# Theory of ultrafast dynamics in superconductors
## How to enhance pairing with light

Michael A. Sentef

*lab.sentef.org*

Max-Planck Institute for the Structure and Dynamics of Matter, Hamburg

IMS 2017 Workshop, Dresden

Emmy Noether-Programm
Deutsche Forschungsgemeinschaft
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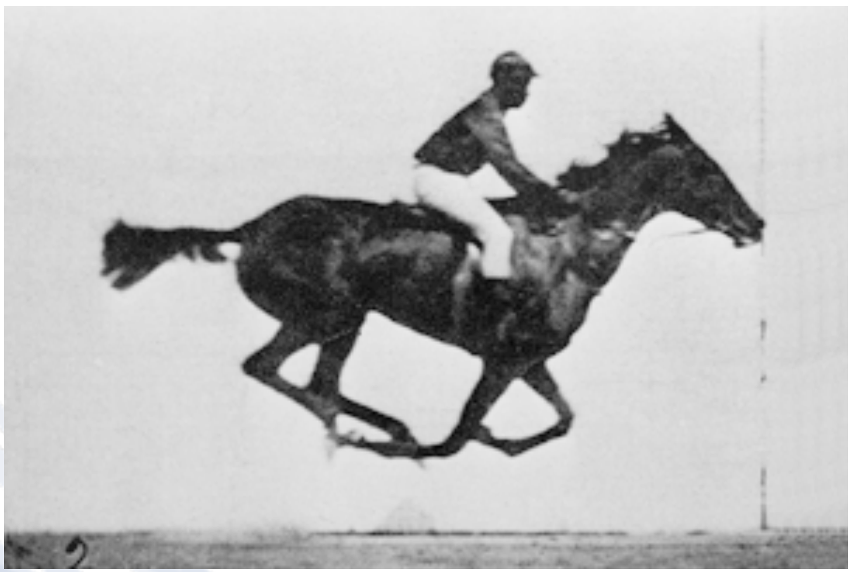
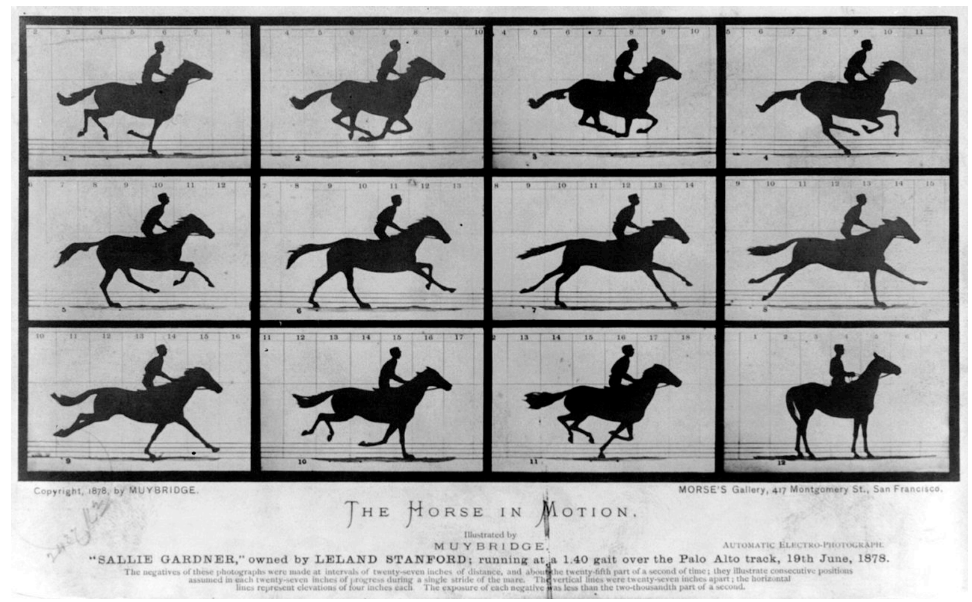
# Pump-probe spectroscopy (1887)

- stroboscopic investigations of dynamic phenomena

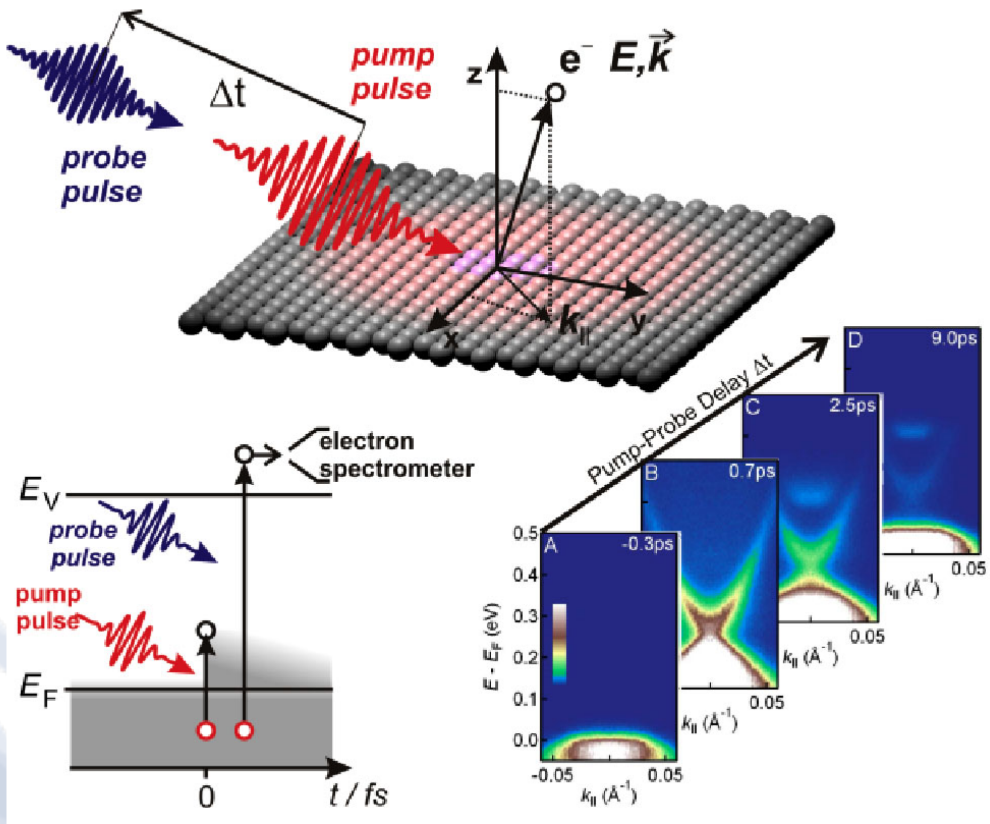




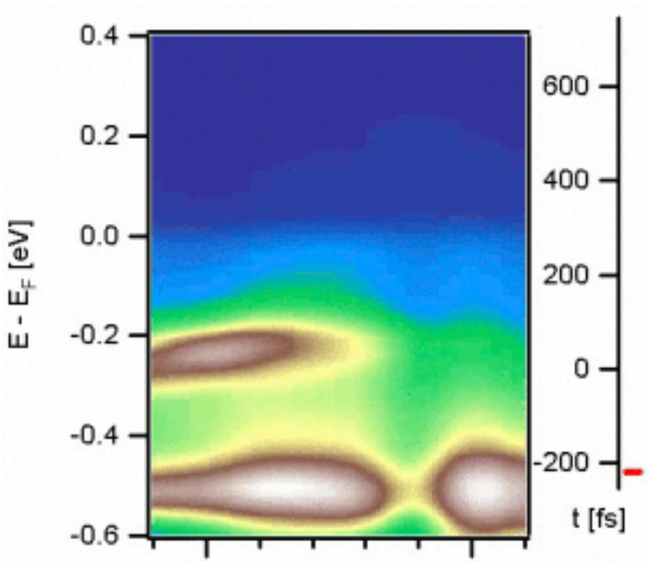

*Muybridge 1887*

# Pump-probe spectroscopy (today)

- stroboscopic investigations of dynamic phenomena




TbTe3 CDW metal




*Simulations of time-resolved ARPES: PRX 3, 041033 (2013)*

*Image courtesy: J. Sobota / F. Schmitt*

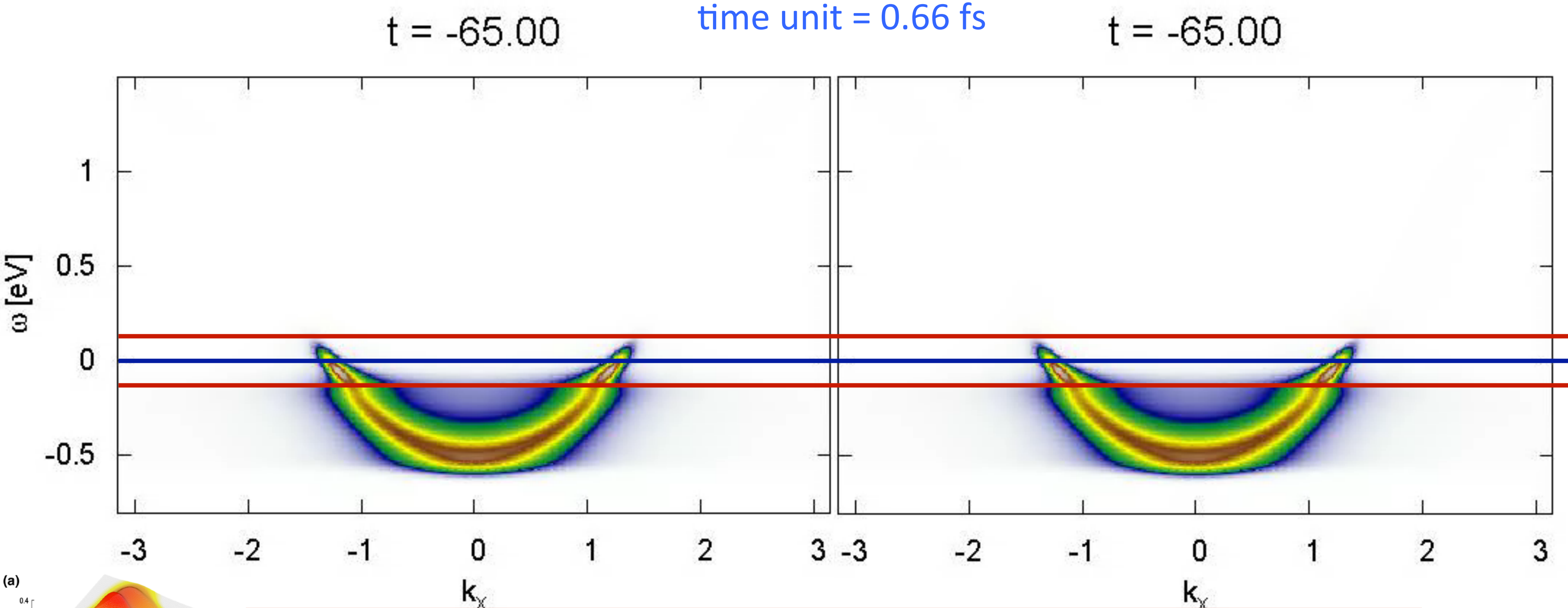
# Simulation of time-resolved ARPES

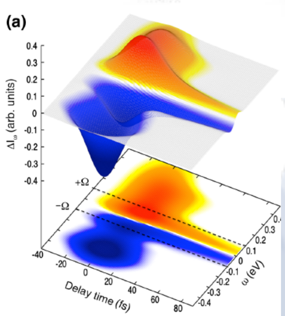
*PRX 3, 041033 (2013)*

Weak pump

Strong pump

time unit = 0.66 fs




boson window effect for fast versus slow relaxation

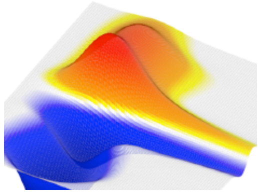
nonlinear response for strong pump

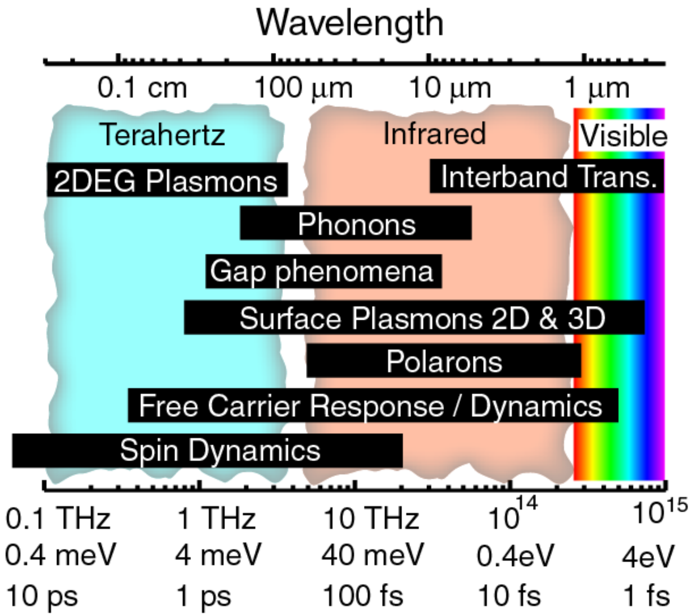
# Ultrafast Materials Science today

*Understanding the nature of quasiparticles*

- Relaxation dynamics
- Control of couplings

*PRL 111, 077401 (2013)*
*PRX 3, 041033 (2013)*
*PRB 87, 235139 (2013)*
*PRB 90, 075126 (2014)*
*Nature Commun. 7, 13761 (2016)*
*PRB 95, 024304 (2017)*
*PRB 95, 205111 (2017)*

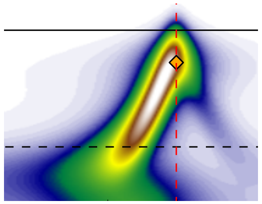





*Understanding ordered phases*

- Collective oscillations
- Competing orders

*PRB 92, 224517 (2015)*
*PRB 93, 144506 (2016)*
*PRL 118, 087002 (2017)*



*Creating new states of matter*

- Floquet topological states

*Nature Commun. 6, 7047 (2015)*
*Nature Commun. 8, 13940 (2017)*

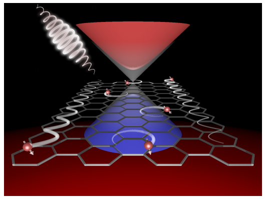


*Image courtesy:*
*D. Basov*

# Outline

## How to enhance pairing with light

- Part I: Laser-controlled competing orders

  Selective melting of a competing order enhances superconductivity

  *PRL 118, 087002 (2017)*

- Part II: Light-enhanced electron-phonon coupling

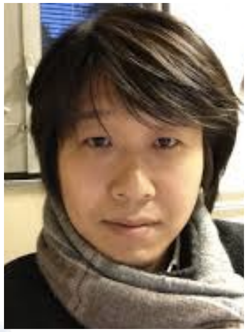
  Resonant excitation of IR phonon enhances electron-phonon coupling

  *PRB 95, 024304 (2017) - experiment*

  *PRB 95, 205111 (2017) - theory*

# I Theory of laser-controlled competing orders

*Phys. Rev. Lett. 118, 087002 (2017)*



Akiyuki Tokuno   Antoine Georges   Corinna Kollath
Palaiseau/Paris/Geneva   University of Bonn

# Nonequilibrium superconductivity

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**Why?**

- understand ordering mechanisms
- control ordered states: ultrafast switching
- induce new states of matter

**How to control?**

- laser near resonance with collective modes

**Generic mechanism to control competing orders with light?**

**Recent theories on laser-controlled couplings and competing orders:**
Akbari et al., EPL 101, 17003 (2013); Moor et al., PRB 90, 024511 (2014); Fu et al., PRB 90, 024506 (2014); Dzero et al., PRB 91, 214505 (2015); Tsuji&Aoki, PRB 92, 064508 (2015); Cea et al., PRB 93, 180507 (2016); Kemper et al., PRB 92, 224517 (2015); Sentef et al., PRB 93, 144506 (2016); Krull et al., Nat. Commun. 7, 11921 (2016); Patel&Eberlein, PRB 93, 195139 (2016); Knap et al., PRB 94, 214504 (2016); Komnik&Thorwart EPJB 89, 244 (2016); Coulthard et al., 1608.03964; Kennes et al., Nat. Physics (2017), doi:10.1038/nphys4024; Sentef, 1702.00952; Babadi et al. 1702.02531; Murakami et al.,1702.02942; Mazza&Georges, 1702.04675; Dehghani&Mitra, 1703.01621 ...

# Experimental motivation: competing orders are everywhere

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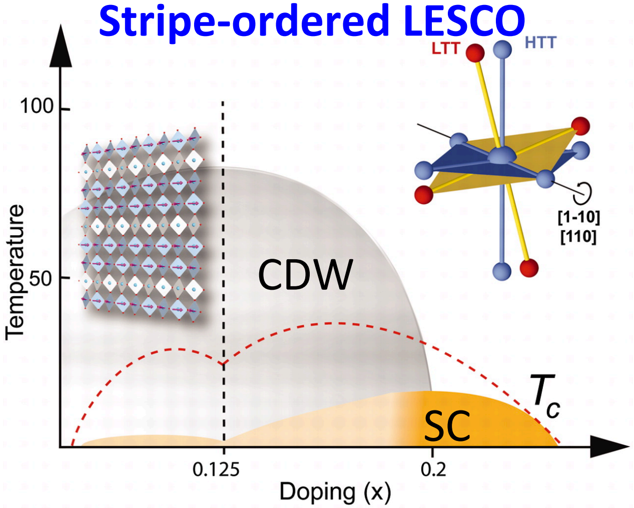
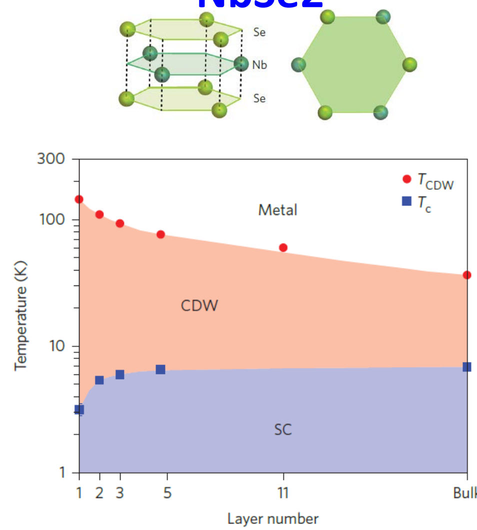
### Stripe-ordered LESCO



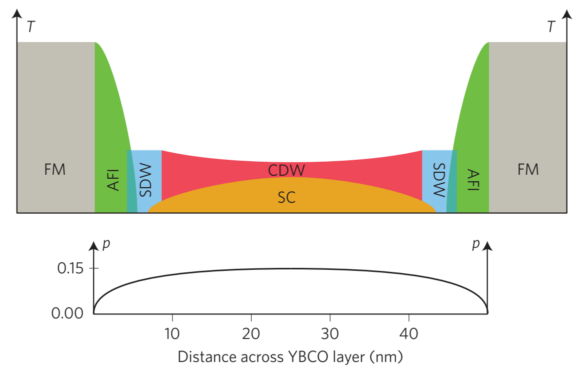

*D. Fausti et al.,*
*Science, 331, 189 (2011)*

### NbSe2




*X. Xi et al., Nat. Nanotechnol. 10, 765 (2015)*

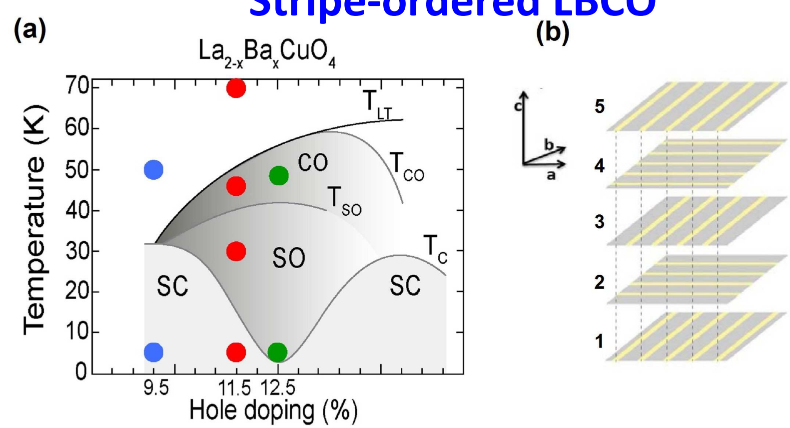
### YBCO-LCMO heterostructure




*A. Frano et al.,*
*Nat. Mater. 15, 831 (2016)*

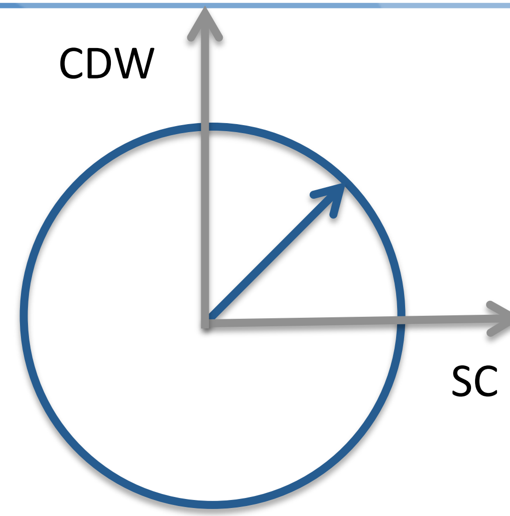
### Stripe-ordered LBCO

*D. Nicoletti et al.,*
*PRB 90, 100503 (2014)*



# Competing orders

- attractive -U Hubbard model
- degeneracy of SC and CDW at particle-hole symmetry (SU(2))
- SO(4) symmetry (SC, CDW, eta pairing)




VOLUME 63, NUMBER 19 PHYSICAL REVIEW LETTERS 6 NOVEMBER 1989

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

Chen Ning Yang

C. N. Yang

S.-C. Zhang

Reprinted from Mod. Phys. Lett. B4 (1990) 759–766


**$SO_4$ SYMMETRY IN A HUBBARD MODEL**

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

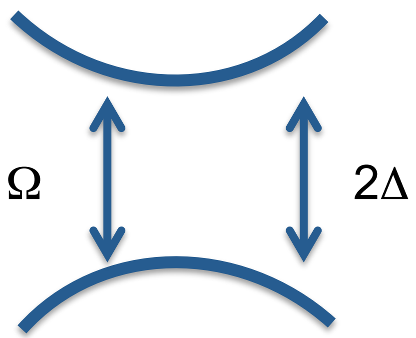
and

S. C. ZHANG
*IBM Research Division, Almaden Research Center, San Jose, CA 95120-6099, USA*

also see: Demler, Hanke, Zhang, SO(5) theory of antiferromagnetism and dSC, RMP 76, 909 (2004)

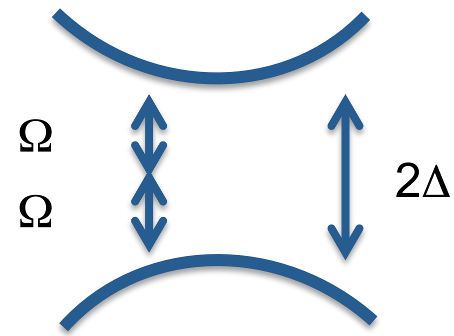
# Driven SC/CDW: Gauge field coupling

CDW ~ A
1-photon resonance

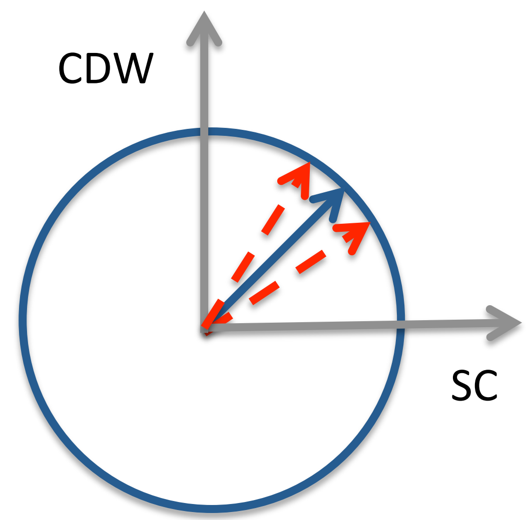



SC ~ A$^2$
2-photon resonance

Tsuji&Aoki, PRB 92, 064508 (2015)
Cea et al., PRB 93, 180507 (2016)




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



# Model

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

2D square lattice + attractive U + mean-field decoupling

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

# Mean-field equations

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Equations of motion for electronic driving:

$$i\partial_t n_k = -\Delta_{SC}(f_k - f_k^*) + \Delta_{CDW}(g_k - g_k^*) - \Delta_\eta^* \eta_k + \Delta_\eta \eta_k^*,$$

$$i\partial_t f_k = \Delta_{SC}(1 - (n_k + n_{-k})) + (\epsilon(k-A) + \epsilon(k+A)) f_k + \Delta_{CDW}(\eta_k + \eta_{k+Q}) - \Delta_\eta (g_k^* + g_{-k}^*),$$

$$i\partial_t g_k = \Delta_{CDW}(n_k - n_{k+Q}) - 2\epsilon(k-A) g_k + \Delta_{SC}(\eta_k^* - \eta_{k+Q}) + \Delta_\eta f_k^* - \Delta_\eta^* f_{k+Q},$$

$$i\partial_t \eta_k = \eta_k(\epsilon(k-A) - \epsilon(k+A)) + \Delta_{CDW}(f_k + f_{k+Q}) - \Delta_{SC}(g_{-k} + g_k^*) - \Delta_\eta (n_k + n_{-(k+Q)} - 1).$$

eta pairing provides coupling

nonlinear equations:
self-consistency in real time

$$\Delta_{SC} = U \sum_k f_k,$$

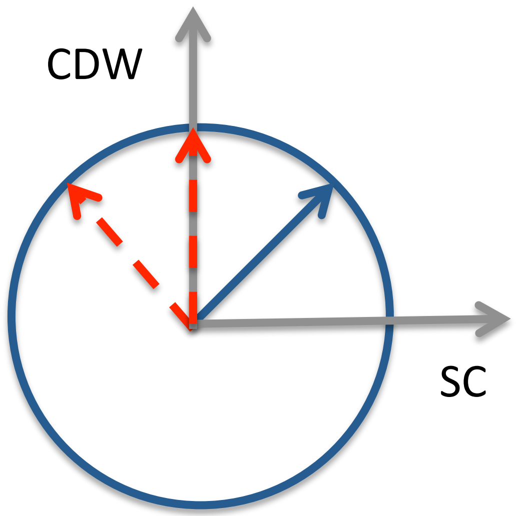
$$\Delta_{CDW} = U \sum_k g_k,$$

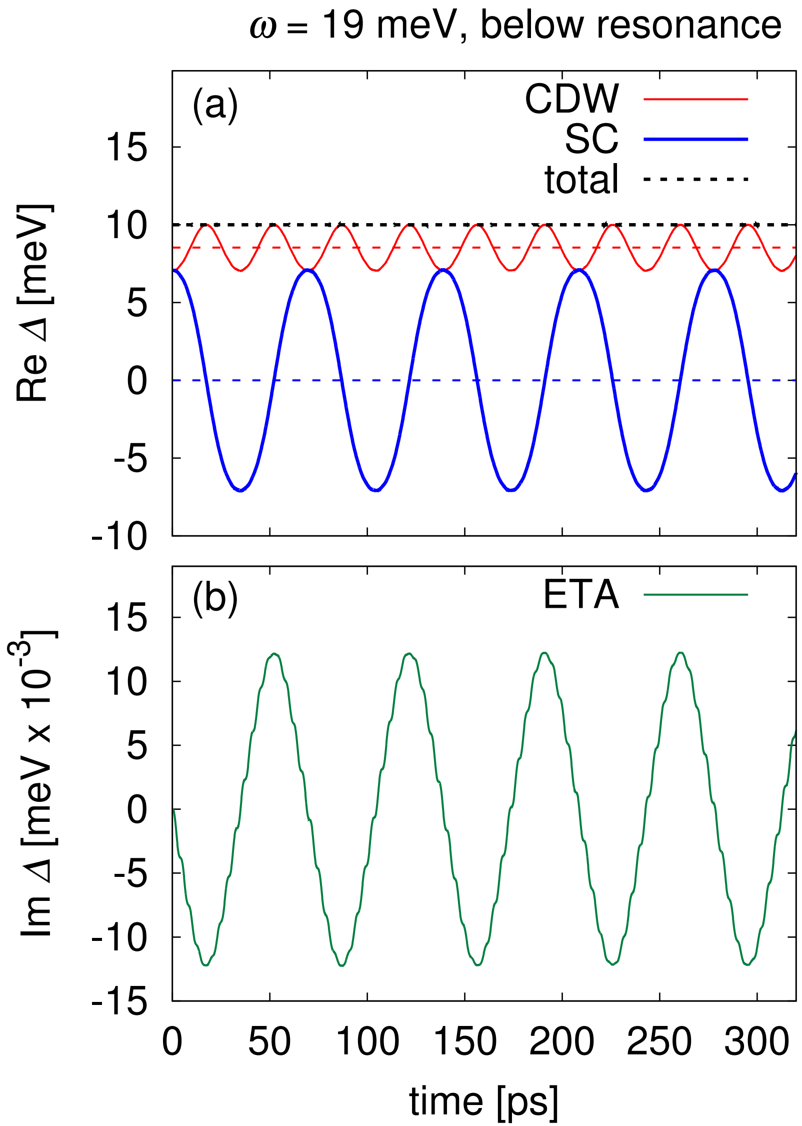
$$\Delta_\eta = U \sum_k \eta_k.$$

Nonequilibrium:
Periodic driving field: $A(t) = A_{max} \sin(\omega t)(e_x + e_y)$
$A_{max} = 5 \times 10^{-5}$, $E_{max} \sim 10\text{-}100$ V/cm – weak fields!

# Gap resonance – coexisting initial state

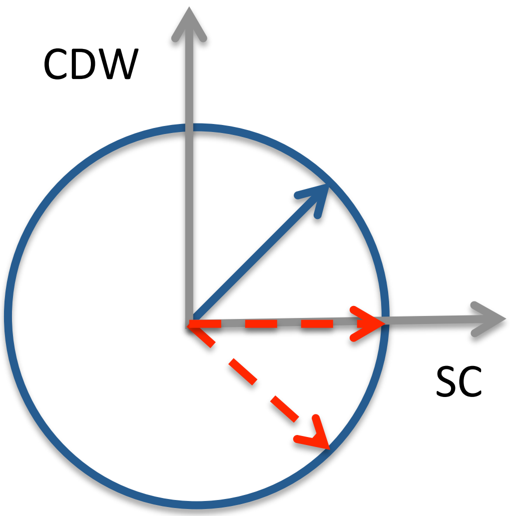
Below resonance:
SC down, CDW up

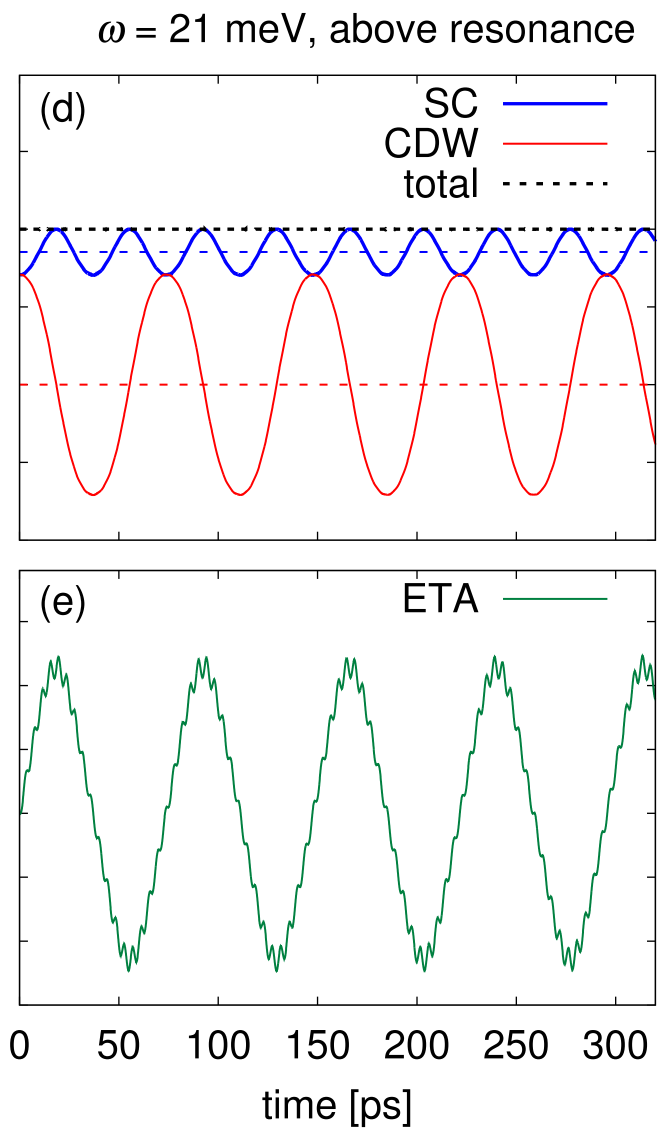


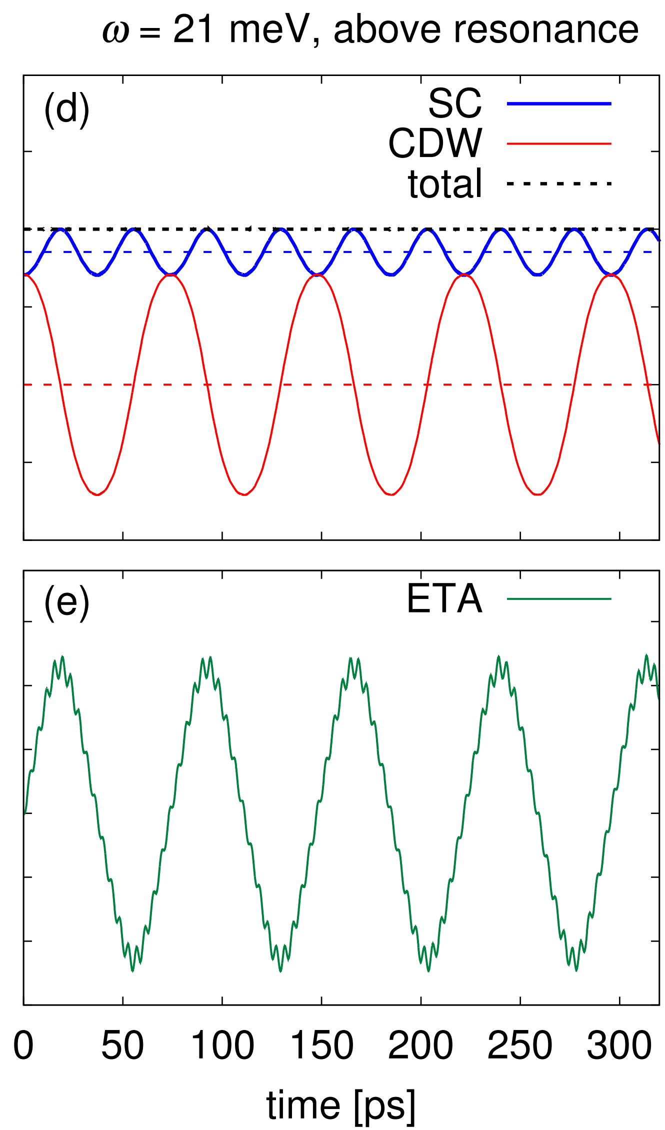
# Gap resonance – coexisting initial state

Above resonance:
SC up, CDW down



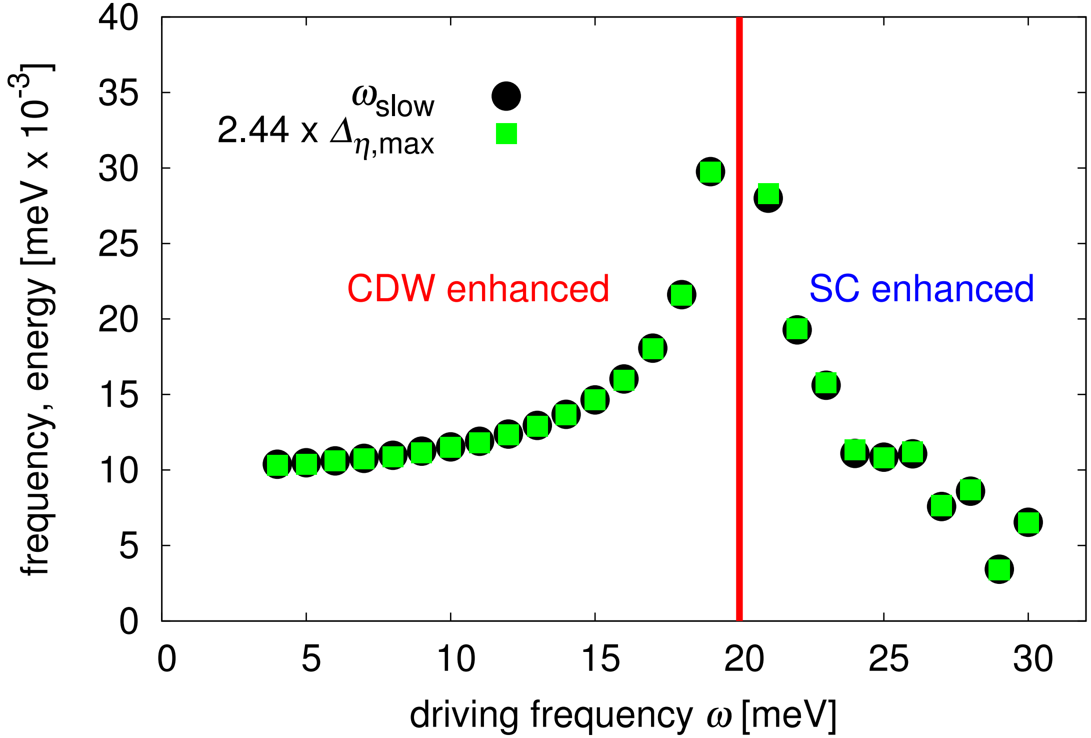



# Gap resonance – coexisting initial state


**„Floquet time crystal“ without many-body localization??**

# Gap resonance




collective mode frequency set by light-induced eta pairing amplitude, which gives „mass“ to collective mode

resonance at photon frequency $\omega=2\Delta$ = single-particle gap

# Gap resonance – why?

$$\mathrm{Im}\eta_{\vec{k},2}(t) = 2A_{\vec{k},0}\int_0^t \eta_{\vec{k},1}(t')\sin(\omega t')dt'$$
$$= \frac{2A_{\vec{k},0}^2\Delta_0 g_{\vec{k},0}t}{4E_{\vec{k}}^2 - \omega^2} + \eta_{\vec{k},2,\mathrm{osc}}(t),$$

short time expansion: leading contribution resonant for light-induced eta pairing – sign change when crossing $\omega=2\Delta$

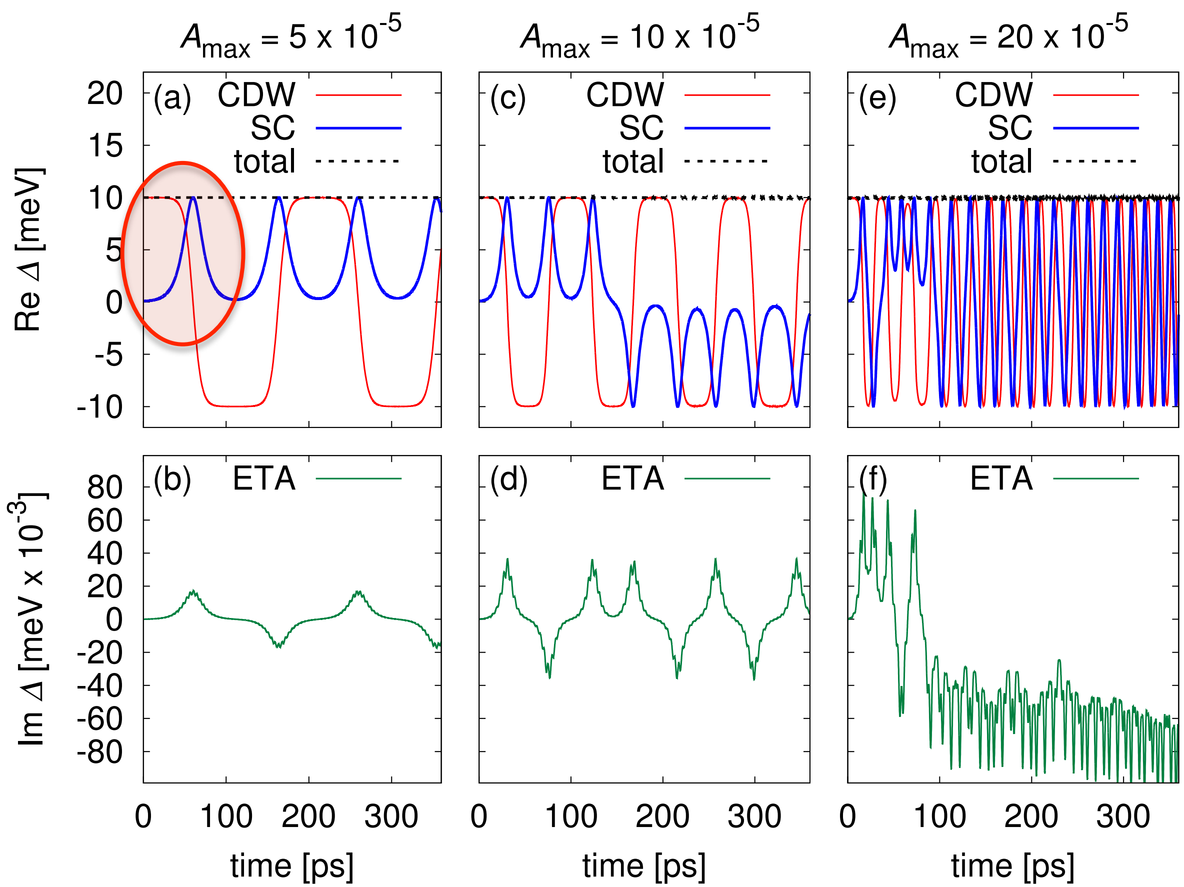
-> this triggers the dynamics between SC and CDW

generic mechanism for coexisting, non-commuting orders!

# Inducing superconductivity

99% CDW initial state

Drive slightly above gap




SC comes alive!

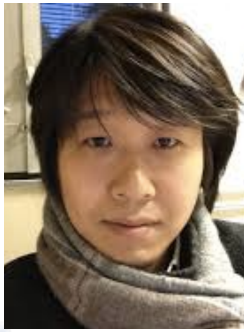
Irregular behavior for stronger driving

# Summary I

Tight-binding model + time-dependent mean-field theory:

- laser-controlled switching between SC/CDW
- path to understanding of light-induced superconductivity and light-induced CDW in systems with competing orders?

*Phys. Rev. Lett. 118, 087002 (2017)*



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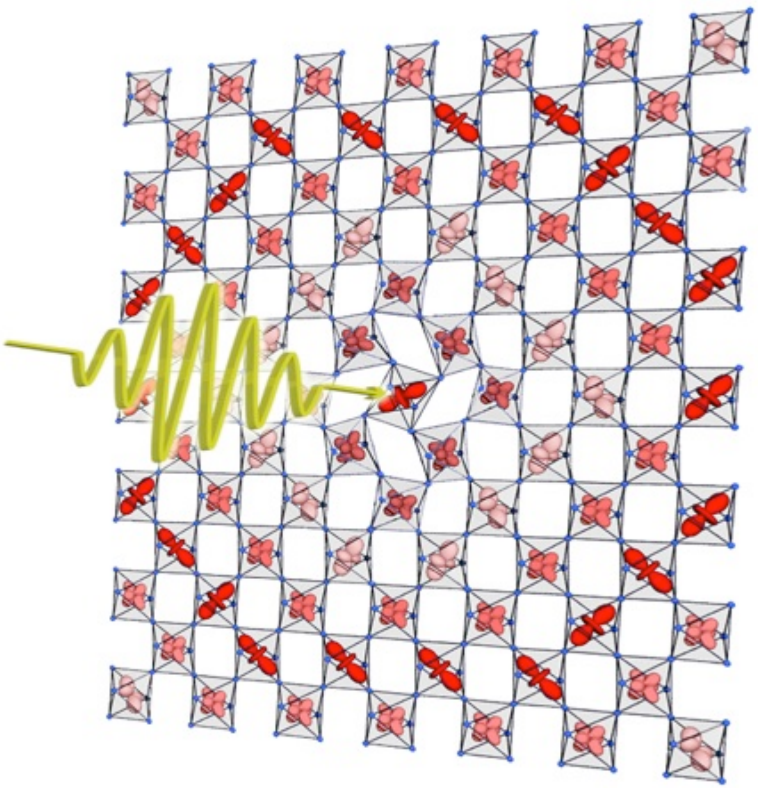
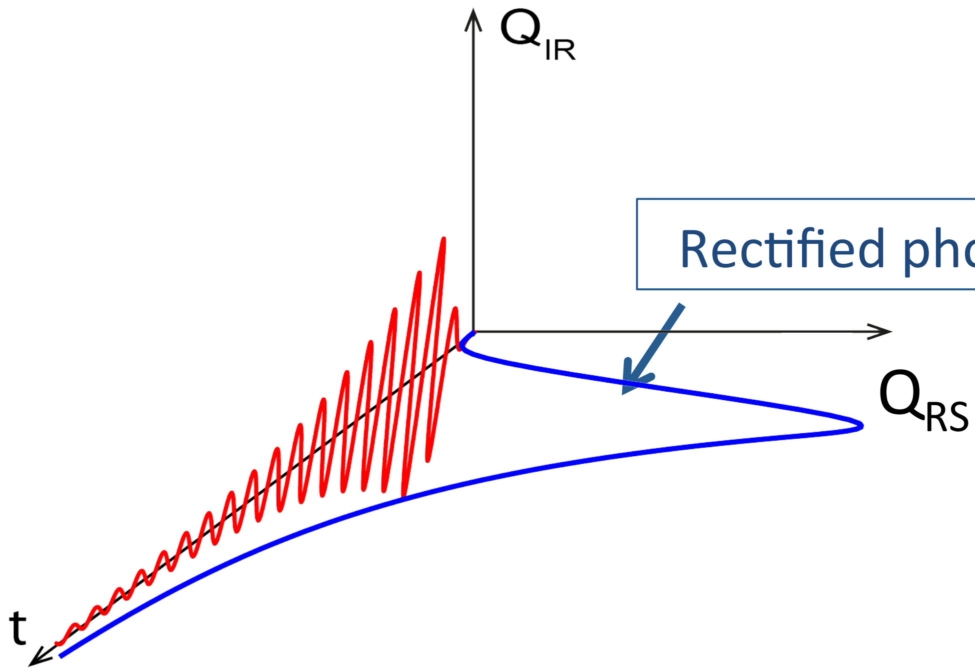
Palaiseau/Paris/Geneva   University of Bonn

# Resonant excitation of crystal lattice



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

# Classical nonlinear phononics




Rectified phonon field → directional force

Simplest model: classical dynamics

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

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

„nonlinear phononics“

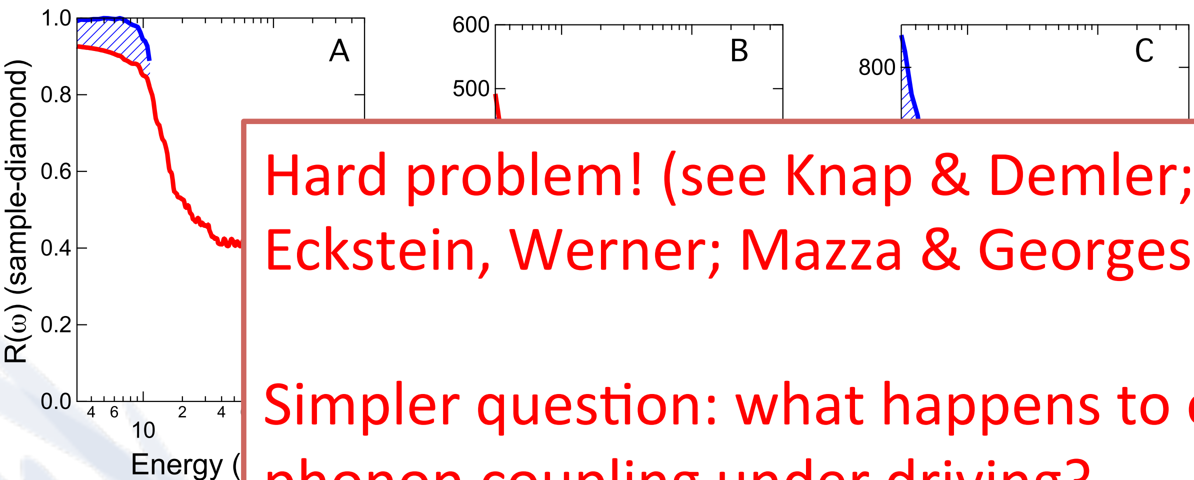
$$H = A Q_{IR}^2 Q_{RS}$$

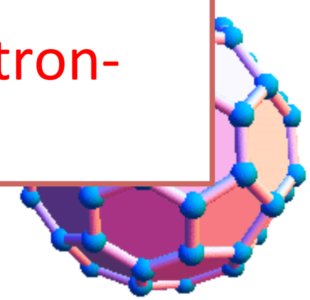
*M. Först et al., Nature Physics 7, 854 (2011)*

# Light-induced superconductivity?

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

Lattice control of reflectivity in $K_3C_{60}$



Hard problem! (see Knap & Demler; Murakami, Eckstein, Werner; Mazza & Georges ... )

Simpler question: what happens to electron-phonon coupling under driving?

# Electron-boson coupling (bilinear)

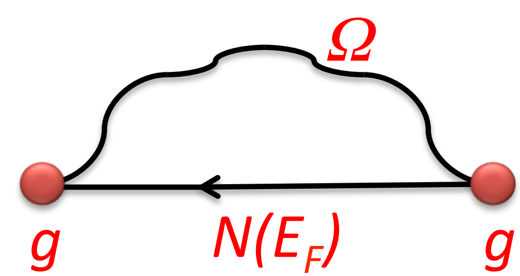
## Holstein model (minimal version):

$$H = \sum_k \epsilon(k) c_k^\dagger c_k + \Omega \sum_i b_i^\dagger b_i - g \sum_i c_i^\dagger c_i (b_i + b_i^\dagger)$$

Electrons (Fermi gas/liquid)

Bosons (e.g., Einstein phonon)

Electron-boson coupling

$\Sigma =$ 


Migdal-Eliashberg theory
boson-mediated pairing

# II Dynamically enhanced coupling

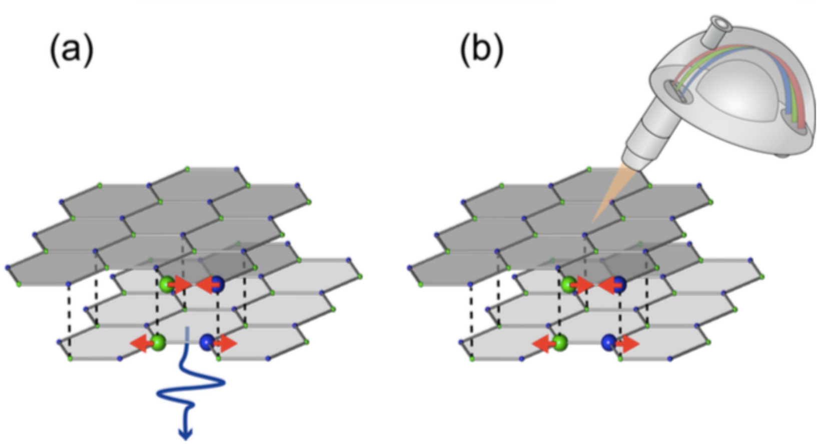
## Enhanced electron-phonon coupling in graphene with periodically distorted lattice

E. Pomarico, M. Mitrano, H. Bromberger, M. A. Sentef, A. Al-Temimy, C. Coletti, A. Stöhr, S. Link, U. Starke, C. Cacho, R. Chapman, E. Springate, A. Cavalleri, and I. Gierz

Phys. Rev. B **95**, 024304 – Published 13 January 2017

*PRB 95, 024304 (2017)*

enhanced electron-phonon for pump on resonance with IR phonon



# Dynamically enhanced coupling?

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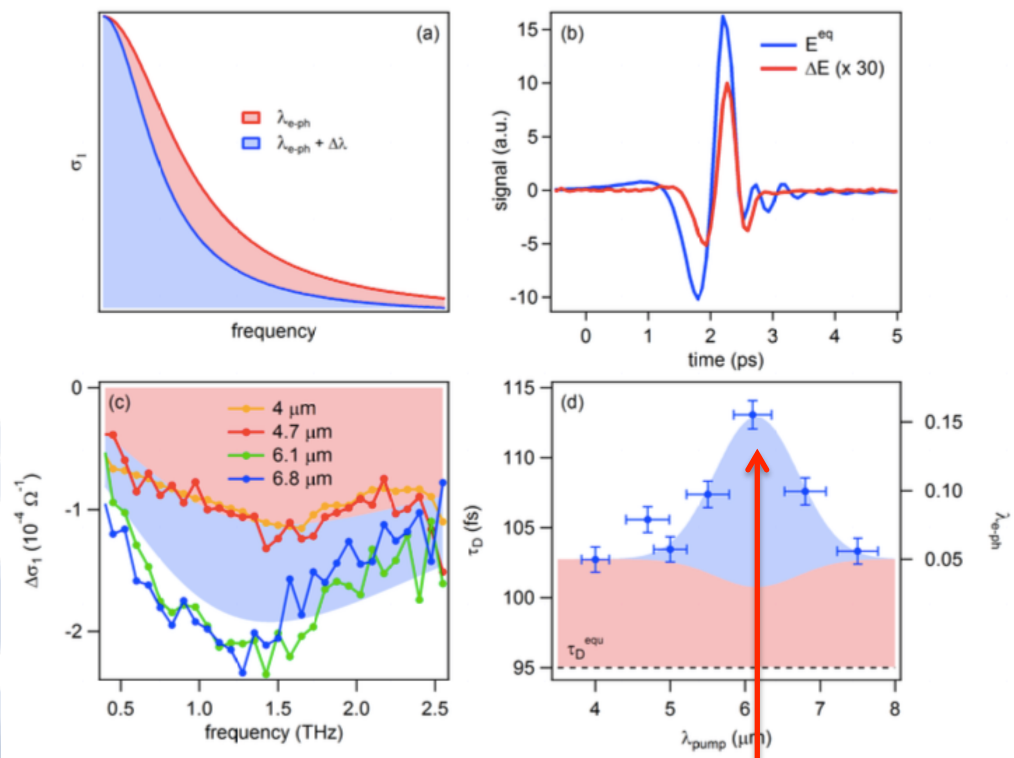
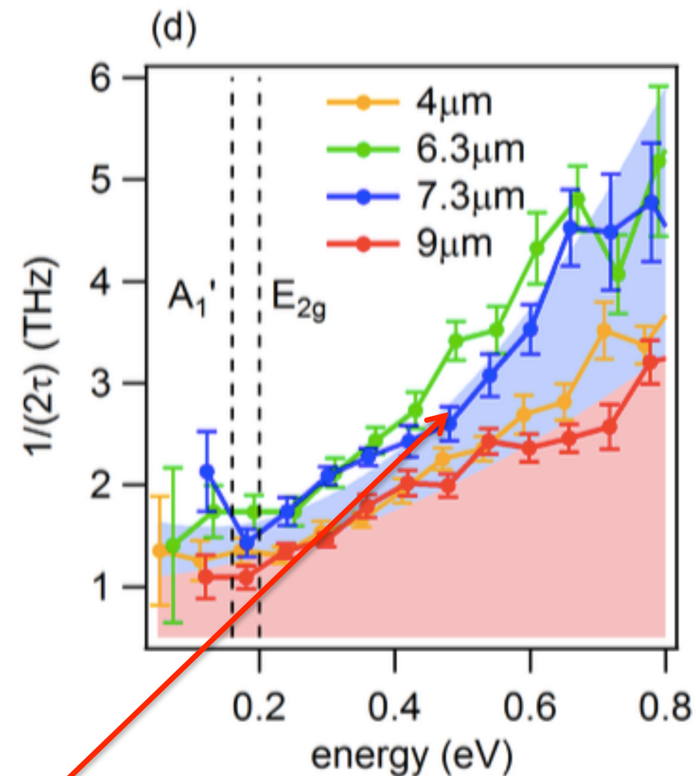
## Enhanced electron-phonon coupling in graphene with periodically distorted lattice

*PRB 95, 024304 (2017)*

transient reduction of THz Drude weight




enhanced tr-ARPES relaxation




3-fold enhancement of effective $\lambda_{el\text{-}ph}$! Why?

# Quantum nonlinear phononics

*PRB 95, 205111 (2017)*

$$\hat{H}(t) = -J\sum_{\sigma}(c^\dagger_{1,\sigma}c_{2,\sigma} + c^\dagger_{2,\sigma}c_{1,\sigma}) + g_2 \sum_{\sigma,l=1,2} \hat{n}_{l,\sigma}(b_l + b_l^\dagger)^2 + \Omega \sum_{l=1,2} b_l^\dagger b_l + F(t)\sum_{l=1,2}(b_l + b_l^\dagger),$$

also cf.
Kennes et al.,
Nature Physics 13, 479 (2017),
1609.03802

electron-occupation dependent squeezing of the phonon;
$g_2$ can be positive or negative in materials
-> mode hardening or softening

Idea: Drive nonlinearly coupled phonon and look at electronic response

Drive: $$F(t) = F\sin(\omega t),$$

Response: time-resolved spectral function

$$I(\omega, t_0) = \mathrm{Re}\int dt_1\, dt_2\, e^{i\omega(t_1-t_2)} s_{t_1,t_2,\tau}(t_0) \times \Big[\langle\psi(t_2)|c^\dagger_{1,\uparrow}\mathcal{T}e^{-i\int_{t_1}^{t_2} H(t)dt} c_{1,\uparrow}|\psi(t_1)\rangle + + \langle\psi(t_1)|c_{1,\uparrow}\mathcal{T}e^{-i\int_{t_2}^{t_1} H(t)dt} c^\dagger_{1,\uparrow}|\psi(t_2)\rangle\Big],$$

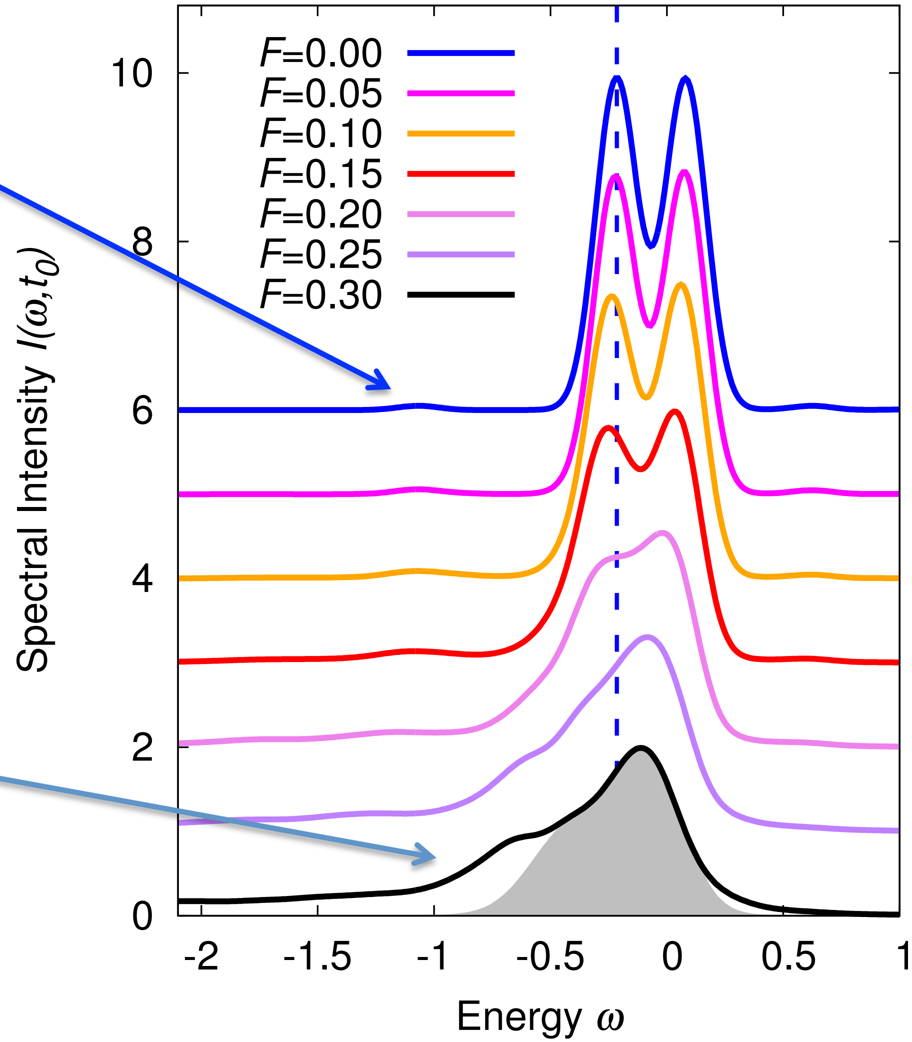
# Time-resolved electronic spectrum

*PRB 95, 205111 (2017)*

Here: g2=−0.05 < 0
Does not matter for light-enhanced coupling

Reduced coherence peaks with stronger driving

Looks like enhanced el-ph coupling



# Field scaling

*PRB 95, 205111 (2017)*

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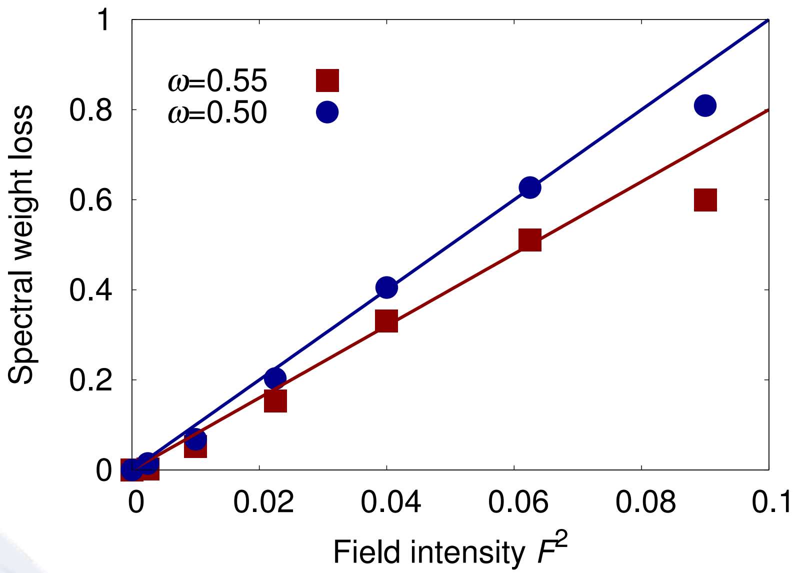
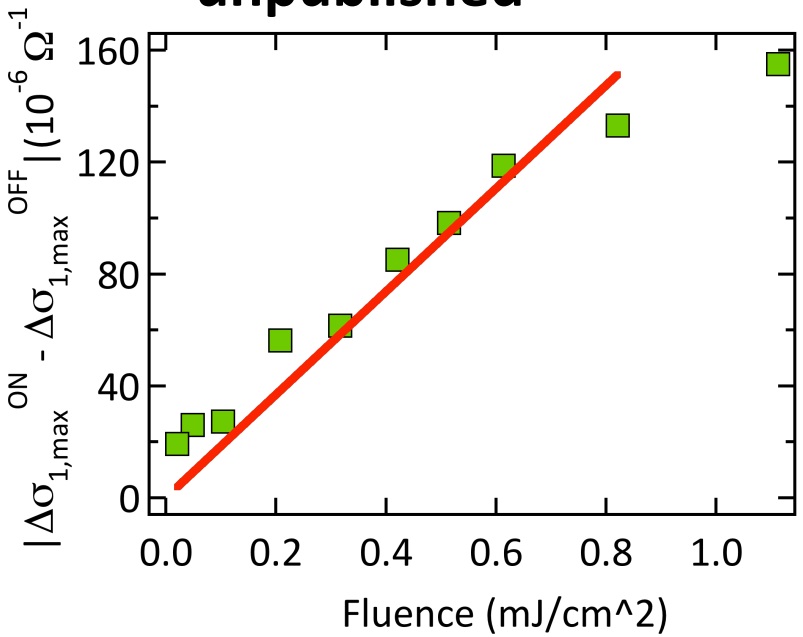
**Theory**




**Data by E. Pomarico, unpublished**




Scaling of coherent spectral weight loss: proportional to field intensity F^2 consistent with experiments

# Quantum nonlinear phononics

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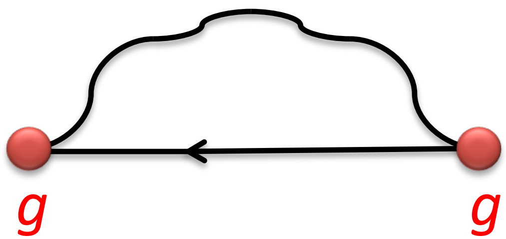
Forced coherent oscillation $\langle \hat{x}_l(t) \rangle \propto F \sin(\omega t)$

Coupling term in „mean-field“: $g_2 \hat{n}_l (b_l \langle b_l(t) \rangle + b_l^\dagger \langle b_l^\dagger(t) \rangle)$

~F ~F

$\Sigma =$




Migdal-Eliashberg theory

$$\Sigma(t,t') = i g(t) g^*(t') G(t,t') D(t,t')$$

effectively time-dependent vertex, g^2 ~ F^2
=> light-induced coupling, effective lambda scales ~ F^2

# Enhanced double occupancy: attraction?

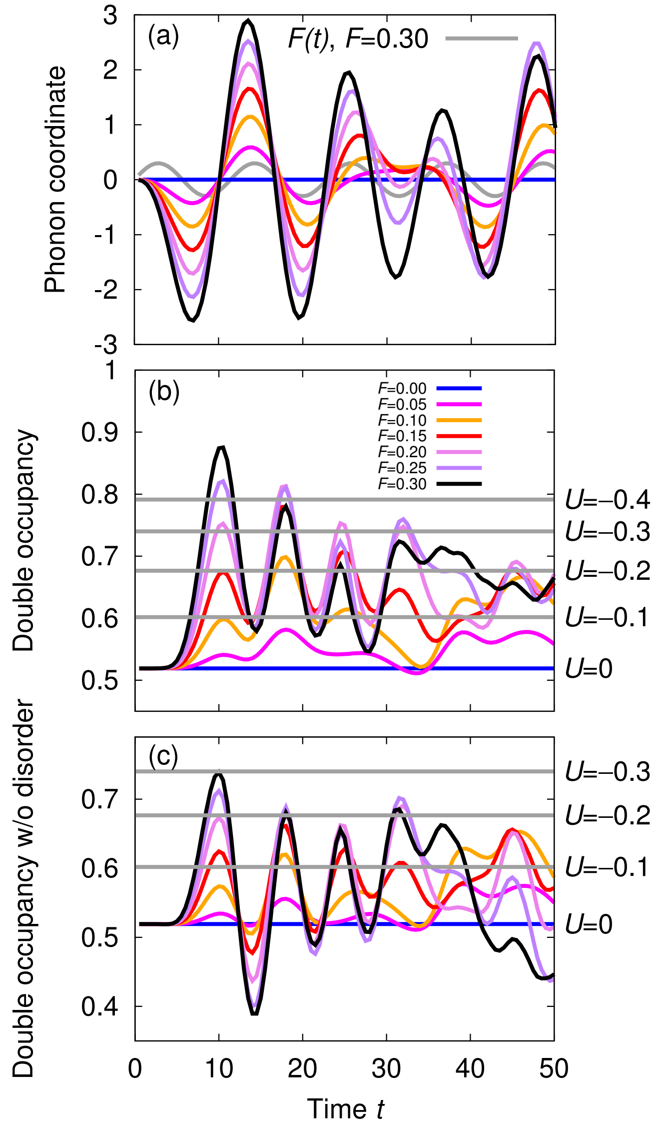
Coherent phonon oscillation

Enhanced double occupancy (above „random“ value)

Contribution from „disorder“ term (localization)

$$g_2 \hat{n}_l 2 b_l^\dagger b_l$$

subtracting disorder contribution: effective attraction when drive is not too strong



# Summary II

- enhanced electron-phonon coupling in phononically driven bilayer graphene

*PRB 95, 024304 (2017)*

E. Pomarico

I. Gierz

A. Cavalleri

Exact solution of electron-phonon model system:

- theoretical proposal: nonlinear el-ph coupling as mechanism behind this enhancement

*PRB 95, 205111 (2017)*