Femtosecond coherent nonequilibrium electronic ordering and topological defect dynamics in Charge Density Waves.

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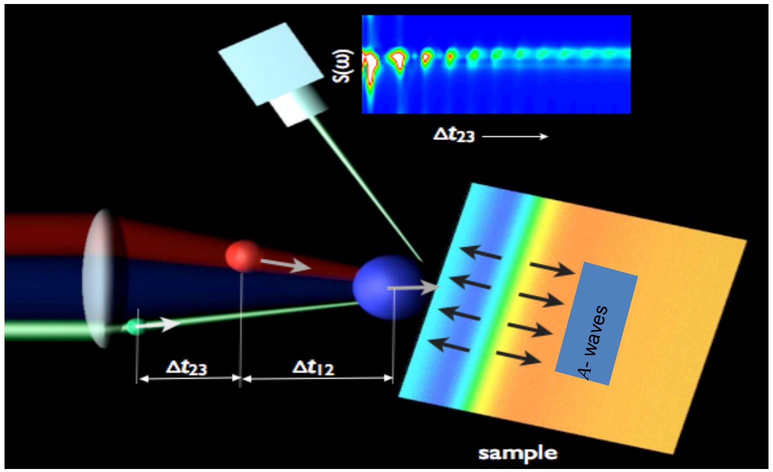
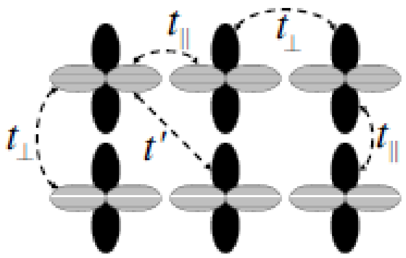
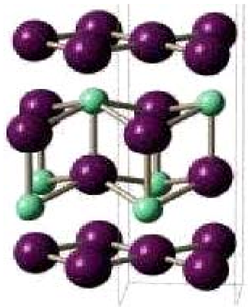


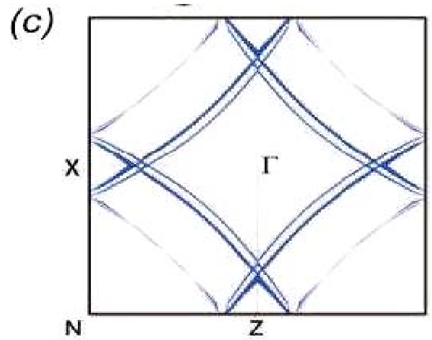
Nonlinear "femto-second" optics
- time resolution **~10^0fs<10^{-13}** sec
Well shorter than pico-second scale of phonon's periods **$2\pi/\omega_0$~1ps=10^{-12}** sec .

Real-time probe of evolving phonon modes and of electronic relaxation

Layered compounds with Charge Density Waves: Tb(Dy)Te_3, $TaSe_2$, $K_{0.3}MoO_3$

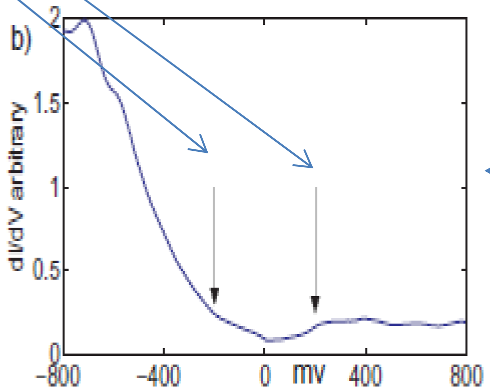


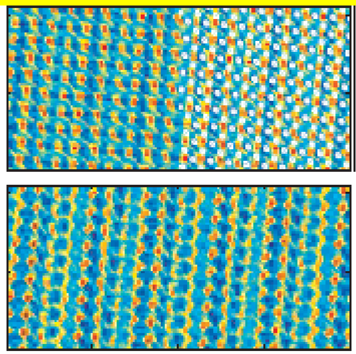




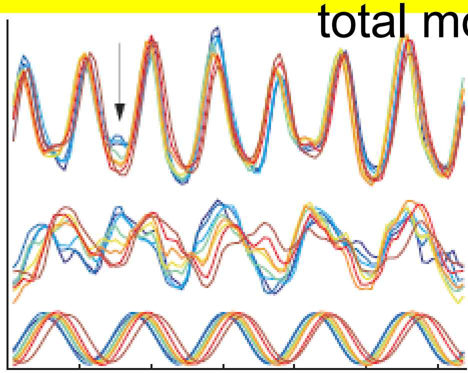
Fermi surfaces from ARPES experiments

Strongly oriented molecular orbitals →
hidden one-dimensionality in two orthogonal directions →
flattened, nearly nested Fermi surfaces →
instability towards an incommensurate CDW, wave number Q=0.7→
gap closes some ½ of the Fermi surface



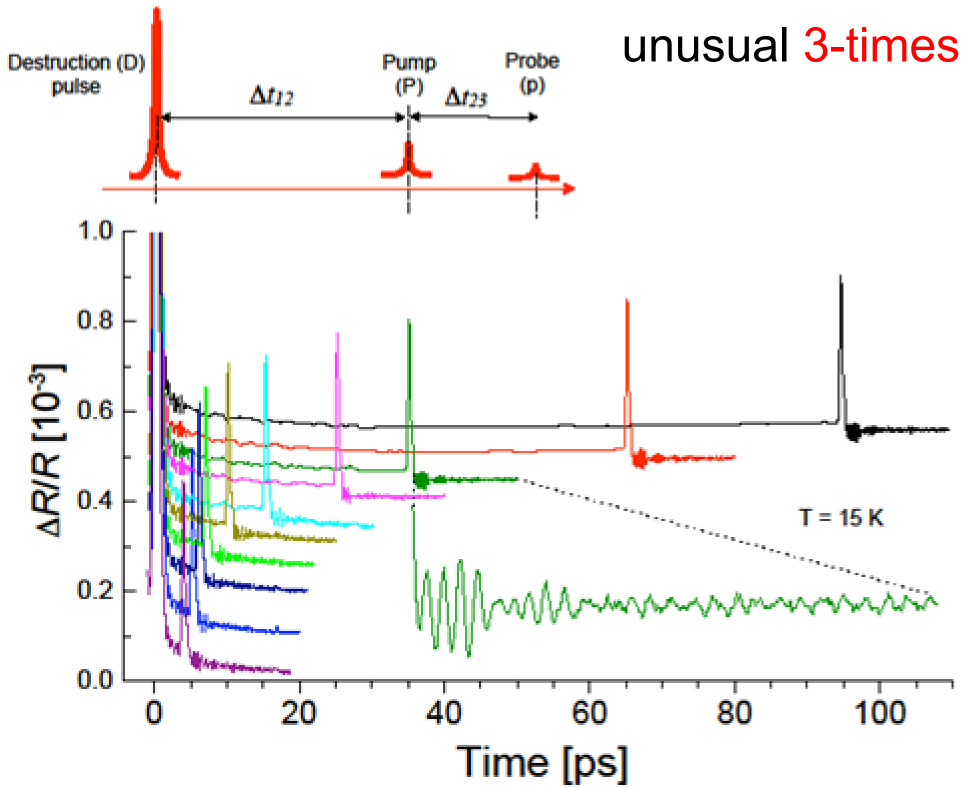


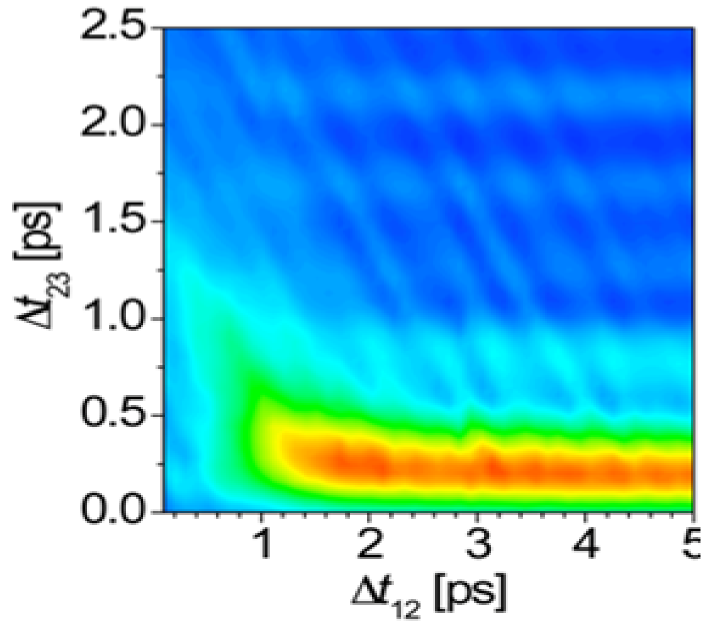
total modulation



extracted CDW overlap

STM Studies of TbTe3: Evidence for a fully Incommensurate Charge Density Wave *A. Fang, N. Ru, I.R. Fisher, and A. Kapitulnik* unusual 3-times experiment





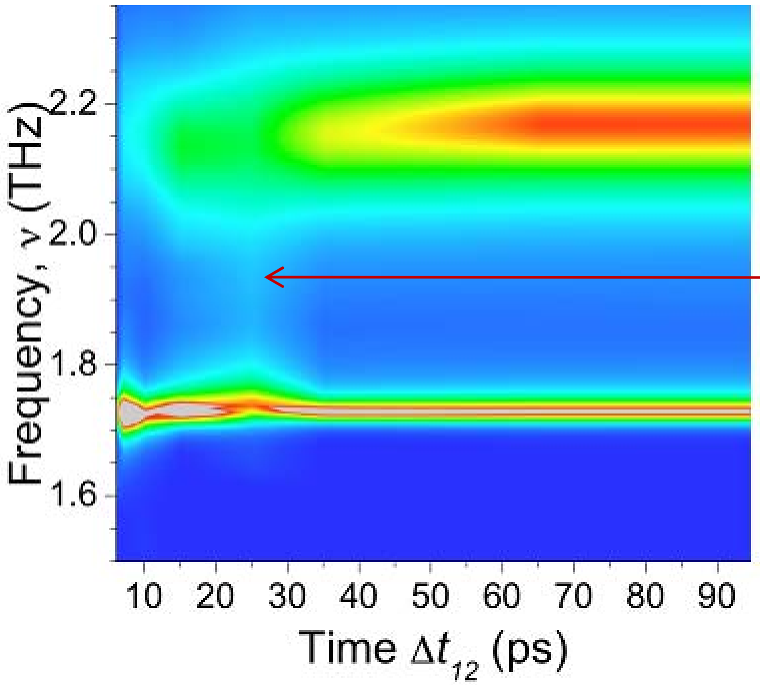
$t=t_1$ –"destruction D" pulse strongly perturbs the electronic system causing weakly attenuating "pendulum" oscillations of the CDW.

$t_2=t_1+\Delta t_{12}$ – "Pump P" pulse weakly perturbs the evolving non-equilibrium state.

$t_3=t_2+\Delta t_{23}$ – "probe p" pulse probes the reaction of the still evolving system to the Pump pulse.

The way all have started; no fine structure yet

Fast Fourrier Tranform FFT : $t_{23} \to \omega$ at a given t_{12}

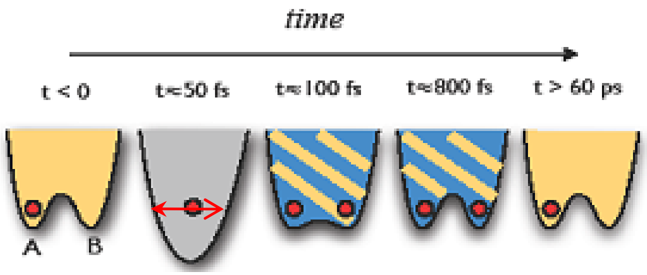


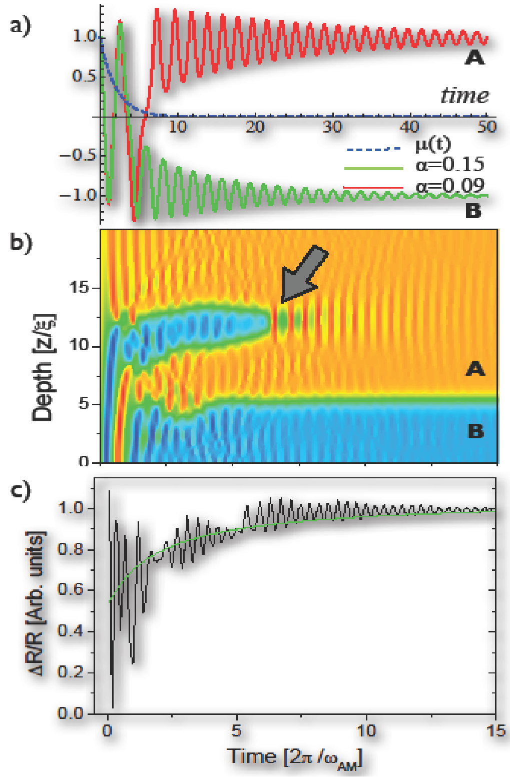
Amplitude mode AM of the order parameter

Inter-mode interaction when one gains a width

An auxiliary mode noninteracting with electrons

Original challenge:
Why the AM sets in at times x10-100 longer then the relaxation time of electrons ~2ps?
Suspected: creation of topological defects, hence links to dynamical phase transitions in cosmology and cold atoms





Time evolution of the ground state energy U profile as function of the order parameter A.

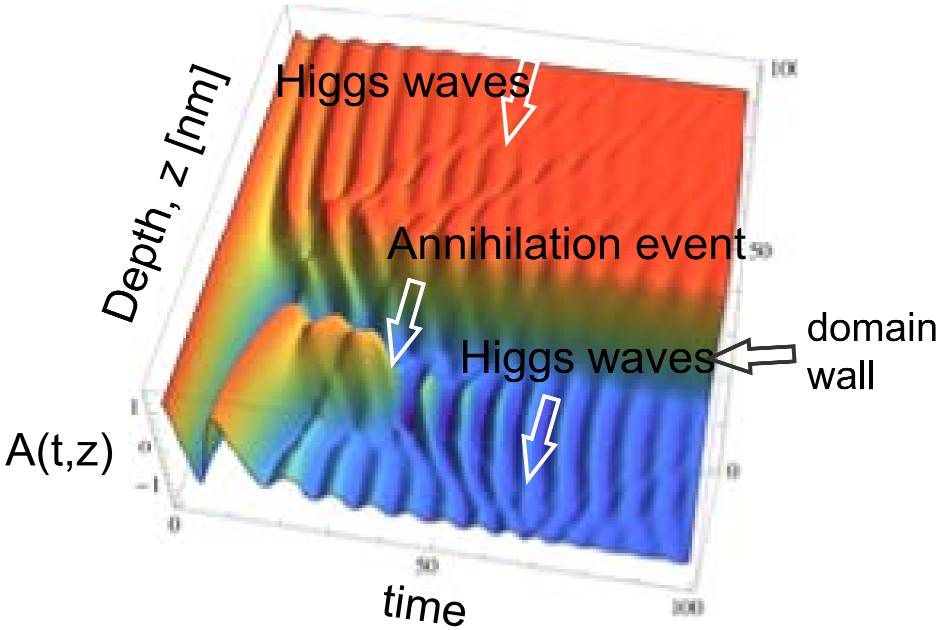
Red dot - the state of the system.

Blue/orange potential signifies the pendulum oscillating regime spanning both signs of A.

$$\frac{1}{\omega_0^2}\partial_t^2 A + \frac{\alpha}{\omega_0}\partial_t A - \xi^2\partial_z^2 A + A^3 - A\left(1 - \eta\exp(-\frac{t}{\tau} - \frac{z}{\lambda})\right) = 0$$

z – coordinate from the surface towards the sample depth.
ξ – inter-plane coupling length
λ – light's penetration depth.
η≈2 – destruction strength – *the only adjustable parameter*

Inhomogeneous laser excitation



Calculated order parameter as a function of depth, and of time after quench:

A(z; t) as a function of depth z and waiting time t_{12}.

Ripples in the space-time texture are due to annihilation event at 3.5 ps.

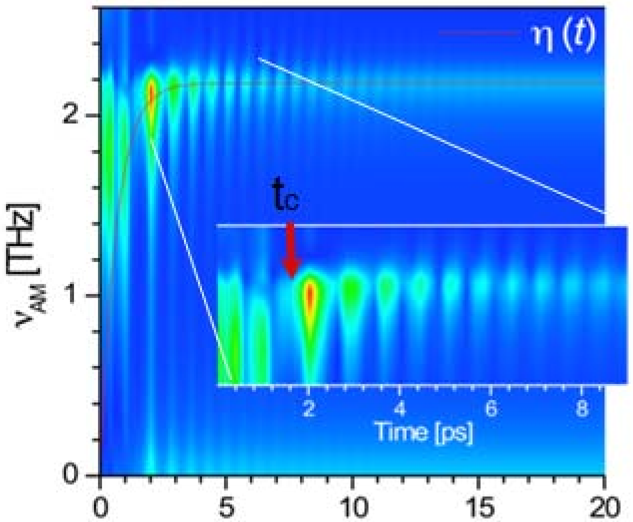
The wave reaches the surface at around 6ps.

Theory versus experiment: homogeneous – z-independent regime

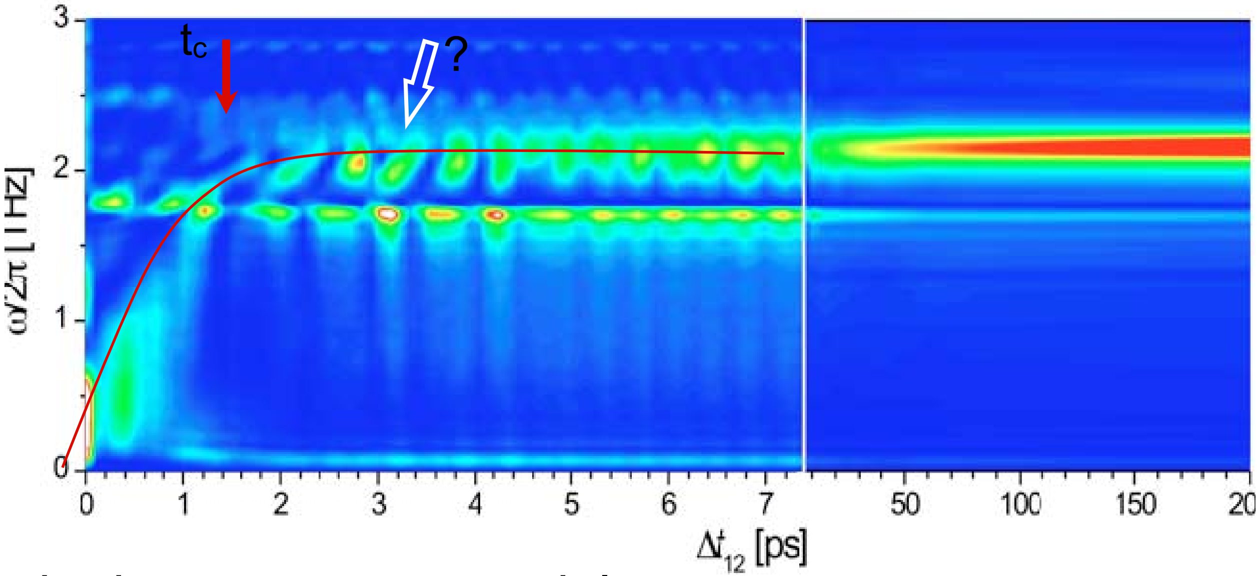
FFT power spectra of the data as a function of t_{12}. Note:

1. non-periodic fluctuations of intensity around the transition at t_c=1.5 ps
2. asymmetric line shapes near t_{12}=3.5ps as the domain wall reaches the surface (white arrows).

Numerical homogeneous solution



Experimental spectrum



Features reproduced by the homogeneous model:

- order parameter fluctuations
- slowing down below t_c
- softening of the amplitude mode AM

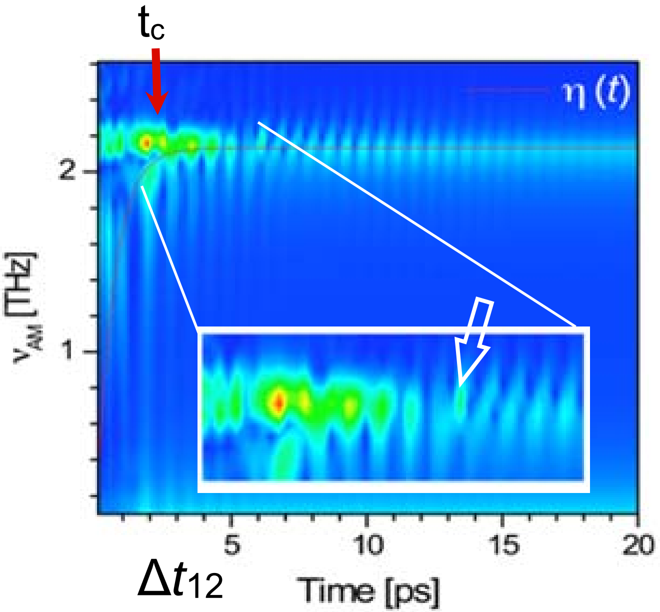
Theory vs Experiment

Note : order parameter fluctuations, slowing down at t_c , softening off the amplitude mode AM.

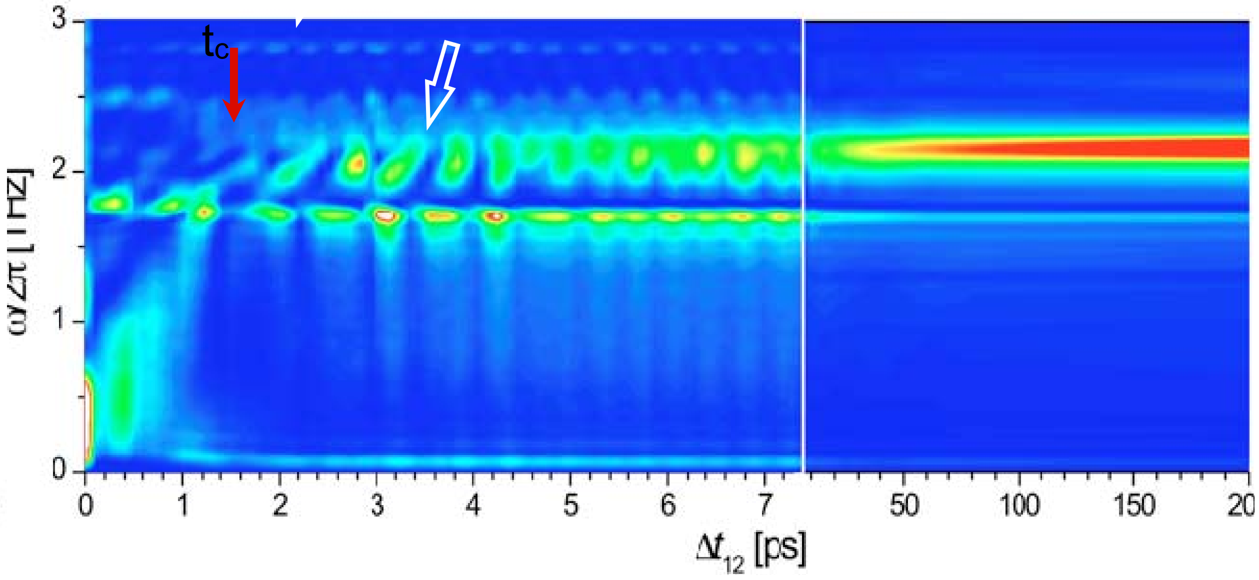
WHITE ARROWS - distortion due to the Higgs wave.

Red arrows - slowing down below the critical time of the transition t_c.

Numerical solution integrated over z



Experimental spectrum



FFT power spectra of the data as a function of t_{12}. Note:

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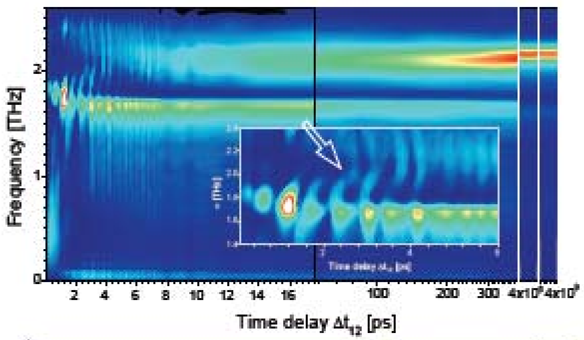
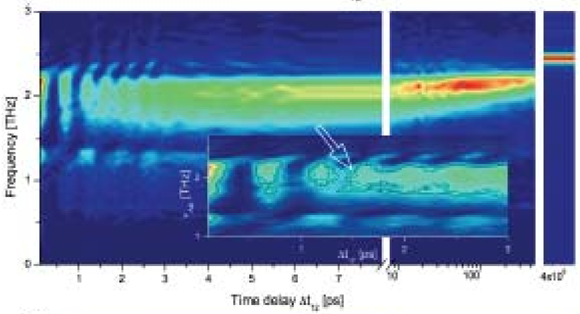
Universality of the dynamics: extensions from $TbTe_3$ to $DyTe_3$, $TaSe_2$ and $K_{0.3}MoO_3$.

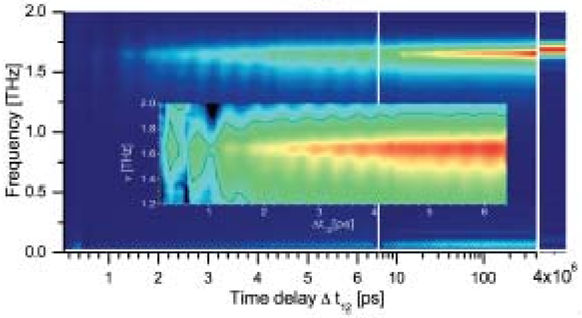
Universal features:
(1) subpicosecond gap recovery →
(2) slowing down of the order parameter fluctuations through the transition
(3) creation of multiple domains →
(4) coherent topological defect annihilation →
(5) incoherent dynamics.



DyTe$_3$



TaSe$_2$



Universality of the dynamics: extensions from TbTe$_3$ to DyTe$_3$, TaSe$_2$ and K$_{0.3}$MoO$_3$.

Universal features:

(1) subpicosecond gap recovery

(2) slowing down of the order parameter fluctuations through the transition

(3) creation of multiple domains

(4) coherent topological defect annihilation

(5) incoherent dynamics.

K$_{0.3}$MoO$_3$: Waves are not observed and are not predicted for parameters of this material - all frequencies are in-phase.

Power spectra of oscillatory responses obtained directly from data by FFTs.

Inserts show details around the critical time of the transition t_c.

Arrows point to distortions of the spectra - diagonal spots in the $\omega - \Delta t_{12}$ plots, when Higgs-waves arising from domain wall annihilation reach the surface.

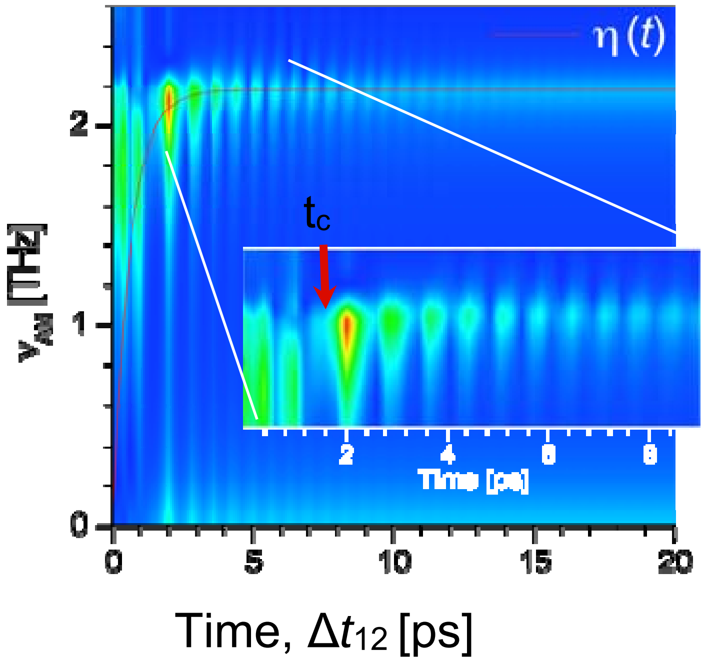
Calculated spectral response

Equation of motion without gradient term, appropriate for a uniform system:

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

The solution gives the time-evolution of the spectrum through the transition:

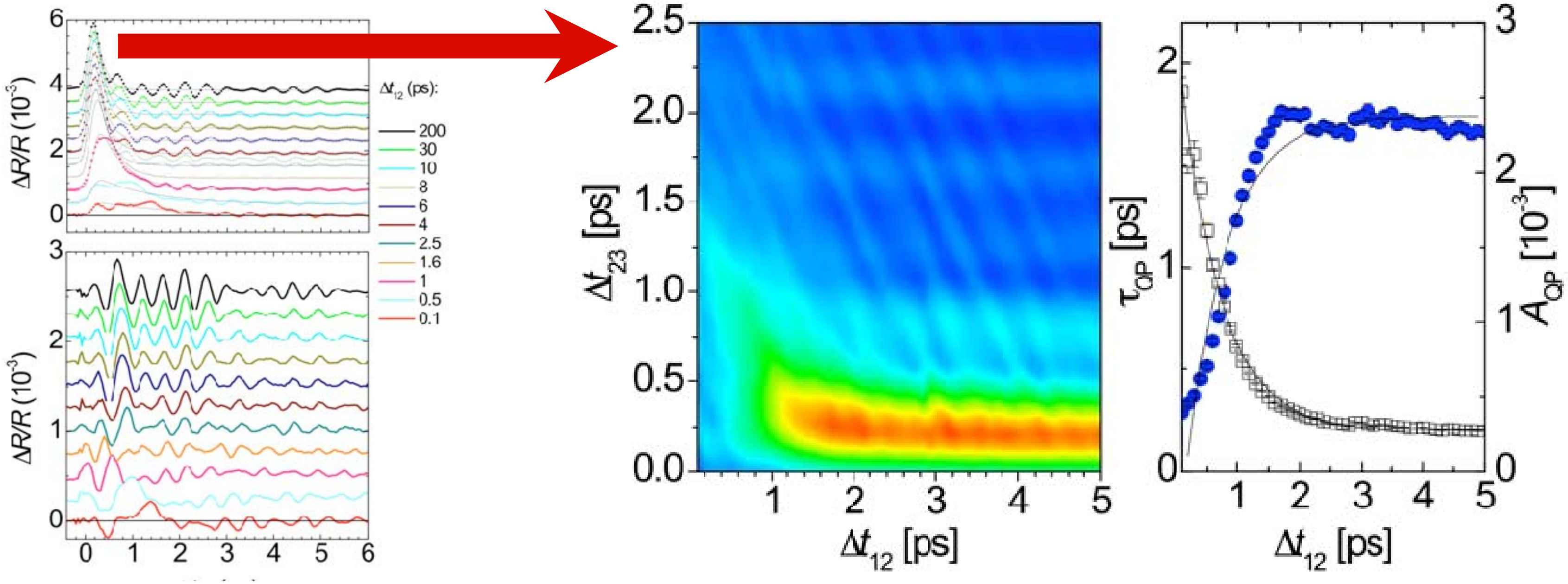
Calculated ΔR/R based on the homogeneous solution



Experimental parameters:
τ=650fs
$\omega_0/2\pi$=2.2THz
α=0.1
η=2exp(-t/τ)

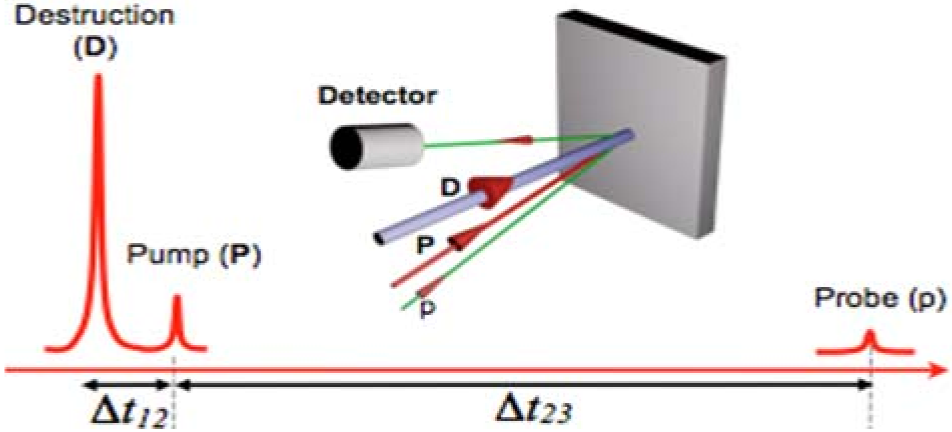
Quasi-particle (fermion)dynamics: gap recovery

Gap recovery time τ = 650 fs

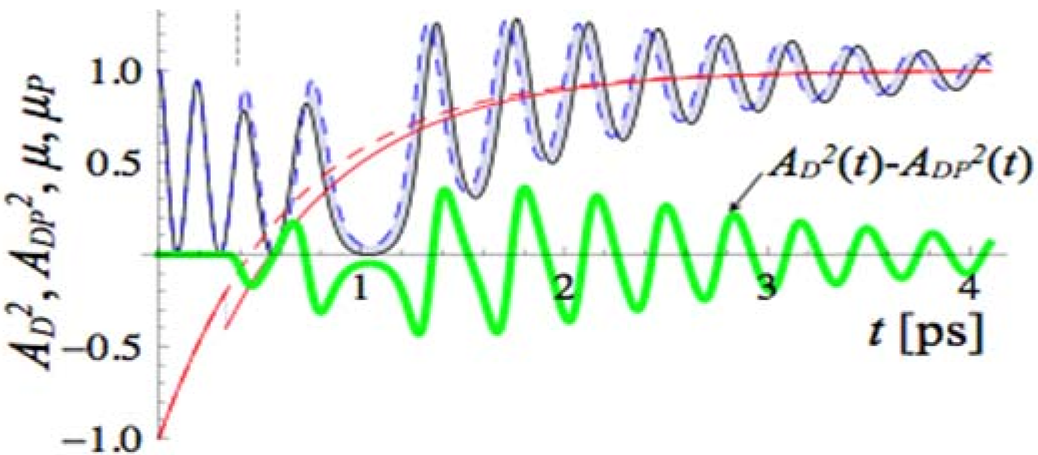


Raw transient reflectivity data $\Delta R/R$ for different delays Δt_{12}. $\Delta R/R$ with the QP response subtracted.

The QP response $\Delta R/R_{QP}$ after the quench as a function of t_{12}. The ripples arise from space-time
fluctuations of the OP.
The QP lifetime QP and the amplitude of the QP response Aqp as a function of Δt_{12}.
A single exponential fit to both data sets gives $t_{QP} = t_{AQP}$ = 650fs.

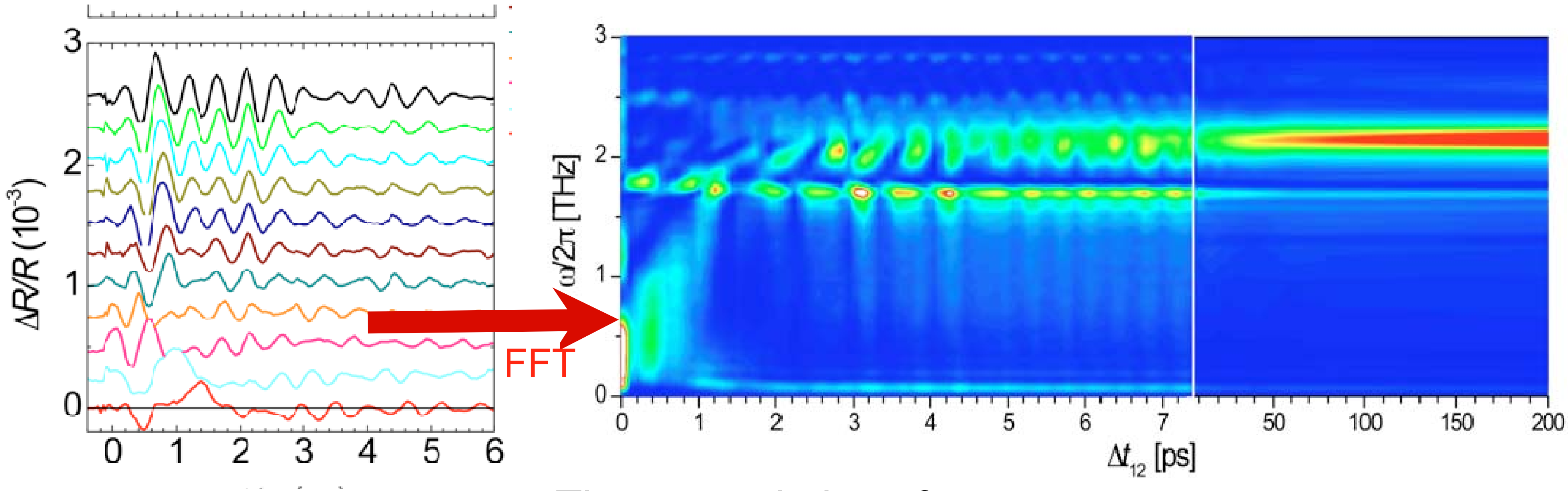


Destruction
(D)
Pump (P)
Detector
D
P
p
Probe (p)
Δt12
Δt23

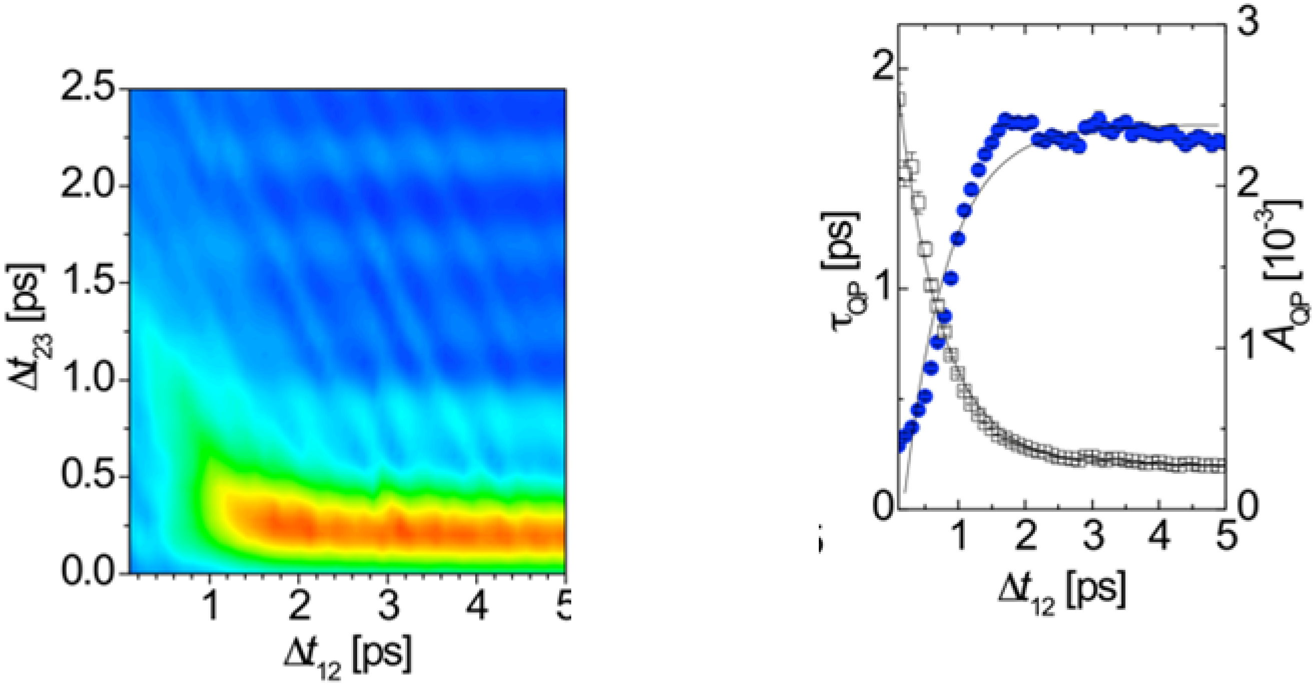


1.0
0.5
-0.5
-1.0
1
2
3
4
t [ps]
A_D^2, A_DP^2, μ, μ_P
A_D^2(t)-A_DP^2(t)

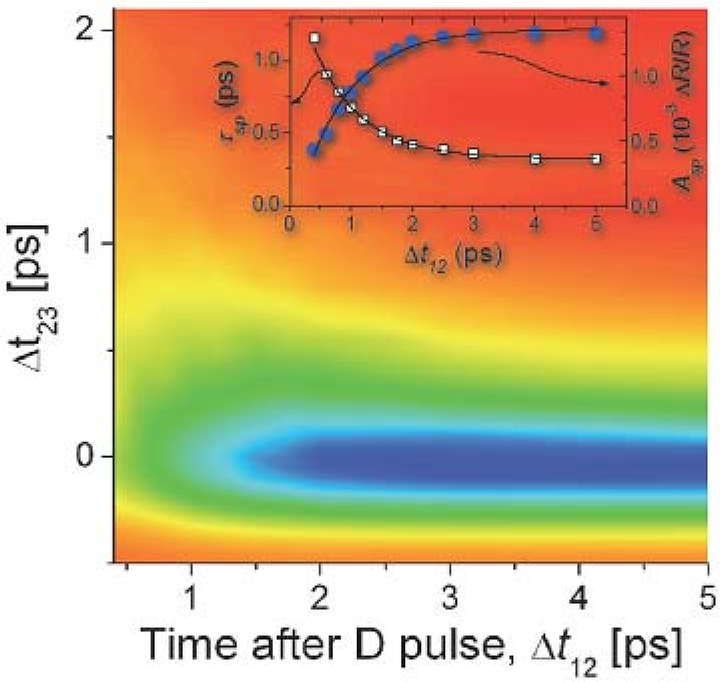
Evolution of the collective mode spectrum with time after quench



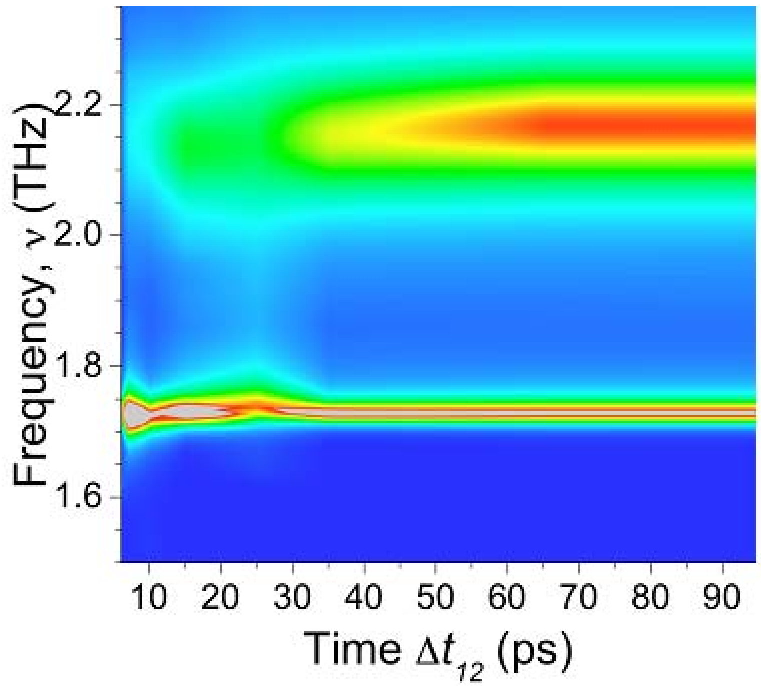
The most obvious feature:
coherent order parameter fluctuations



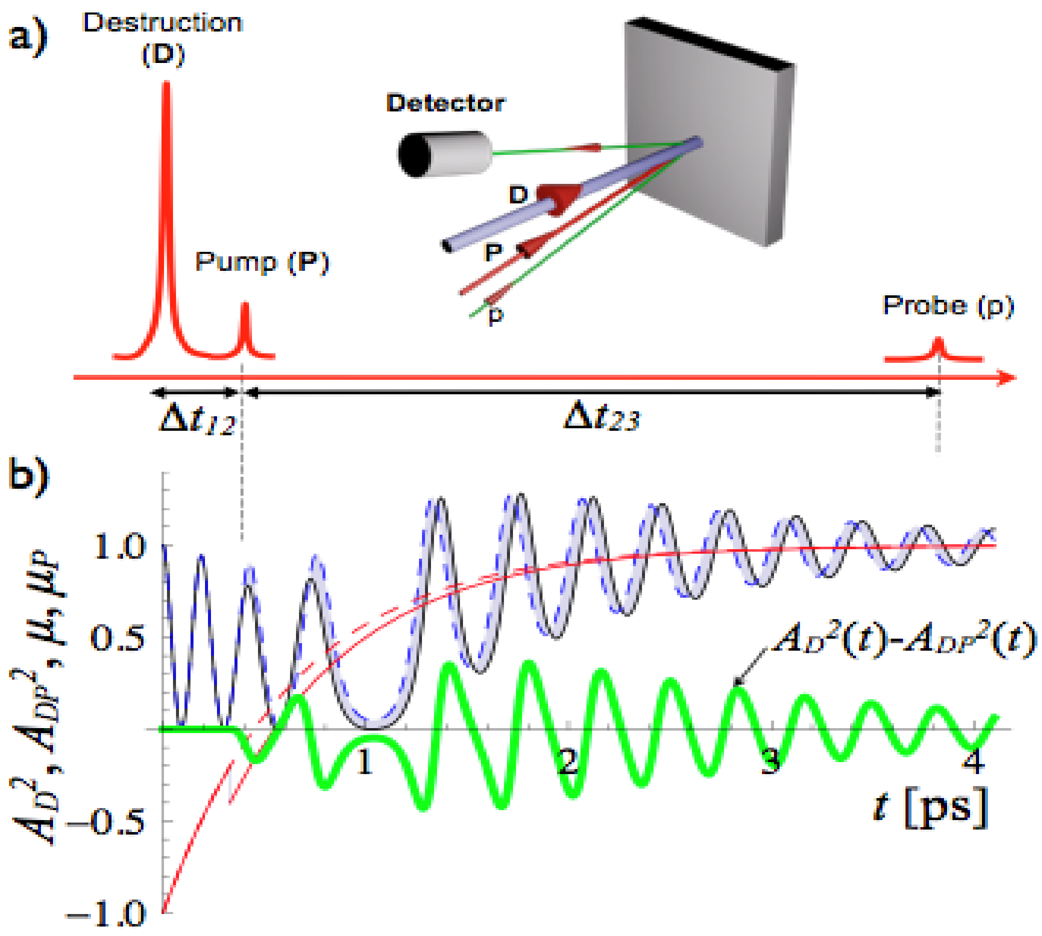
2.5
2.0
1.5
1.0
0.5
0.0
Δt23 [ps]
1
2
3
4
5
Δt12 [ps]
2
1
0
τQP [ps]
3
2
1
0
AQP [10-3]
1
2
3
4
5
Δt12 [ps]



Time after D pulse, Δt12 [ps]
Δt23 [ps]
1
2
3
4
5
0
1
2
Δt12 (ps)
0.0
0.5
1.0

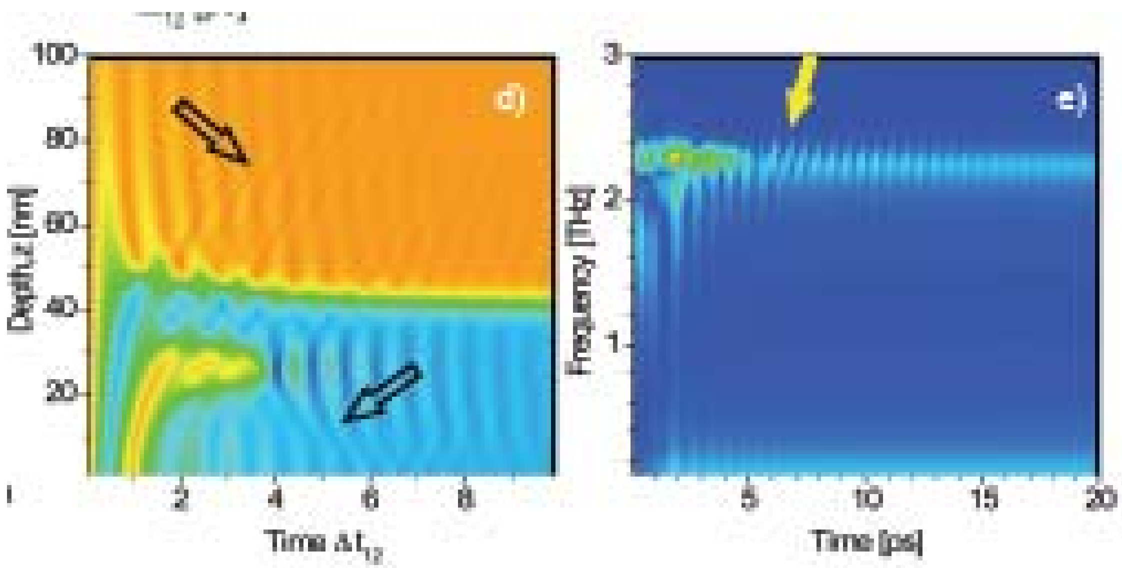


Frequency, ν (THz)
Time Δt12 (ps)
1.6
1.8
2.0
2.2
10
20
30
40
50
60
70
80
90



a) Destruction (D) pulse quenches the system, pump- probe (P-p) sequence probes the reflectivity at a later time t_{12}.

b) Control parameters mu (solid) and mu_P (dashed). Predicted oscillations of $A^2(t)$ with and without the P pulse are shown by the dashed and solid oscillatory curves. Predicted $\Delta R(t)$ is shown by the green curve.



100
80
60
40
20
Depth z [nm]
2
4
6
8
Time Δt$_{12}$
d)
3
2
1
Frequency [THz]
5
10
15
20
Time [ps]
e)