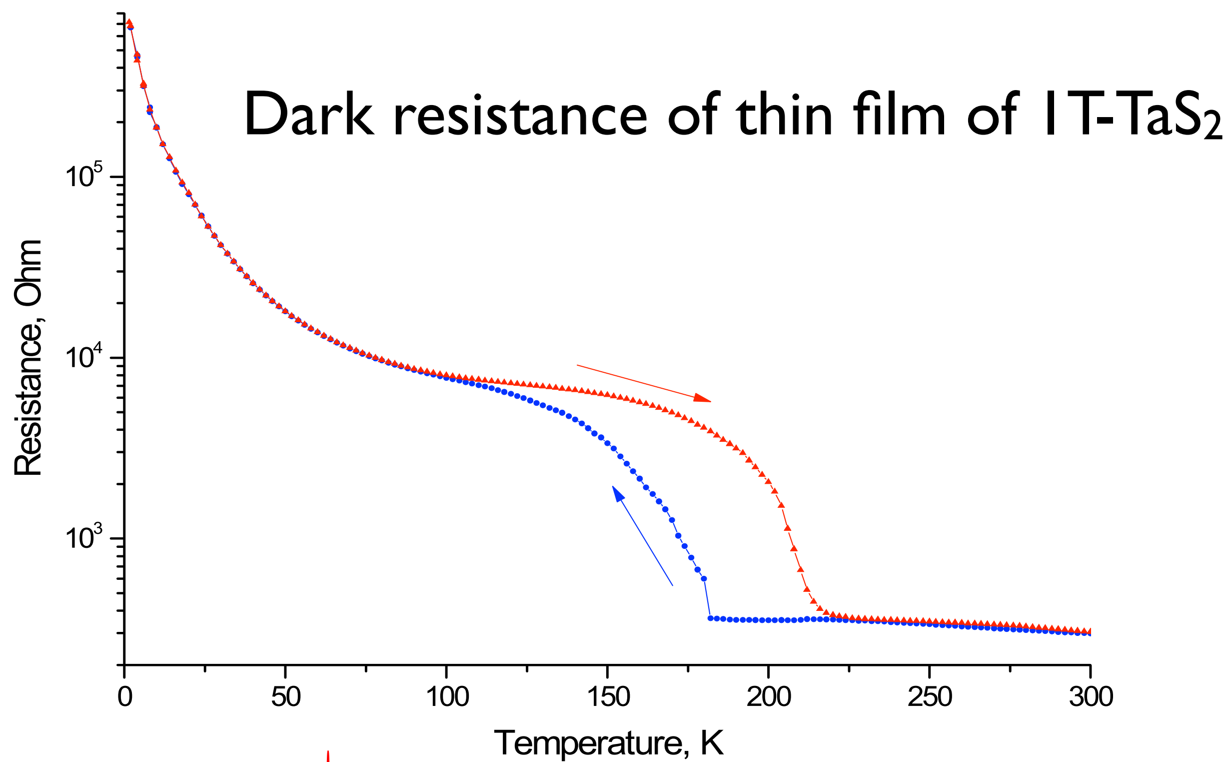


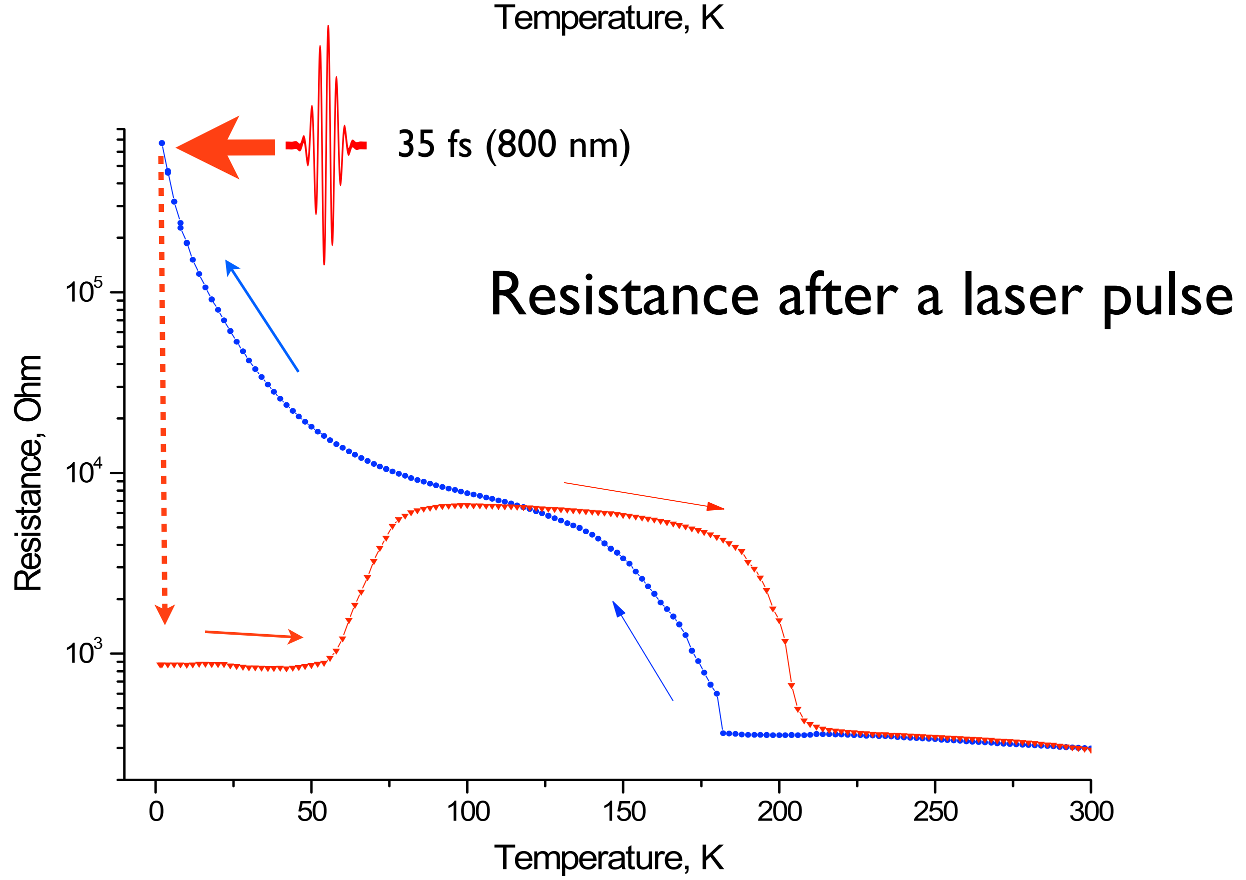
Li et al. EPL 2012

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

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

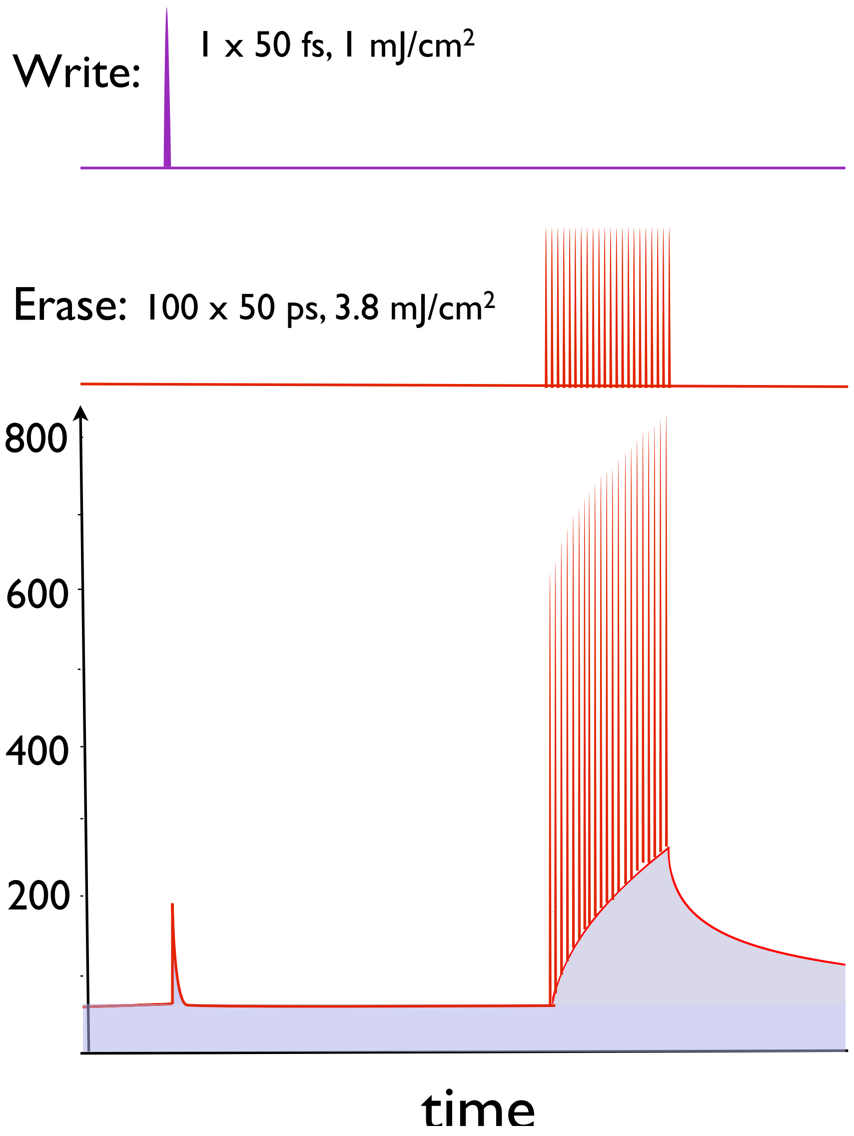


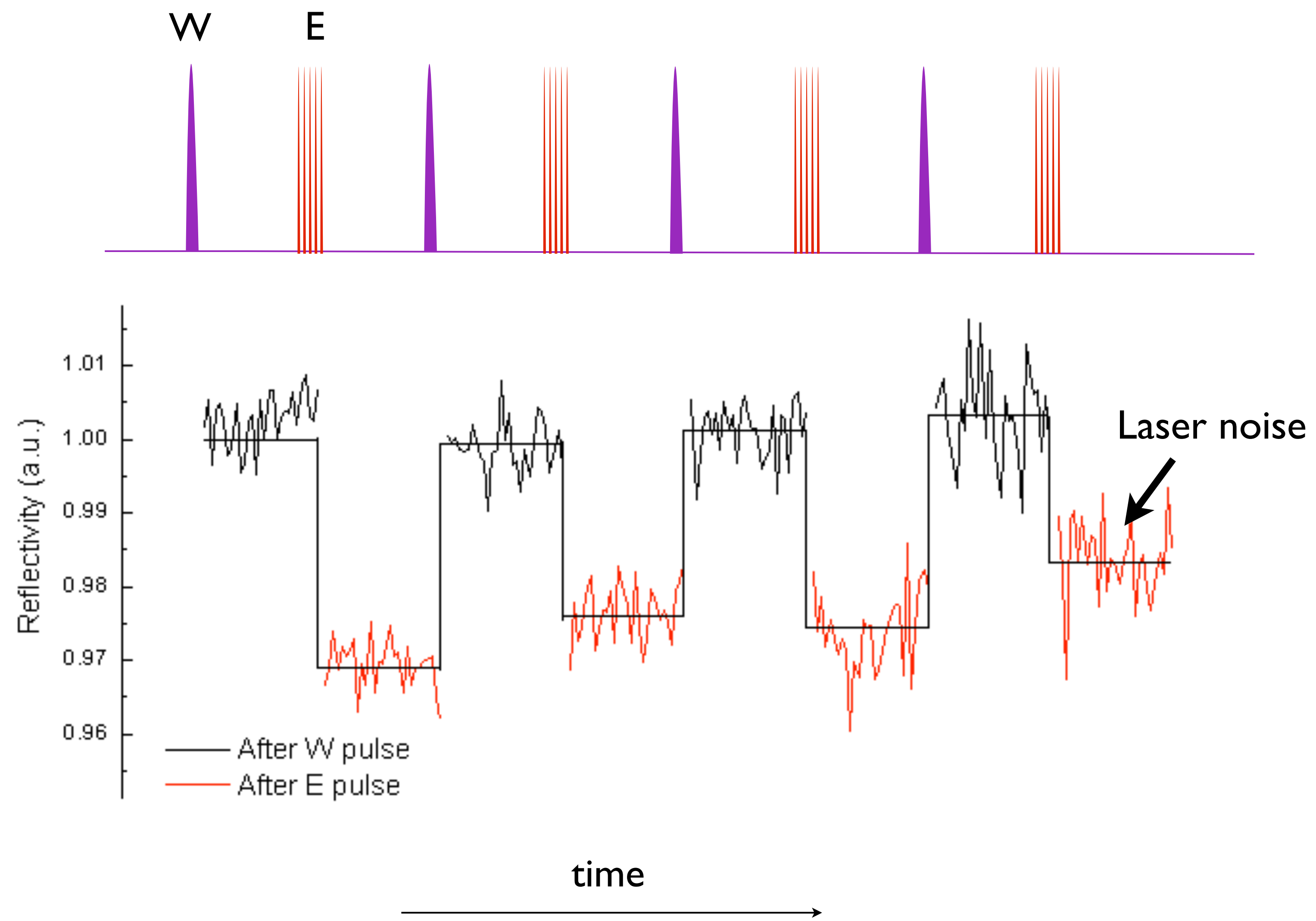




Stojchevska et al, Science **344**, 177 (2014).

Reflectivity switching by laser pulses

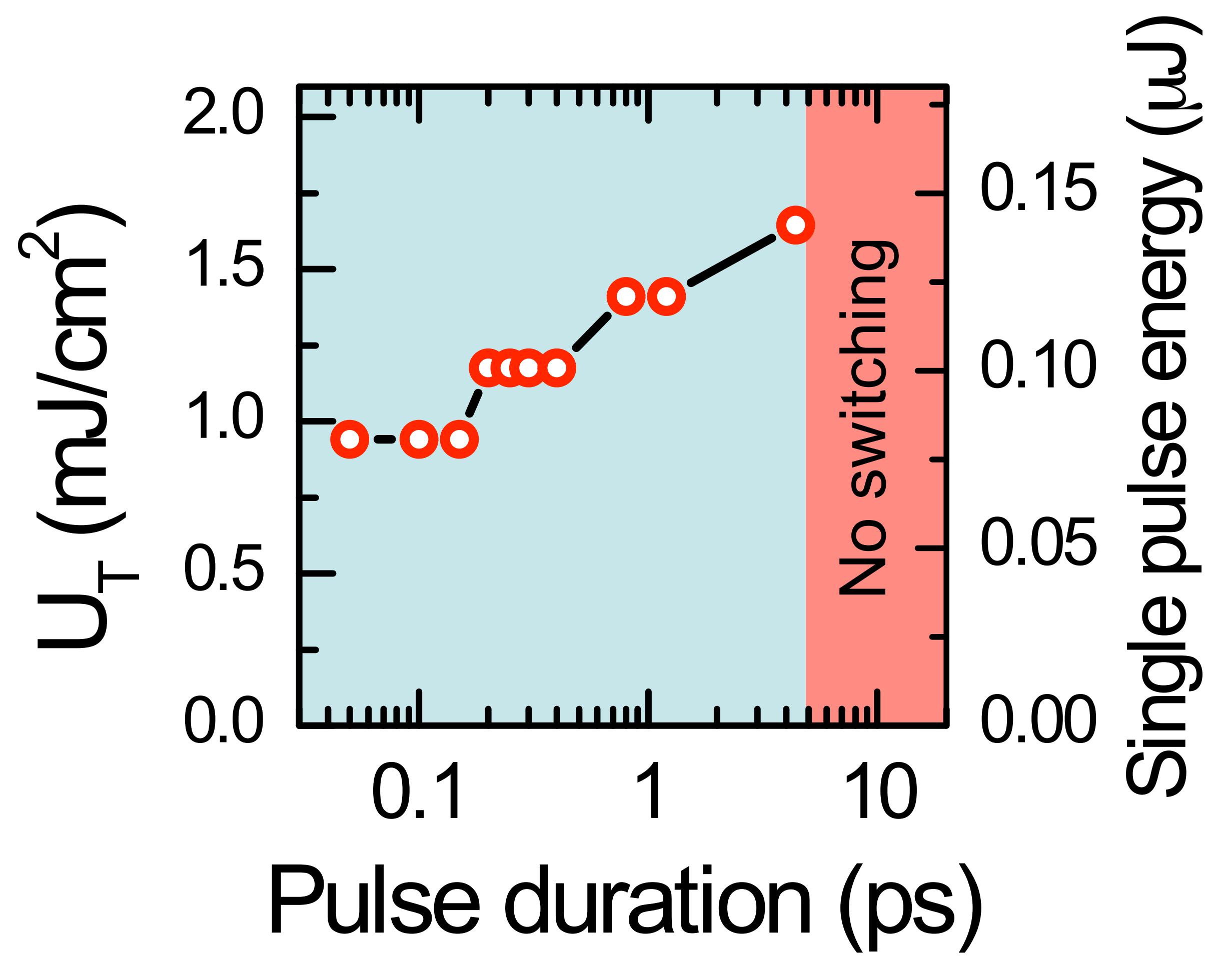




Switching is accompanied by a change in dielectric constant.

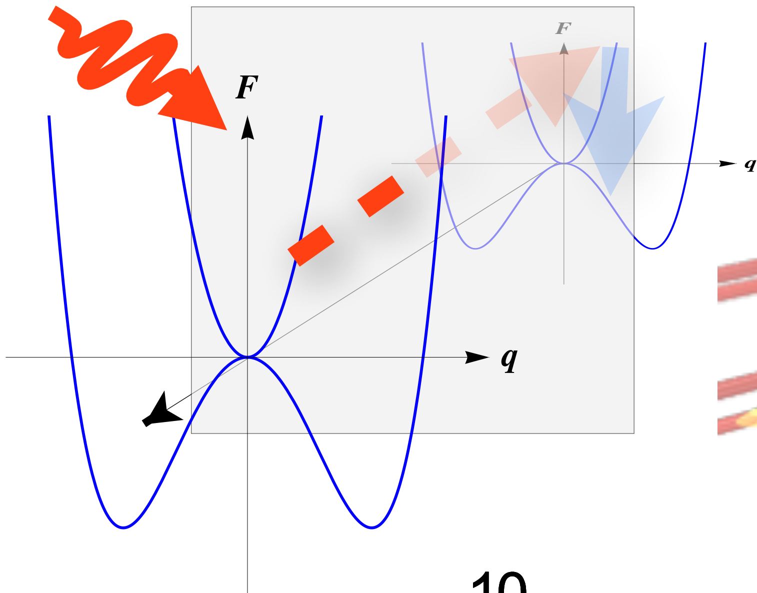
ΔR = 5% at 800 nm (1.5 eV)

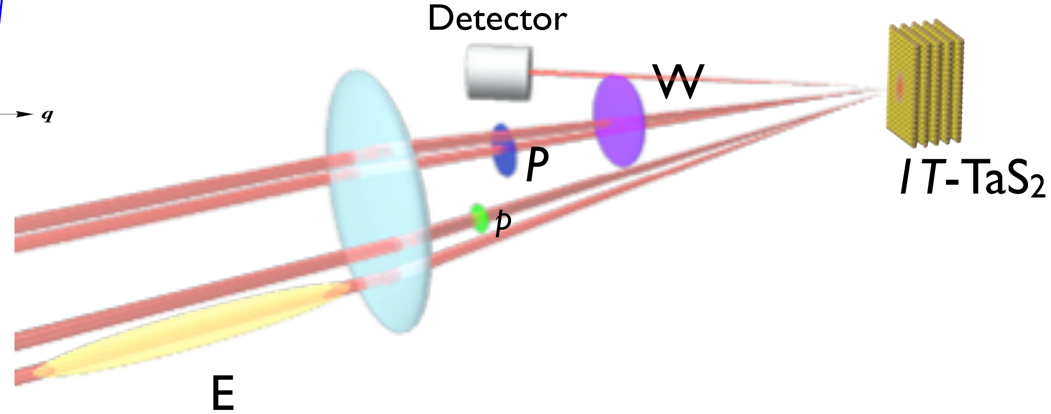
Switching <u>only</u> occurs for short pulses $\tau_L < 4$ ps



Stojchevska et al, Science **344**, 177 (2014).

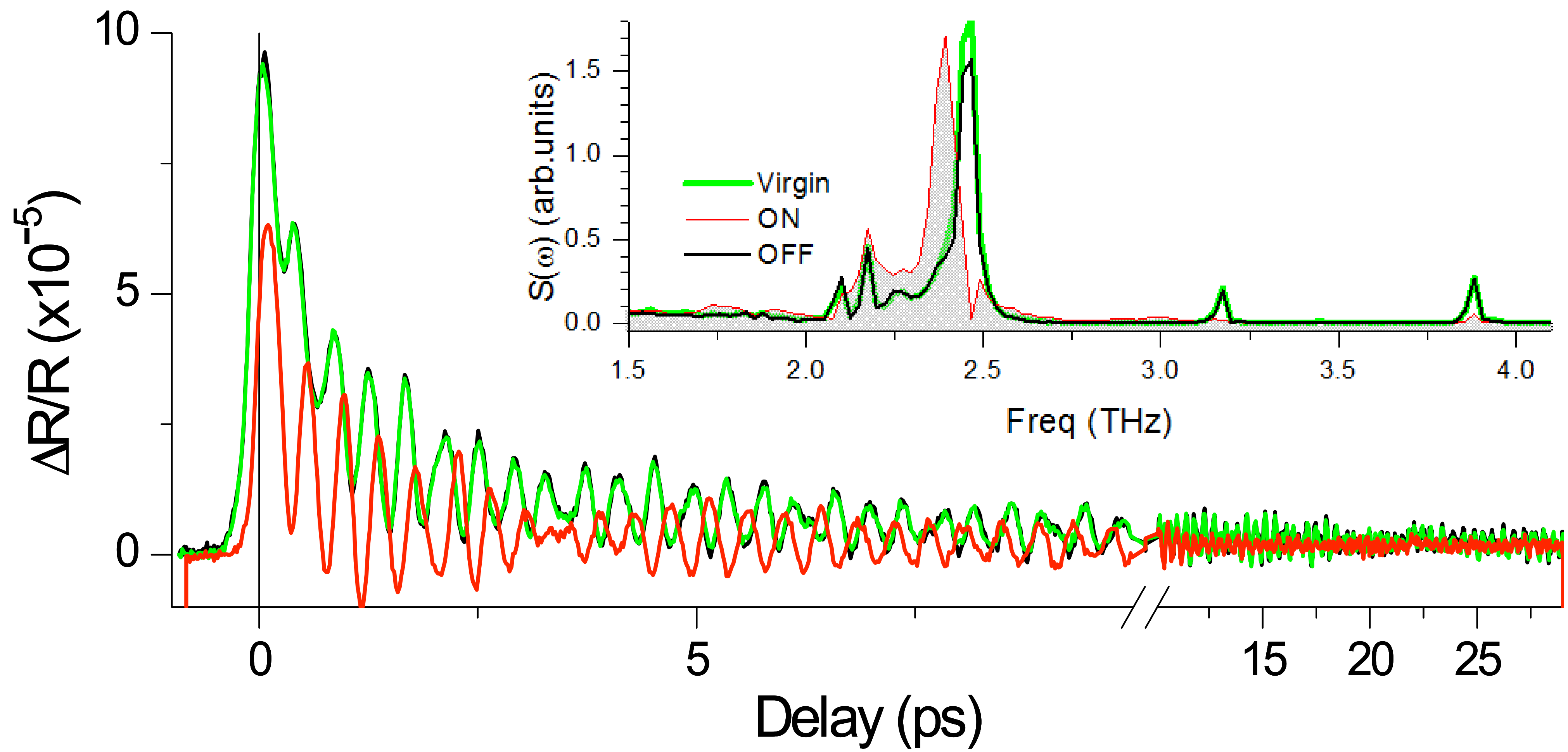
1T-TaS_2: Collective mode switching





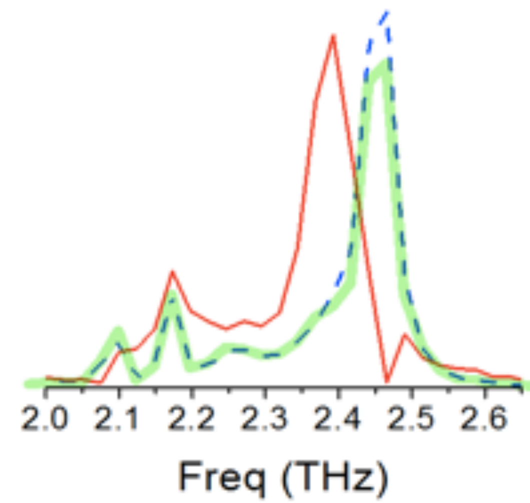
W = 50 fs “write”
E = 50 ps “erase”
P = “pump” (50 fs)
p = “probe” (50 fs)

Ljupka Stojchevska

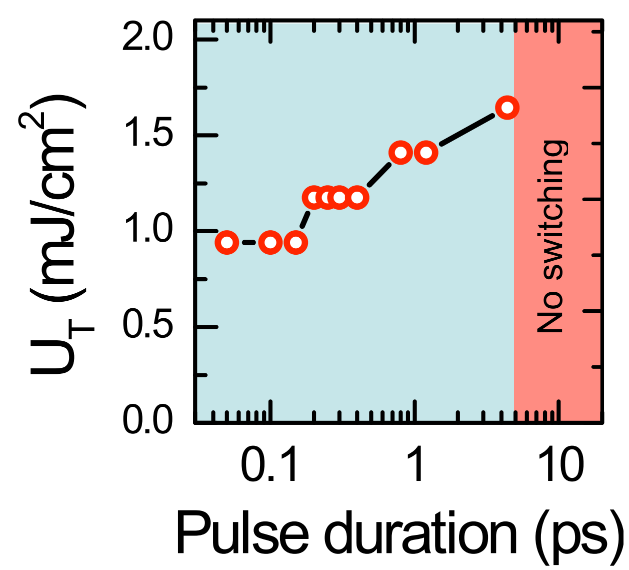


3 unusual characteristics of the hidden state

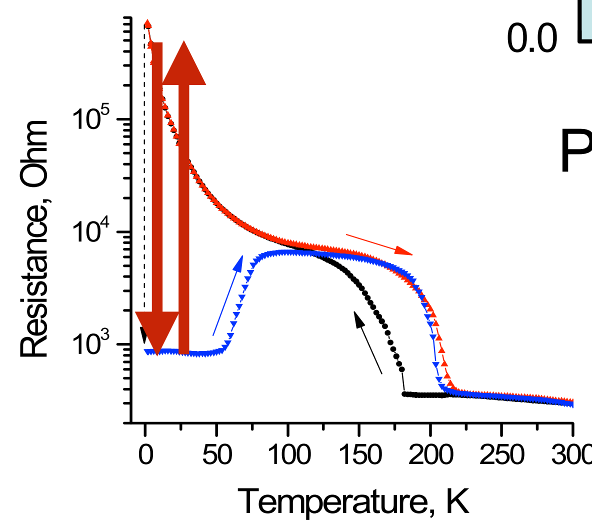
1. Narrow collective mode peak - implies <u>long range order</u> with no intermediate states, or inhomogeneity



2. Switching is achieved <u>only</u> with sub-5 ps pulses

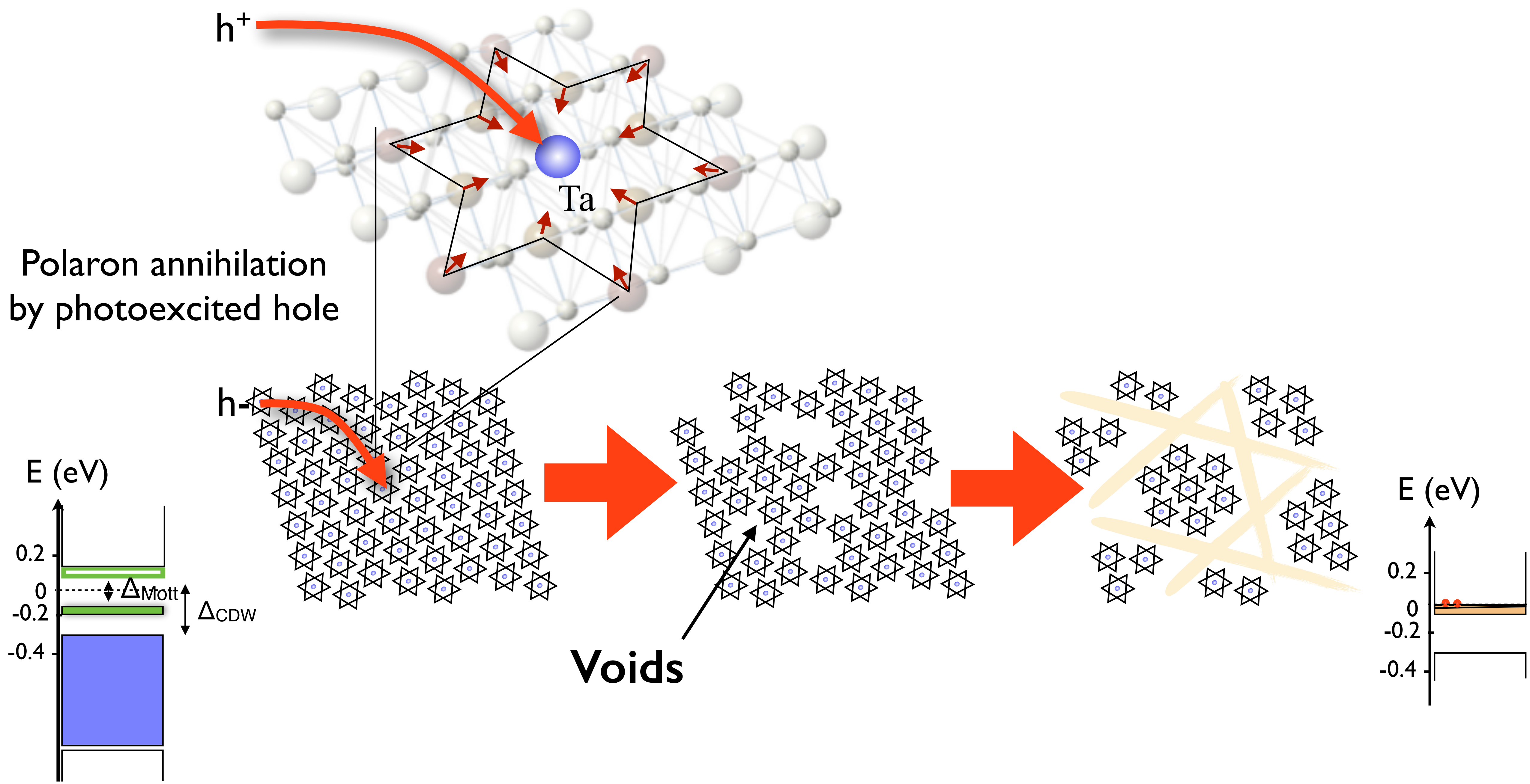


3. Reproducible metal-Insulator bistability (<u>not</u> sample dependent)



Photo"doping" and ordering of voids

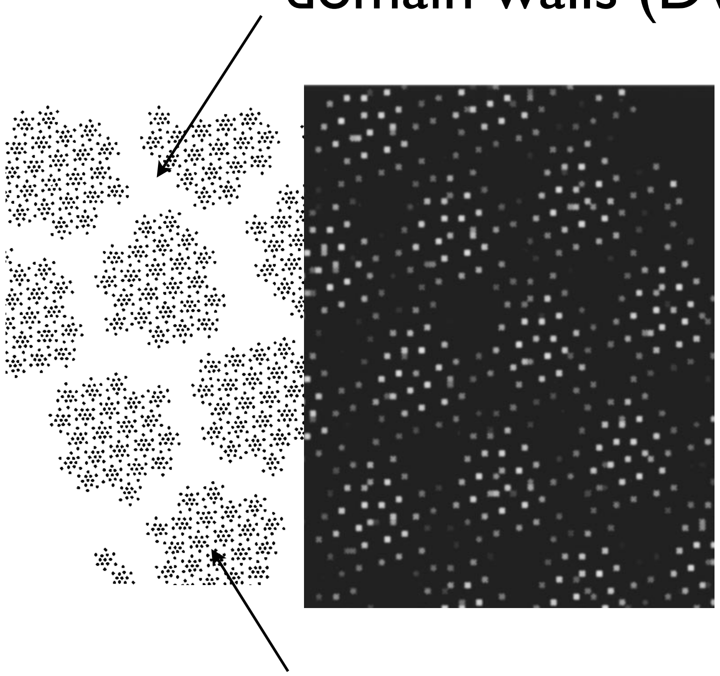
The addition of a h+ to the C structure annihilates a polaron, creating a defect.



The nearly-commensurate state of $1T$-TaS_2

McMillan (1975), Nakanishi et al (1977), Serguei Brazovskii, (2013)

domain walls (DW)



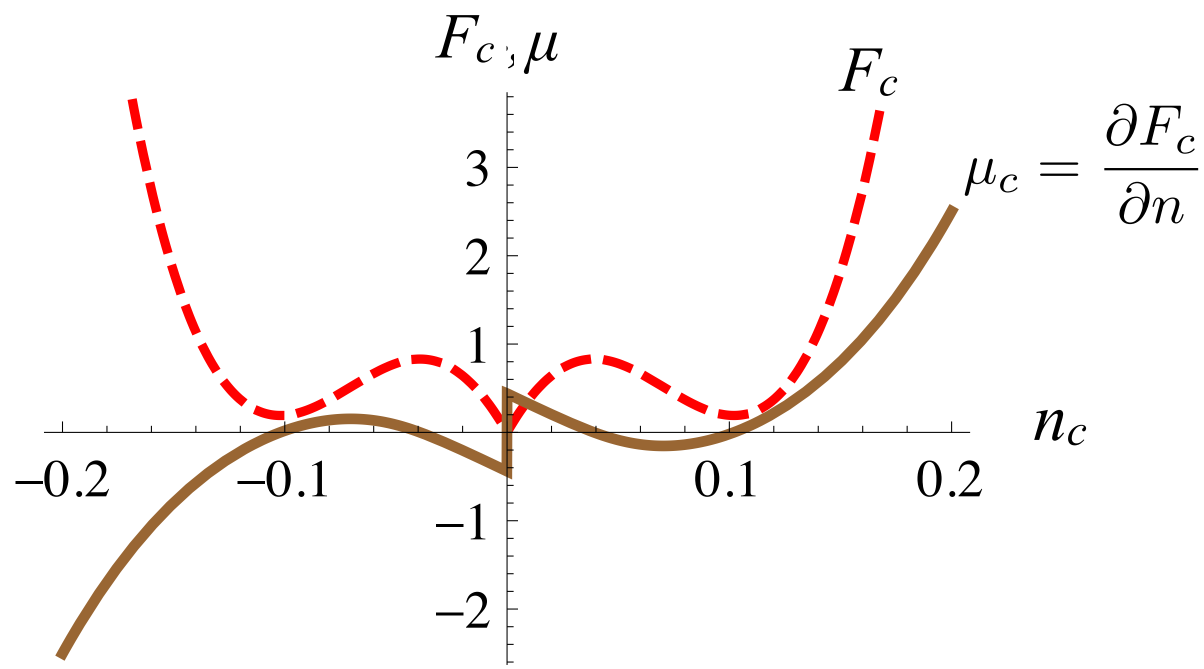
Insulating patches of polarons

Free energy:

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

Where $\quad n_c = n_h - n_e$

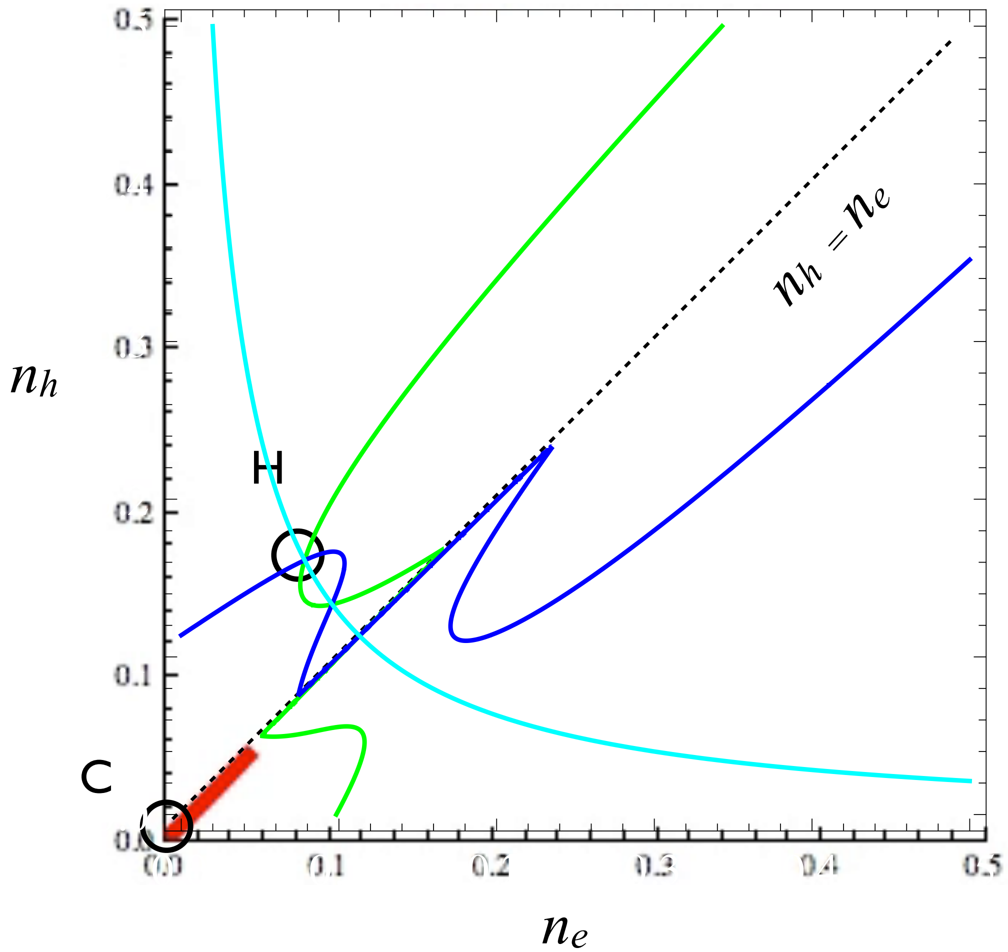
Free energy and chemical potential:

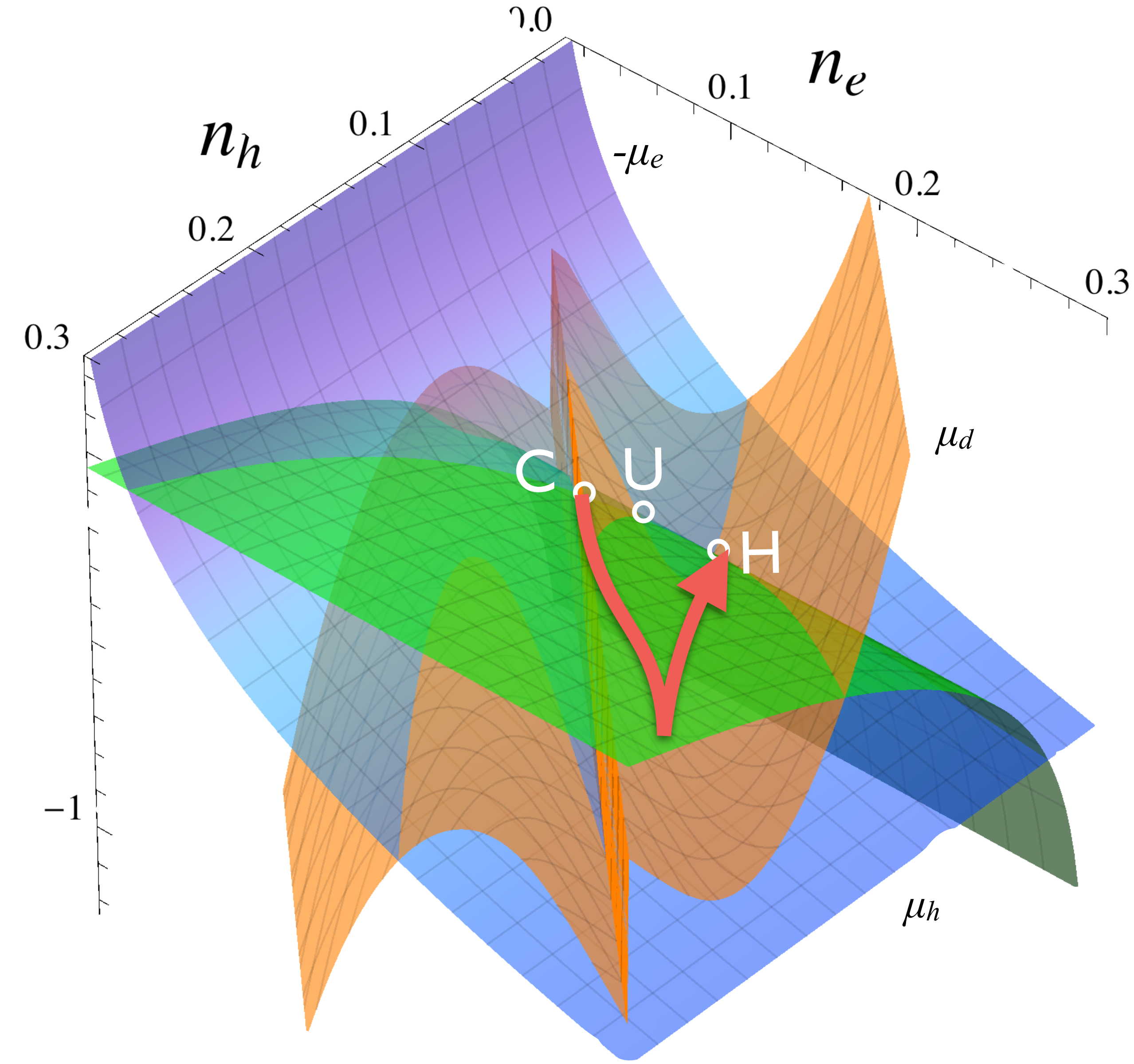


Calculated trajectory

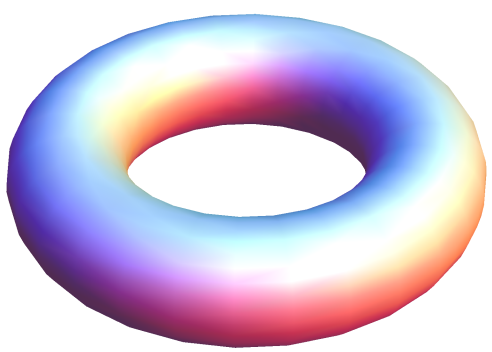
Laser pulse energy **above** threshold ($U_W > U_T$):

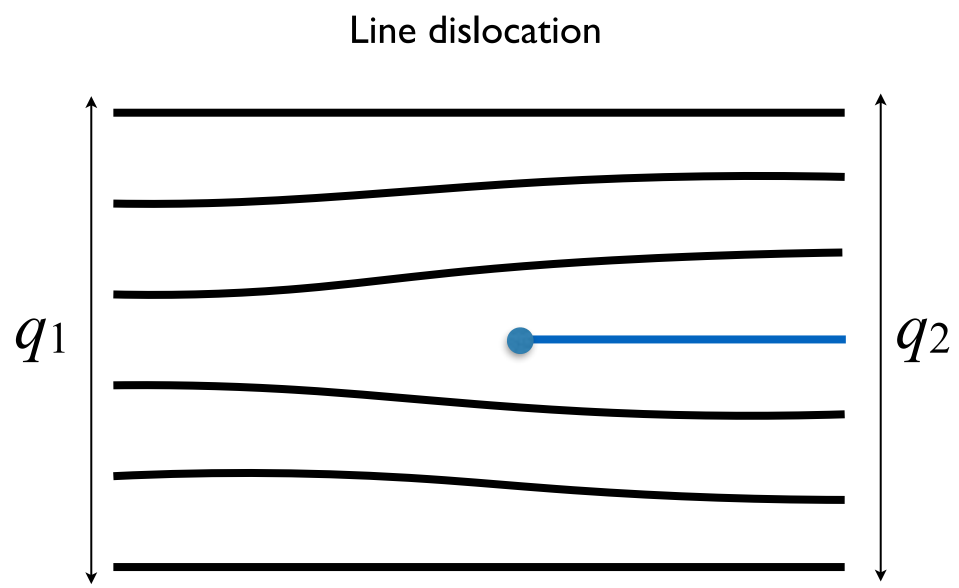
System trajectory (parametric plot)

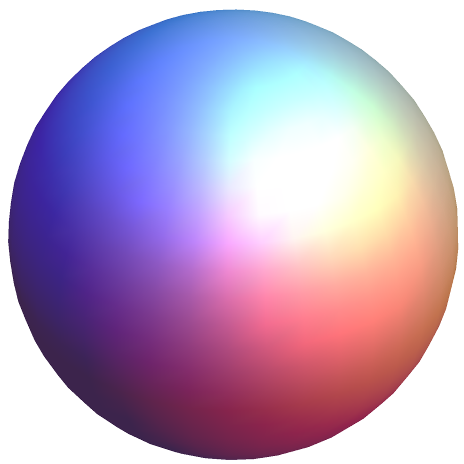




Why is the H state so stable?

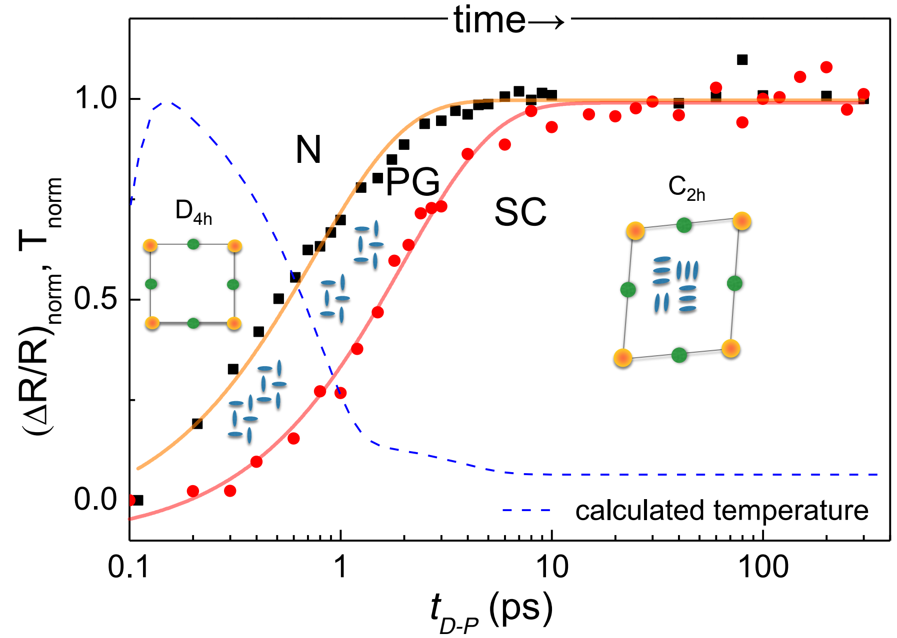




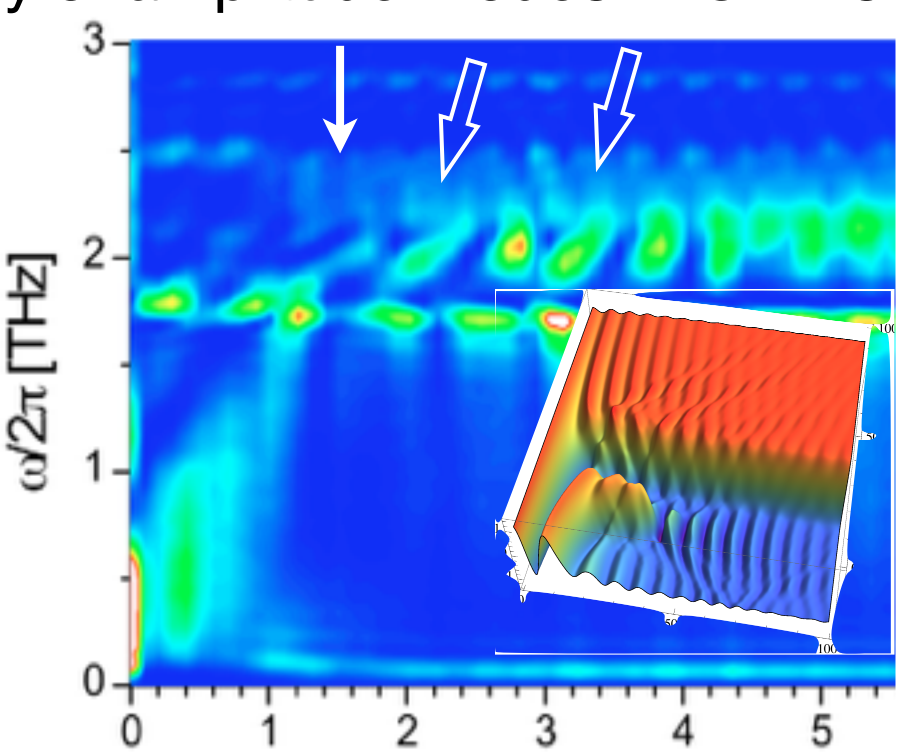


Conclusions

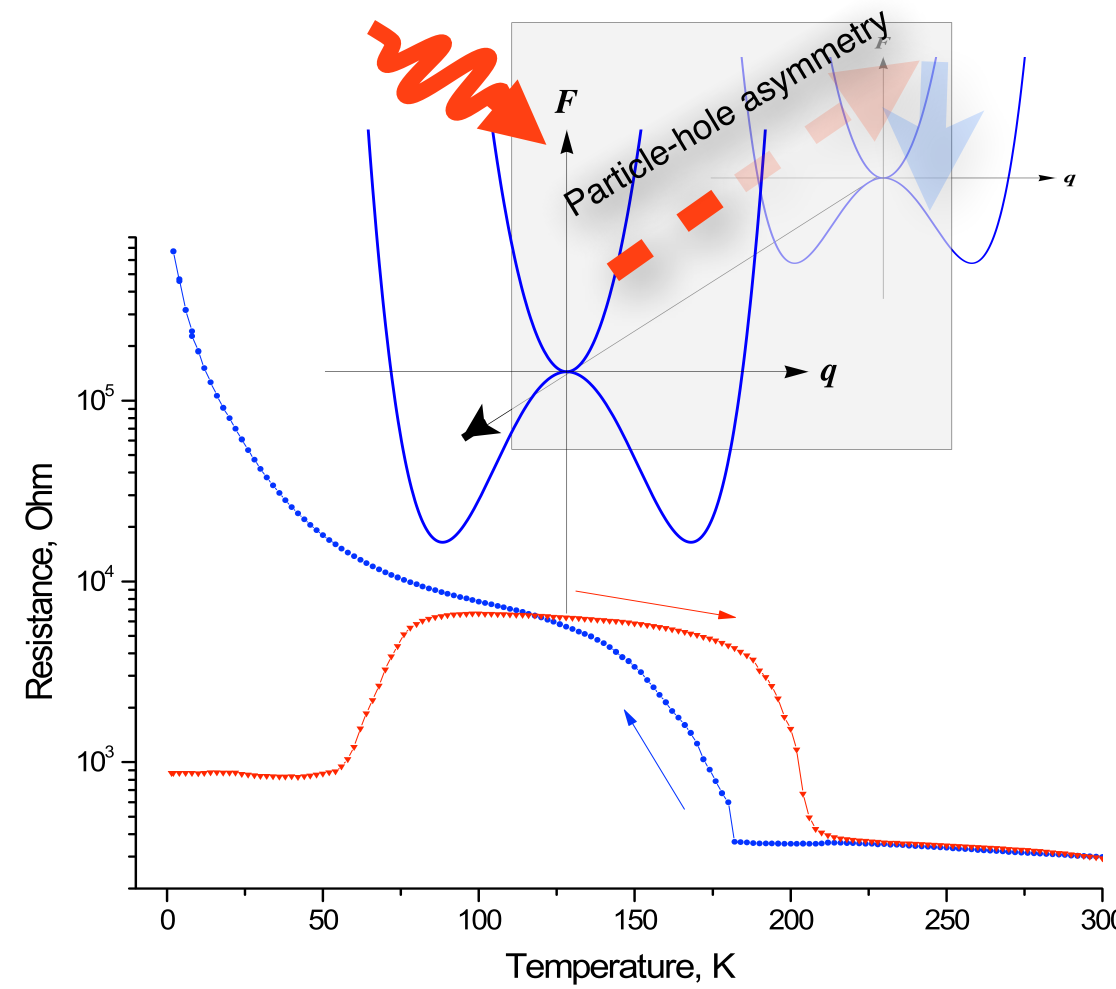
No Higgs oscillations in HTS



Plenty of amplitude modes in CDWs



Hidden states of matter



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