

# Theory of laser-driven nonequilibrium superconductivity

*PRB 92, 224517 (2015)*

*PRB 93, 144506 (2016)*

Theory collaborators:

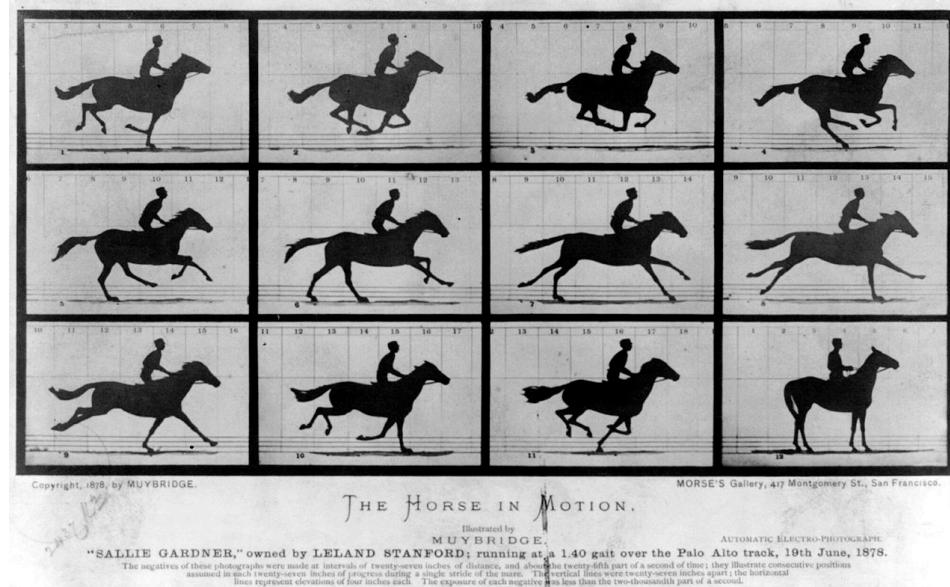
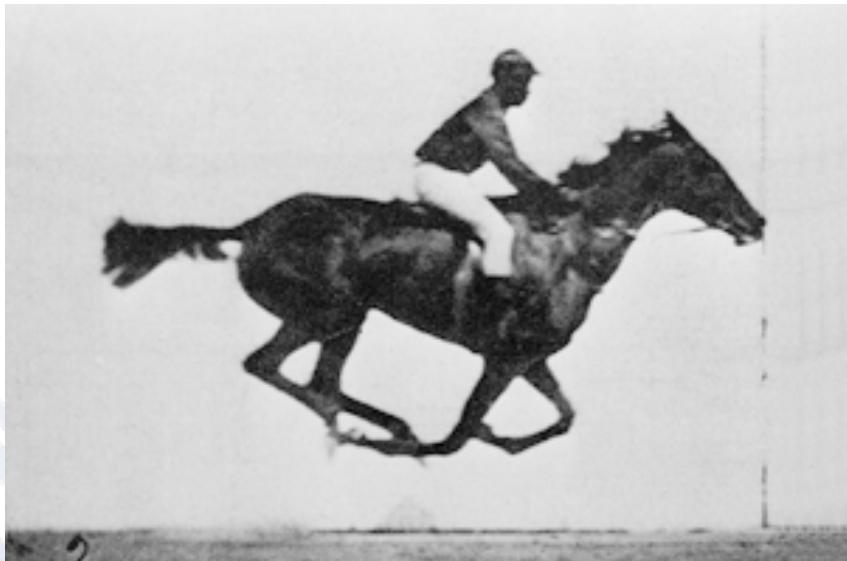
A. F. Kemper, B. Moritz, J. K. Freericks, T. P. Devereaux,  
A. Georges, C. Kollath

Michael Sentef

New3SC Bled, September 14, 2016

# Pump-probe spectroscopy (1887)

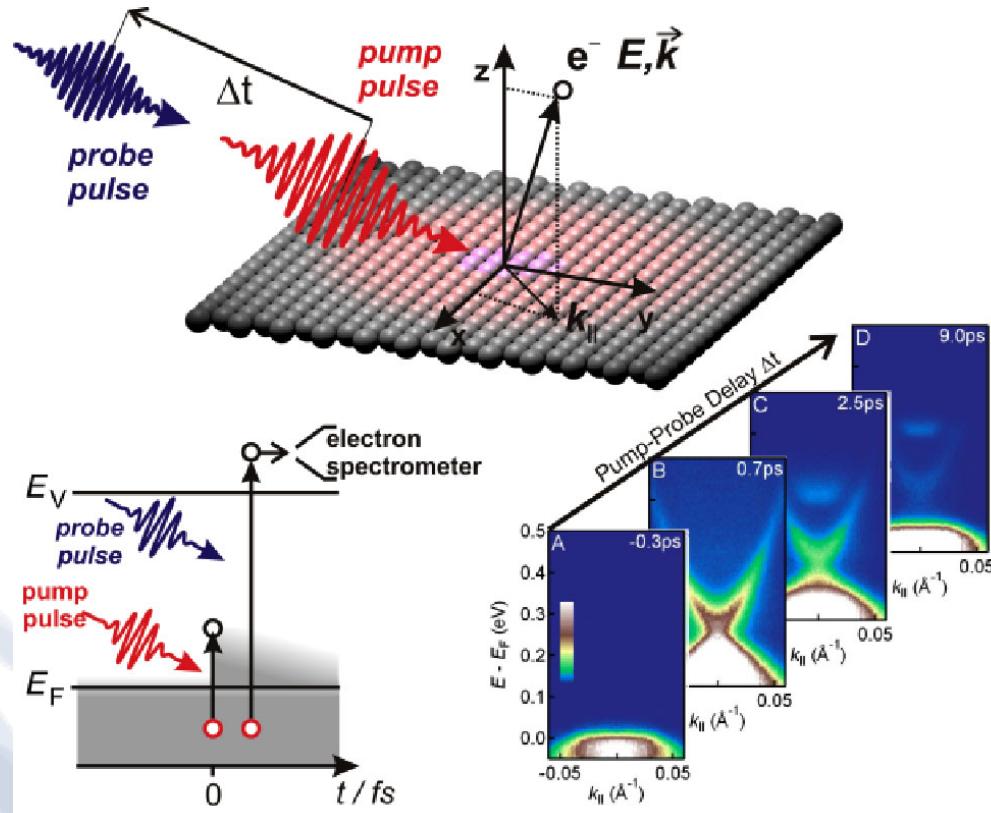
- stroboscopic investigations of dynamic phenomena



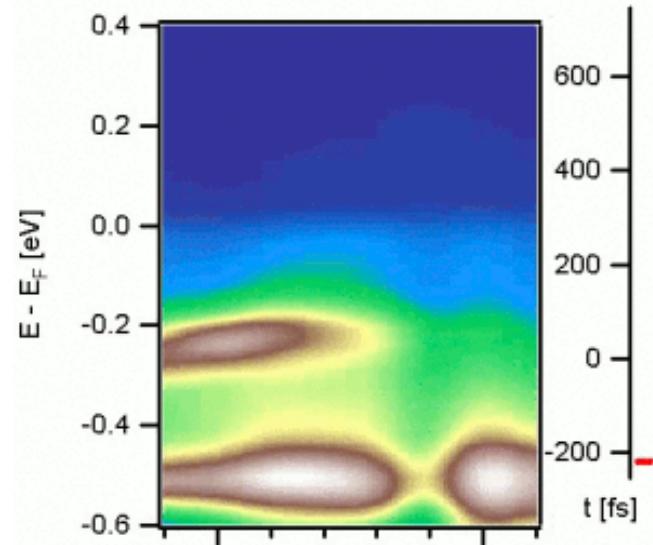
Muybridge 1887

# Pump-probe spectroscopy (today)

- stroboscopic investigations of dynamic phenomena



TbTe<sub>3</sub> CDW metal



J. Sobota et al., PRL 108, 117403 (2012)  
F. Schmitt et al., Science 321, 1649 (2008)  
Image courtesy: J. Sobota / F. Schmitt

# Ultrafast Materials Science

## *Understanding the nature of quasiparticles*

### ▪ Relaxation dynamics

PRL 111, 077401 (2013)

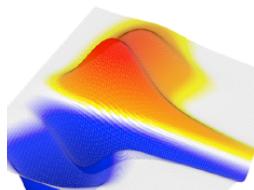
PRX 3, 041033 (2013)

PRB 87, 235139 (2013)

PRB 90, 075126 (2014)

arXiv:1505.07055

arXiv:1607.02314

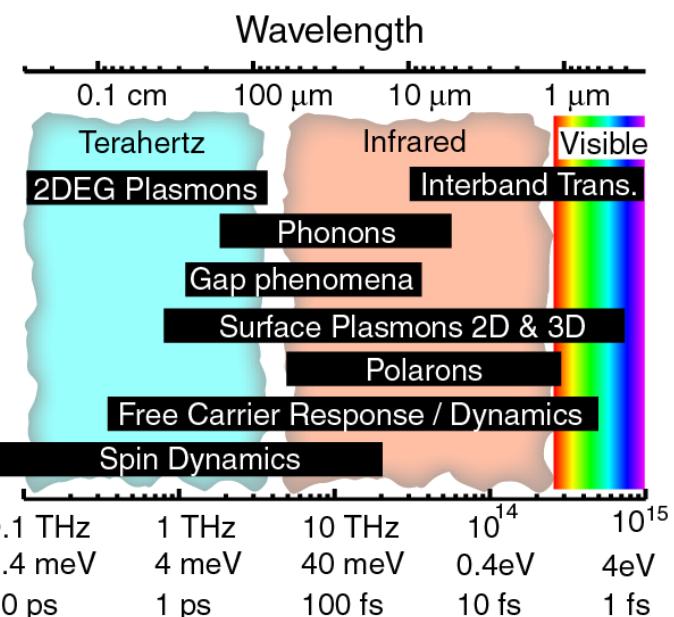
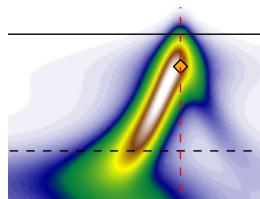


## *Understanding ordered phases*

### ▪ Collective oscillations ▪ Competing order parameters

PRB 92, 224517 (2015)

PRB 93, 144506 (2016)



## *Creating new states of matter*

### ▪ Photo-induced phase transitions ▪ Floquet topological states

Nature Commun. 6, 7047 (2015)

arXiv:1604.03399

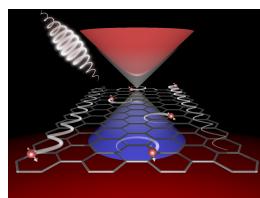


Image courtesy:  
D. Basov

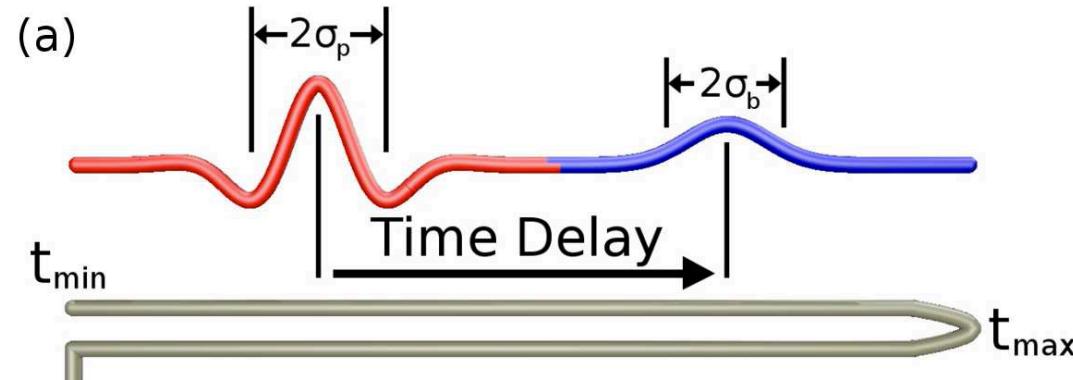
# Outline

- Keldysh Green functions
- Ordered states: Driven superconductors
  - Higgs amplitude mode oscillations for optical pumping (1.5 eV laser)  
*PRB 92, 224517 (2015)*
  - light-enhanced superconductivity via hopping control  
*PRB 93, 144506 (2016)*

# Non-Equilibrium Keldysh Formalism

Beyond BCS

$$G_{\mathbf{k}}(t_k, t(\omega)) G_{\mathbf{k}}^0(t) G_{\mathbf{k}}^0(+\omega) dt_1 dt_2 G_{\mathbf{k}}^{00}(t_1, t_2) \Sigma(t_1, t_2) G_{\mathbf{k}}(t_2, t')$$



$t_{\min} - i\beta$

...and about its history

System knows about its thermal initial state...

self-energy  $\Sigma$ :  
electron-electron scattering  
electron-phonon scattering  
...

Include the effects of driving field through time-dependent electronic dispersion

$$\varepsilon(k) \rightarrow \varepsilon(k, t)$$

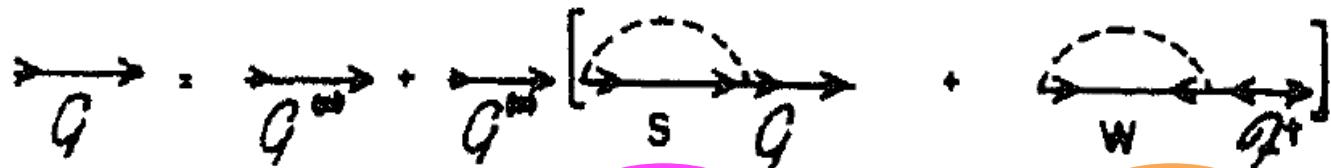
# Model and Method

$$\mathcal{H} = \sum_{\mathbf{k}\sigma} \epsilon(\mathbf{k}, t) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_{\mathbf{q}, \gamma} \Omega_\gamma b_{\mathbf{q}, \gamma}^\dagger b_{\mathbf{q}, \gamma} - \sum_{\mathbf{q}, \gamma, \sigma} g_\gamma c_{\mathbf{k}+\mathbf{q}\sigma}^\dagger c_{\mathbf{k}\sigma} (b_{\mathbf{q}, \gamma} + b_{-\mathbf{q}, \gamma}^\dagger)$$

- electrons (2D square latt.) + spectrum of phonons + el-ph coupling (Holstein)
- Migdal-Eliashberg (1st Born) + phonon heat bath approximation

superconductor:

normal



anomalous



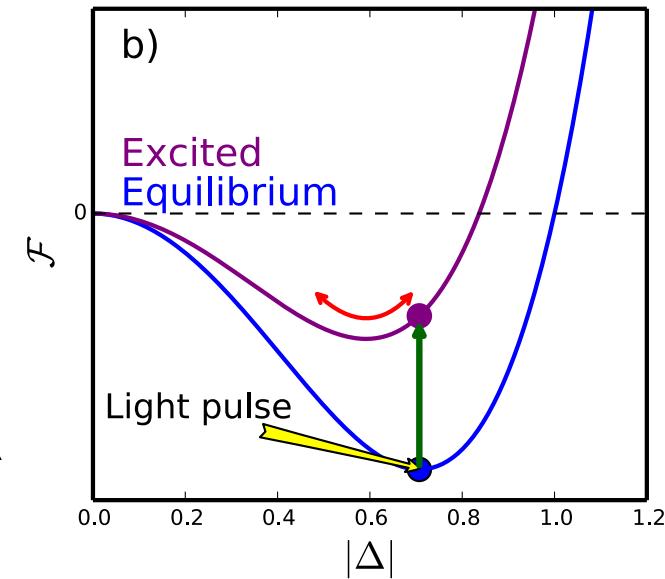
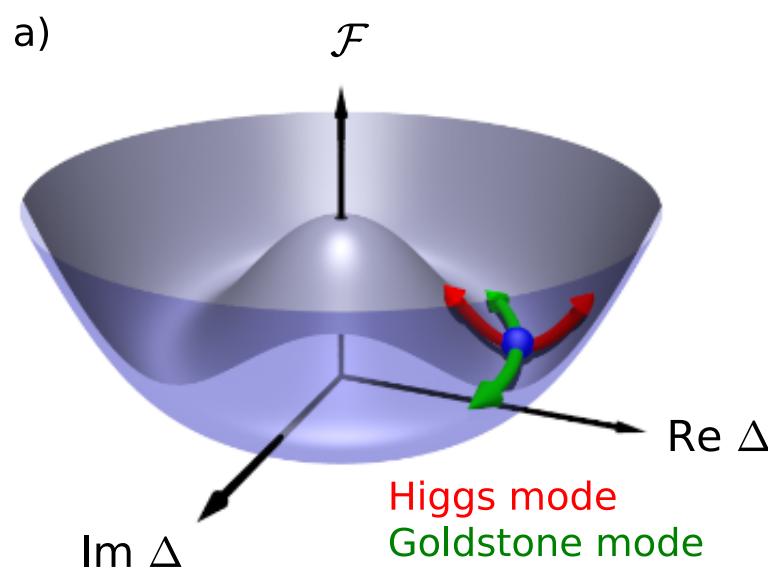
order parameter  $\Delta$ ,  
condensate dynamics

el-ph single-particle  
scattering

also see:

Murakami, Werner et al., PRB 93, 094509 (2016)

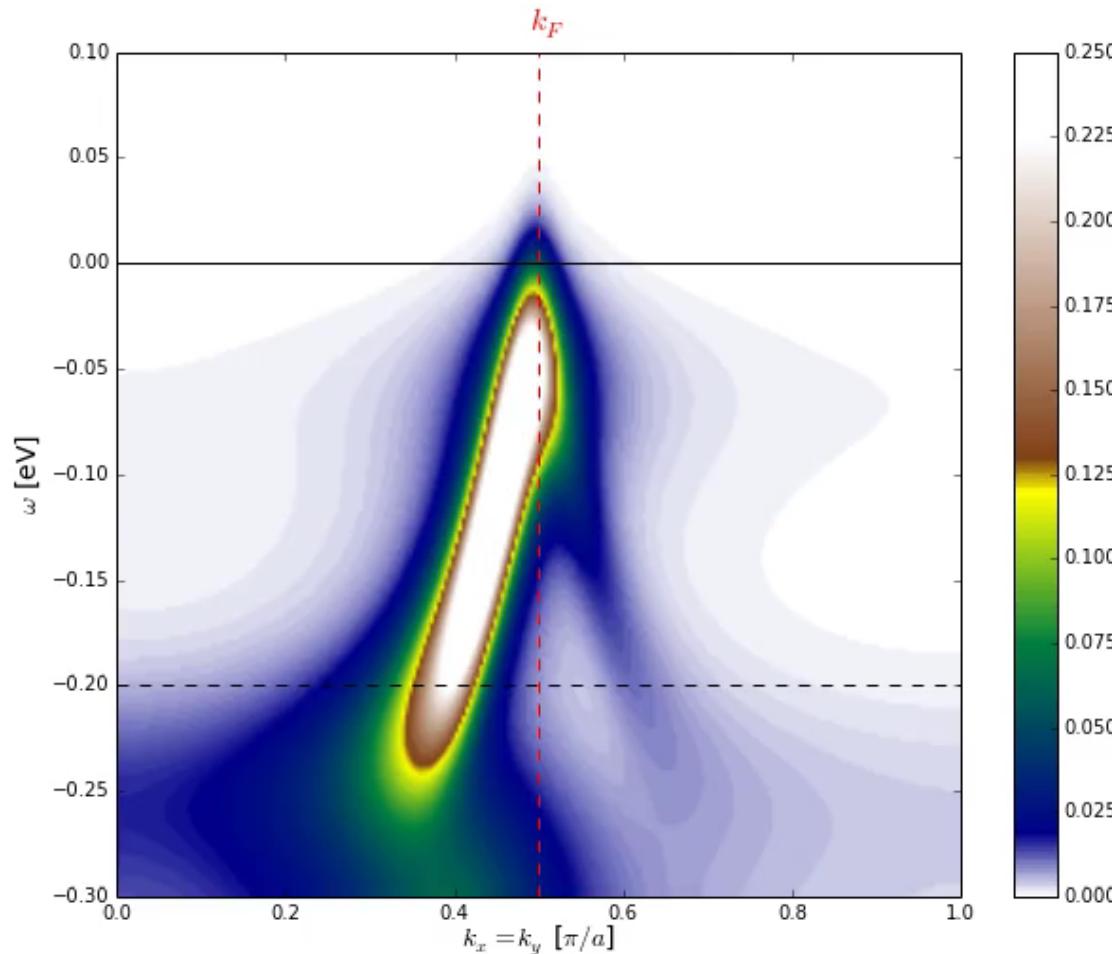
# Higgs amplitude mode



Include the effects of driving field through Peierls substitution

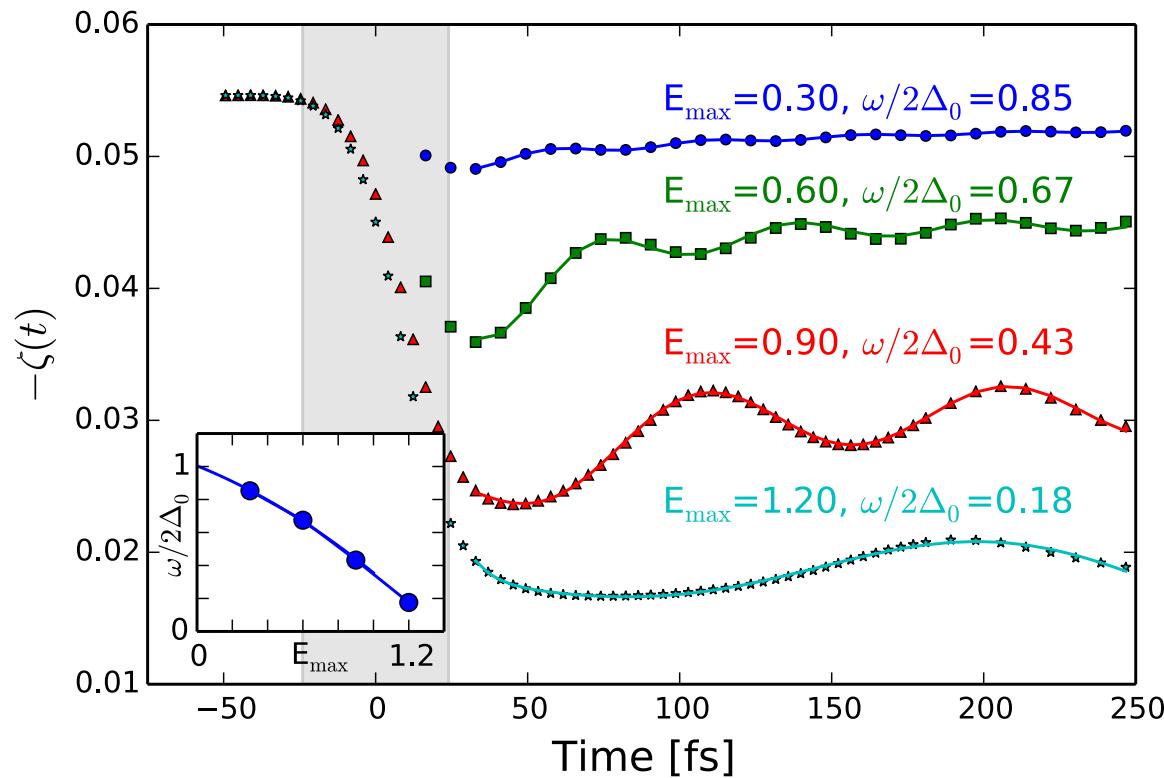
$$\mathbf{k} \rightarrow \mathbf{k} - e\mathbf{A}(t)$$

# Oscillations in photocurrent



# Amplitude mode oscillations

PRB 92, 224517  
(2015)



Amplitude (“Higgs”) mode oscillations predicted in time-resolved ARPES  
Reduced order parameter sets oscillation frequency  
Dissipation: Exciting Higgs even far away from gap resonance

Optics: Matsunaga et al., Phys. Rev. Lett. 111, 057002 (2013), Science 2014 [10.1126/science.1254697]

Theory: Volkov & Kogan 1974, Barankov PRL 2004, Yuzbashyan PRL 2006, Tsuji PRL 2013, Murakami PRB 2016, Schnyder, Manske et al., PRB 2011, Krull, Manske et al., PRB 2014

# How to enhance boson-mediated SC?

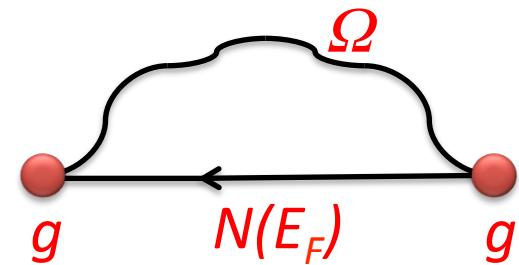
- BCS theory – plain vanilla SC (weak coupling)

$$\Delta \approx 2\hbar\Omega_c \exp(-1/V_0 N(E_F))$$



- effective attraction  $V_0 \sim g^2/(\hbar \Omega)$
- e-boson coupling  $g$
- boson frequency  $\Omega$
- electronic DOS  $N(E_F)$

$$\Sigma =$$



Migdal-Eliashberg theory  
boson-mediated pairing

# How to enhance boson-mediated SC?

- **nonlinear phononics**  $Q^2Q$ : resonant excitation of vibrational modes – effects?

## 1. tune model parameters

- e-boson coupling  $g$
  - boson frequency  $\Omega$
  - electronic DOS  $N(E_F)$
- }  $\alpha^2F$  – Eliashberg function

Gedankenexperiment (what if?)

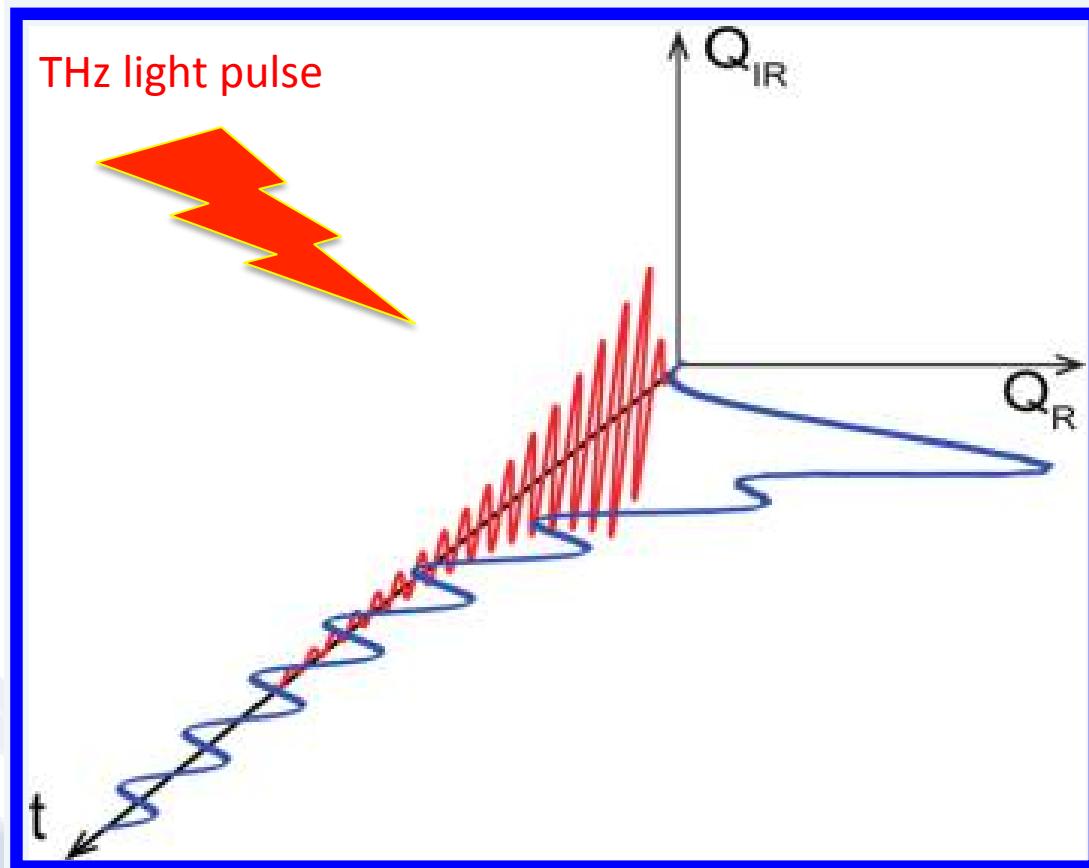
## 2. dynamical effect

- effective Hamiltonian (e.g., Floquet)

*also see: Knap et al., arXiv:1511.07874,*

*Patel & Eberlein PRB 93, 195139 (2016), Komnik & Thorwart arXiv:1607.03858*

# Classical lattice dynamics



$$\ddot{Q}_{\text{IR}} + \Omega_{\text{IR}}^2 Q_{\text{IR}} = \frac{e^* E_0}{\sqrt{M_{\text{IR}}}} \sin(\Omega_{\text{IR}} t) F(t)$$

$$\ddot{Q}_{\text{RS}} + \Omega_{\text{RS}}^2 Q_{\text{RS}} = A Q_{\text{IR}}^2$$

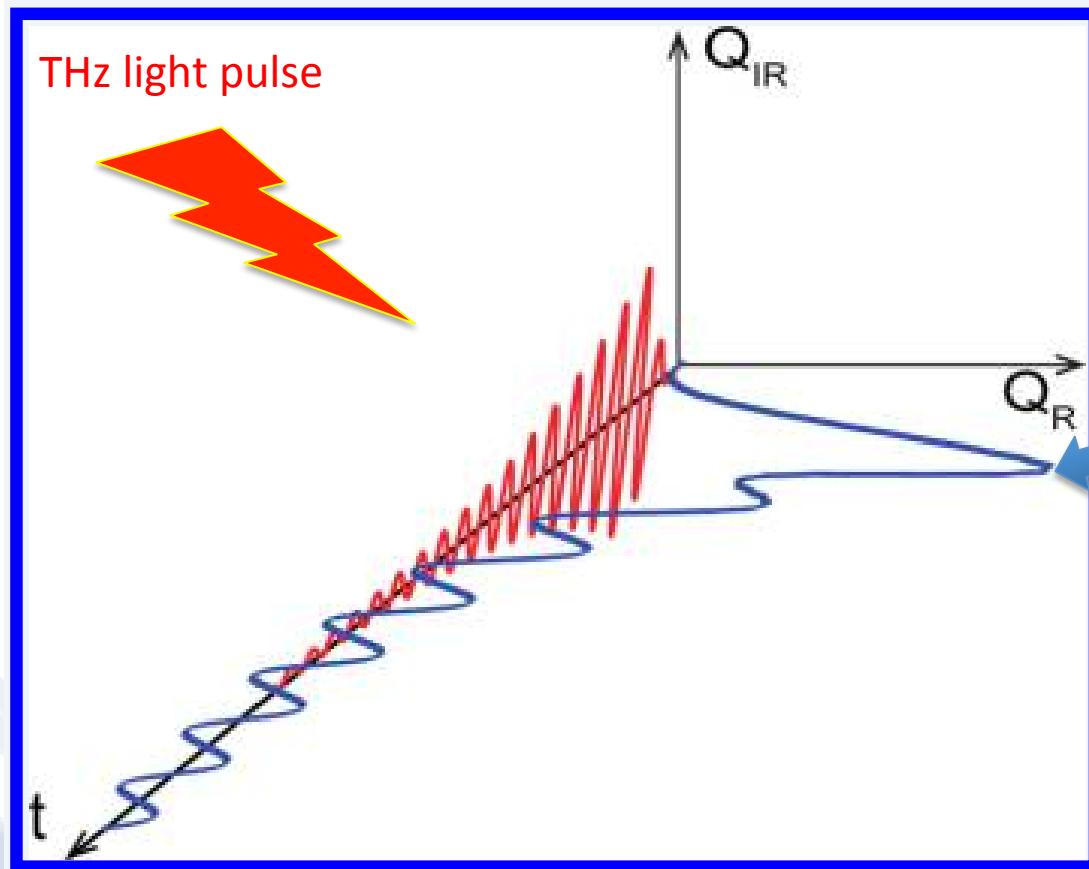
Rectification of a second (Raman) phonon via coherent driving of a first (IR) phonon

„Nonlinear phononics“

M. Först et al., *Nature Physics* 7, 854 (2011)

A. Subedi, A. Cavalleri, A. Georges, *PRB* 89, 220301R (2014)

# Classical lattice dynamics



Displaced Raman mode assumed to lead to temporal change of electronic hopping

Gedankenexperiment (what if?)

Include the effects of driving field through time-dependent electronic dispersion

$$\varepsilon(k) \rightarrow \varepsilon(k, t)$$

„Nonlinear phononics“

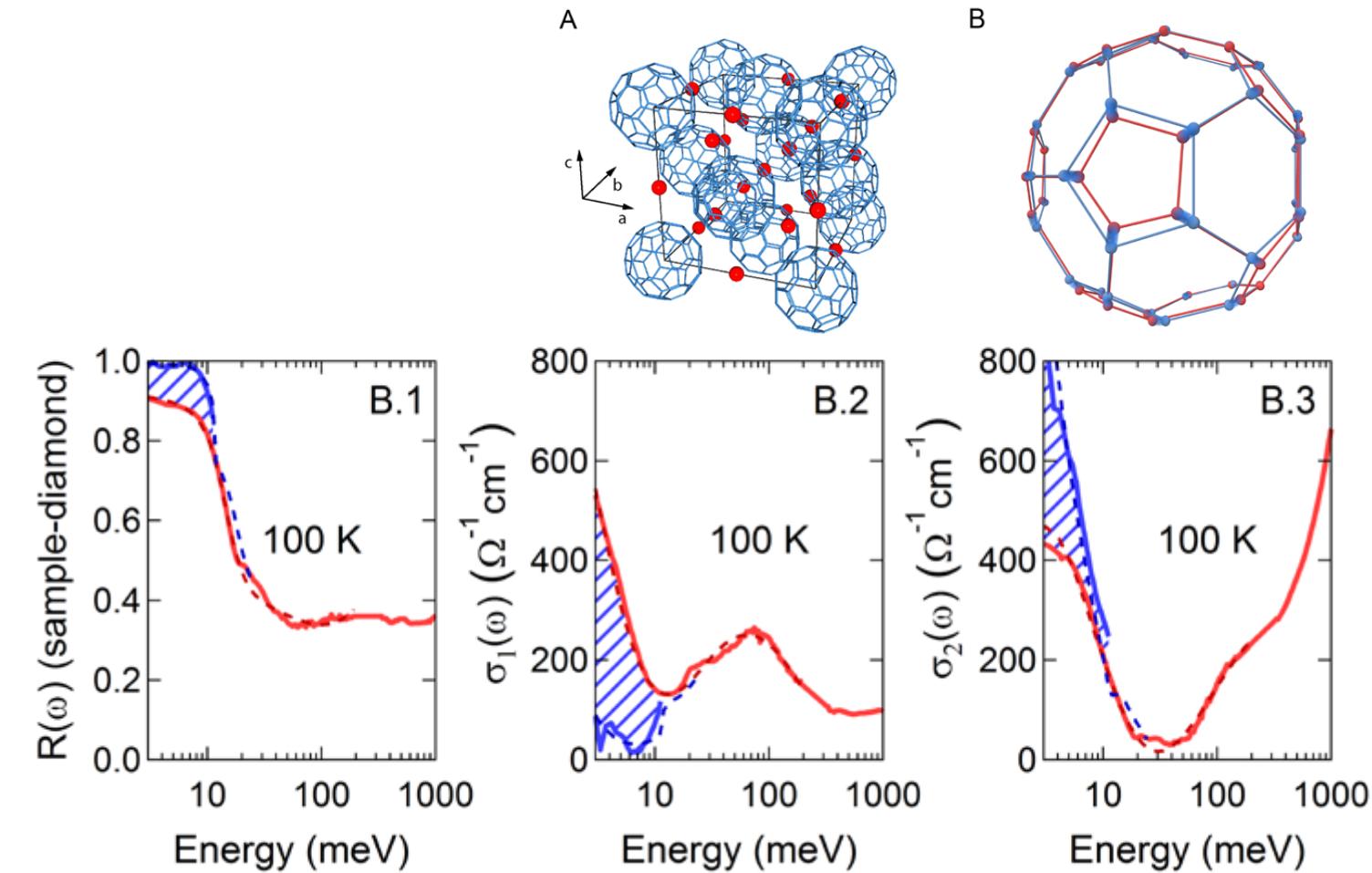
M. Först et al., *Nature Physics* 7, 854 (2011)

A. Subedi, A. Cavalleri, A. Georges, *PRB* 89, 220301R (2014)

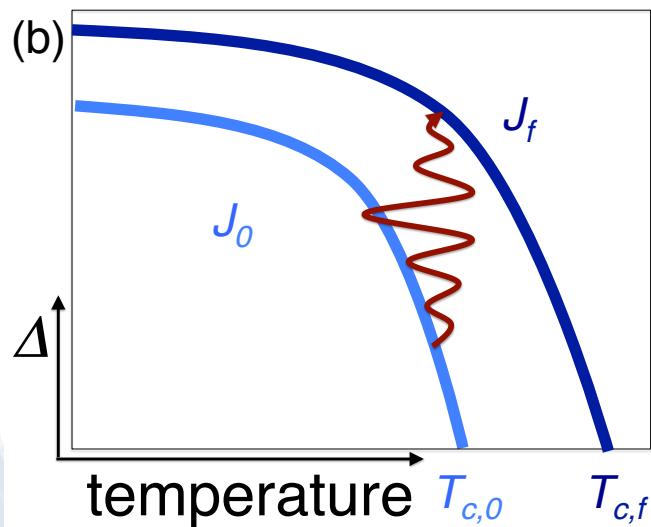
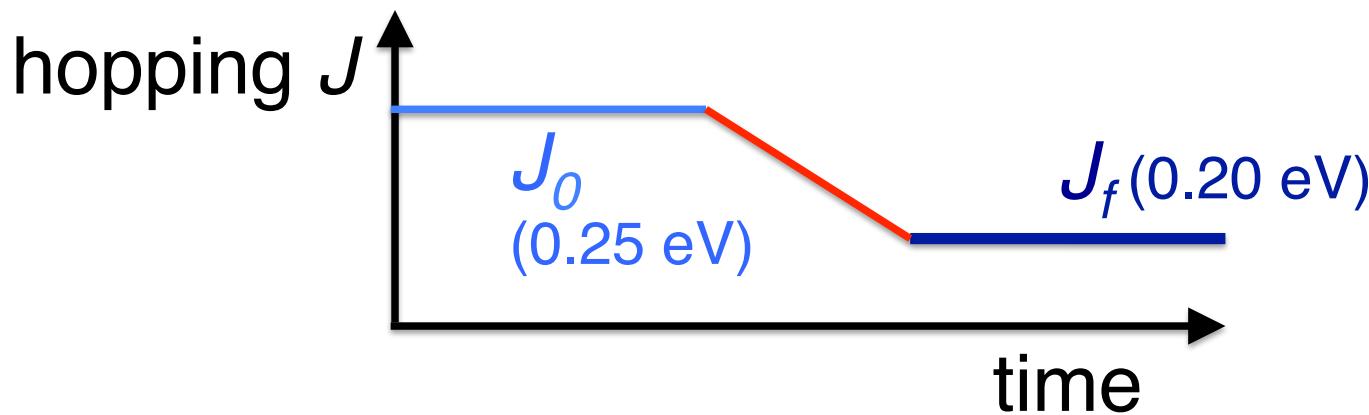
# Experimental motivation

„Possible light-induced superconductivity in K<sub>3</sub>C<sub>60</sub> at high temperature“

M. Mitrano et al., Nature 530, 461 (2016)

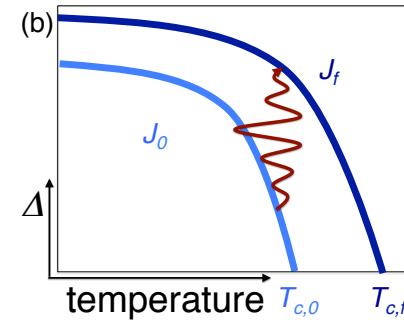
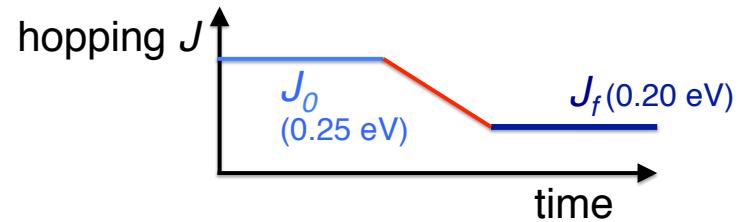
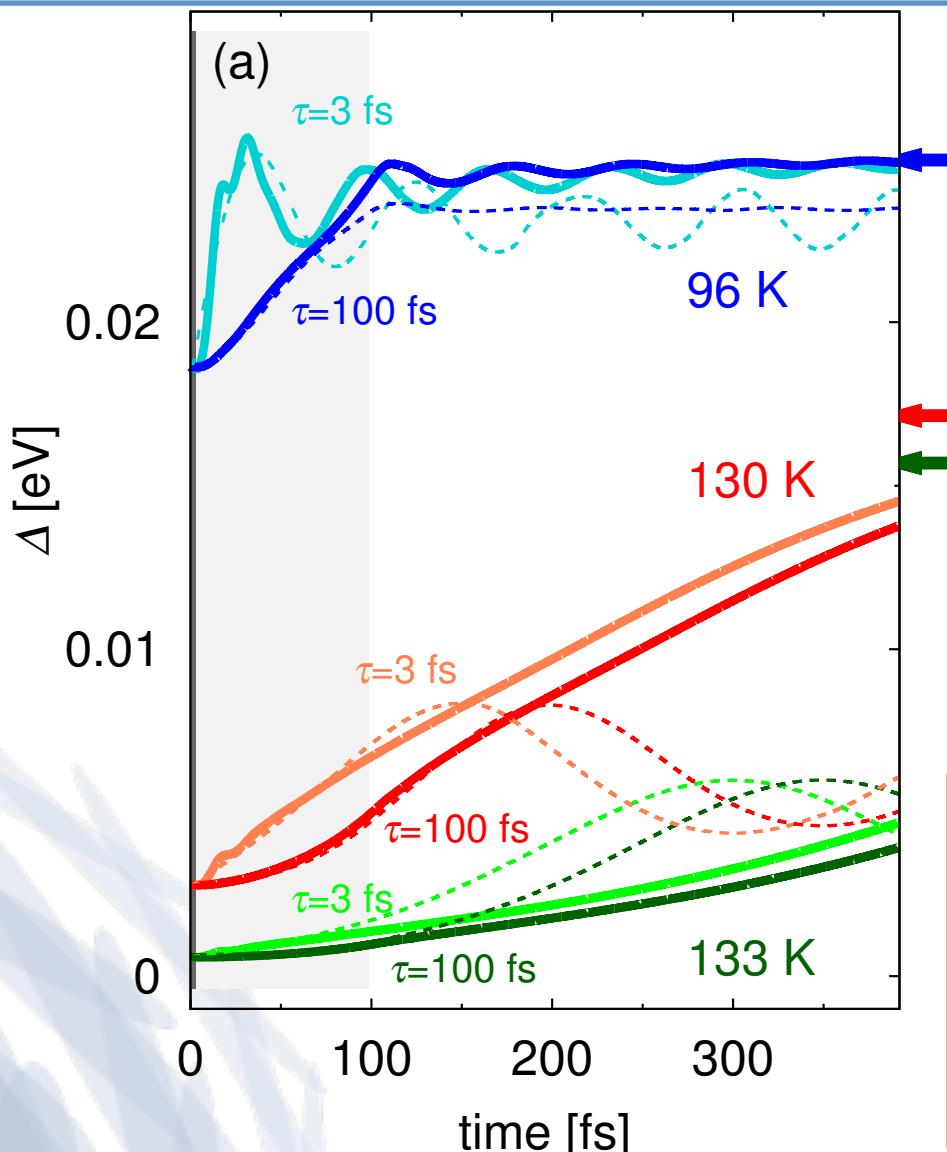


# Simplest model: hopping ramp



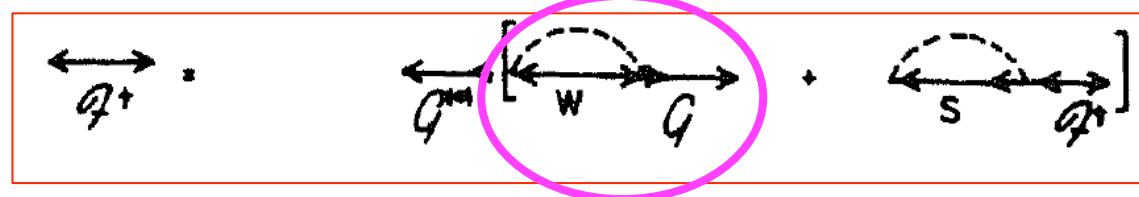
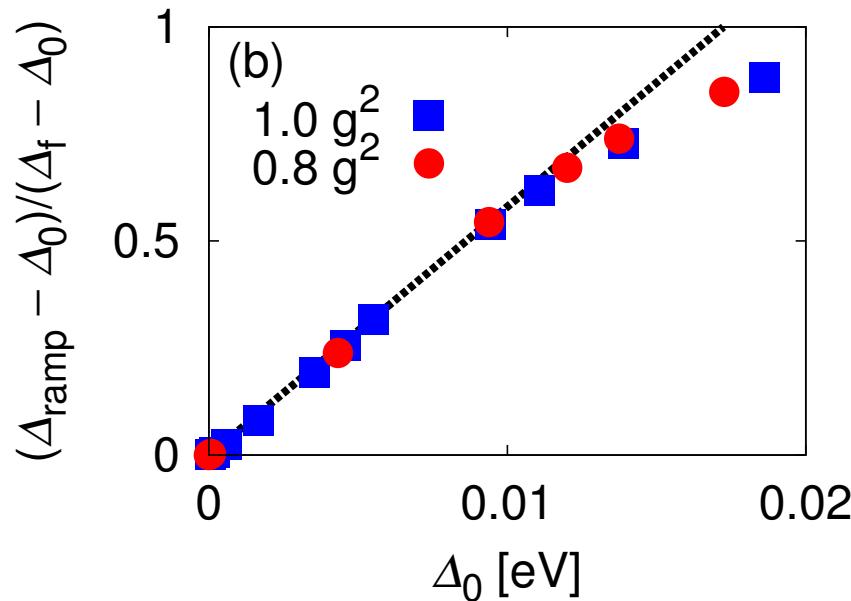
Equilibrium picture:  
Enhancement of SC via  
enhanced DOS at Fermi  
energy

# Superconductor evolution



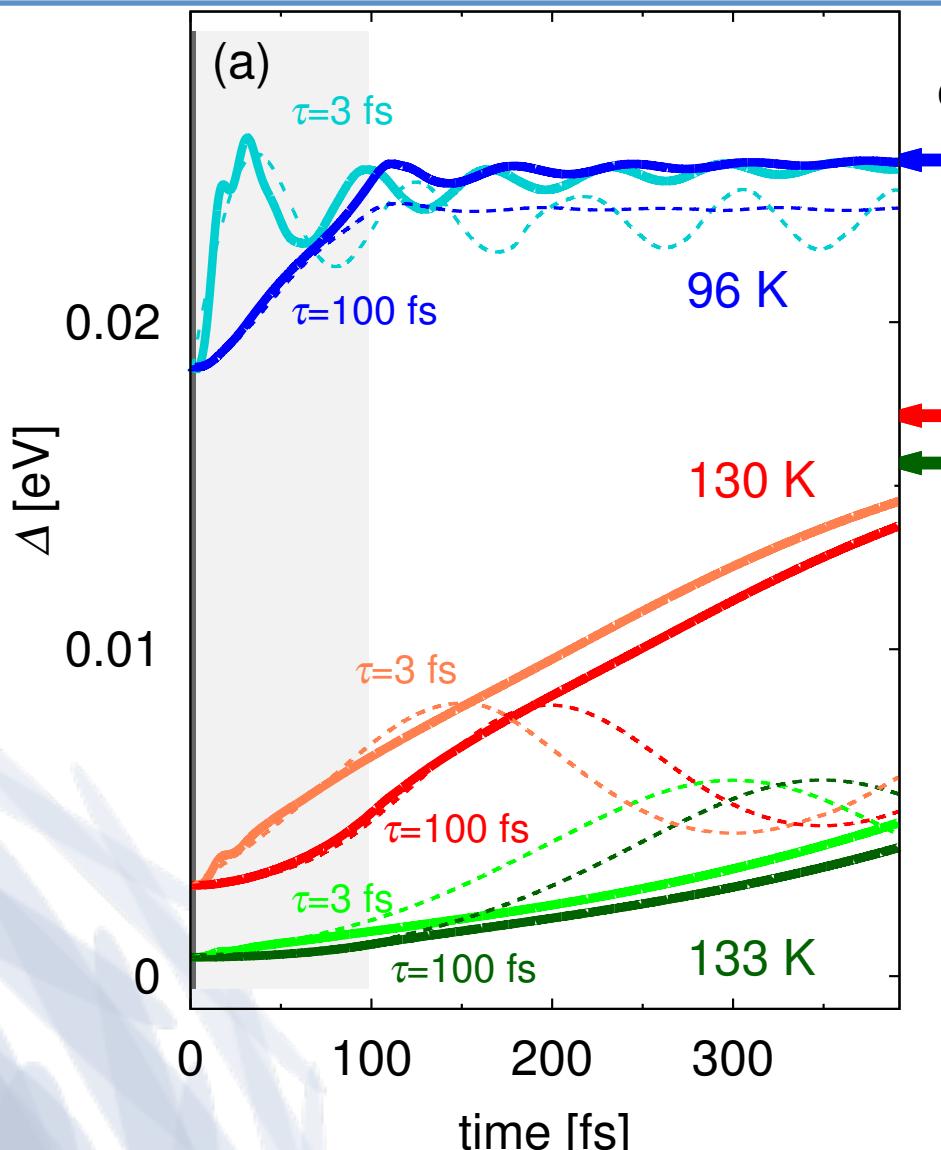
Enhancement of SC  
strongly depends on  
initial thermal state

# Enhancement during ramp

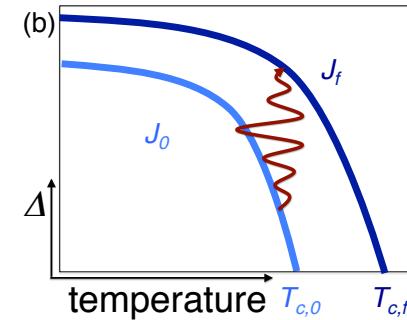
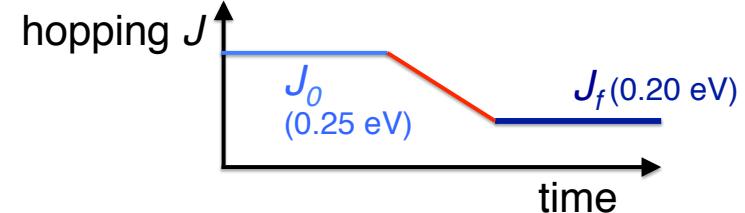


Order parameter enhancement  $\sim \Delta_0$   
time scale to fully enhance long-range order diverges at  $T_c$ !

# Superconductor evolution



dashed: no dissipation (BCS only)



Dissipation helps  
enhancement for fast  
ramps

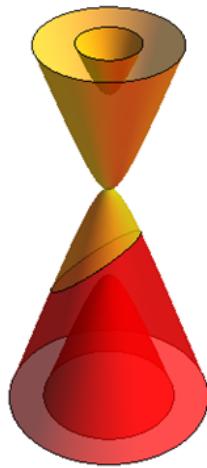
# Dynamically enhanced coupling?!

I. Gierz et al,

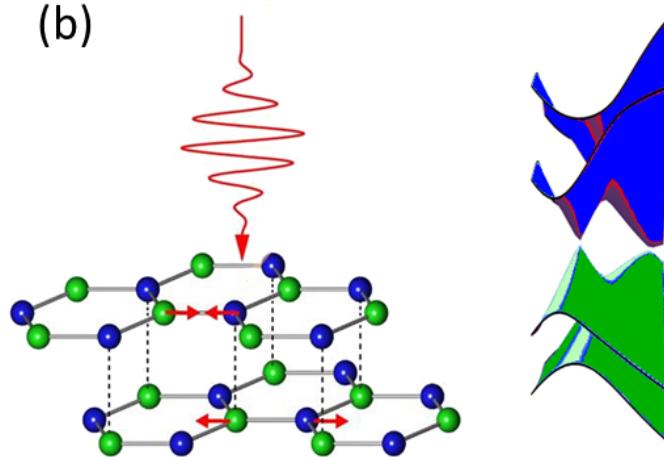
arXiv:1607.02314

enhanced electron-phonon coupling in a periodically distorted graphene lattice driven on resonance with IR phonon

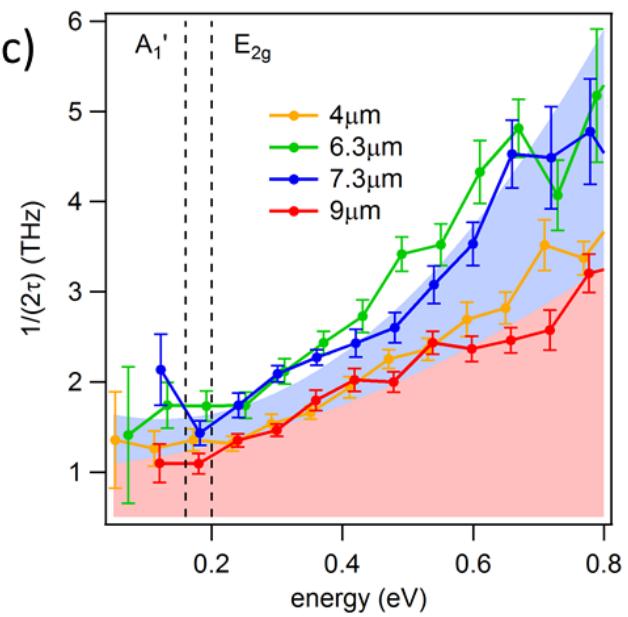
(a)



(b)



(c)



# Summary part 1

- Amplitude mode oscillations in pumped SC

*PRB 92, 224517 (2015)*

- Light-enhanced SC via nonlinear phononics

*PRB 93, 144506 (2016)*



A. F. Kemper



T. P. Devereaux



B. Moritz



J. K. Freericks



A. Georges



C. Kollath



GEOGETOWN UNIVERSITY



COLLÈGE  
DE FRANCE  
1530



universität bonn

# Theory of laser-controlled competing orders



Akiyuki Tokuno, Antoine Georges, Corinna Kollath  
(Paris/Bonn)

## Why?

- understand ordering mechanisms
- control ordered states
- induce new states of matter

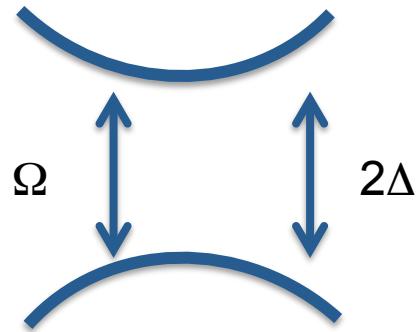
## How?

- resonance with something

Is there a generic mechanism to control ordered states?

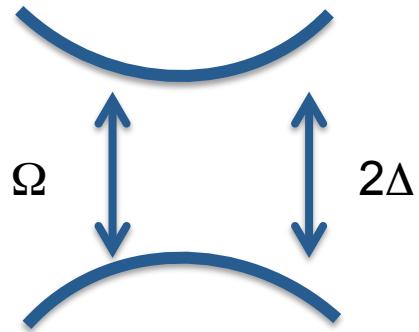
# Driven SC/CDW

CDW  $\sim A$   
1-photon resonance

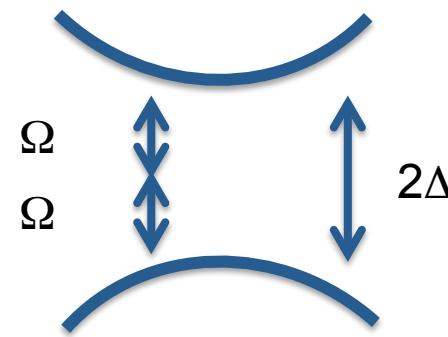


# Driven SC/CDW

CDW  $\sim A$   
1-photon resonance

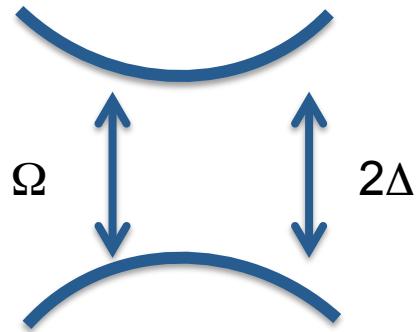


SC  $\sim A^2$   
2-photon resonance

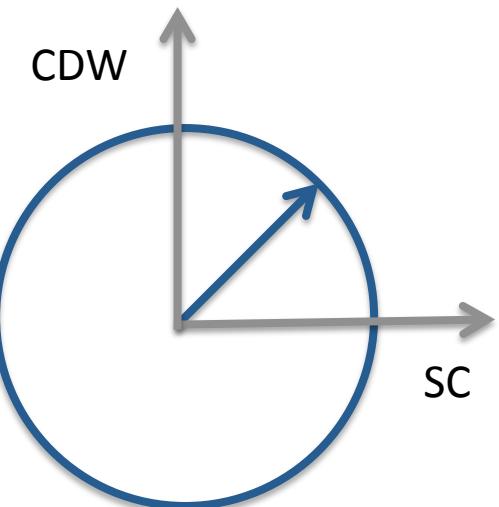
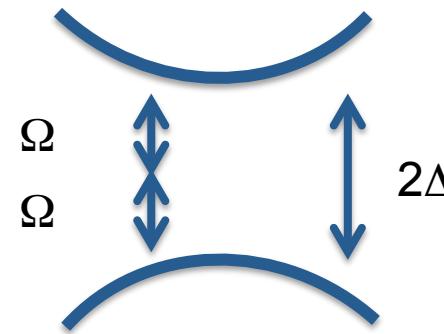


# Driven SC/CDW

CDW  $\sim A$   
1-photon resonance

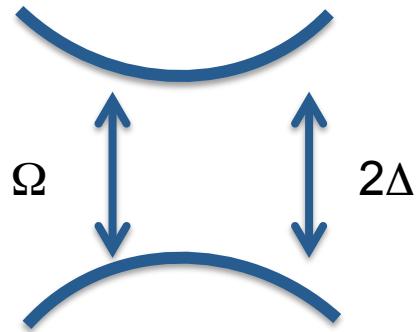


SC  $\sim A^2$   
2-photon resonance

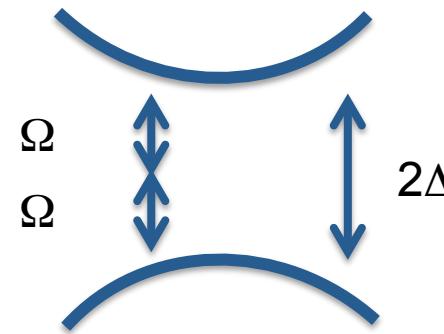


# Driven SC/CDW

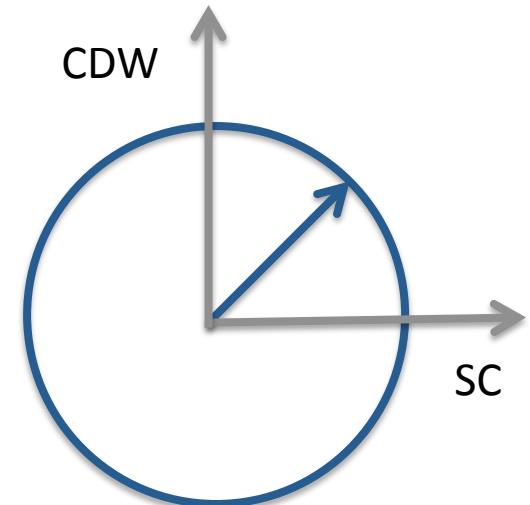
CDW  $\sim A$   
1-photon resonance



SC  $\sim A^2$   
2-photon resonance

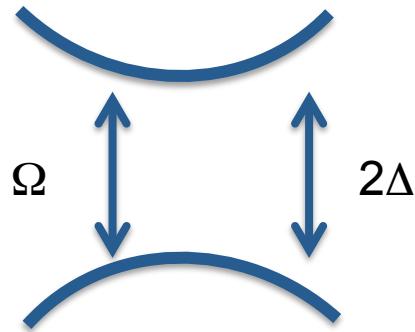


... laser lifts SC/CDW degeneracy

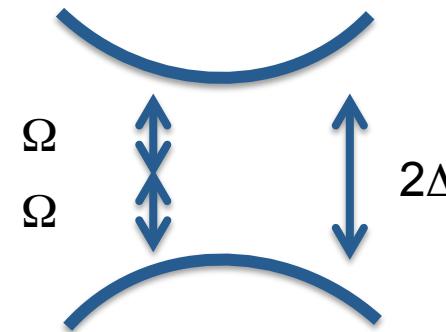


# Driven SC/CDW

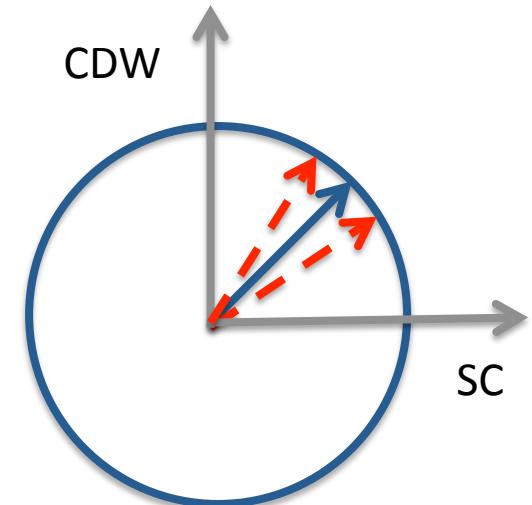
CDW  $\sim A$   
1-photon resonance



SC  $\sim A^2$   
2-photon resonance

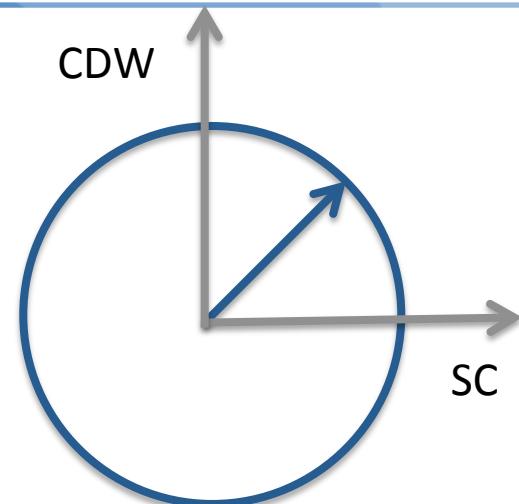


... laser lifts SC/CDW degeneracy  
... Goldstone-like collective mode?



# Competing orders

- attractive -U Hubbard model
- degeneracy of SC and CDW at perfect nesting
- SO(4) symmetry (SC, CDW, eta pairing)



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PHYSICAL REVIEW LETTERS

6 NOVEMBER 1989

## $\eta$ Pairing and Off-Diagonal Long-Range Order in a Hubbard Model

Chen Ning Yang

C. N. Yang  
(1957 Nobel for parity violation in weak interaction)



Reprinted from Mod. Phys. Lett. B4 (1990) 759–766  
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## $SO_4$ SYMMETRY IN A HUBBARD MODEL

CHEN NING YANG

Institute for Theoretical Physics, State University of New York,  
Stony Brook, NY 11794-3840, USA

and

S. C. ZHANG

IBM Research Division, Almaden Research Center,  
San Jose, CA 95120-6099, USA

# Simplistic Model

$$H = \sum_{k\sigma} \epsilon(k) n_{k\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} = H_J + H_U,$$
$$\epsilon(k) = -2J(\cos(k_x) + \cos(k_y)),$$

attractive U + mean-field decoupling

$$\Delta_{SC} = U \sum_k f_k, \quad f_k \equiv \langle c_{-k\downarrow} c_{k\uparrow} \rangle \quad (\text{SC}),$$
$$\Delta_{CDW} = U \sum_k g_k, \quad g_k \equiv \frac{1}{2} \sum_{\sigma} \langle c_{k\sigma}^{\dagger} c_{k+Q\sigma} \rangle \quad (\text{CDW}),$$
$$\Delta_{\eta} = U \sum_k \eta_k. \quad \eta_k \equiv \langle c_{-(k+Q)\downarrow} c_{k\uparrow} \rangle \quad (\eta \text{ pairing}).$$

# Mean-field Hamiltonian

$$H_{MF} = \sum_k \begin{pmatrix} c_{k\uparrow}^\dagger \\ c_{k+Q\uparrow}^\dagger \\ c_{-k\downarrow} \\ c_{-(k+Q)\downarrow} \end{pmatrix}^T \begin{pmatrix} \epsilon(k - A) & \Delta_{CDW}^* & \Delta_{SC} & \Delta_\eta \\ \Delta_{CDW} & \epsilon(k + Q - A) & \Delta_\eta & \Delta_{SC} \\ \Delta_{SC}^* & \Delta_\eta^* & -\epsilon(k + A) & -\Delta_{CDW} \\ \Delta_\eta^* & \Delta_{SC}^* & -\Delta_{CDW}^* & -\epsilon(k + Q + A) \end{pmatrix} \begin{pmatrix} c_{k\uparrow} \\ c_{k+Q\uparrow} \\ c_{-k\downarrow}^\dagger \\ c_{-(k+Q)\downarrow}^\dagger \end{pmatrix}$$

4x4 matrix: SO(4) algebra

$$\begin{aligned} \Delta_{SC} &= U \sum_k f_k, & f_k &\equiv \langle c_{-k\downarrow} c_{k\uparrow} \rangle \quad (\text{SC}), \\ \Delta_{CDW} &= U \sum_k g_k, & g_k &\equiv \frac{1}{2} \sum_\sigma \langle c_{k\sigma}^\dagger c_{k+Q\sigma} \rangle \quad (\text{CDW}), \\ \Delta_\eta &= U \sum_k \eta_k. & \eta_k &\equiv \langle c_{-(k+Q)\downarrow} c_{k\uparrow} \rangle \quad (\eta \text{ pairing}). \end{aligned}$$

# Mean-field equations

$$[G_k^<(t, t')]_{\alpha\beta} = +i\langle [\Psi_k^\dagger(t')]_\beta [\Psi_k(t)]_\alpha \rangle.$$

$$i\partial_t G_k^<(t, t) = [H_{MF}(k, t), G_k^<(t, t)].$$

$$\begin{aligned} i\partial_t n_k &= -\Delta_{SC}(f_k - f_k^*) + \Delta_{CDW}(g_k - g_k^*) - \Delta_\eta^* \eta_k + \Delta_\eta \eta_k^*, && \text{eta pairing provides coupling} \\ i\partial_t f_k &= \Delta_{SC}(1 - (n_k + n_{-k})) + (\epsilon(k - A) + \epsilon(k + A))f_k + \Delta_{CDW}(\eta_k + \eta_{k+Q}) - \Delta_\eta(g_k^* + g_{-k}^*), \\ i\partial_t g_k &= \Delta_{CDW}(n_k - n_{k+Q}) - 2\epsilon(k - A)g_k + \Delta_{SC}(\eta_k^* - \eta_{k+Q}) + \Delta_\eta f_k^* - \Delta_\eta^* f_{k+Q}, \\ i\partial_t \eta_k &= \eta_k(\epsilon(k - A) - \epsilon(k + A)) + \Delta_{CDW}(f_k + f_{k+Q}) - \Delta_{SC}(g_{-k} + g_k^*) - \Delta_\eta(n_k + n_{-(k+Q)} - 1). \end{aligned}$$

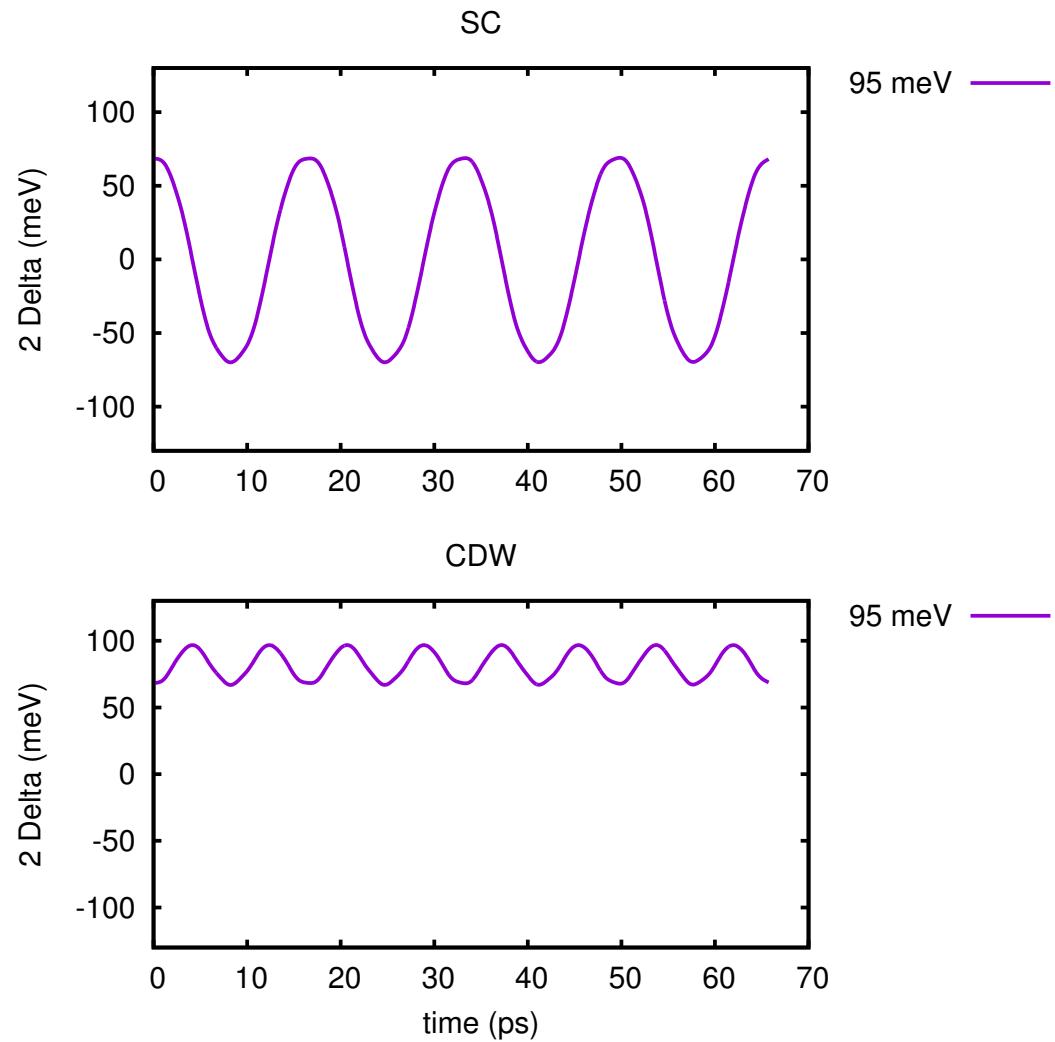
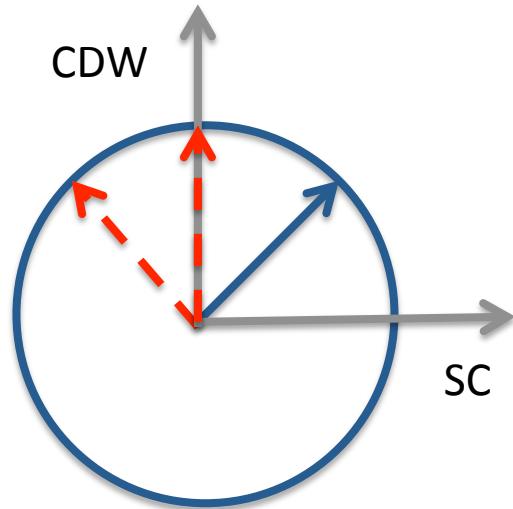
nonlinear equations + self-consistency:

$$\begin{aligned} \Delta_{SC} &= U \sum_k f_k, \\ \Delta_{CDW} &= U \sum_k g_k, \\ \Delta_\eta &= U \sum_k \eta_k. \end{aligned}$$

# Laser hits degenerate orders

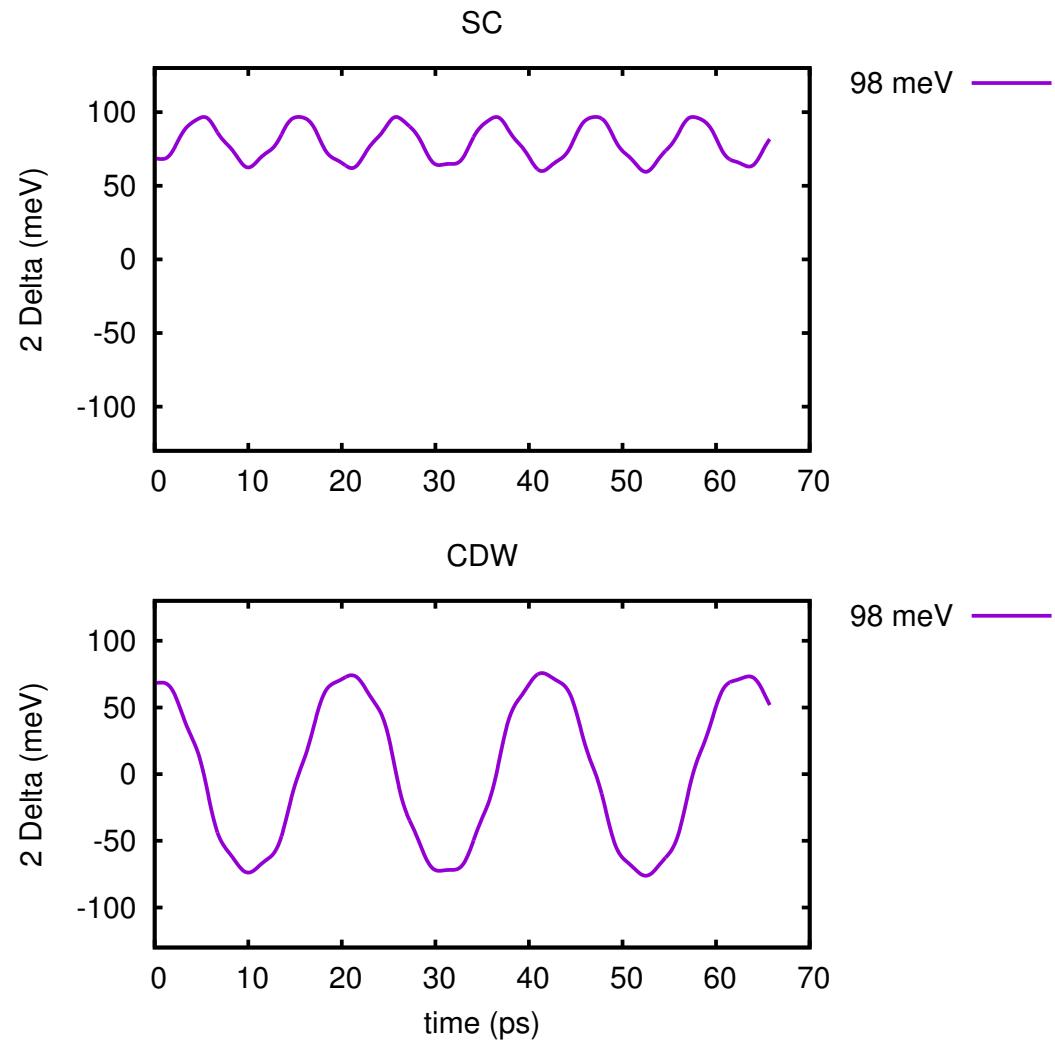
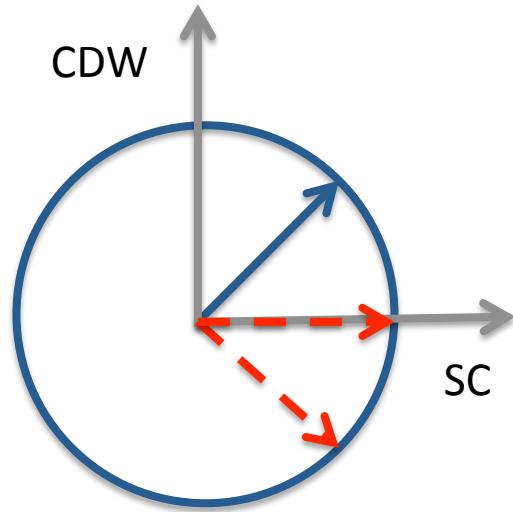
# Gap resonance – cw driving

Below resonance:  
SC down, CDW up



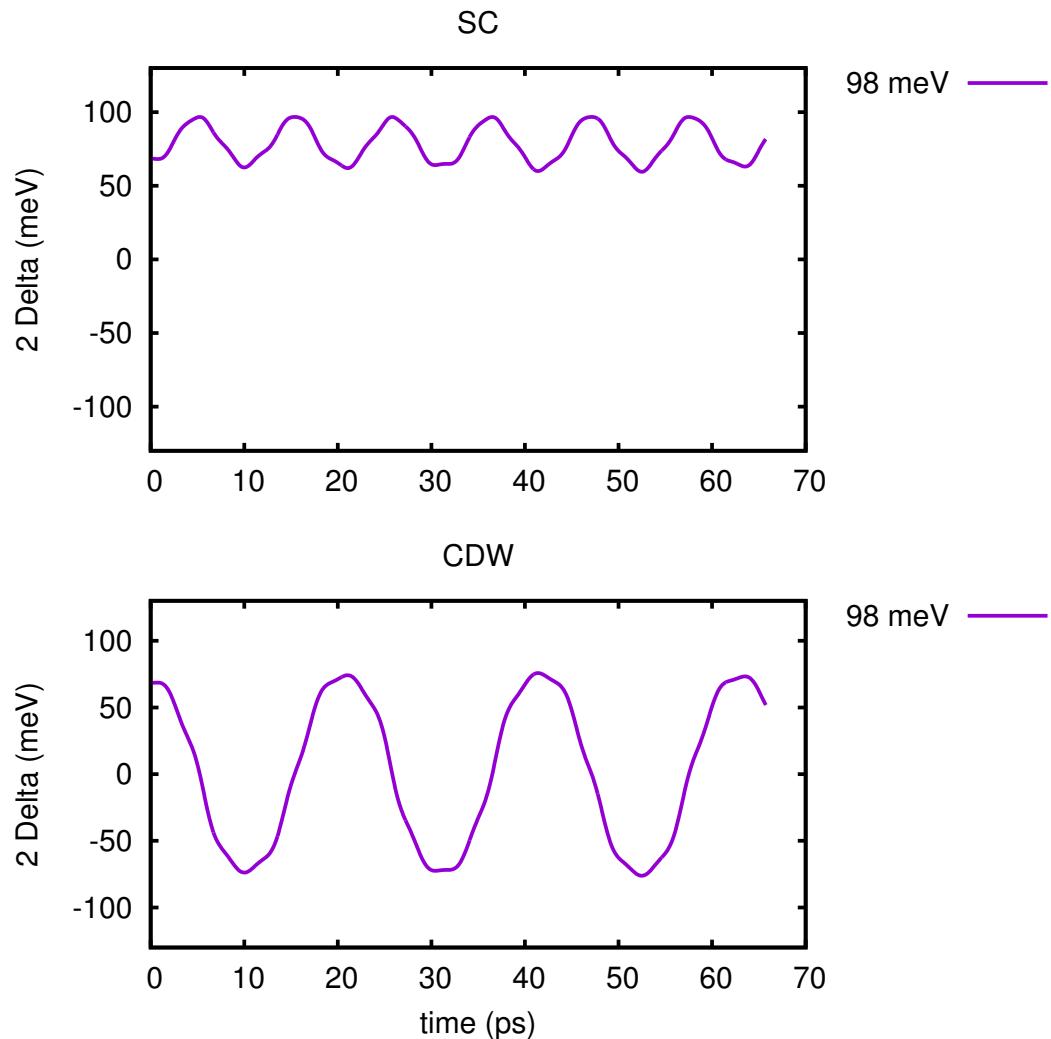
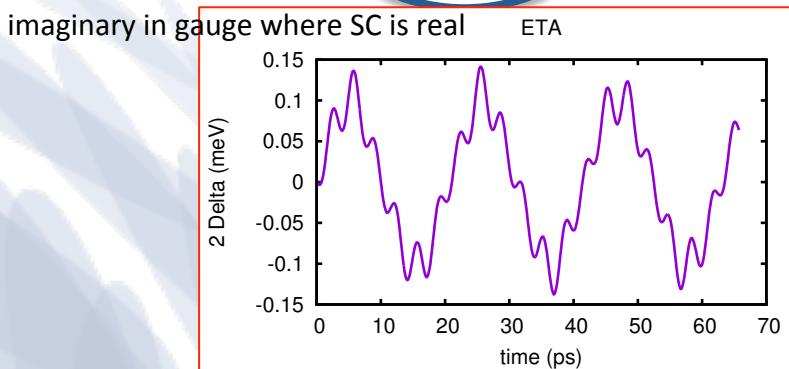
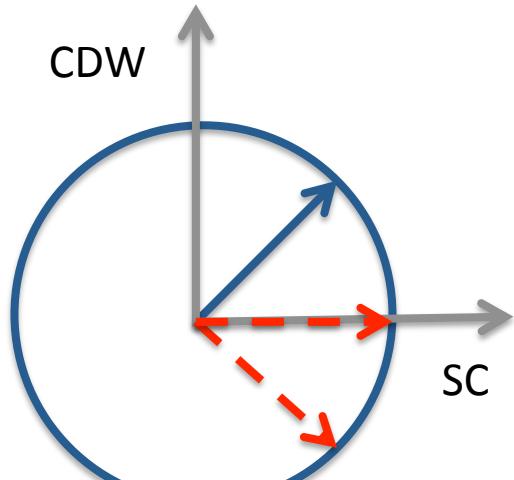
# Gap resonance – cw driving

Above resonance:  
SC up, CDW down

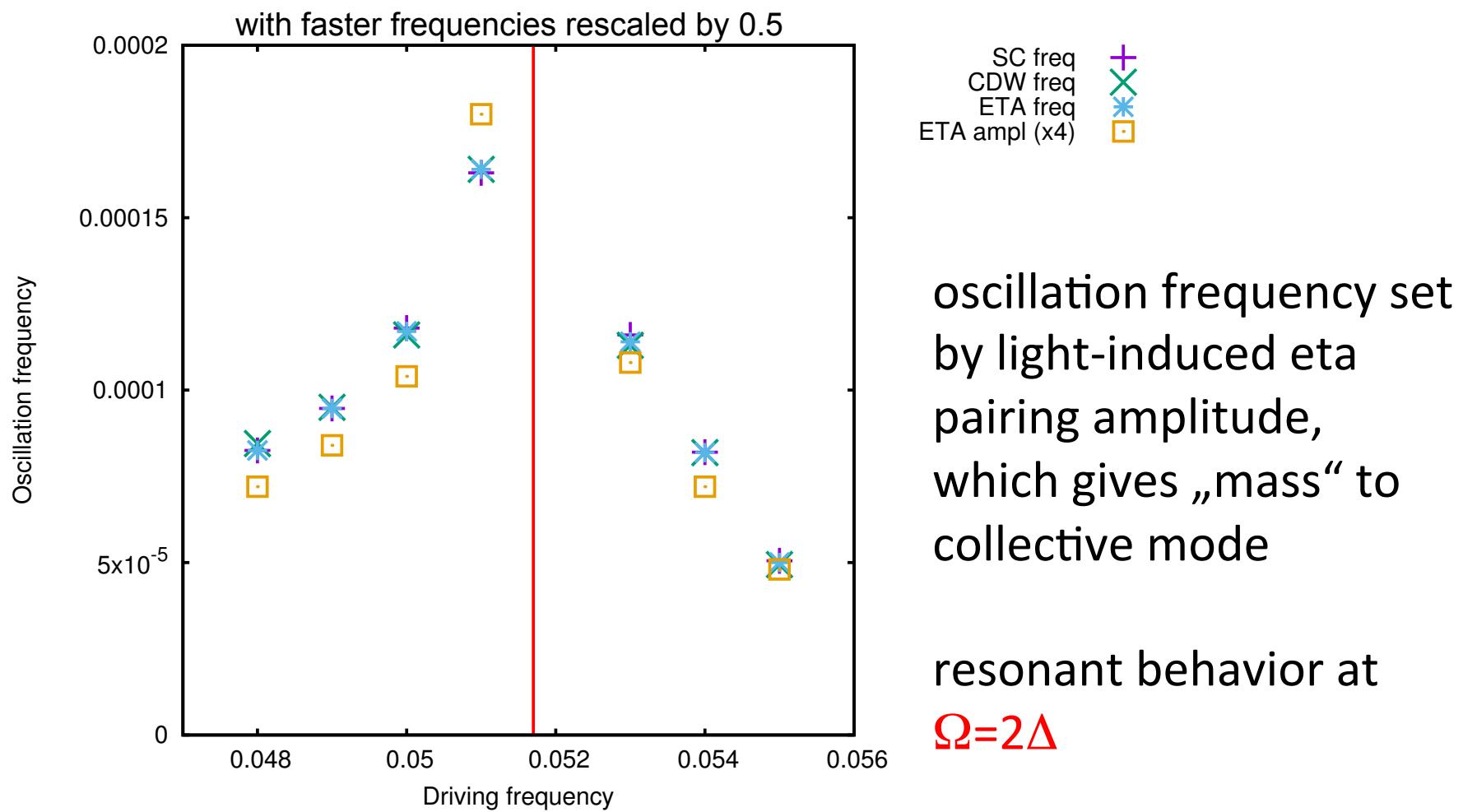


# Gap resonance – cw driving

Above resonance:  
SC up, CDW down



# Gap resonance – cw driving

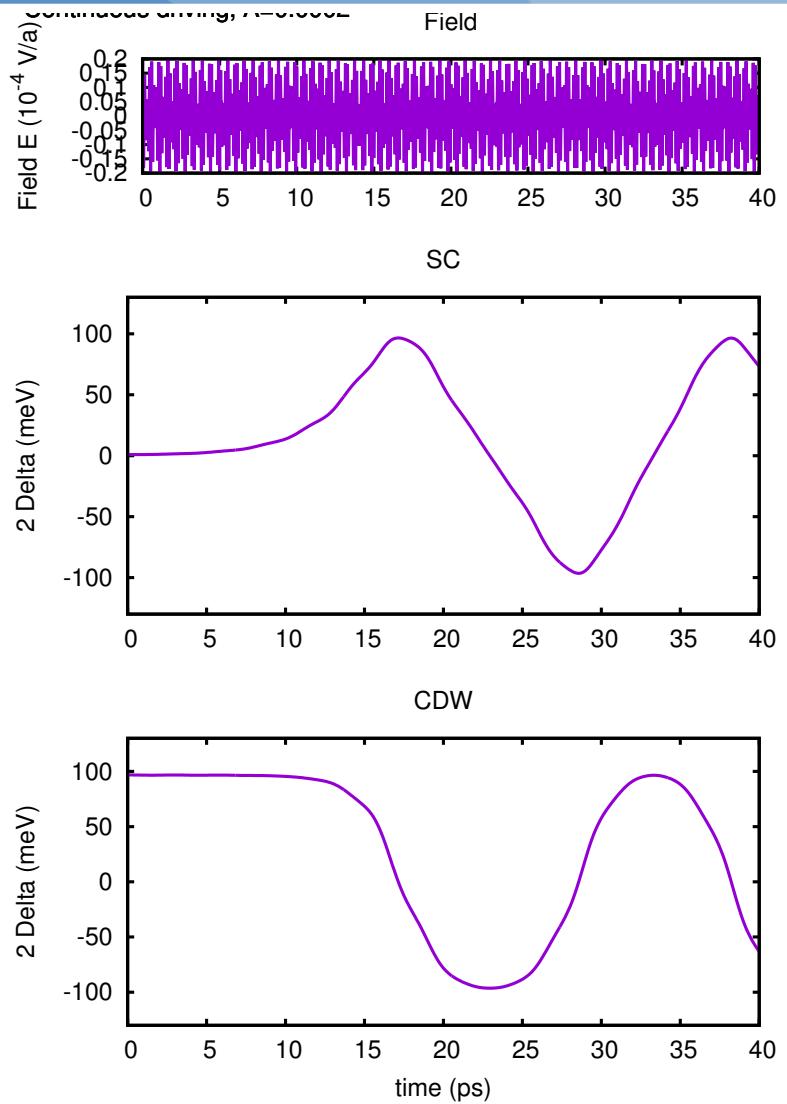


# Can we bring SC alive?

# Can we bring SC alive?

CDW initial state

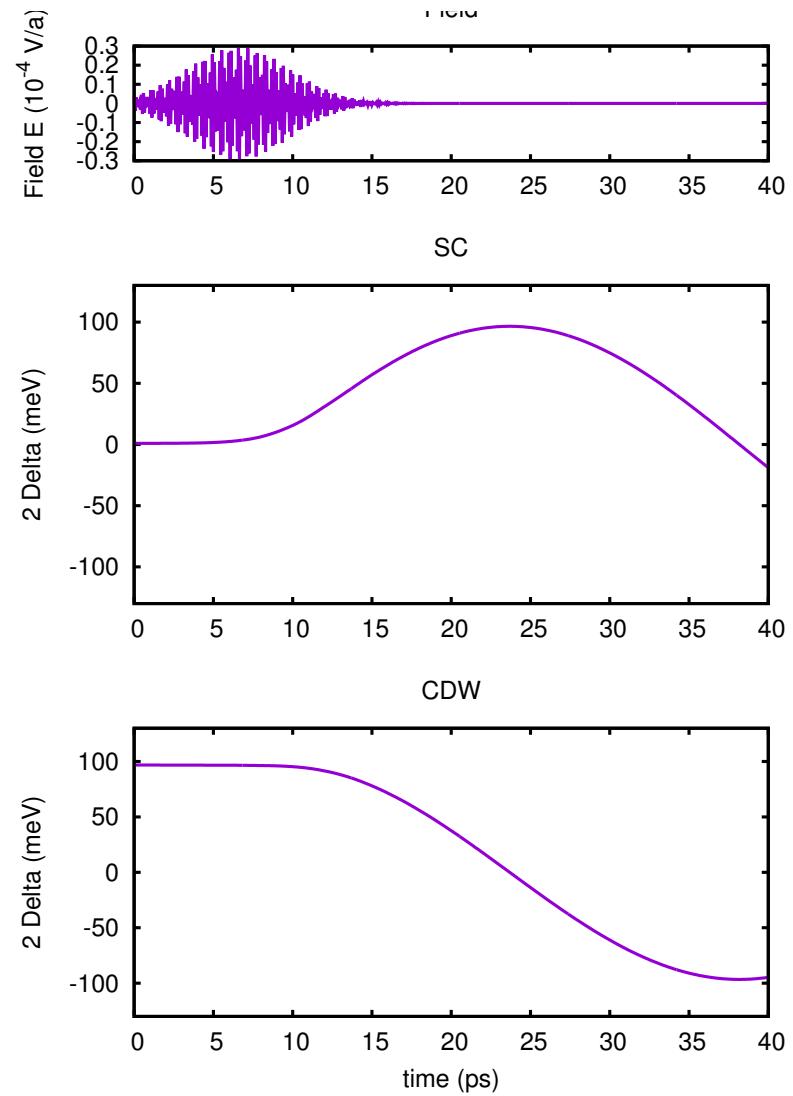
SC comes alive!



# Can we bring SC alive? – pulsed field

CDW initial state

SC comes alive!



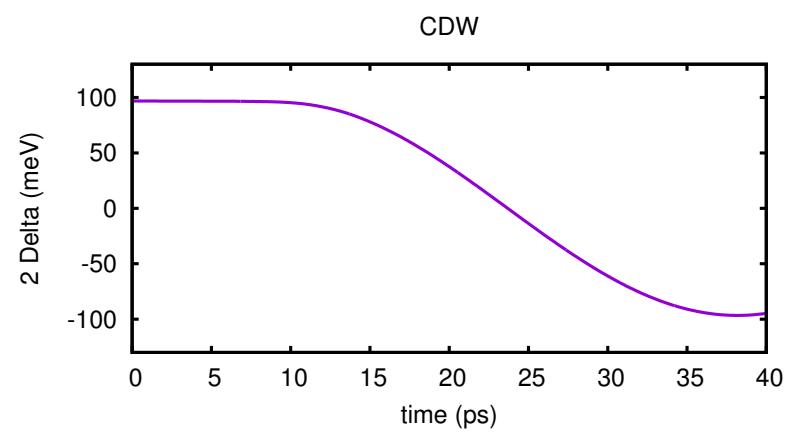
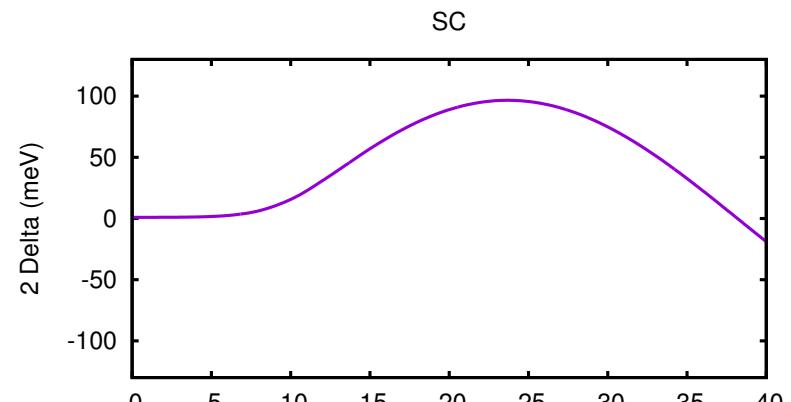
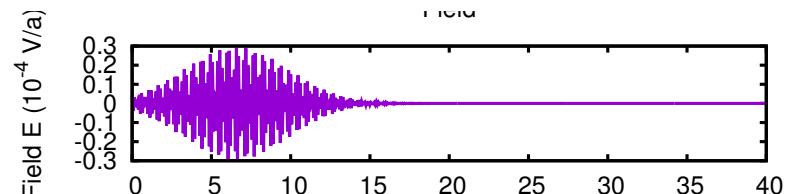
# What controls the dynamics?

Short times: laser control

Long times: dissipation?

## Questions:

1. Can we use dissipation to stabilize SC?
2. Nondegenerate case?



# Summary part 2

- laser-controlled switching between SC/CDW
- light-induced eta pairing and a collective mode
- light-induced long-lived superconductivity possible?

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