



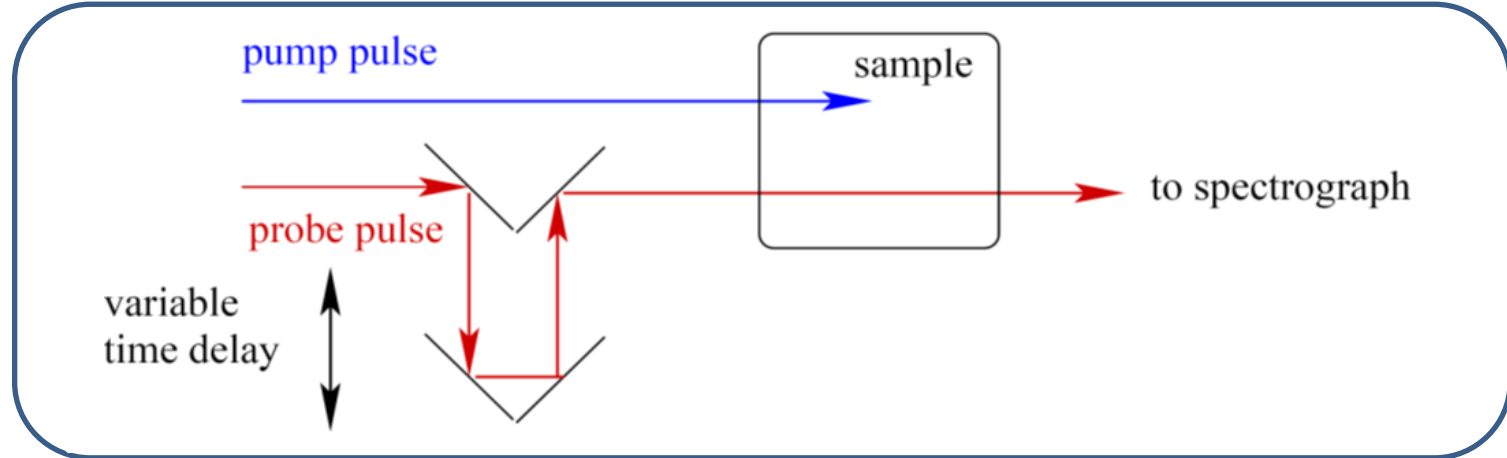
Max Planck Institute for Solid State Research

Novel Non-Equilibrium Dynamics in Superconductors: Induced Superconductivity and Higgs Modes

Nikolaj Bittner

Holger Krull (TU Dortmund+MPI), Andreas Schnyder (MPI),
Takami Tohyama (Tokyo US), and Dirk Manske (MPI)
Exp.: Stefan Kaiser (MPI)

Introduction: pump-probe technique



» Pump-probe spectroscopy

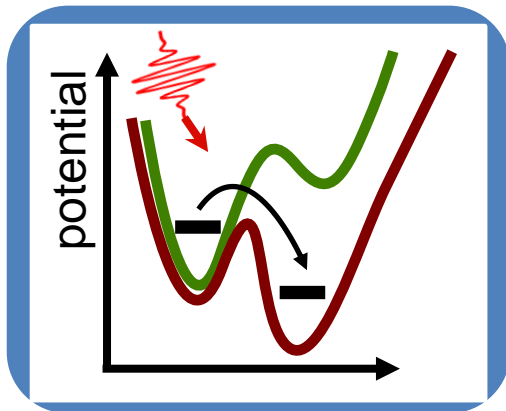
- ⇒ **Pump pulse:** excitation of the system with an **intense pulse**. **Induces dynamics.**
- ⇒ **Probe pulse:** measuring with **less intense pulse** after time delay Δt .

Makes „snapshots“ of a nonequilibrium state

» Two kinds of information can be extracted:

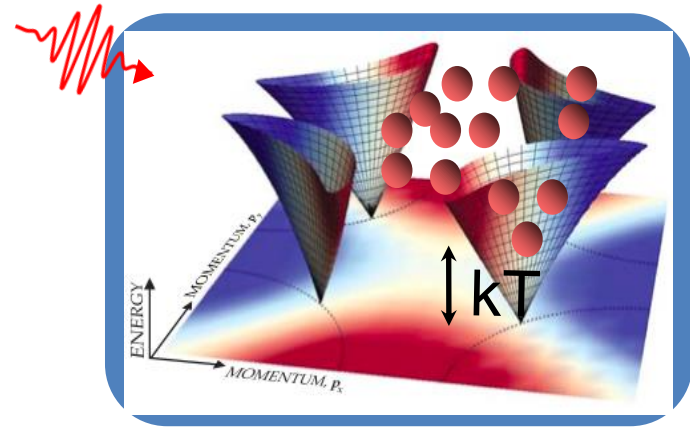
- ⇒ **Time domain:** conductivity change $\Delta\sigma$, depending on time delay Δt
- ⇒ **Energy domain:** change in the conductivity spectra

optical control



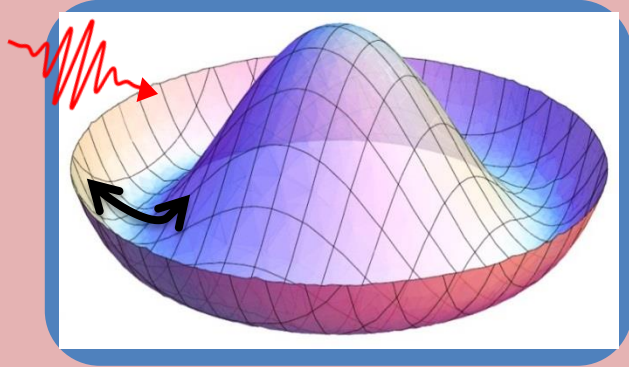
- » New transient ground state

nonequilibrium spectroscopy



- » Recovery of SC condensate
- » Cooper pair formation dynamics
- » SC glue

New! Higgs spectroscopy



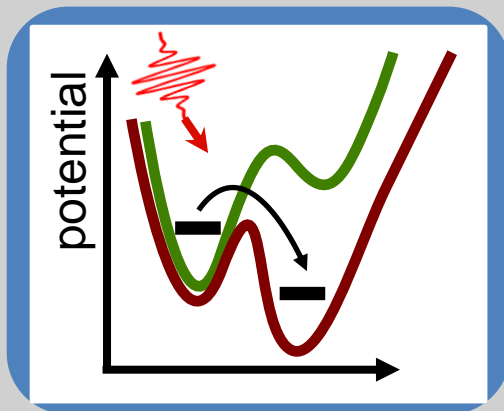
» Direct detection of the SC gap

Part 2

Order parameter oscillations in Superconductors

*H. Krull, N. Bittner, G. S. Uhrig, D. Manske, and A.P. Schnyder,
Nat. Commun. 7:11921 (2016)*

optical control



» New transient ground state

Part 1

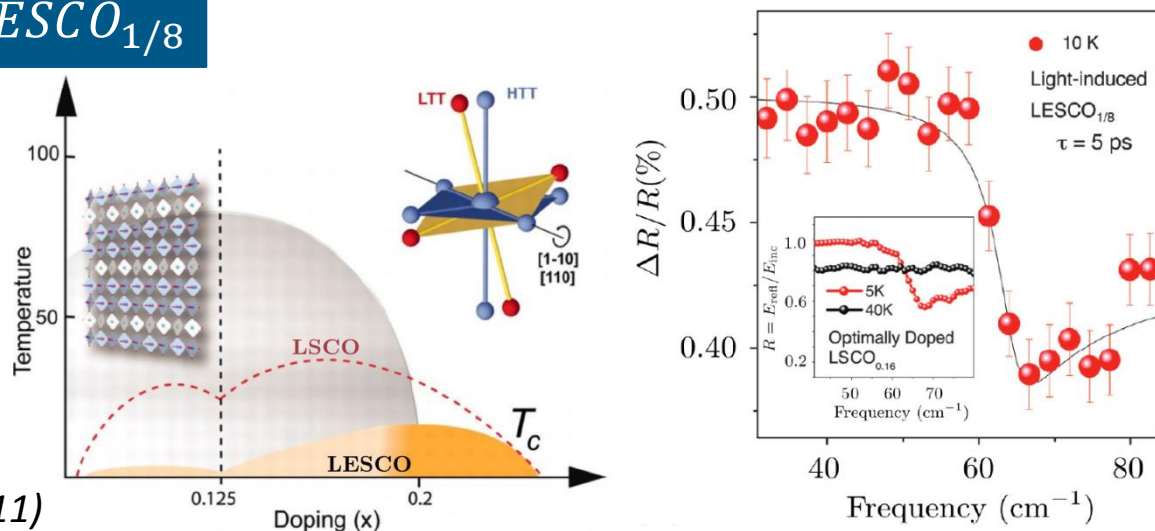
Induced Superconductivity in the extended Hubbard Model

N. Bittner, T. Tohyama, and D. Manske, in preparation

Motivation: Induced superconductivity?

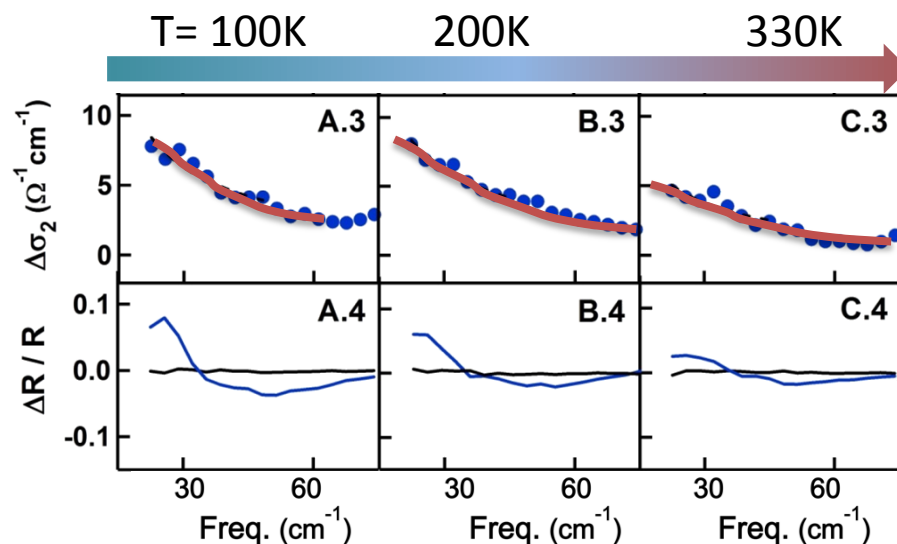
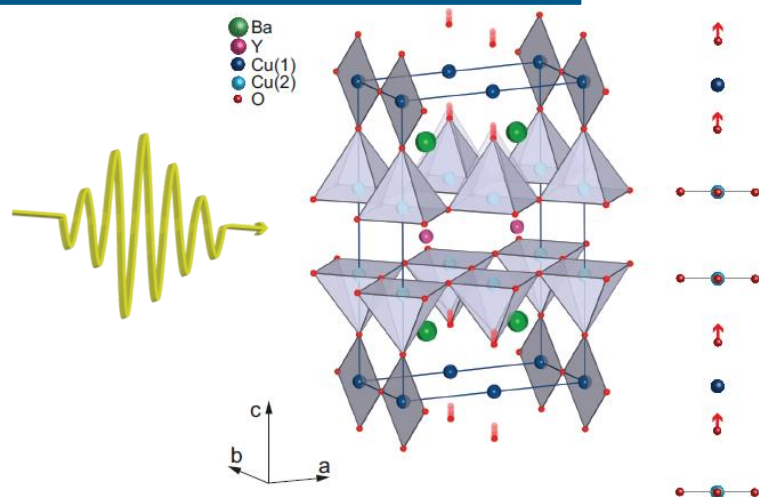
I. Stripe-Ordered Cuprate $LESCO_{1/8}$

Melting of the
competing order



D. Fausti et al. Science **331**, 189 (2011)

II. Bilayer Cuprate $YBCO$



A. Cavalleri et al. Nature, **516**, 71 ; S. Kaiser et al. PR B, **89**, 184516 (2014)

- » Suppression of the **competing order**

*Z. M. Raines et al., PR B **91**, 184506 (2015)*

*A. A. Patel and A. Eberlein, PR B **93**, 195139 (2016)*

- » **Amplification of the superconducting correlations**

(e.g. due to the increase of the DoS near the Fermi level)

*M. A. Sentef et al., PR B **93**, 144506 (2016)*

- » **Nonlinear phononics.** Driving of Raman modes leads to the cubic coupling to an infrared phonon mode, which causes induced superconductivity

*R. Mankowski et al., Nature **516**, 71 (2014)*

→ Induced Superconductivity after pumping?

→ Is the underlying physics special for cuprates?

→ Transient Meissner effect? Can we calculate this?

1D Hubbard model in equilibrium

Our strategy: » calculate correlation function
 » calculate response function

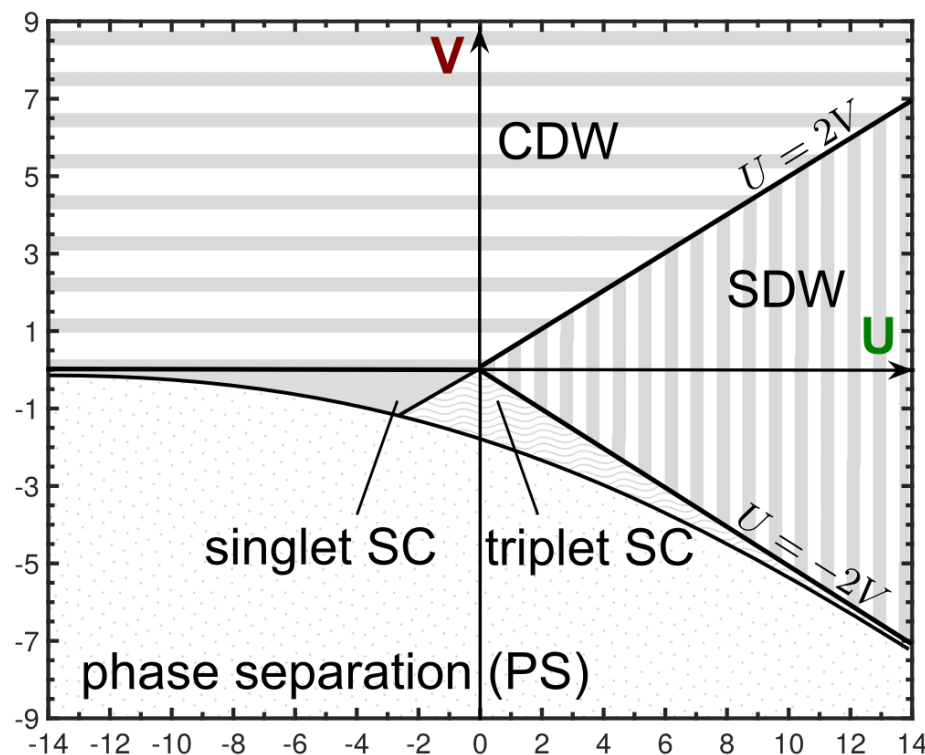
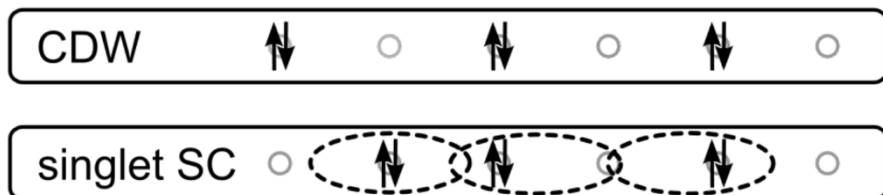
1D Hubbard model in a nutshell

Extended Hamilton operator

$$\hat{H}_0 = \underbrace{-t_h \sum_{i,\sigma} (\hat{c}_{i,\sigma}^\dagger \hat{c}_{i+1,\sigma} + H.c.)}_{\text{Kinetic term}} + \underbrace{U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}}_{\text{On-site interaction}} + \underbrace{V \sum_{\langle i,j \rangle} \hat{n}_i \hat{n}_j}_{\text{Nearest neighbor interaction}}$$

Phase diagram

- » A “toy” model
- » Well studied in equilibrium
- » Very rich phase diagram



J.Solyom, Adv. Phys. **28**, 201 (1979)

J.Voit, PR B **45**, 4027 (1992), *Rep.Prog.Phys.* **58**, 977 (1995)

Two types of quenching

Extended Hamilton operator

$$\hat{H}_0 = \underbrace{-t_h \sum_{i,\sigma} (\hat{c}_{i,\sigma}^\dagger \hat{c}_{i+1,\sigma} + H.c.)}_{\text{Kinetic term}} + \underbrace{U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}}_{\text{On-site interaction}} + \underbrace{V \sum_{\langle i,j \rangle} \hat{n}_i \hat{n}_j}_{\text{Nearest neighbor interaction}}$$

Time dependence

I) Interaction quench

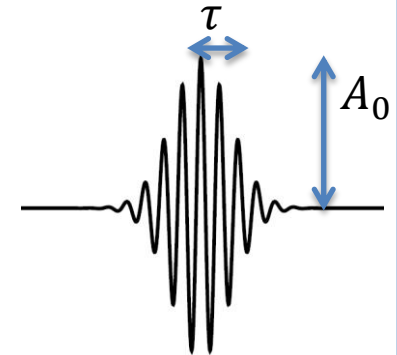
$$U \rightarrow U + \Delta U \cdot \Theta(t - t_0)$$

$$V \rightarrow V + \Delta V \cdot \Theta(t - t_0)$$

II) Pulse quench

$$A(t) = A_0 e^{-(t-t_0)^2/2\tau^2} \cos[\omega_0(t - t_0)]$$

Peierls substitution $t_h \rightarrow t_h \cdot e^{iA(t)}$



Time dependent exact diagonalization

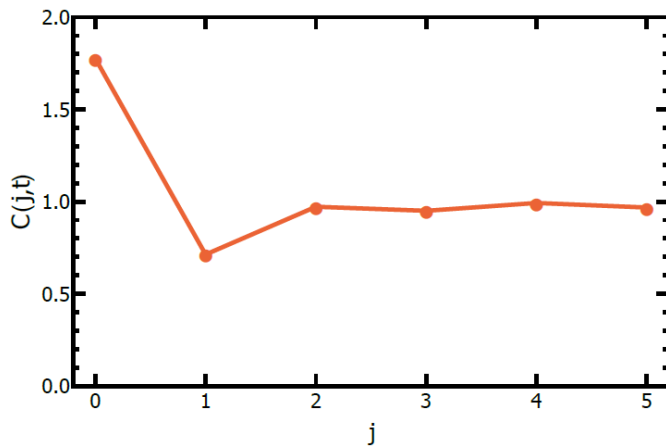
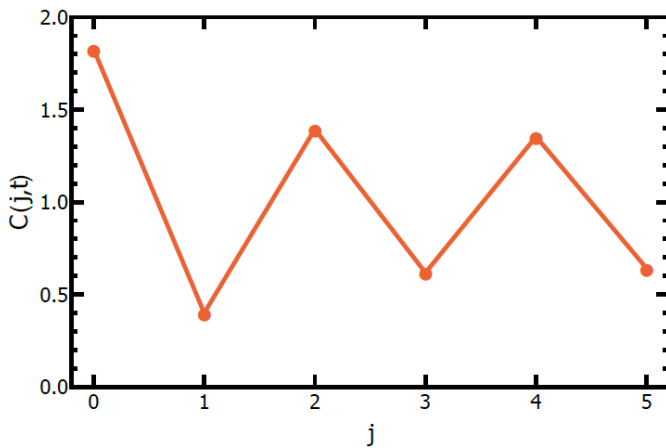
→ Lanczos method in time intervals δt

→ Stepwise approximation of $|\psi(t)\rangle$

$$|\psi(t + \delta t)\rangle \simeq e^{-i\hat{H}(t)\delta t} |\psi(t)\rangle \simeq \sum_{l=1}^M e^{-i\epsilon_l \delta t} |\phi_l\rangle \langle \phi_l | \psi(t)\rangle$$

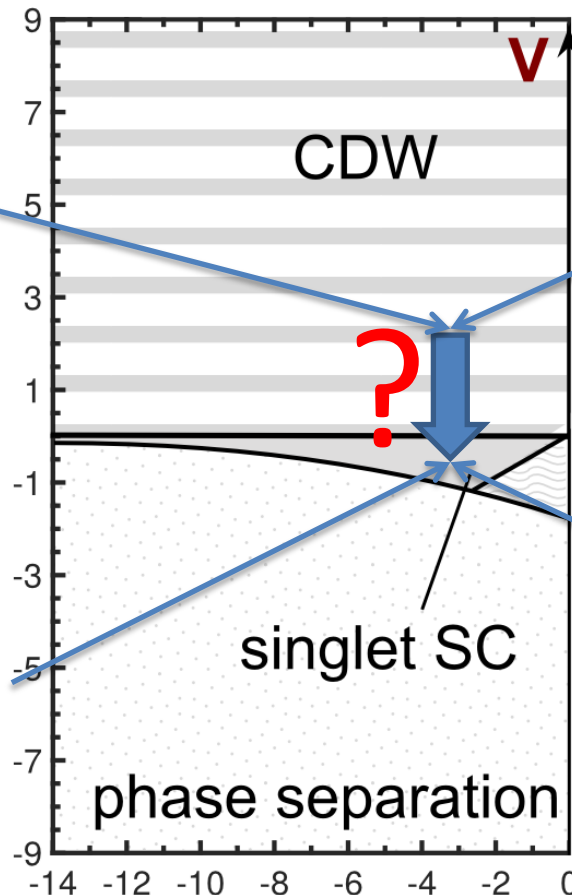
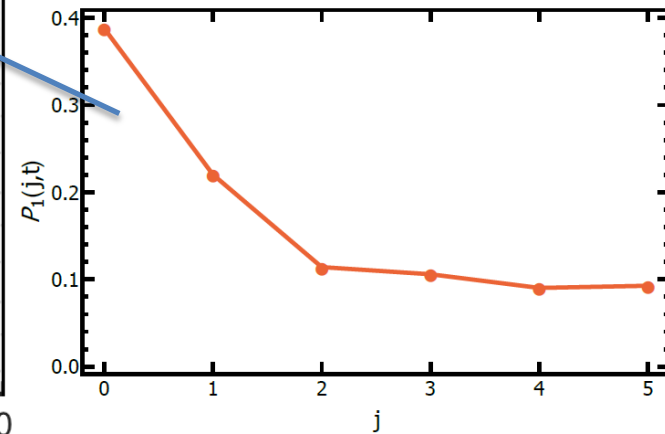
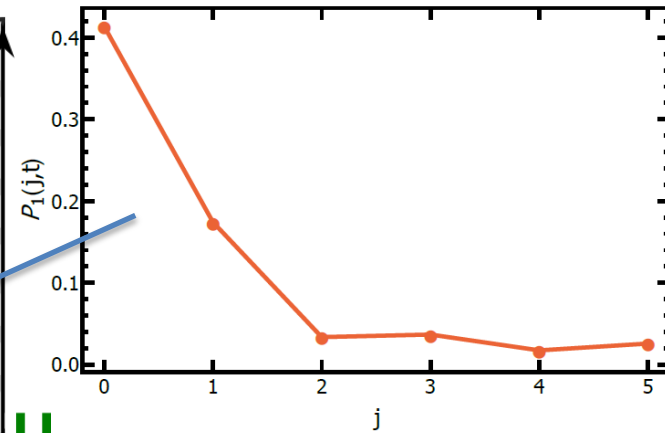
Density-density correlation function

$$C(j) = \frac{1}{L} \sum_{l=0}^{L-1} \langle \psi | \hat{n}_{l+j} \hat{n}_l | \psi \rangle$$



On-site correlation function

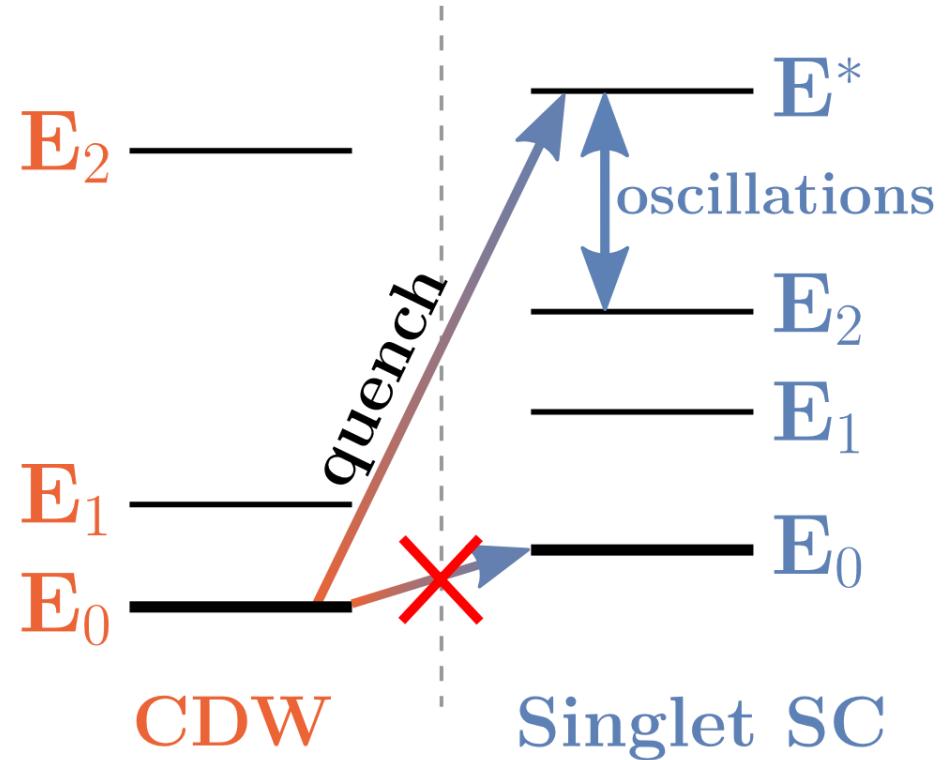
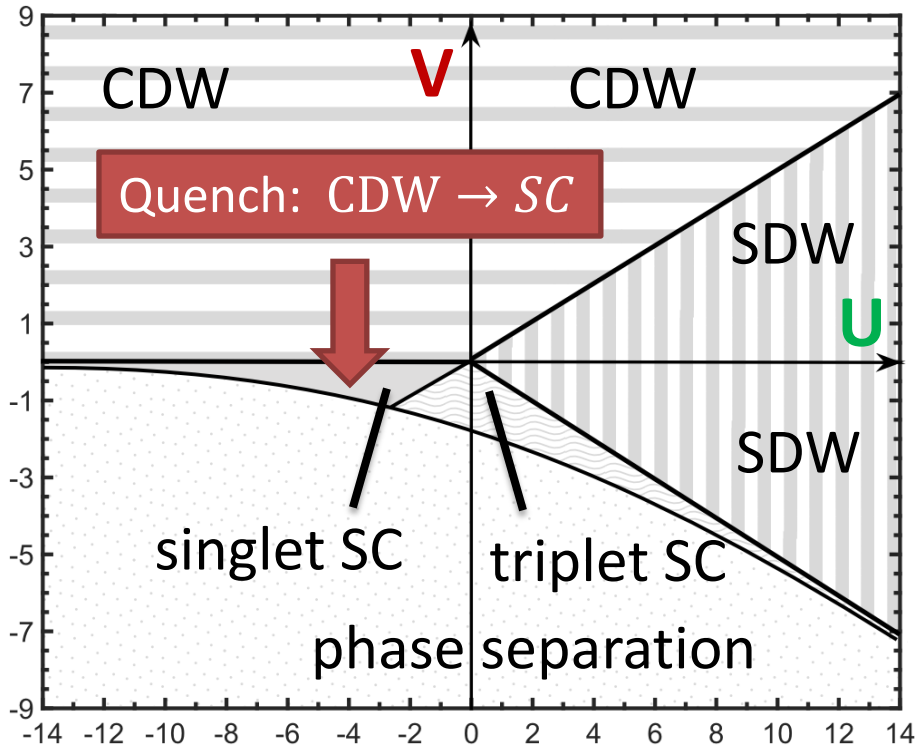
$$P_1(j) = \frac{1}{L} \sum_{l=0}^{L-1} \langle \psi | \hat{c}_{l+j\downarrow}^\dagger \hat{c}_{l+j\uparrow}^\dagger \hat{c}_{l\uparrow} \hat{c}_{l\downarrow} | \psi \rangle$$



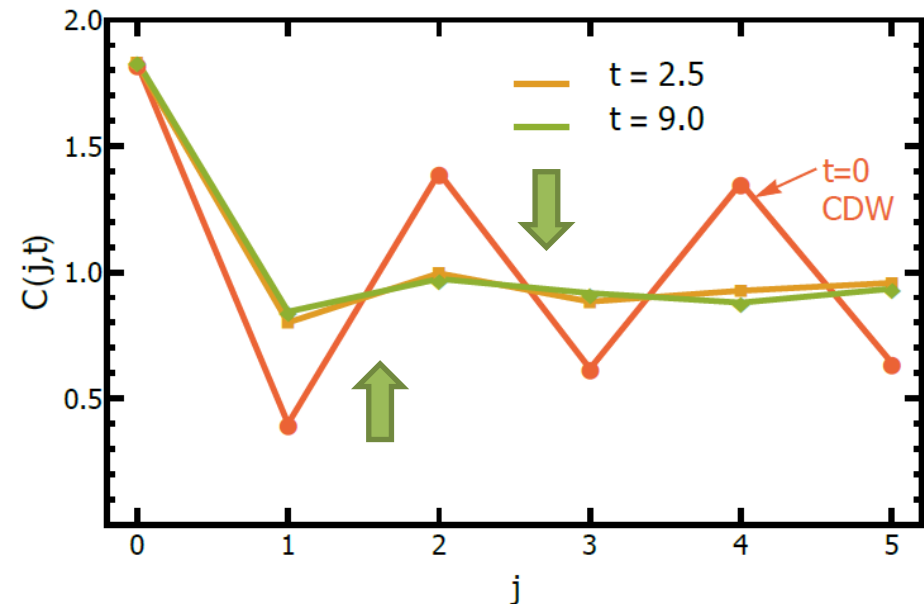
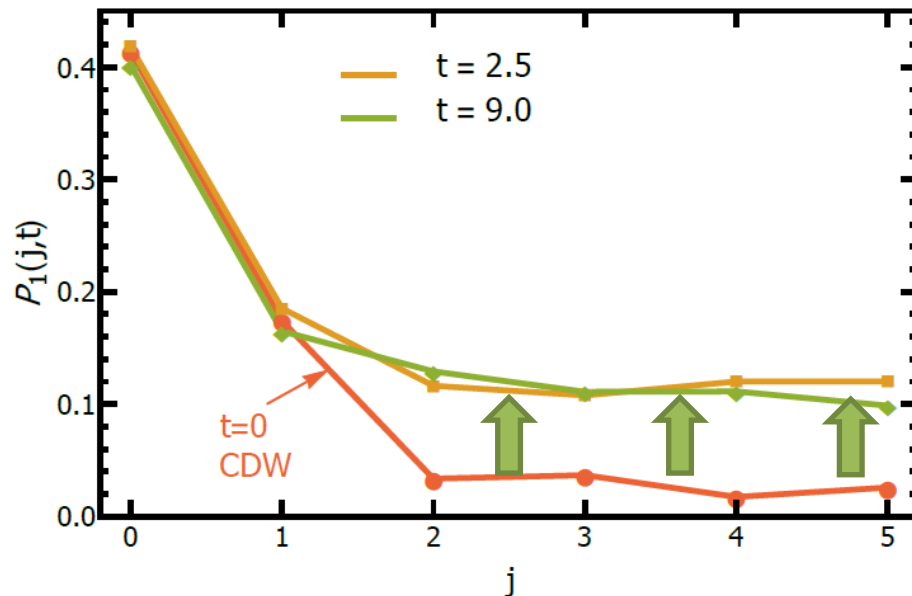
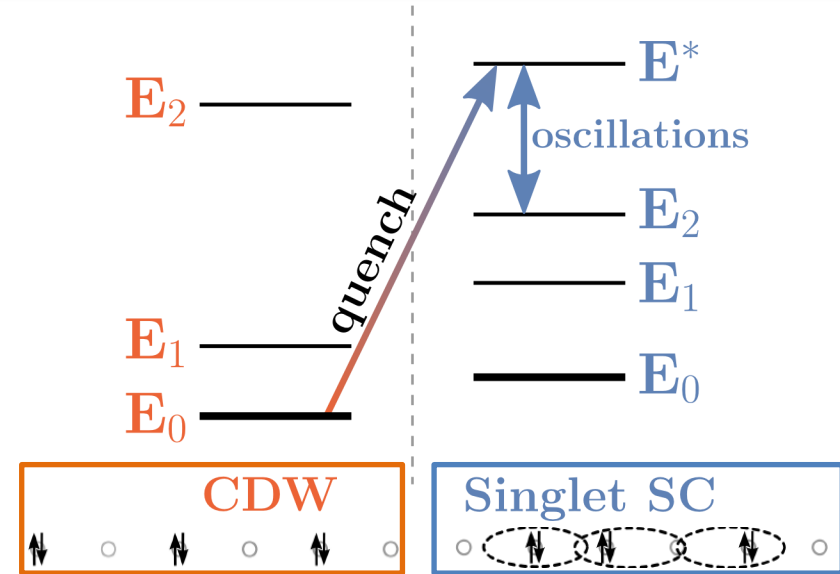
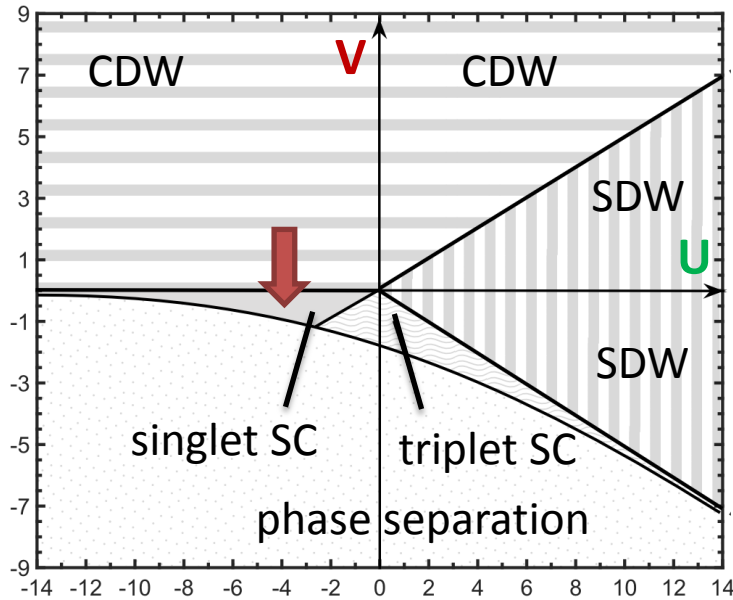
Results

Interaction quench

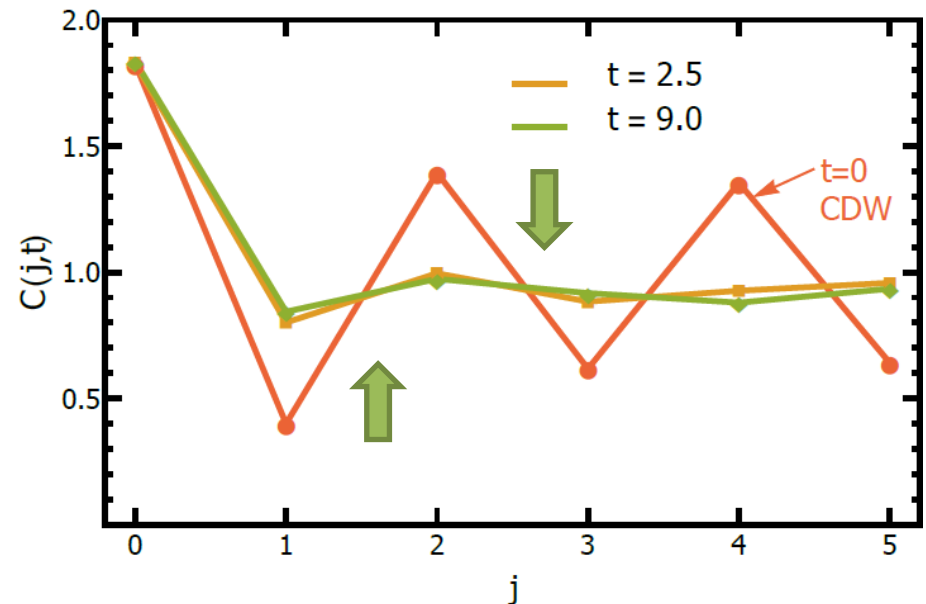
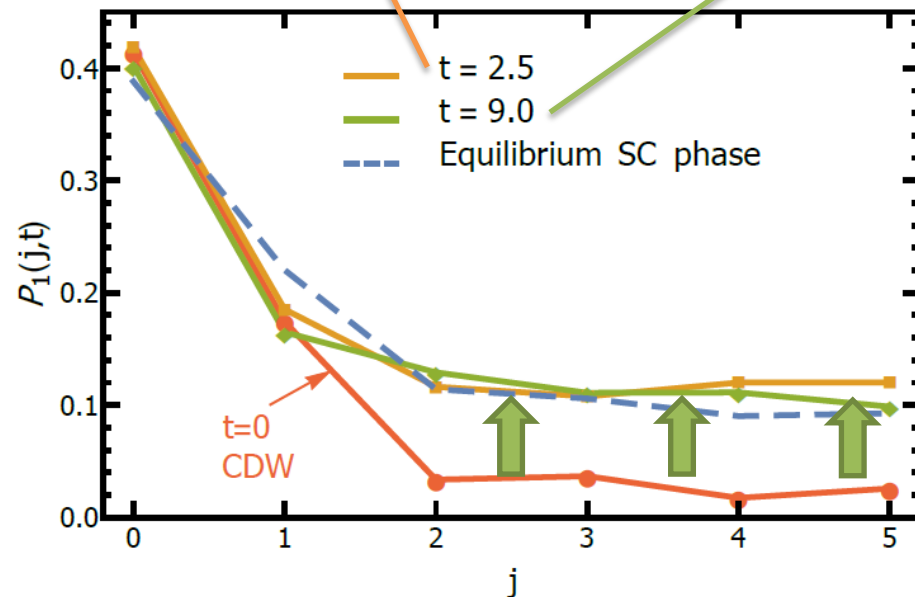
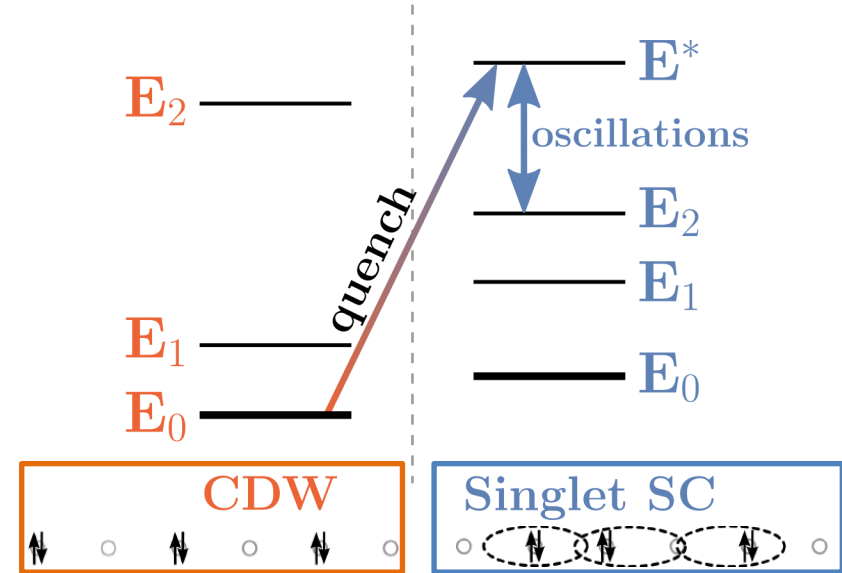
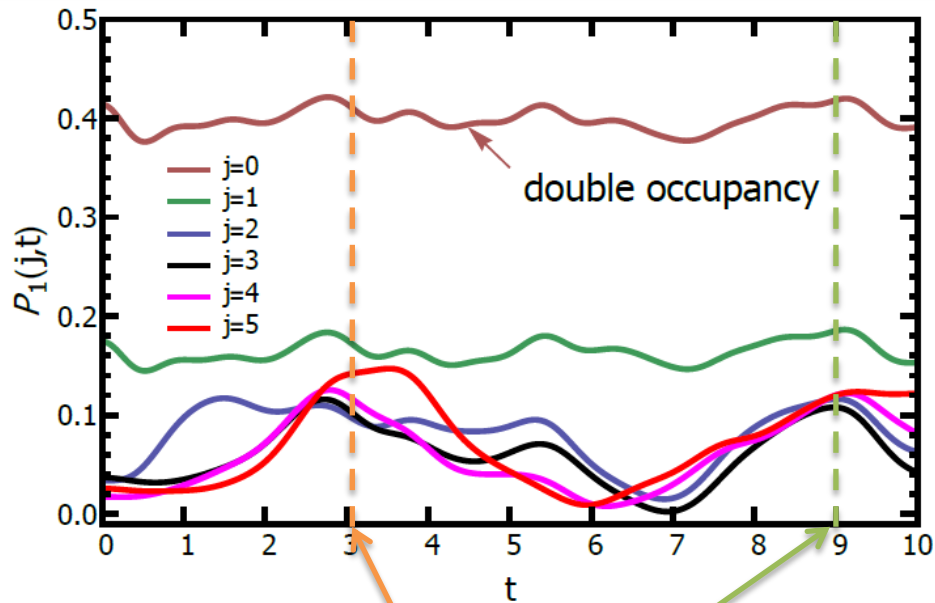
$$V \rightarrow V \cdot \Theta(t - t_0)$$



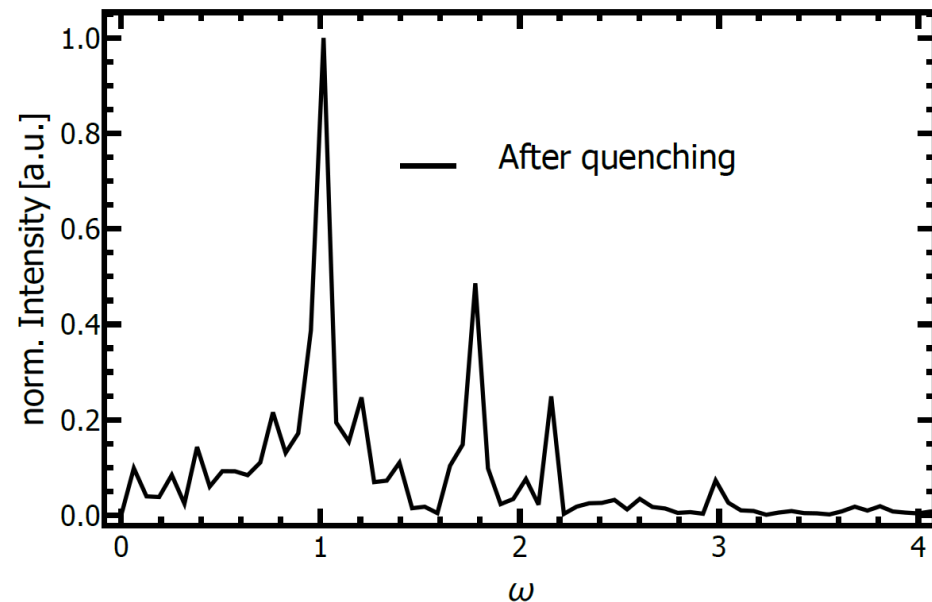
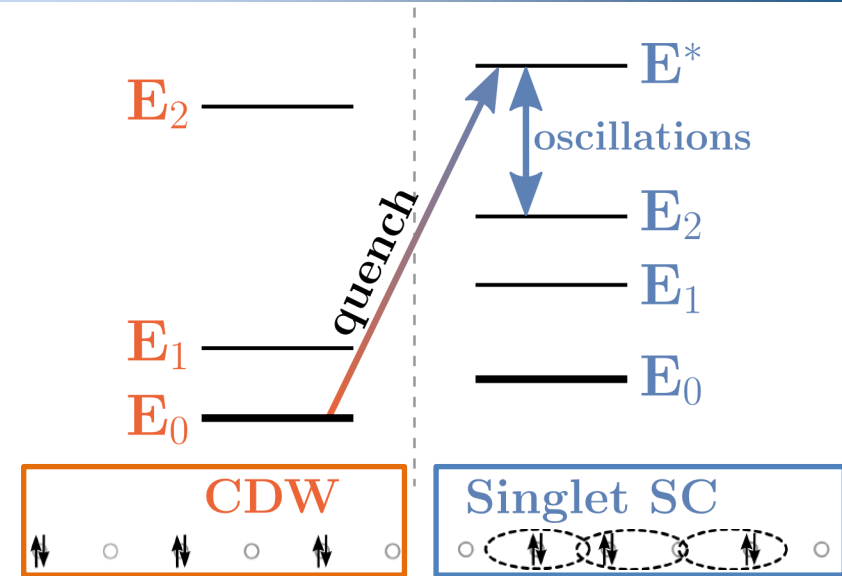
Induced SC? First indications



Induced SC? First indications



Transient Superconductivity

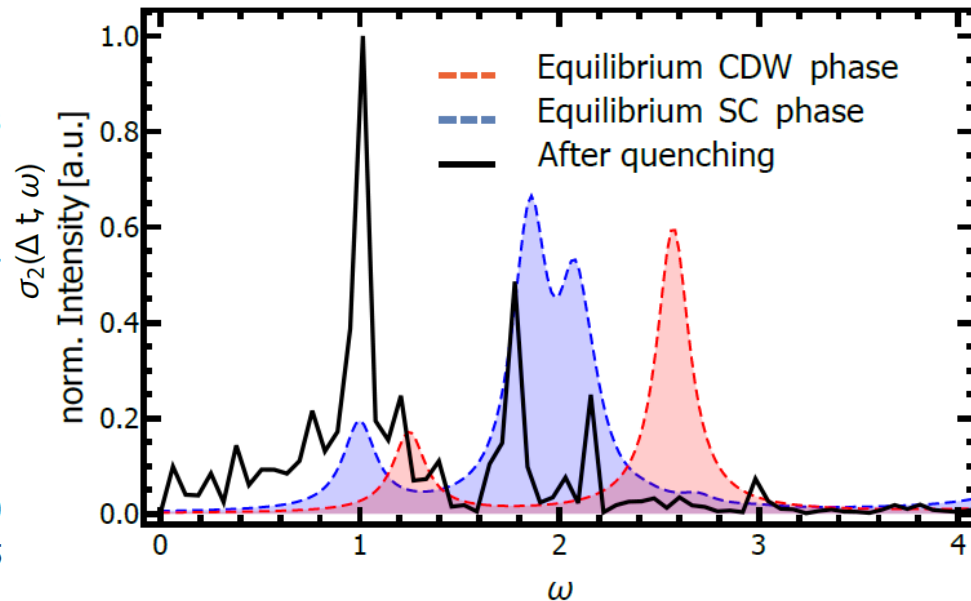
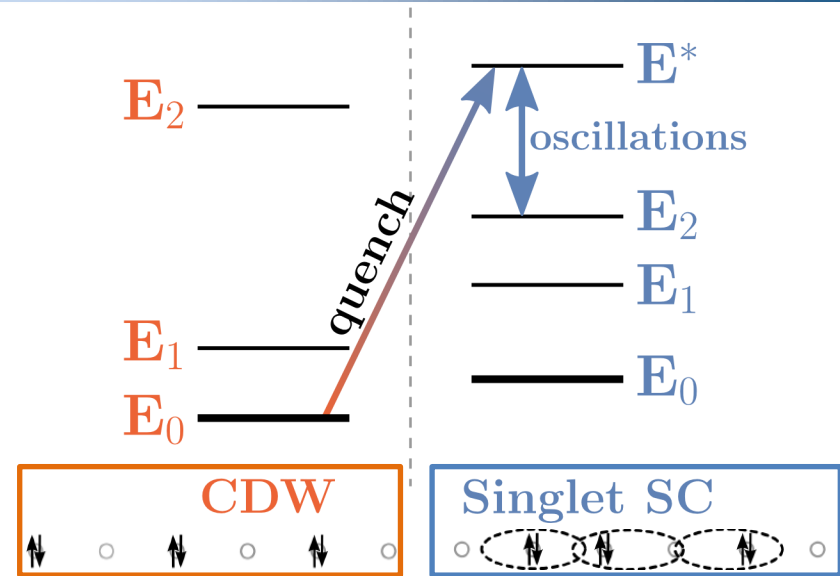
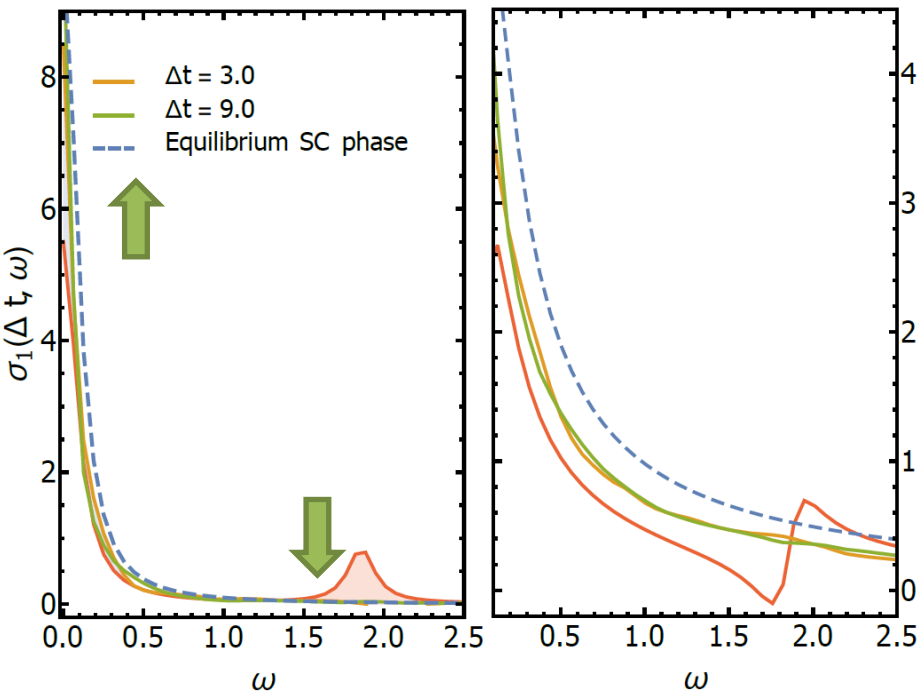


Transient Meissner effect ?



Time-dependent optical conductivity

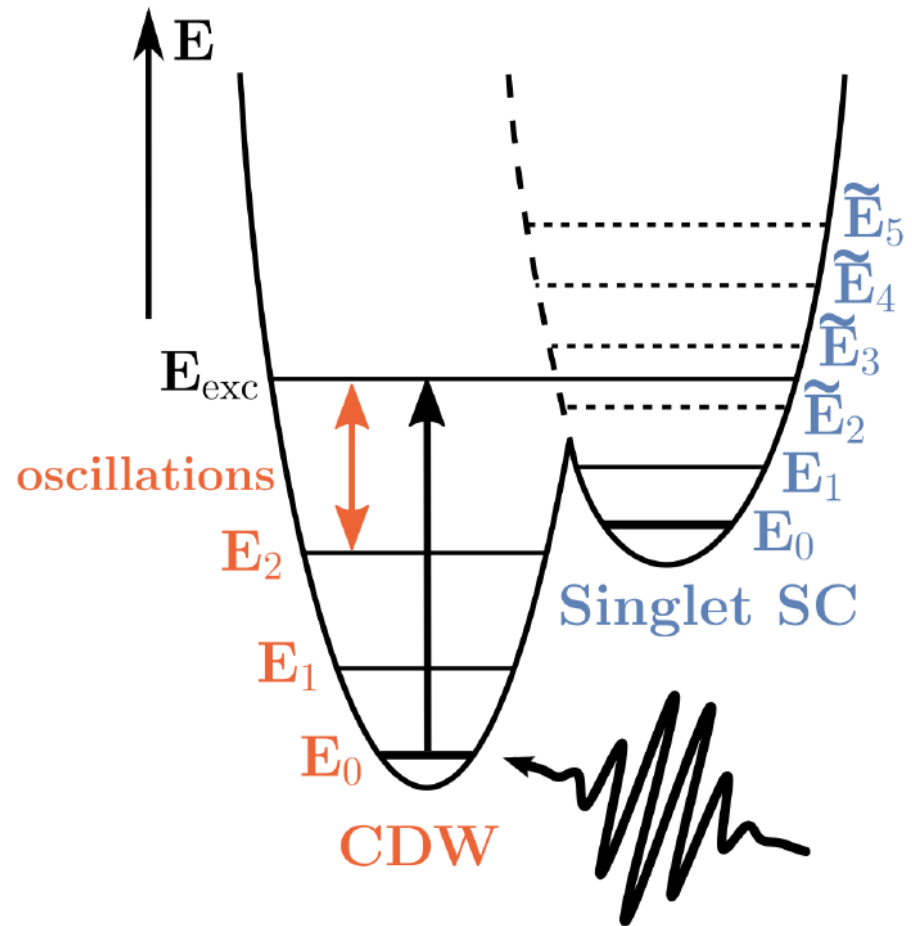
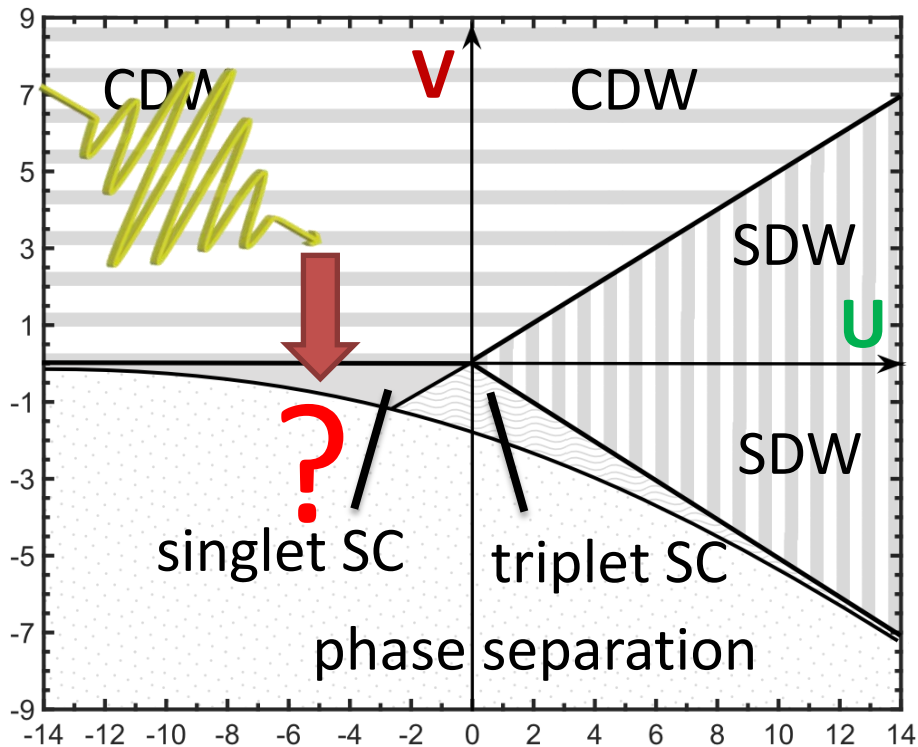
$$\sigma(\Delta t, \omega) = \frac{j_{\text{pr}}(\Delta t, \omega)}{i(\omega + i\eta)L A_{\text{pr}}(\Delta t, \omega)}$$



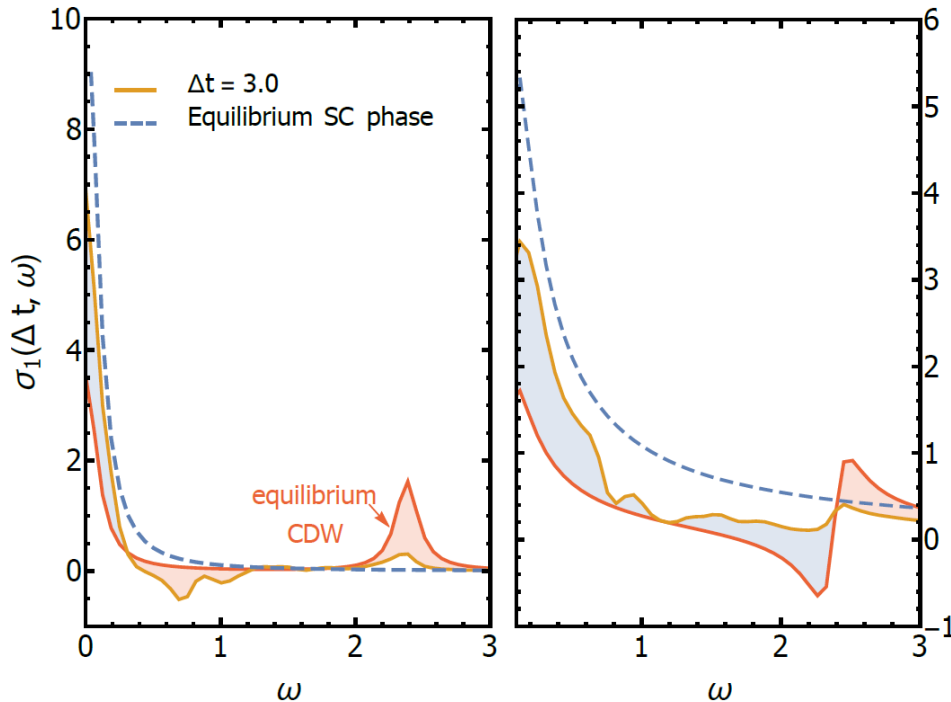
Pulse quench

$$t_h \rightarrow t_h \cdot e^{iA(t)}$$

$$A(t) = A_0 e^{-(t-t_0)^2/2\tau^2} \cos[\omega_0(t-t_0)]$$

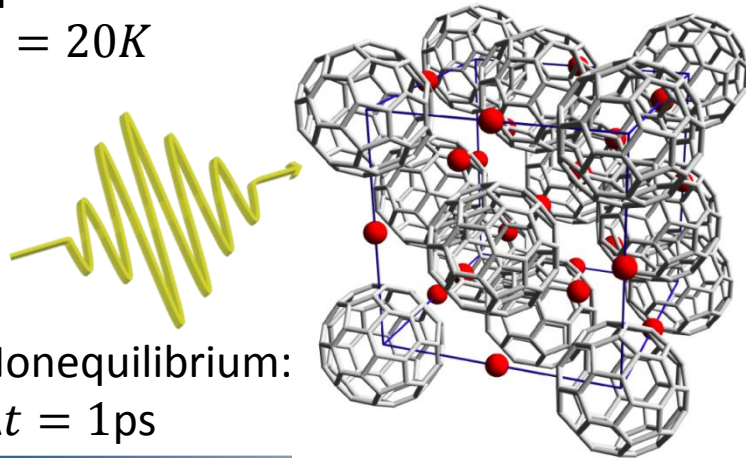


Optical conductivity



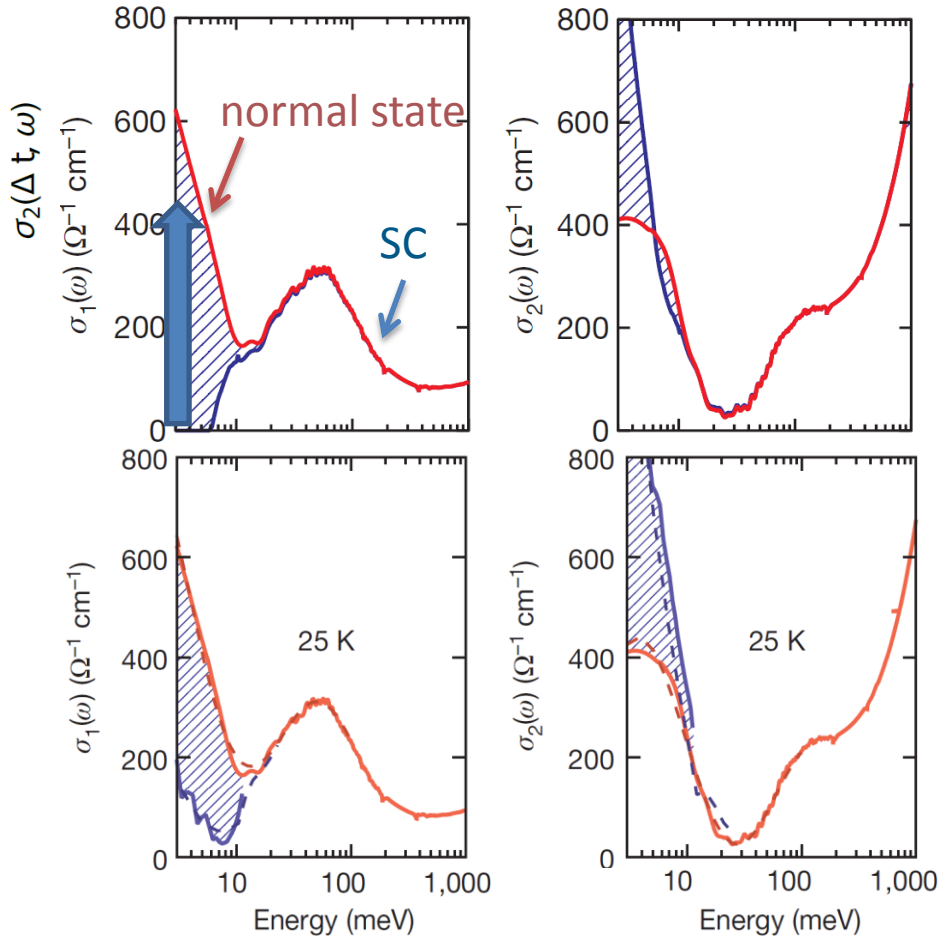
Equilibrium:
 $T_c = 20K$

Nonequilibrium:
 $\Delta t = 1ps$

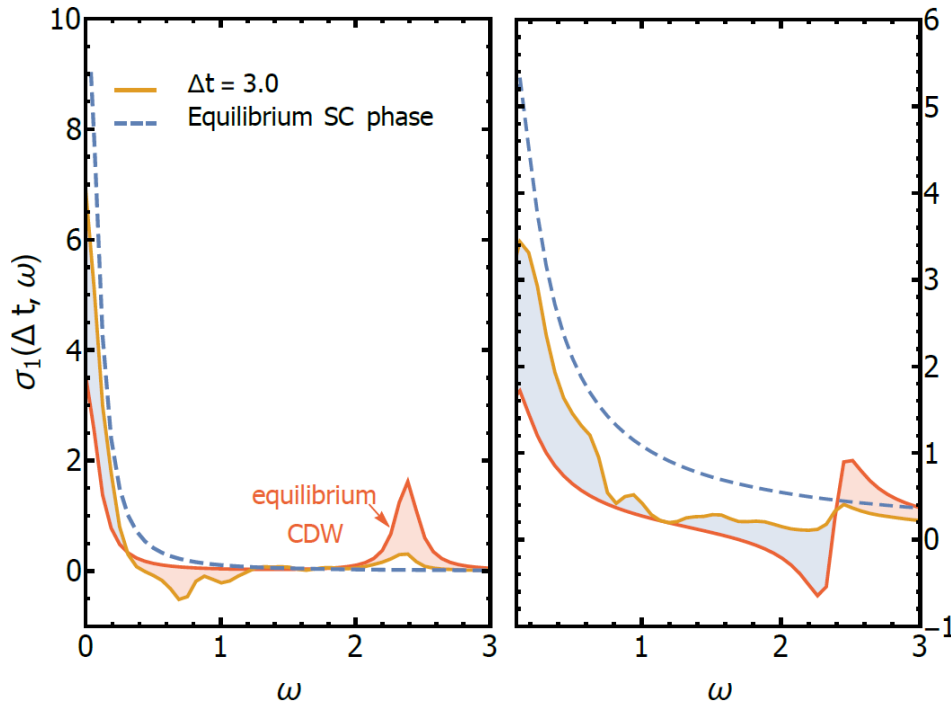


s-wave superconductor K_3C_{60}

Mitrano et.al, Nature **530** (2016)

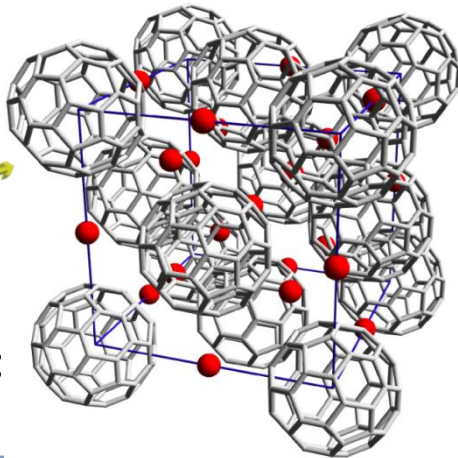


Optical conductivity



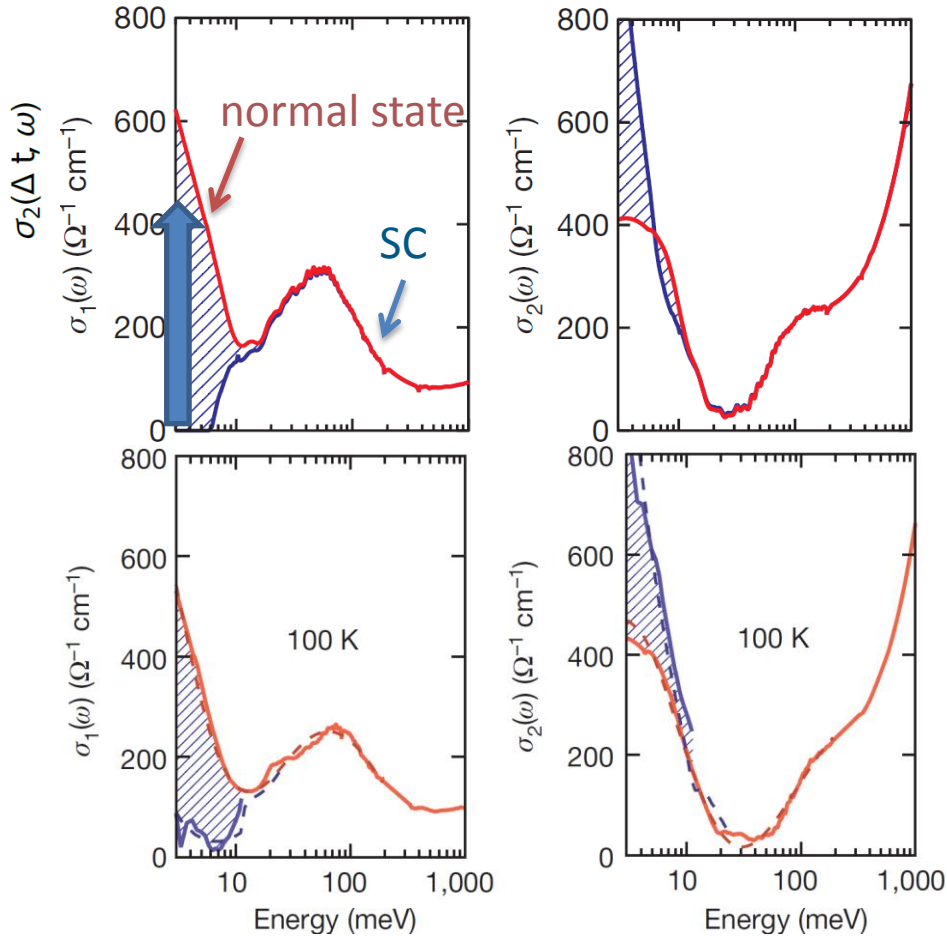
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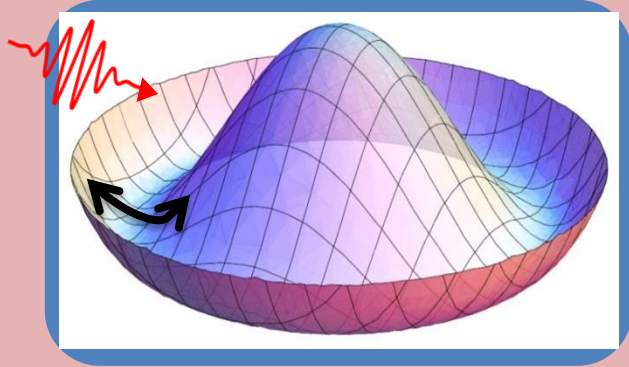


s-wave superconductor K_3C_{60}

Mitrano et.al, Nature **530** (2016)



New! Higgs spectroscopy



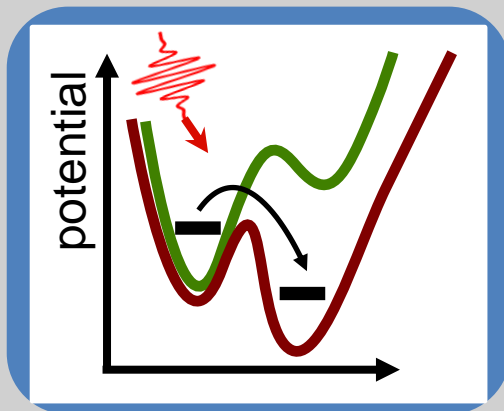
» Direct detection of the SC gap

Part 2

Order parameter oscillations in Superconductors

*H. Krull, N. Bittner, G. S. Uhrig, D. Manske, and A.P. Schnyder,
Nat. Commun. 7:11921 (2016)*

optical control



» New transient ground state

Part 1

Induced Superconductivity in the extended Hubbard Model

N. Bittner, T. Tohyama, and D. Manske, in preparation

One-band superconductor

» Phase (gauge) mode

⇒ Shift to plasma frequency

(Anderson-Higgs mechanism)

P. W. Anderson (1963), P. W. Higgs (1964)

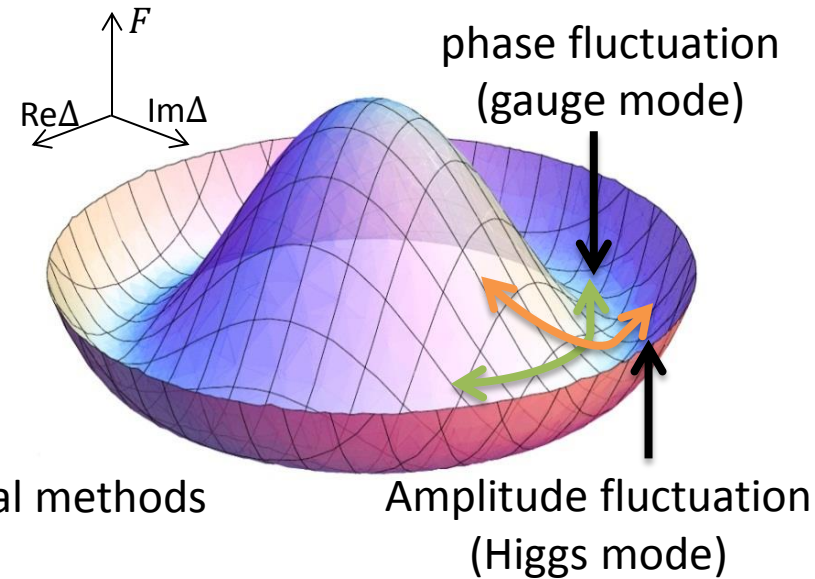
» Amplitude (Higgs) mode

⇒ **lies at 2Δ** . Not detectable in Raman response

⇒ **No charge or dipole moment**. Not seen by optical methods

⇒ indirect detection in **special cases** possible

*M.-A. Measson et al., PR B **89**, 060503 (2014), D. Sherman et al., Nat. Phys. **11**, 188 (2015)*



One-band superconductor

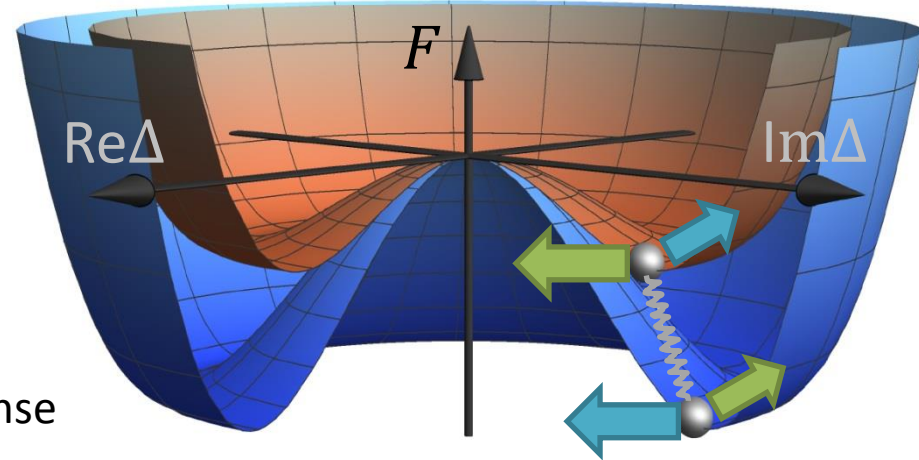
» Phase (gauge) mode

- ⇒ Shift to plasma frequency
(**Anderson-Higgs mechanism**)
P. W. Anderson (1963), P. W. Higgs (1964)

» Amplitude (Higgs) mode

- ⇒ **lies at 2Δ** . Not detectable in Raman response
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*M.-A. Measson et al., PR B **89**, 060503 (2014), D. Sherman et al., Nat. Phys. **11**, 188 (2015)*



Two-band superconductor

$$\omega_L^2 = 4 |\Delta_1| |\Delta_2| \frac{V}{V_{22} - V_{11}V^2}$$

*A. J. Leggett, Prog. Theor. Phys. **36**, 901 (1966)*

» Leggett mode = out-of-phase vibration of the superconducting condensates

- ⇒ **has no analogy** in the one band case
- ⇒ **intrinsic Josephson effect**

- ⇒ Experimental **evidence in Raman response** for MgB_2
*G. Blumberg et al., Phys. Rev. Lett. **99**, 227002 (2007)*

- Direct detection of the Higgs mode in a superconductor?
- Is there a Josephson effect in nonequilibrium?

Method

Shake the condensate!



1. THz pulse
2. short pulse width (non-adiabatic regime)
3. low pulse fluency

» Hamiltonian for a two-band superconductor:

$$\hat{H} = \sum_{l=1}^2 \left[\sum_{k\sigma} \epsilon_{kl} \hat{c}_{k\sigma l}^\dagger \hat{c}_{k\sigma l} + \sum_k \left(\Delta_l \hat{c}_{k\uparrow l}^\dagger \hat{c}_{-k\downarrow l}^\dagger + h.c. \right) \right] \leftarrow \text{BCS}$$

» Assumptions:

- (a) tetragonal lattice
- (b) s-wave order parameters

» Energy gap equations:

$$\begin{aligned} \Delta_1 &= \sum_k [V_{11} \langle \hat{c}_{-k\downarrow 1} \hat{c}_{k\uparrow 1} \rangle + V \langle \hat{c}_{-k\downarrow 2} \hat{c}_{k\uparrow 2} \rangle V_{11}] \\ \Delta_2 &= \sum_k [V_{22} \langle \hat{c}_{-k\downarrow 2} \hat{c}_{k\uparrow 2} \rangle + V \langle \hat{c}_{-k\downarrow 1} \hat{c}_{k\uparrow 1} \rangle V_{11}] \quad \text{with } V = V_{12}/V_{11} \end{aligned}$$

» Hamiltonian for a two-band superconductor **in nonequilibrium**:

$$\hat{H}(t) = \sum_{l=1}^2 \left[\sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}l} \hat{c}_{\mathbf{k}\sigma l}^\dagger \hat{c}_{\mathbf{k}\sigma l} + \sum_{\mathbf{k}} \left(\Delta_l(t) \hat{c}_{\mathbf{k}\uparrow l}^\dagger \hat{c}_{-\mathbf{k}\downarrow l}^\dagger + h.c. \right) \right] \leftarrow \text{BCS}$$

$$-e\hbar \sum_{\mathbf{k}q\sigma l} \frac{(2\mathbf{k} + \mathbf{q}) \cdot \mathbf{A}_q(t)}{2m_l} \hat{c}_{\mathbf{k}+\mathbf{q}\sigma l}^\dagger \hat{c}_{\mathbf{k}\sigma l} + e^2 \sum_{\mathbf{k}q\sigma l} \frac{\sum_{q'} \mathbf{A}_{q-q'}(t) \cdot \mathbf{A}_{q'}(t)}{2m_l} \hat{c}_{\mathbf{k}+\mathbf{q}\sigma l}^\dagger \hat{c}_{\mathbf{k}\sigma l}$$

probe pulse

pump pulse

coupling to the field

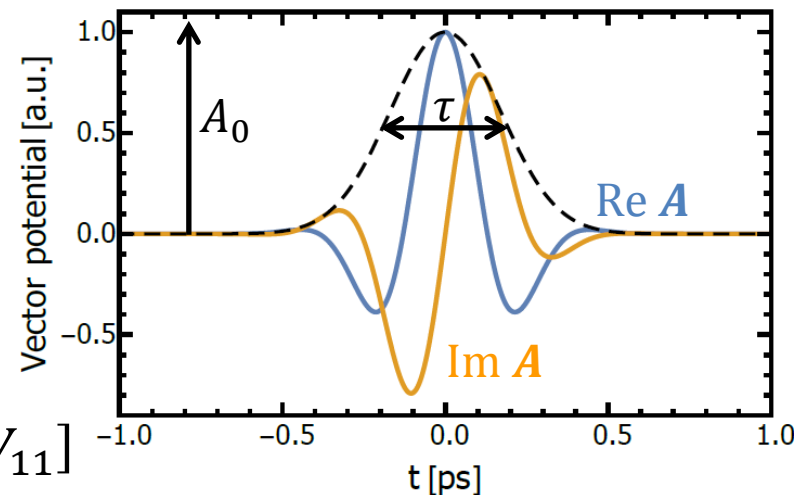
» Assumptions:

- (a) tetragonal lattice
- (b) s-wave order parameters

» Energy gap equations:

$$\Delta_1(t) = \sum_{\mathbf{k}} [V_{11} \langle \hat{c}_{-\mathbf{k}\downarrow 1} \hat{c}_{\mathbf{k}\uparrow 1} \rangle(t) + \mathbf{V} \langle \hat{c}_{-\mathbf{k}\downarrow 2} \hat{c}_{\mathbf{k}\uparrow 2} \rangle(t) V_{11}]$$

$$\Delta_2(t) = \sum_{\mathbf{k}} [V_{22} \langle \hat{c}_{-\mathbf{k}\downarrow 2} \hat{c}_{\mathbf{k}\uparrow 2} \rangle(t) + \mathbf{V} \langle \hat{c}_{-\mathbf{k}\downarrow 1} \hat{c}_{\mathbf{k}\uparrow 1} \rangle(t) V_{11}] \quad \text{with } \mathbf{V} = V_{12}/V_{11}$$



- » Bogoliubov-Valatin transformation (superconducting state):

$$\alpha_{kl}^\dagger = u_{kl} \hat{c}_{k\uparrow l}^\dagger + v_{kl}^* \hat{c}_{-k\downarrow l} \quad \beta_{kl}^\dagger = u_{kl} \hat{c}_{-k\downarrow l}^\dagger - v_{kl}^* \hat{c}_{k\uparrow l}$$



energy gap equation:

$$\Delta_l(t) = \sum_{kj} V_{lj} \left(u_{kj} v_{kj} [1 - \langle \alpha_{kj}^\dagger \alpha_{kj} \rangle - \langle \beta_{kj}^\dagger \beta_{kj} \rangle] + u_{kj}^2 \langle \beta_{kj} \alpha_{kj} \rangle - v_{kj}^2 \langle \alpha_{kj}^\dagger \beta_{kj}^\dagger \rangle \right) (t)$$

- » Derivation of the equations of motions for the Bogoliubov quasiparticle densities:

$$\langle \alpha_{kl}^\dagger \alpha_{kl} \rangle(t), \quad \langle \beta_{kl}^\dagger \beta_{kl} \rangle(t), \quad \langle \beta_{kl} \alpha_{kl} \rangle(t), \quad \langle \alpha_{kl}^\dagger \beta_{kl}^\dagger \rangle(t)$$

- » Density matrix theory:

$$\frac{d}{dt} (\hat{c}_{k_1 l}^\dagger \hat{c}_{k_2 l}) = \frac{i}{\hbar} [\hat{H}, \hat{c}_{k_1 l}^\dagger \hat{c}_{k_2 l}] + \frac{\partial}{\partial t} (\hat{c}_{k_1 l}^\dagger \hat{c}_{k_2 l})$$

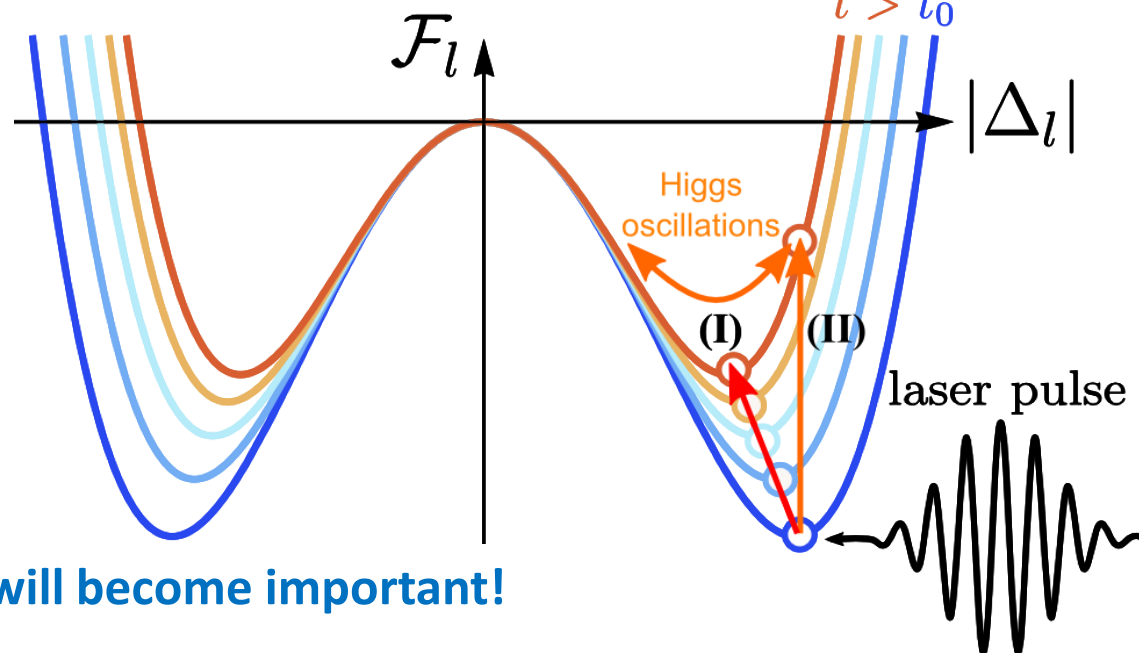
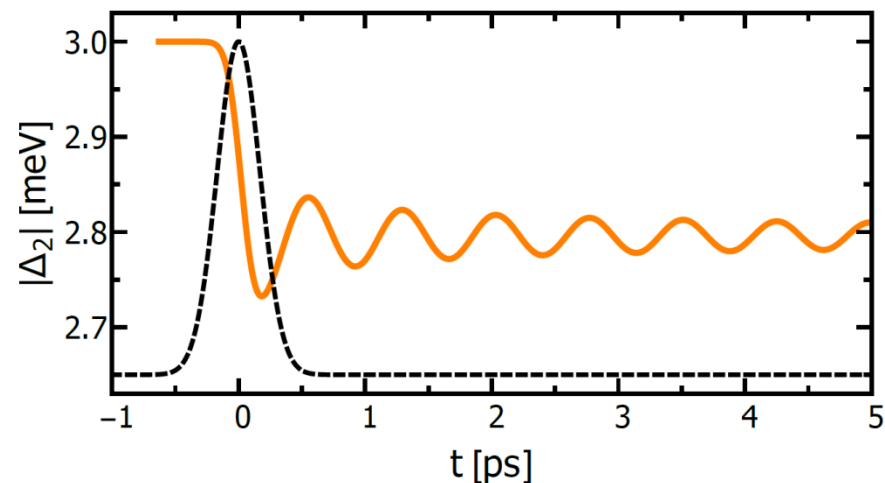
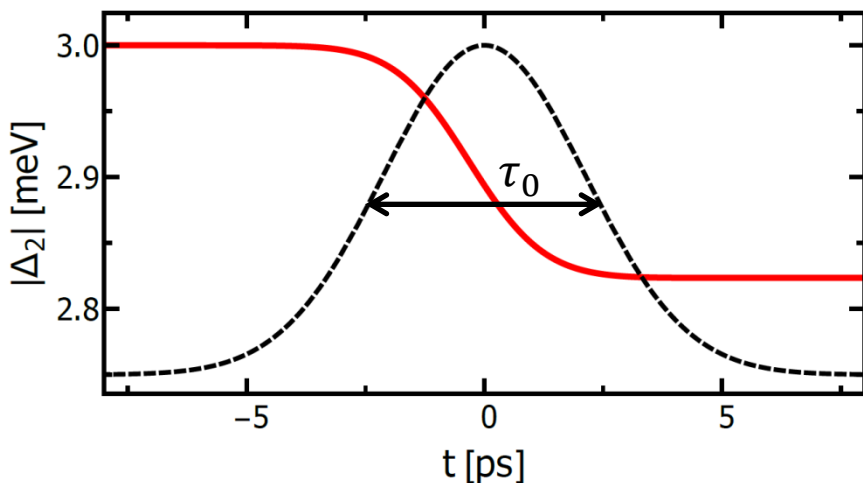


yield equations of motions for the expectation values

- » Numerical solution by using a Runge-Kutta method

Results: two independent bands
(single band solution)

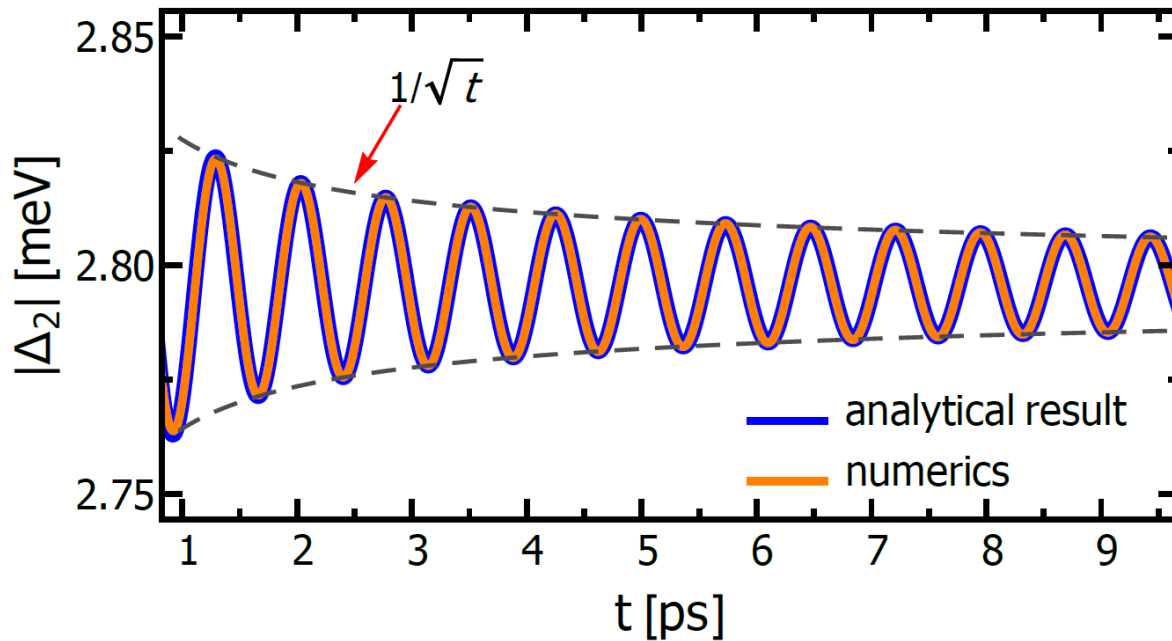
Higgs excitation in nonequilibrium



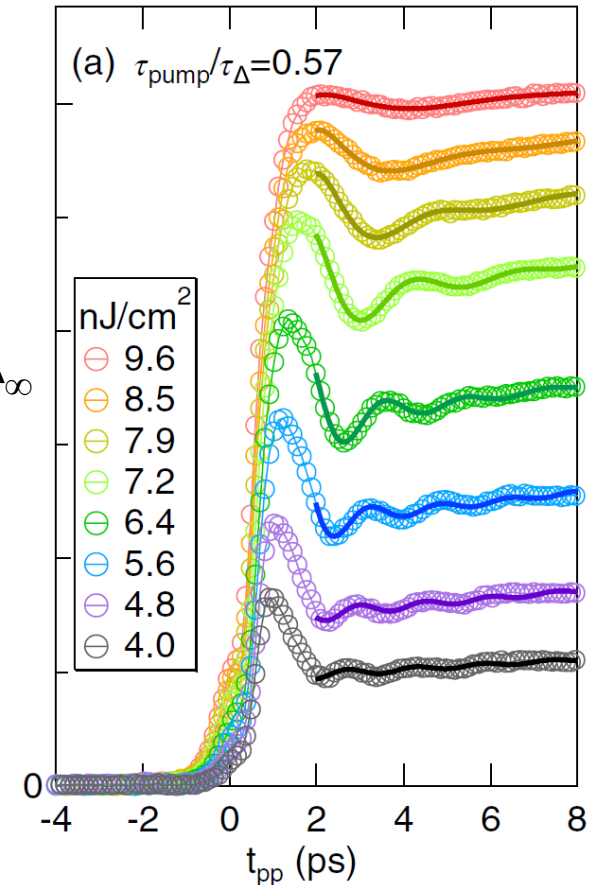
» Gaussian form of the laser pulses

» Pulse duration τ_0 will become important!

Analytic solution possible



→ Δ_∞



Matsunaga et al., PRL 2013

» Analytical expression (**interaction quench**):

$$\frac{|\Delta|}{|\Delta_\infty|} = 1 + \Gamma \frac{\cos\left(\frac{2|\Delta_\infty|}{\hbar} t + \Phi\right)}{\sqrt{t\Delta_\infty}}$$

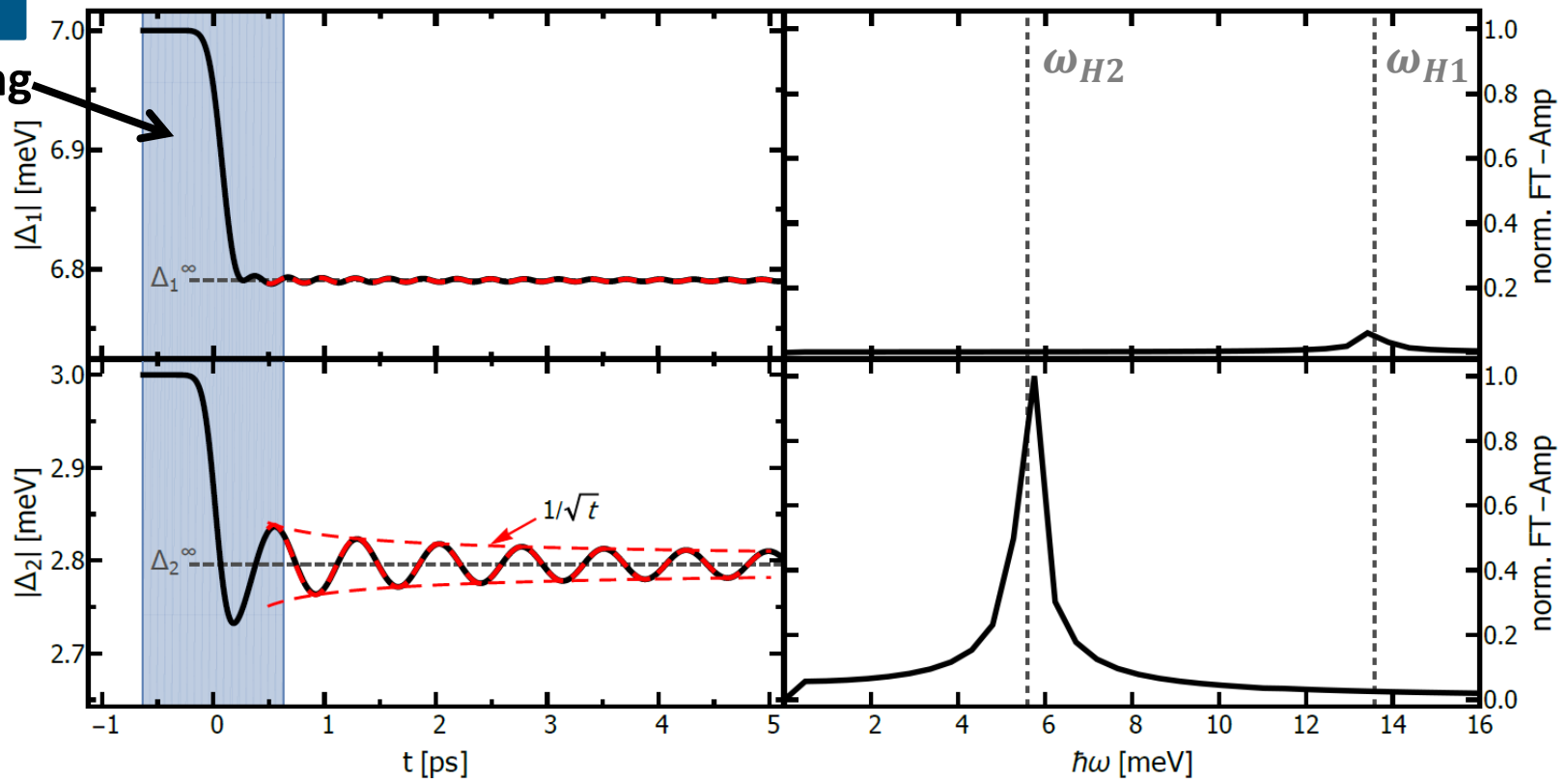
Yuzbashyan, Tsyplatyev, Altshuler, PRL 2006

Results: two coupled bands
(Josephson effect in nonequilibrium)

Leggett mode in nonequilibrium?

$$V = 0$$

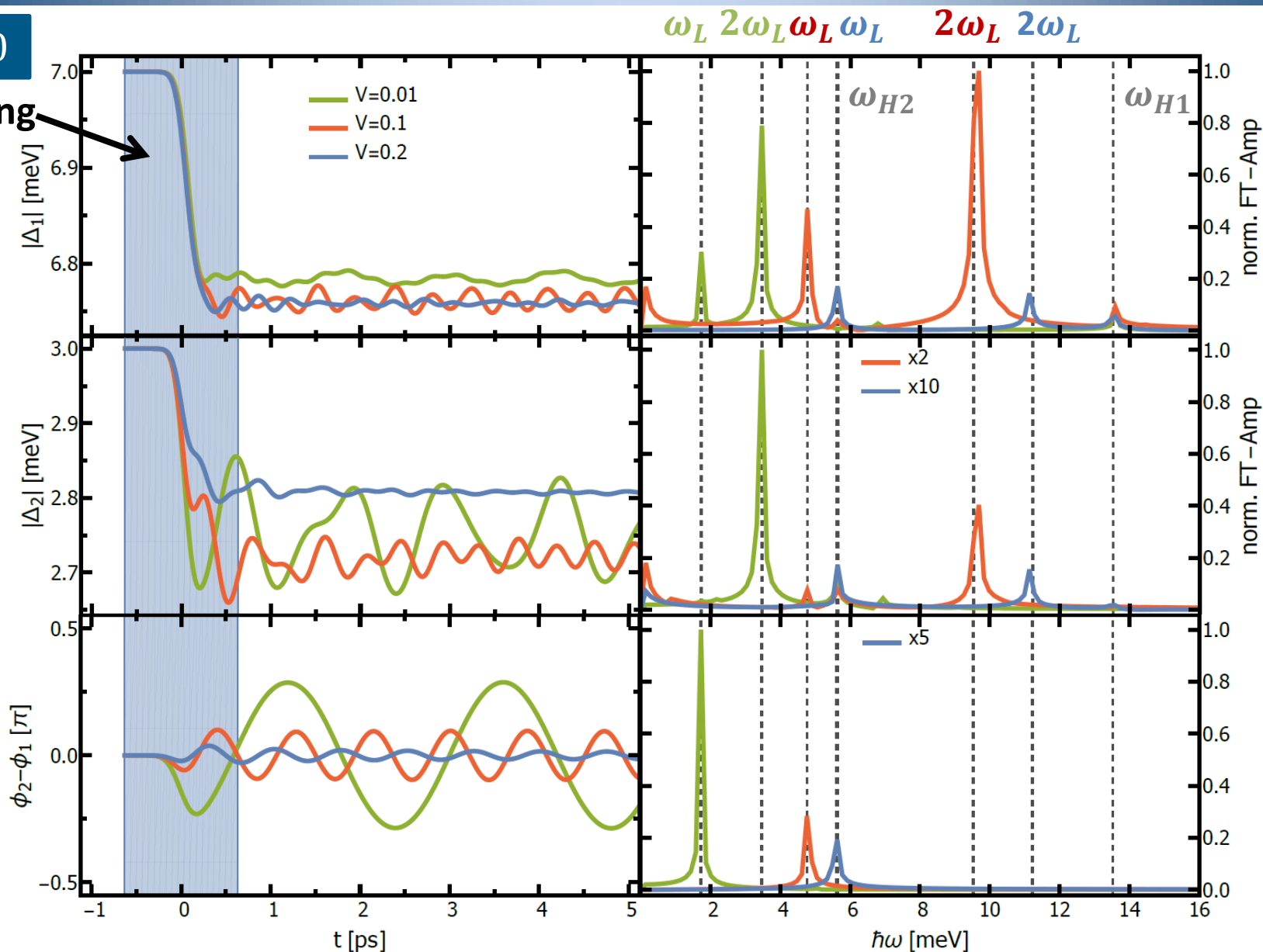
Pumping



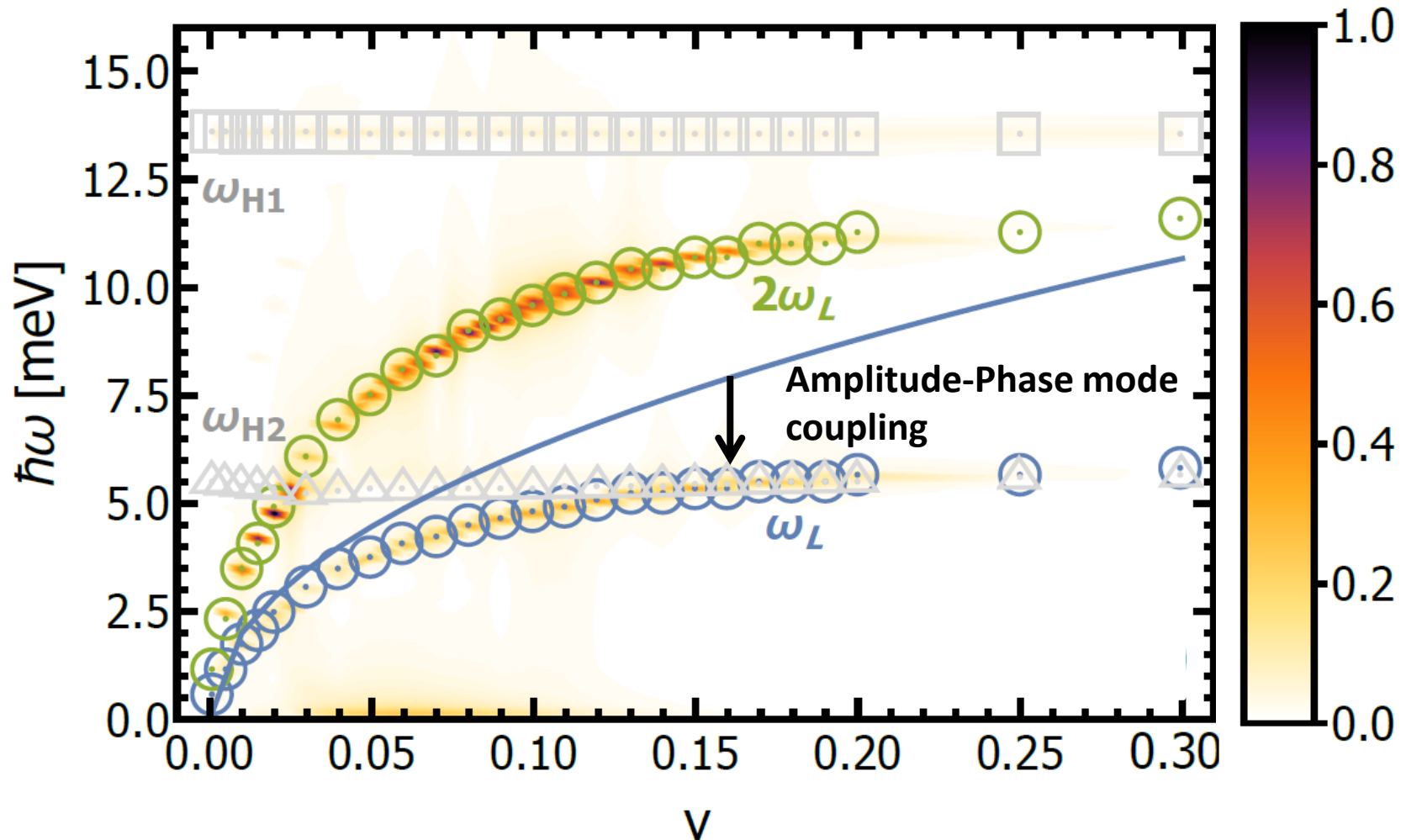
Leggett mode in nonequilibrium?

$V \neq 0$

Pumping



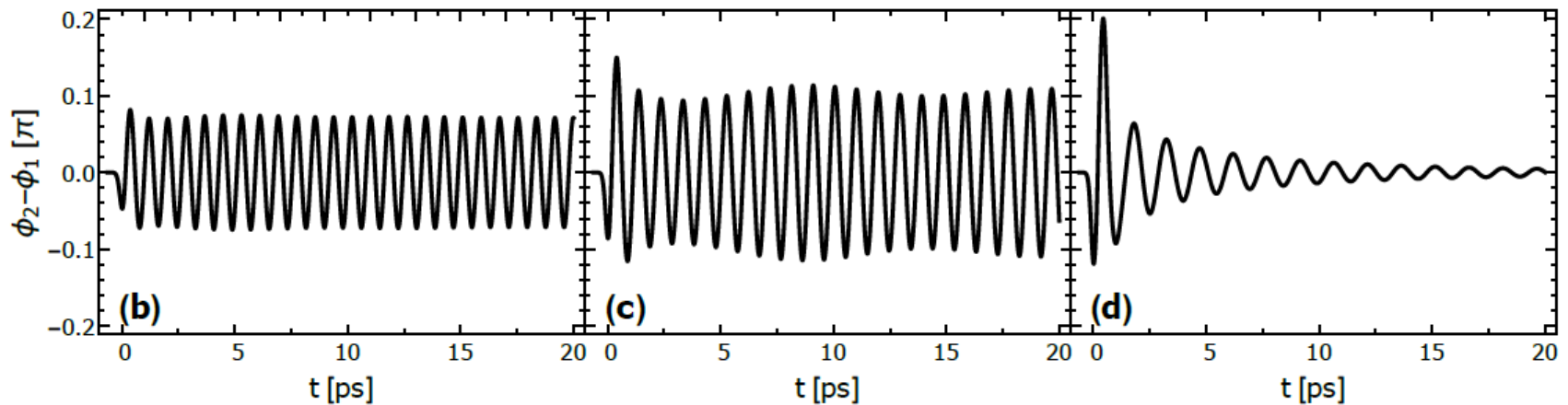
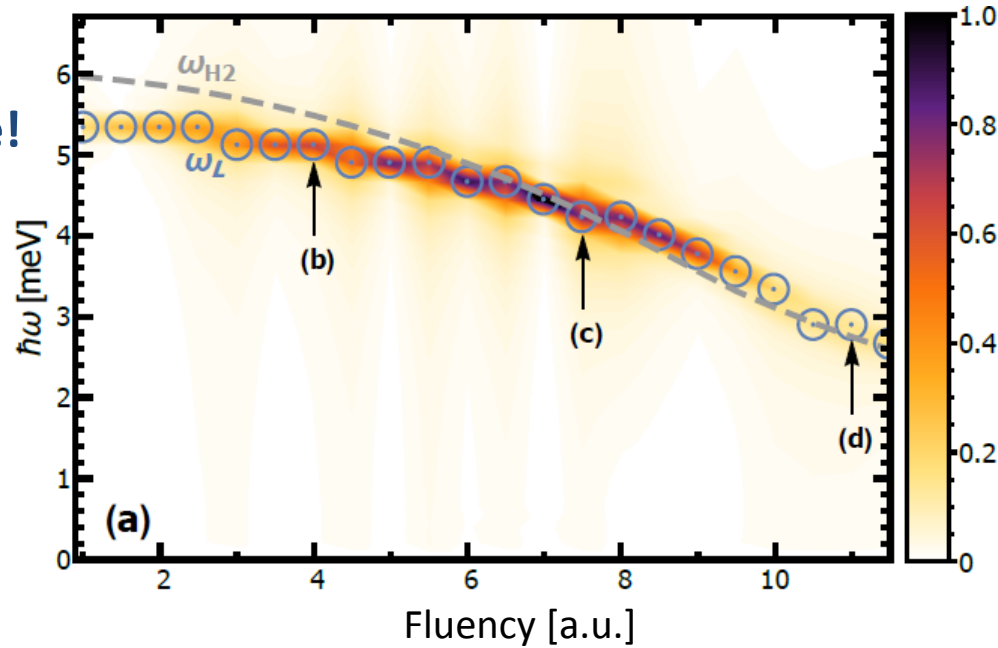
New Leggett mode in nonequilibrium: prediction



- » Amplitude-Phase mode coupling in nonequilibrium
- » Amplitude-channel: harmonic $2\omega_L$ only present in nonequilibrium

Intensity dependence

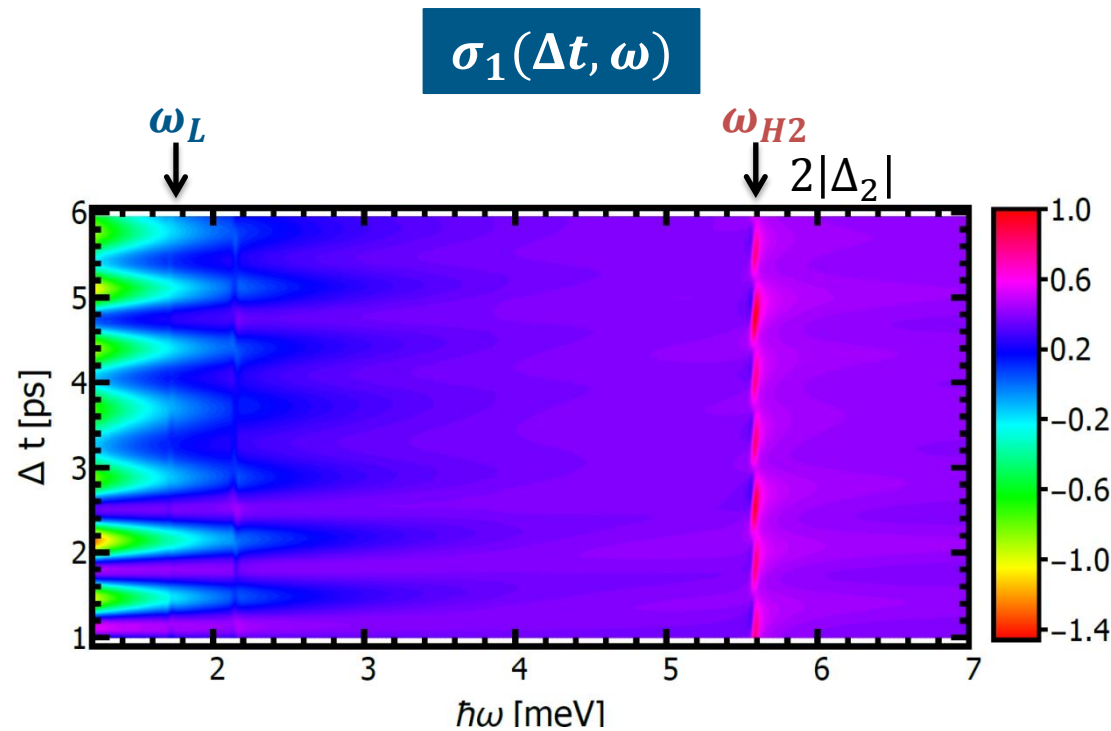
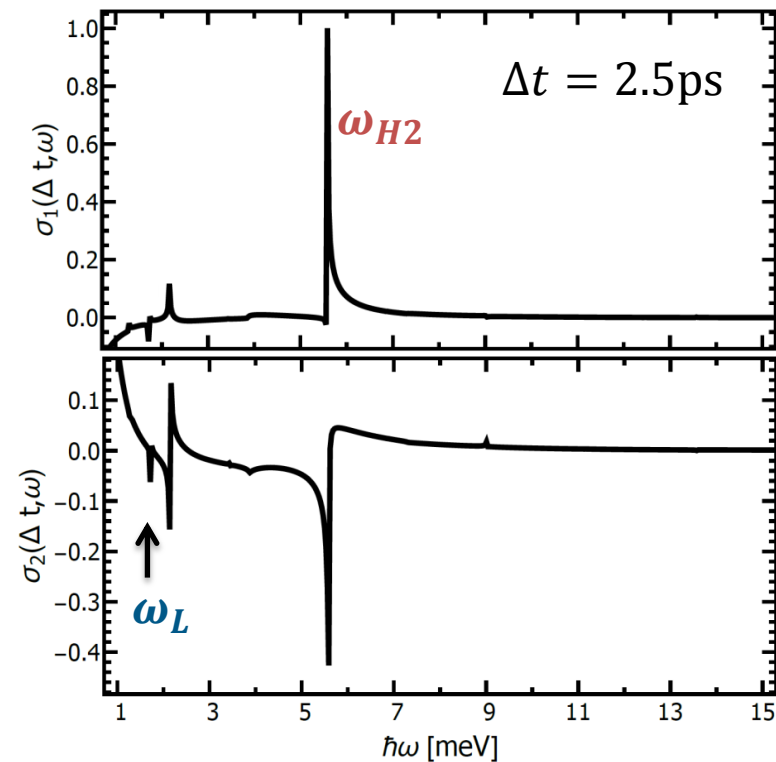
Coherent control
of modes is possible!



- » Pump-probe optical conductivity shows signatures of **non-adiabatic dynamics**

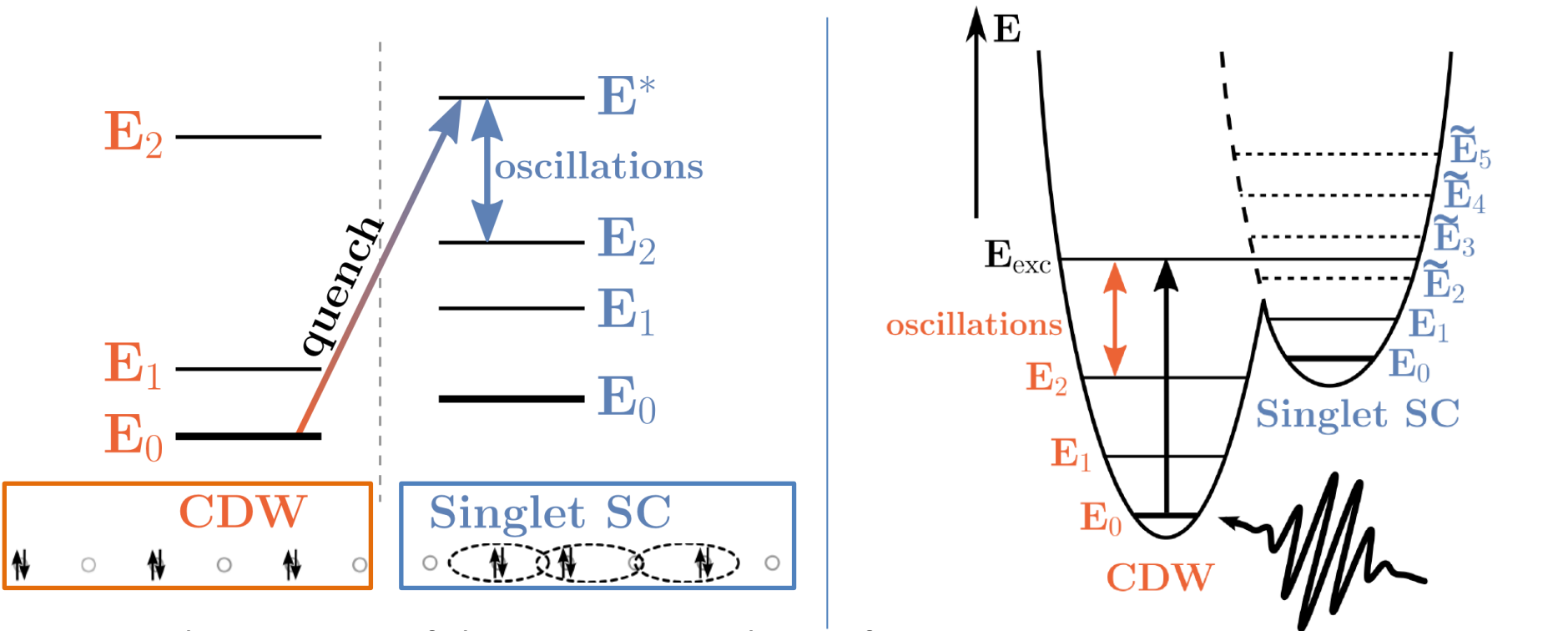
$$\sigma(\Delta t, \omega) = j(\Delta t, \omega) / [i\omega A_{\text{pr}}(\Delta t, \omega)]$$

⇒ **Oscillations** in pump-probe response **as a function of delay time Δt**



Summary (Part 1): Quench vs. Pulse

Time-dependent solution of the extended Hubbard model:



→ Enhancement of the on-site correlation function

→ Transient Meissner effect

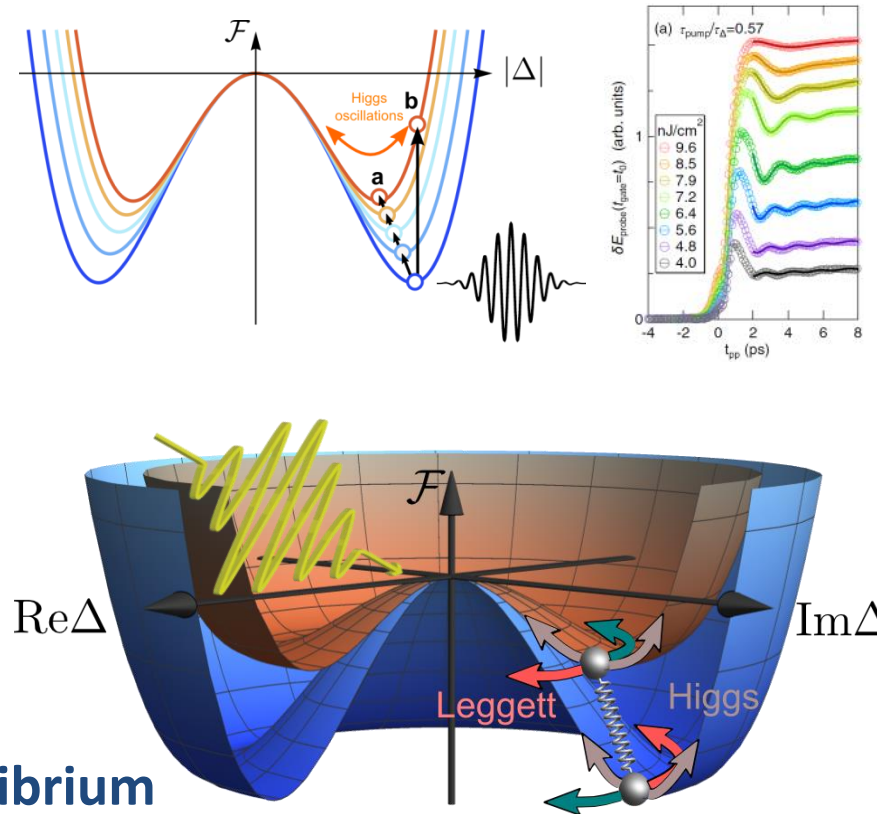
→ Quench: direct into the new phase

→ Pulse: fingerprints of the new phase short after the pulse

Microscopic theory for ultrafast dynamics in superconductors employing Density Matrix Theory:

What happens after the laser pulse?

- case 1: single band
 - » Higgs oscillations in the *non-adiabatic* regime, $\tau_p < \tau_\Delta$
- case 2: two bands (MgB_2): predictions
 - » Josephson coupling: new Leggett mode detected
 - » new dispersion
 - » amplitude-phase coupling in nonequilibrium
 - » higher harmonics can be observed



Thank you for your attention!