Theory of pump-probe spectroscopy:
Ultrafast laser engineering of ordered phases and microscopic couplings

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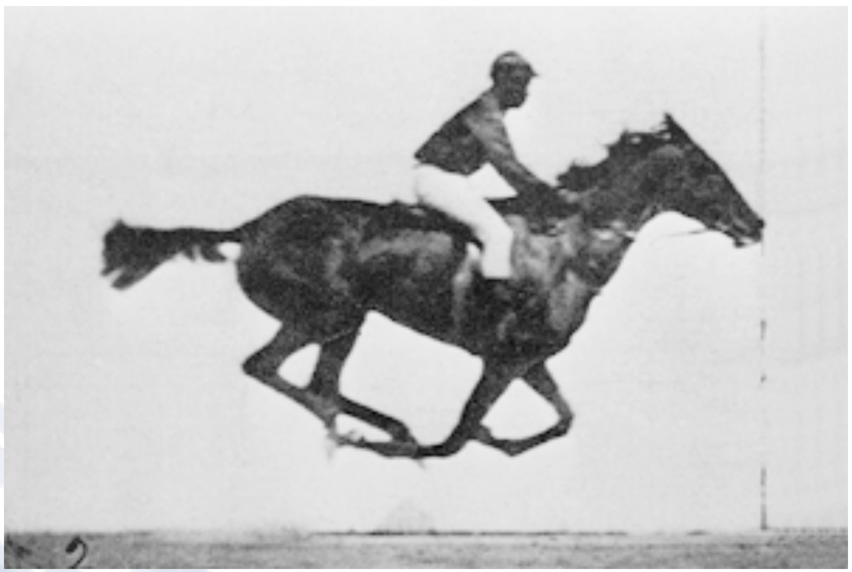
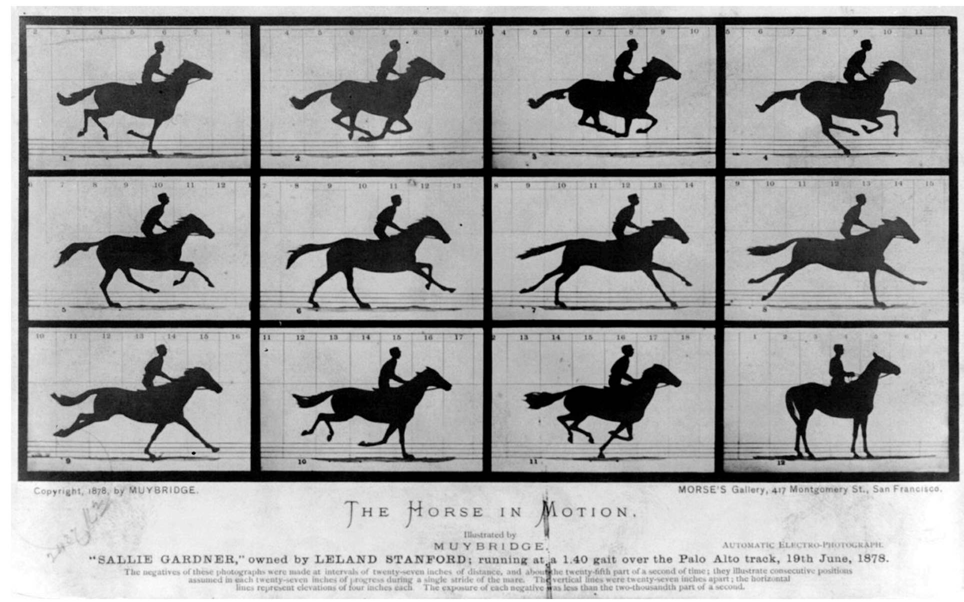
DPG Meeting Berlin, March 2018

Funded through Deutsche Forschungsgemeinschaft
Emmy Noether Programme (SE 2558/2-1)

Pump-probe spectroscopy (1887)

- stroboscopic investigations of dynamic phenomena

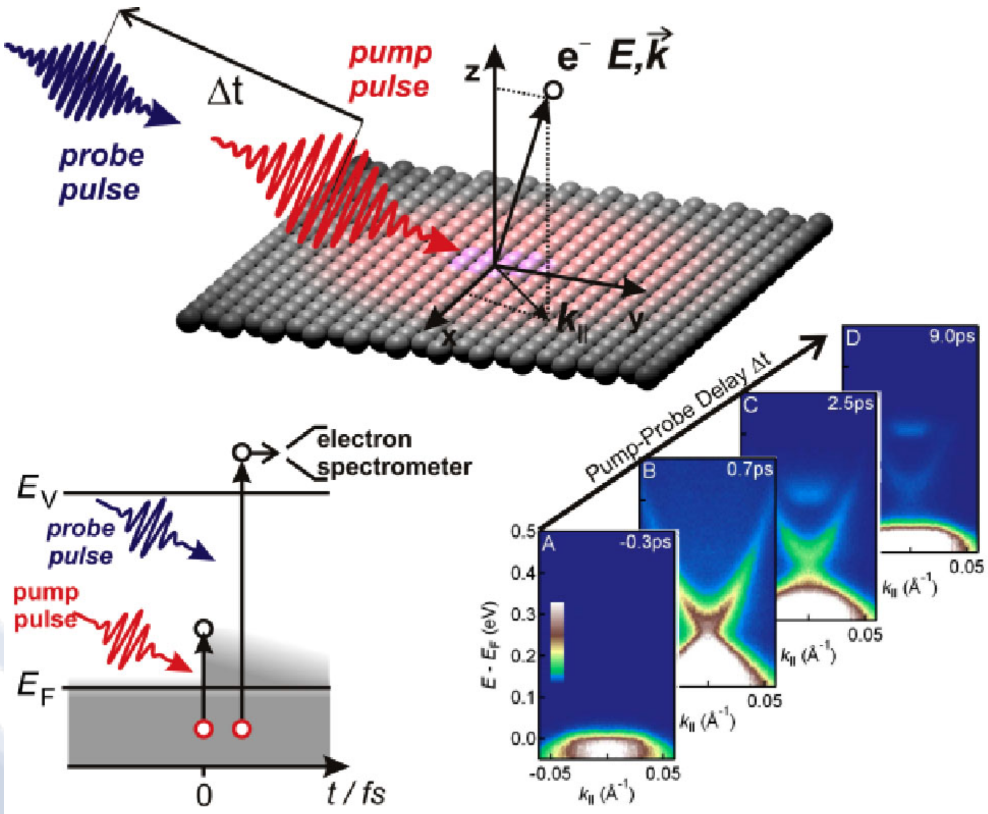




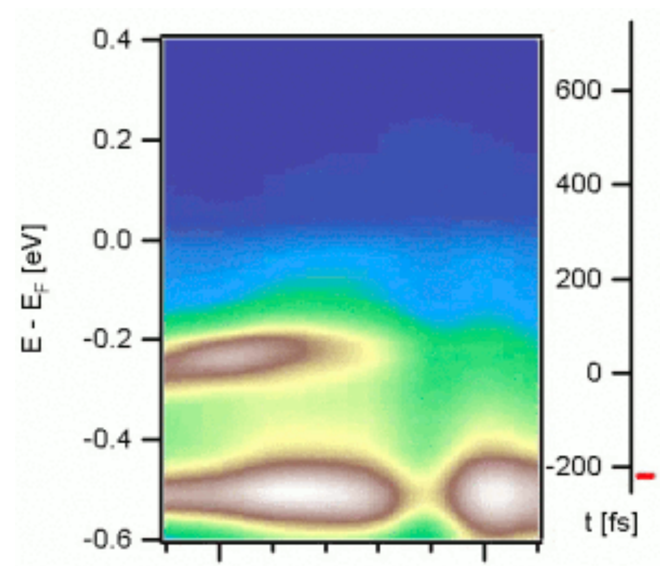
Muybridge 1887

Pump-probe spectroscopy (today)

- stroboscopic investigations of dynamic phenomena



TbTe3 CDW metal



Simulations of time-resolved ARPES: PRX 3, 041033 (2013), PRB 90, 075126 (2014), PRB 92, 224517 (2015), Nature Commun. 7, 13761 (2016)

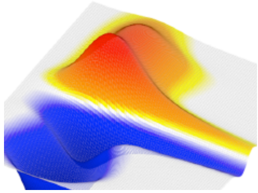
Image courtesy: J. Sobota / F. Schmitt

Ultrafast Materials Science today

Understanding the nature of quasiparticles

- Relaxation dynamics
- Control of couplings

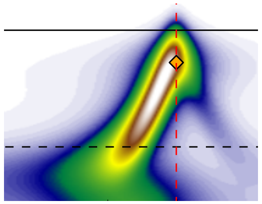
PRL 111, 077401 (2013)
PRX 3, 041033 (2013)
PRB 87, 235139 (2013)
PRB 90, 075126 (2014)
Nature Commun. 7, 13761 (2016)
PRB 95, 024304 (2017)
PRB 95, 205111 (2017)
arXiv:1712.01067
arXiv:1802.09437



Understanding ordered phases

- Collective oscillations
- Competing orders

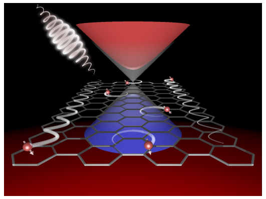
PRB 92, 224517 (2015)
PRB 93, 144506 (2016)
PRL 118, 087002 (2017)



Creating new states of matter

- Floquet topological states

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



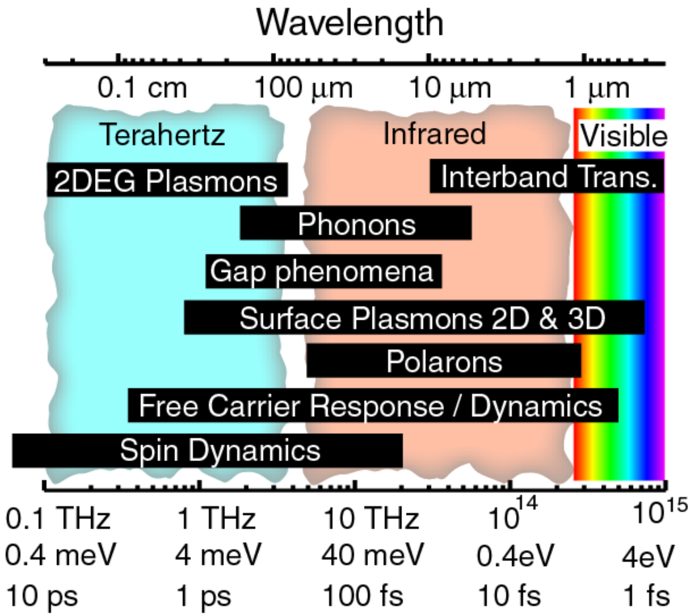


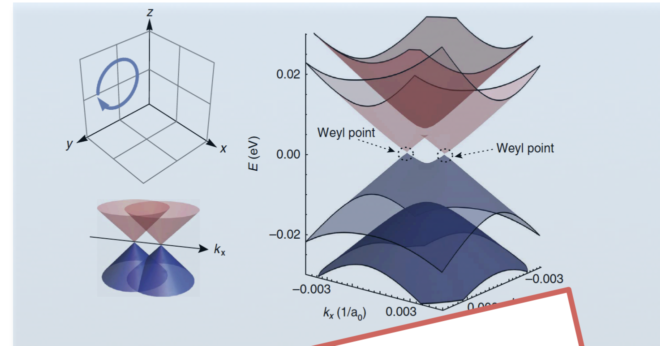
Image courtesy: D. Basov

Outline

Ultrafast laser engineering of

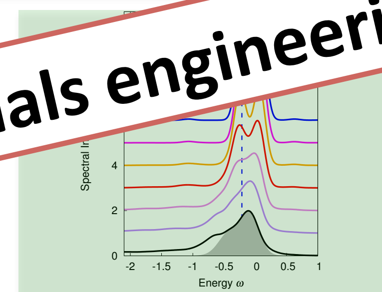
- band structure, topology (Floquet)

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



- electron-phonon coupling (quantum nonlinear phononics)

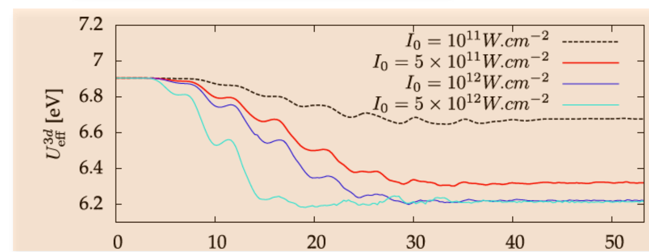
  PRB 95, 024304 (2017)
  PRB 95, 2051



Path towards nonequilibrium materials engineering

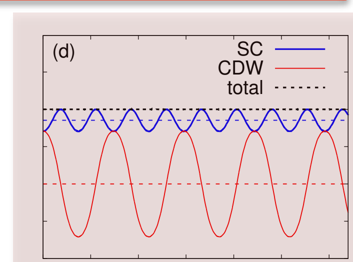
- H... subresonant ...ations in correlated insulators)

  arXiv:1712.01067



- superconductivity

  PRB 92, 224517 (2015)
  PRB 93, 144506 (2016)
  PRL 118, 087002 (2017)
  arXiv:1802.09437



Outline

How to modify couplings with light

- Part I: Light-enhanced electron-phonon coupling

  Resonant excitation of IR phonon enhances electron-phonon coupling

  E: Pomarico et al., PRB 95, 024304 (2017) – experiment (bilayer graphene)

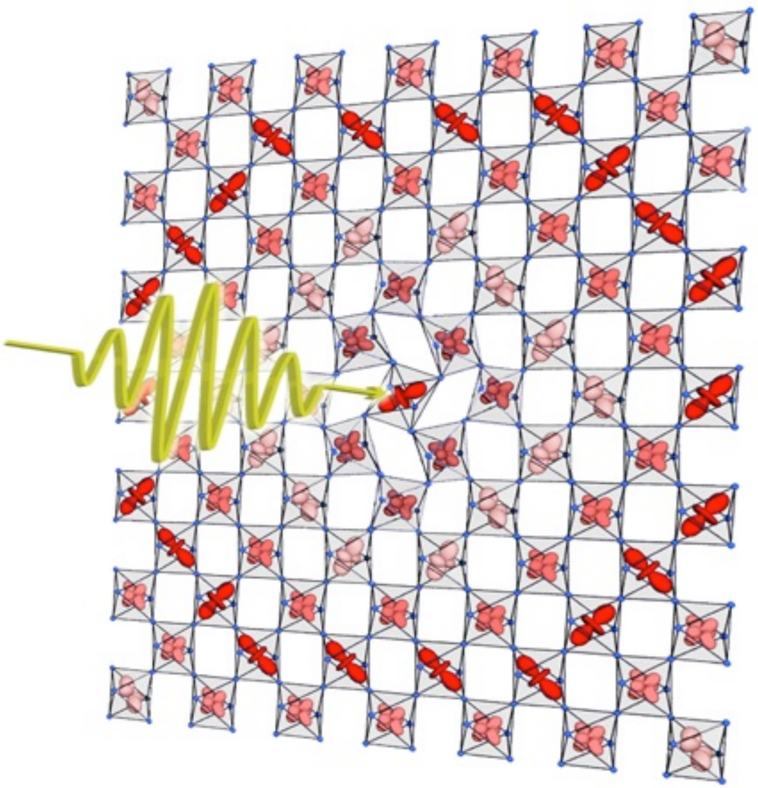
  M. A. Sentef, PRB 95, 205111 (2017) – theory

- Part II: Light-reduced Hubbard U

  Nonresonant laser driving reduces Hubbard U in NiO

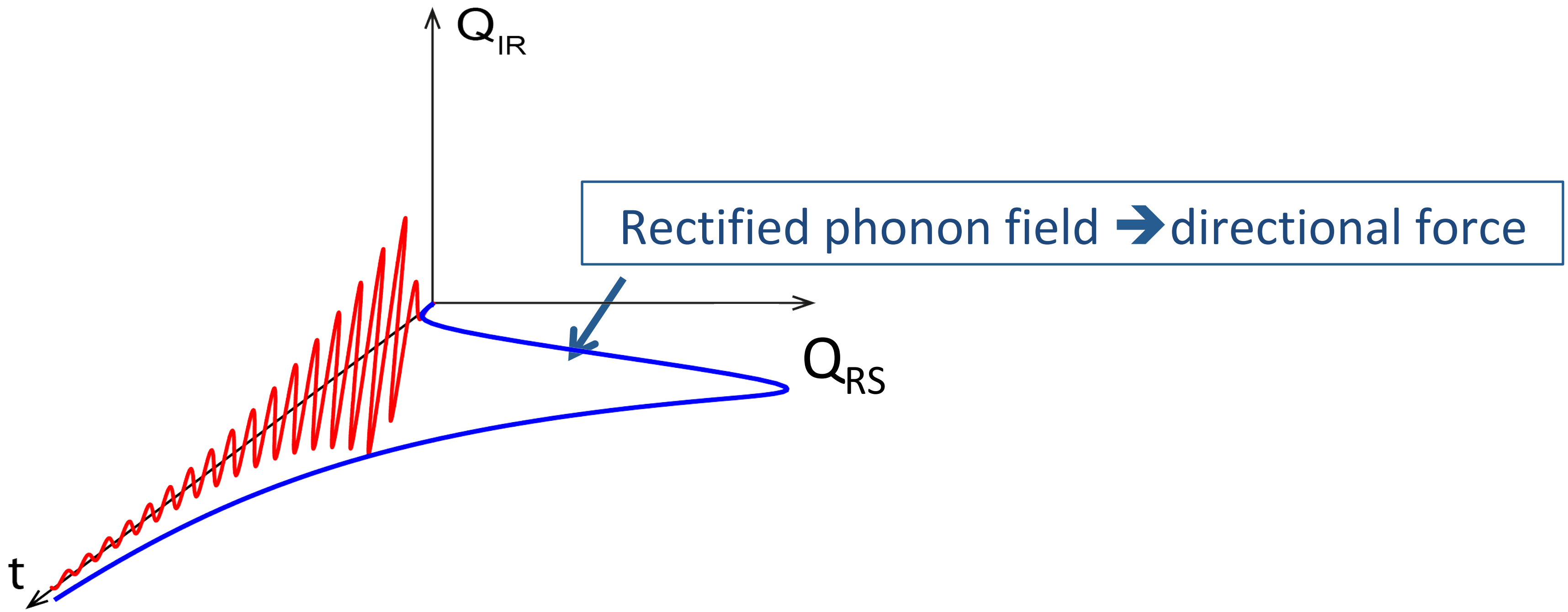
  N. Tancogne-Dejean et al., 1712.01067

I Resonant excitation of crystal lattice



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

Classical nonlinear phononics



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)

Classical nonlinear phononics

Explains a number of observed effects, e.g.,

- structurally induced metal-insulator transitions

  Rini et al., Nature 449, 72 (2007)

- phononic rectification in YBCO

  Mankowsky et al., Nature 516, 71 (2014)

- ferroelectric switching in $LiNbO_3$

  Subedi et al., Phys. Rev. B 89, 220301 (2014)

  Mankowsky et al., Phys. Rev. Lett. 118, 197601 (2017)

Classical mechanistic phonon dynamics **does not** explain all effects in IR-driven materials.

examples: - light-induced superconductivity

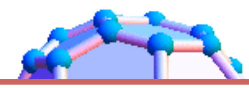
- light-enhanced el-ph coupling

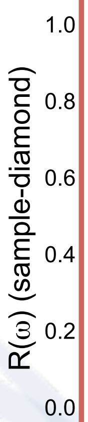
... quantum nature of phonons important?

Light-induced superconductivity?

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

Lattice control of reflectivity in K_3C_{60}





Not (easily) explained by classical nonlinear phononics

Hard problem!

Kennes, Millis, Knap, Demler, Murakami, Eckstein, Werner, Thorwart, Mazza, Georges, Fabrizio, Galitskii, Sentef, Kollath, ...

<u>Simpler question:</u> what happens to electron-phonon coupling under IR driving in a metal?

Dynamically enhanced coupling

mpsd

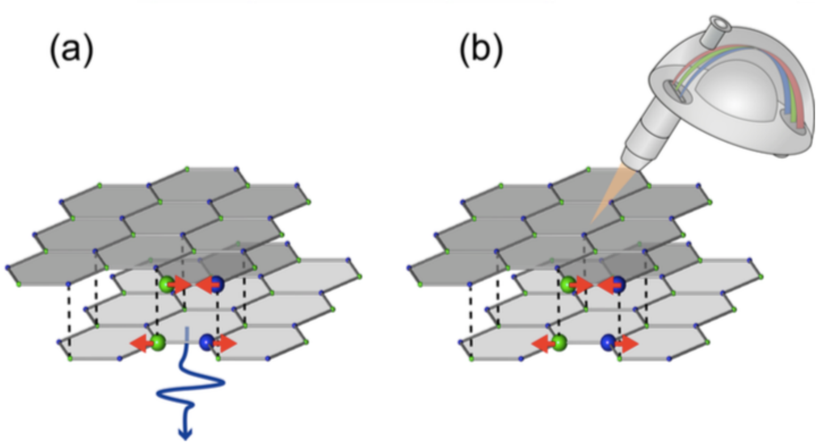
Enhanced electron-phonon coupling in graphene with periodically distorted lattice

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

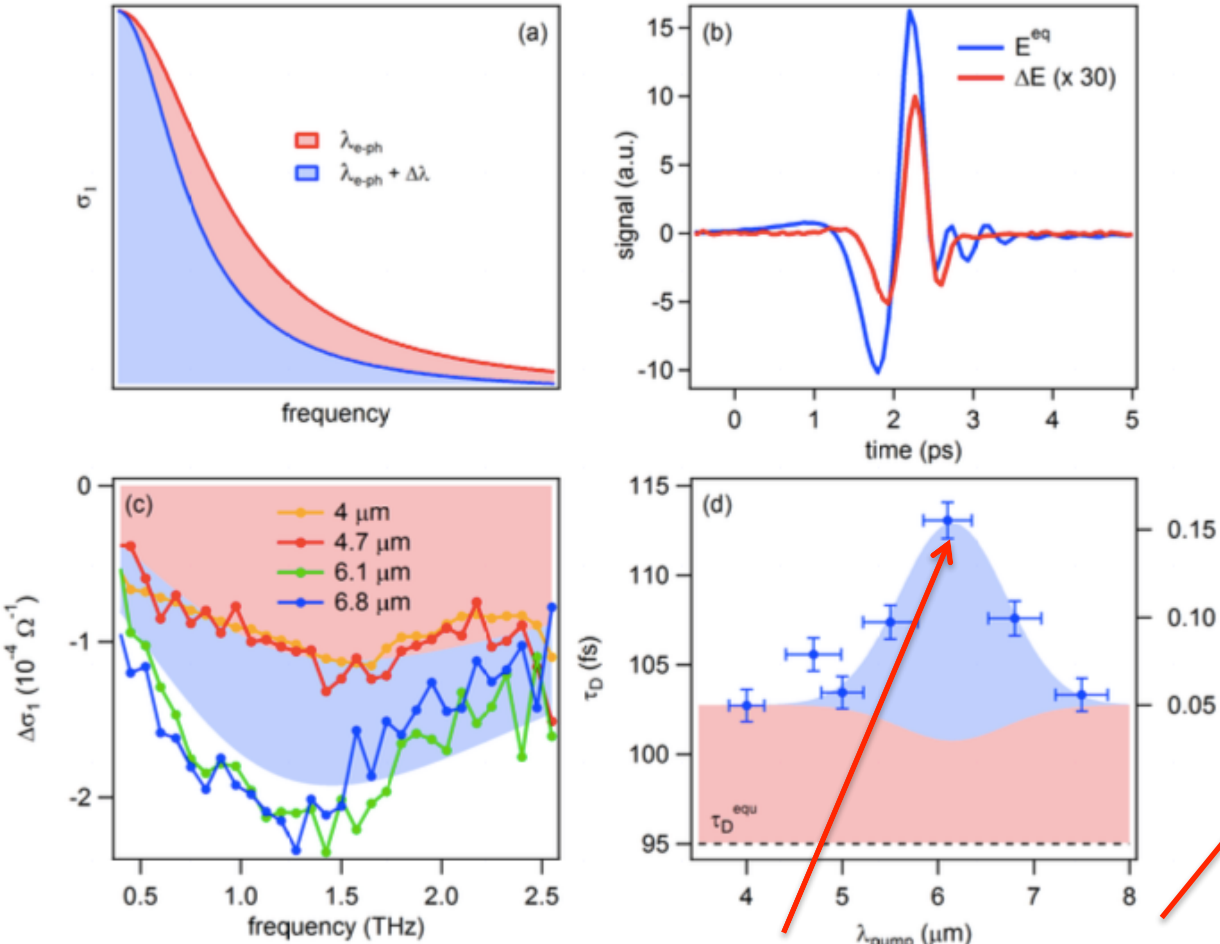


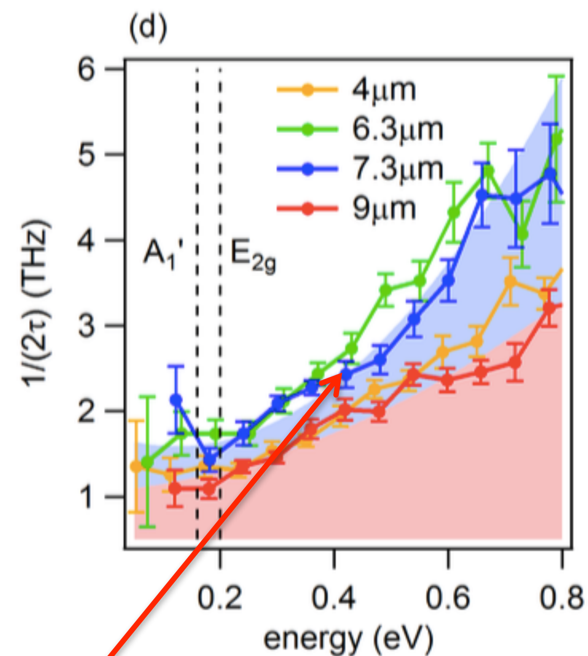
Dynamically enhanced coupling

PRB 95, 024304 (2017)

transient reduction of **THz Drude weight**

accelerated **tr-ARPES relaxation**





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

Quantum nonlinear phononics

mpsd

PRB 95, 205111 (2017)

2-site toy model, solve dynamics exactly

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

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

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

Idea: Drive nonlinearly coupled IR-phonon, analyze electronic response

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

Response: time-resolved spectral function

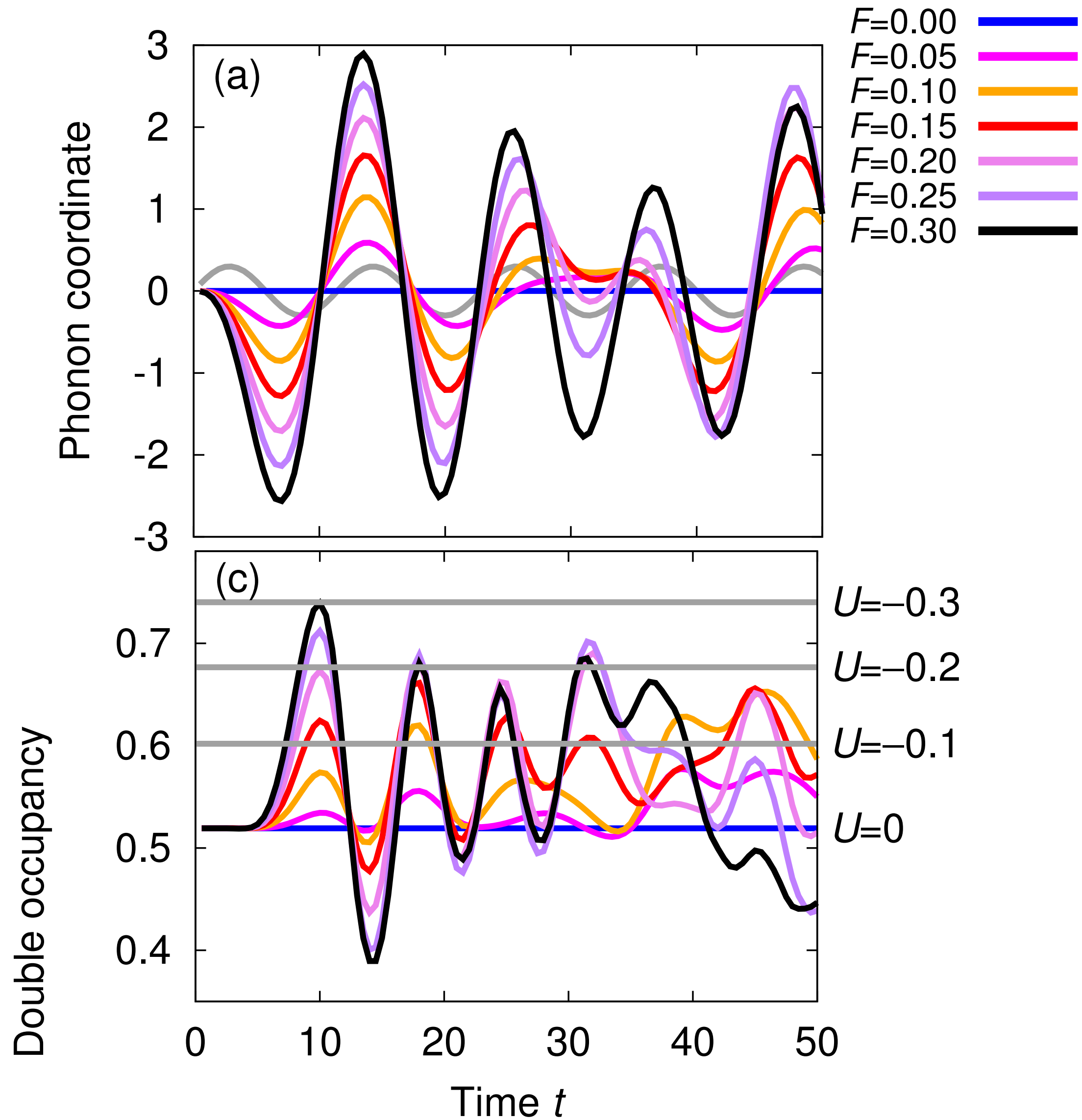
$$I(\omega, t_0) = \mathrm{Re}\int dt_1\, dt_2\, e^{i\omega(t_1-t_2)} s_{t_1,t_2,\tau}(t_0) \times \Big[\langle\psi(t_2)|c^{\dagger}_{1,\uparrow}\mathcal{T}e^{-i\int_{t_1}^{t_2} H(t)dt}c_{1,\uparrow}|\psi(t_1)\rangle + \langle\psi(t_1)|c_{1,\uparrow}\mathcal{T}e^{-i\int_{t_2}^{t_1} H(t)dt}c^{\dagger}_{1,\uparrow}|\psi(t_2)\rangle\Big],$$

IR-driven nonlinear el-ph system

Driving IR phonon with sinusoidal F(t):
coherent phonon oscillation

enhancement of local electronic double occupancy

-> induced el-el attraction



Time-resolved electronic spectrum

PRB 95, 205111 (2017) mpsd

Here: g2=−0.05 < 0

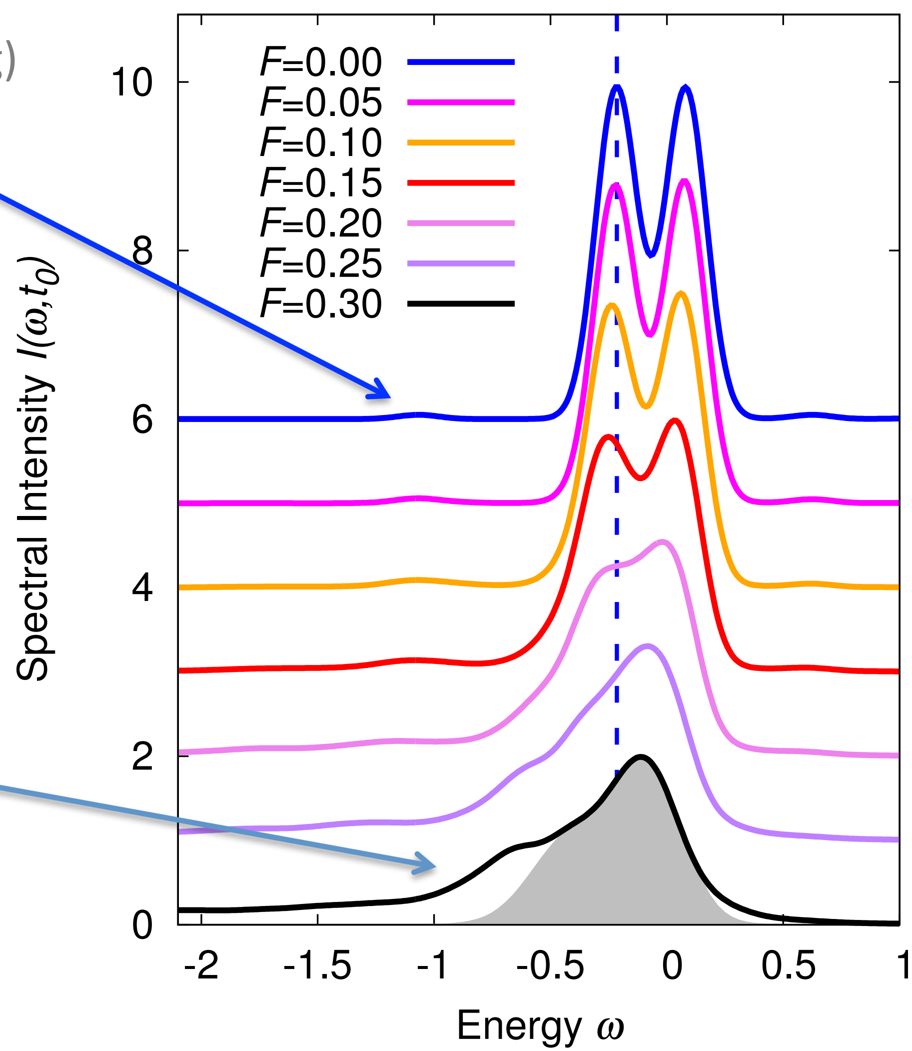
(does not matter for light-enhanced coupling)

2-phonon shakeoff

Reduced coherence peaks with stronger driving

light-enhanced el-ph coupling

light-induced polaron formation



Field scaling

PRB 95, 205111 (2017)

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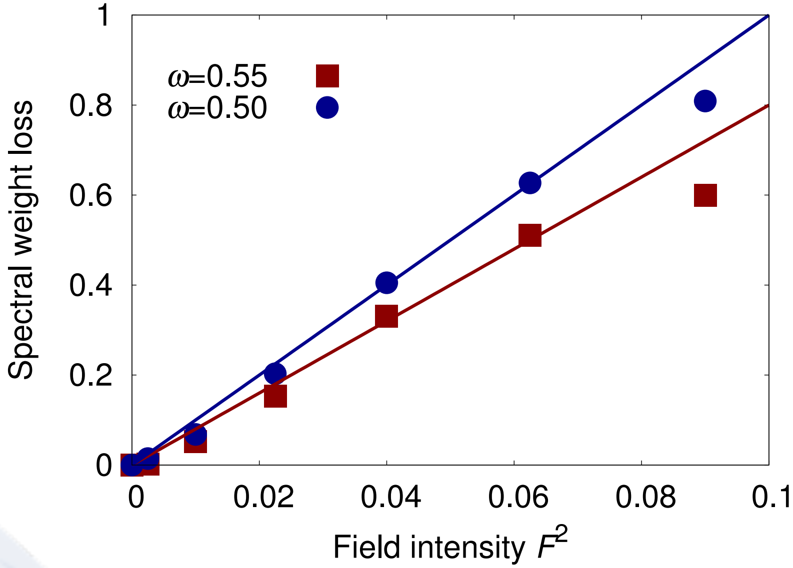
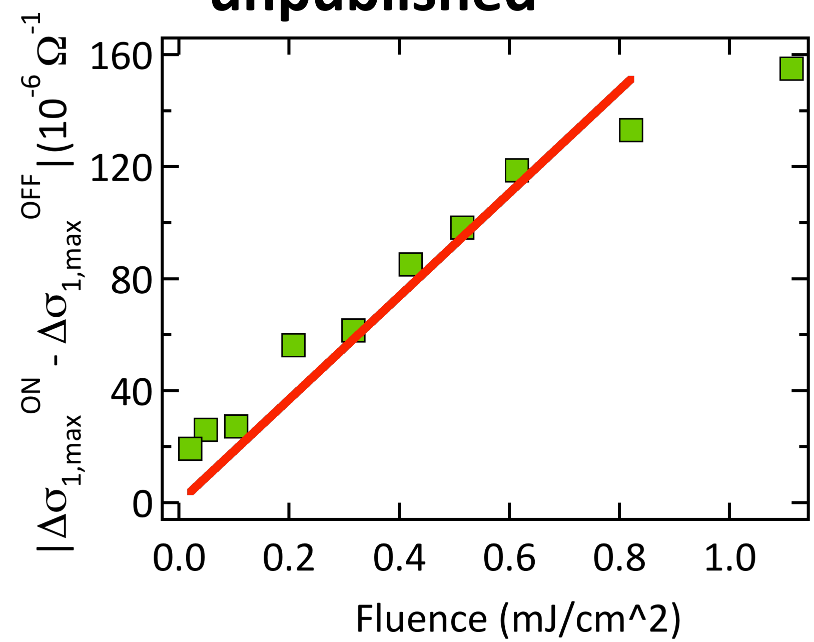
Theory



Data by E. Pomarico, unpublished



Coherence peak weight loss: proportional to field intensity F^2 consistent with experiments

Quantum nonlinear phononics

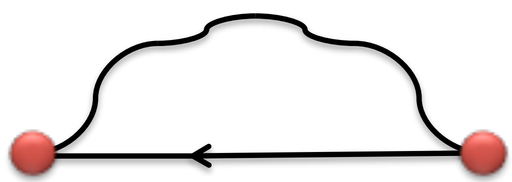
Forced coherent oscillation $\langle \hat{x}_l(t) \rangle \propto F \sin(\omega t)$

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

nonlinear

~F ~F

$\Sigma =$



g(t) g(t)

Migdal-Eliashberg diagram

effective induced linear coupling

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

time-dependent vertex, amplitude g^2 ~ F^2

=> light-induced coupling, lambda scales ~ F^2

Summary I

- enhanced electron-phonon coupling in phononically driven bilayer graphene

PRB 95, 024304 (2017)

E. Pomarico I. Gierz A. Cavalleri

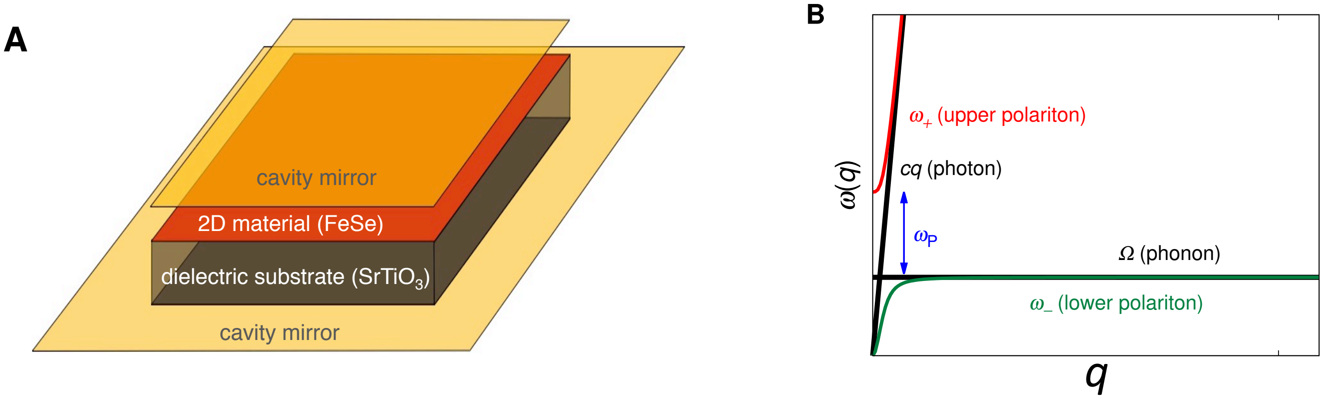
Exact solution of electron-phonon model system:

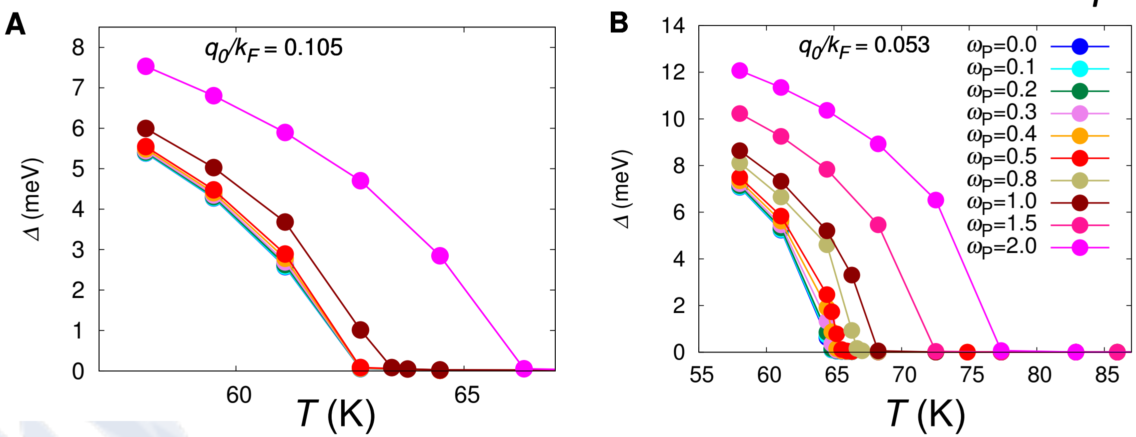
- theoretical proposal: nonlinear el-ph coupling as mechanism behind this enhancement

PRB 95, 205111 (2017)

Cavity QED superconductivity

arXiv:1802.09437





Materials engineering in nanocavities through coupling to quantum light

M. Ruggenthaler A. Rubio

Flavors of phononics

mpsd

light	lattice	coupling	
classical	classical	nonlinear	*Först et al., Nat. Phys. 7, 854 (2011)*

Classical nonlinear phononics: directional forces, structural transitions

classical	quantum	nonli	*...at. Phys. 13, 479 (2017)* *...95, 205111 (2017)*

Quantum nonlinear ... ...ced coupling, light-induced superconductivity?

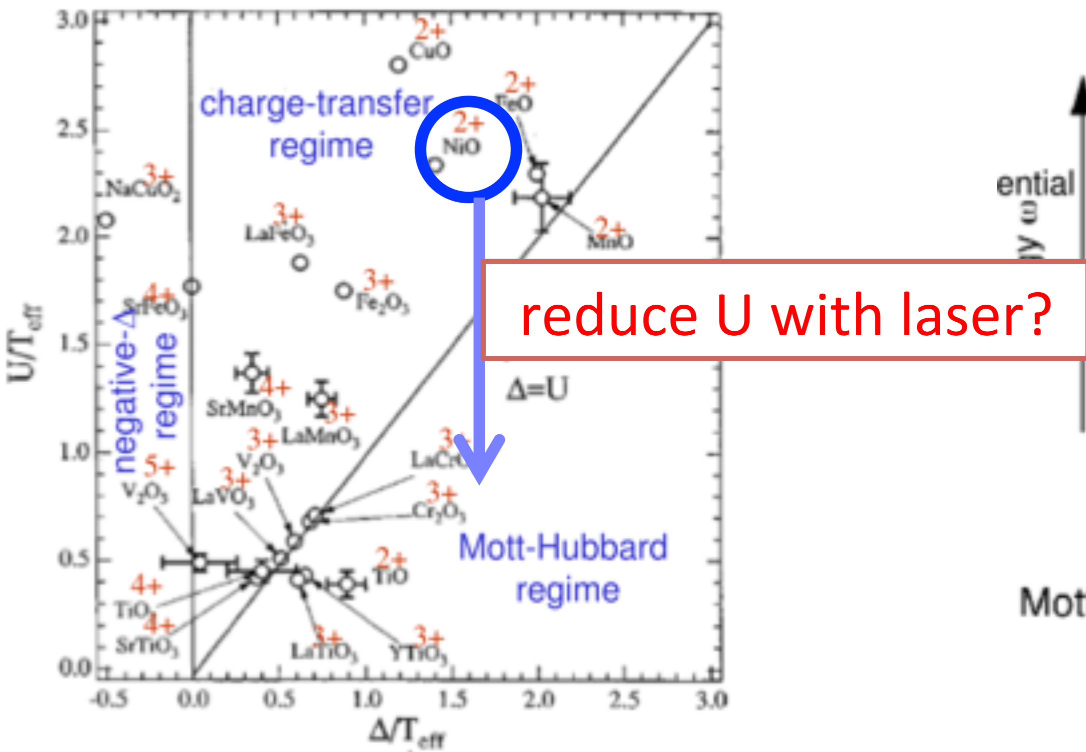
quan	quantum	linear	*arXiv:1802.09437*

Cavity QED phononics: light-enhanced coupling & superconductivity

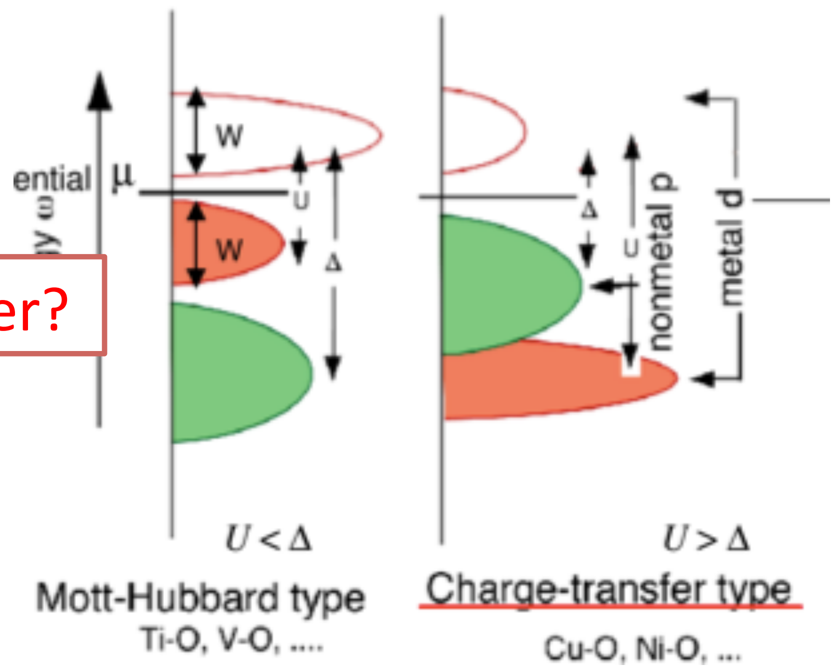
Path towards nonequilibrium materials engineering

II Dynamical modification of Hubbard U

Can we drive a charge-transfer insulator towards a Mott insulator?



Zaanen-Sawatzky-Allen phase diagram



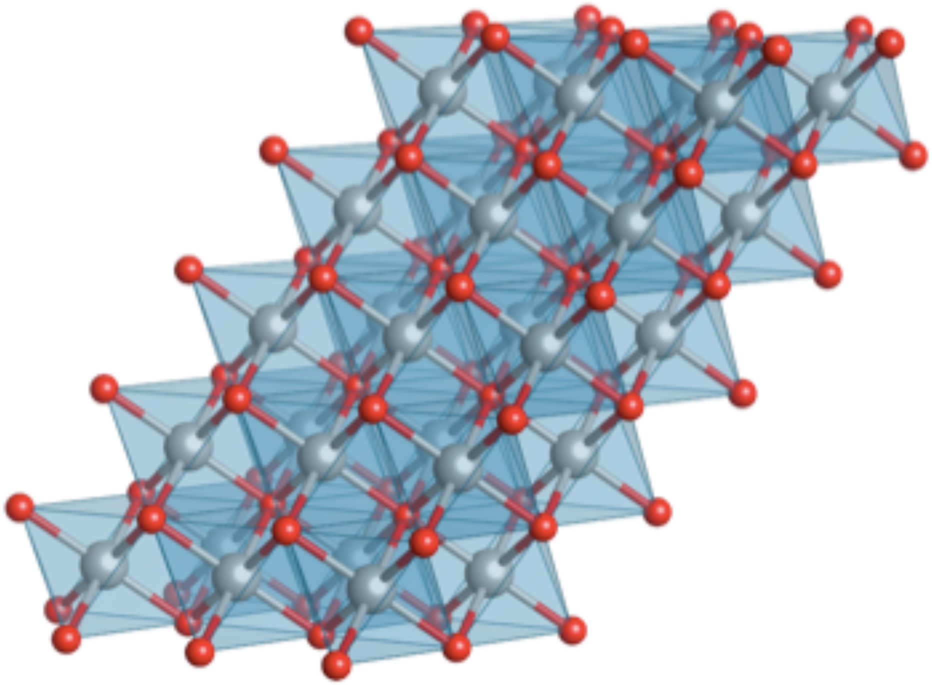
NiO as prototypical charge-transfer insulator

NiO:

Antiferromagnetic type 2

Band gap: ~4 eV (exp.)

Néel temperature: 523K



Time-dependent U with TDDFT+U

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DFT with ab initio and self-consistent Hubbard U (Hybrid functional)

$$E_{\mathrm{DFT+U}}[n, \{n^{I,\sigma}_{mm'}\}] = E_{\mathrm{DFT}}[n] + E_{ee}[\{n^{I,\sigma}_{mm'}\}] - E_{dc}[\{n^{I,\sigma}_{mm'}\}]$$

Electron-electron interaction — Double counting

$$E_{ee} \approx \frac{\bar{U}}{2} \sum_{\{m\},\sigma} N^{\sigma}_{m} N^{-\sigma}_{m'} + \frac{\bar{U}-\bar{J}}{2} \sum_{m \neq m',\sigma} N^{\sigma}_{m} N^{\sigma}_{m'}.$$

Usual expression in DFT+U

occupations — Coulomb integrals

$$E_{ee} = \frac{1}{2} \sum_{\{m\}} \sum_{\alpha,\beta} \bar{P}^{\alpha}_{mm'} \bar{P}^{\beta}_{m''m'''} (mm'|m''m''') - \frac{1}{2} \sum_{\{m\}} \sum_{\alpha} \bar{P}^{\alpha}_{mm'} \bar{P}^{\alpha}_{m''m'''} (mm'''|m''m'),$$

ACBN0 functional
PRX 5,011006 (2015)

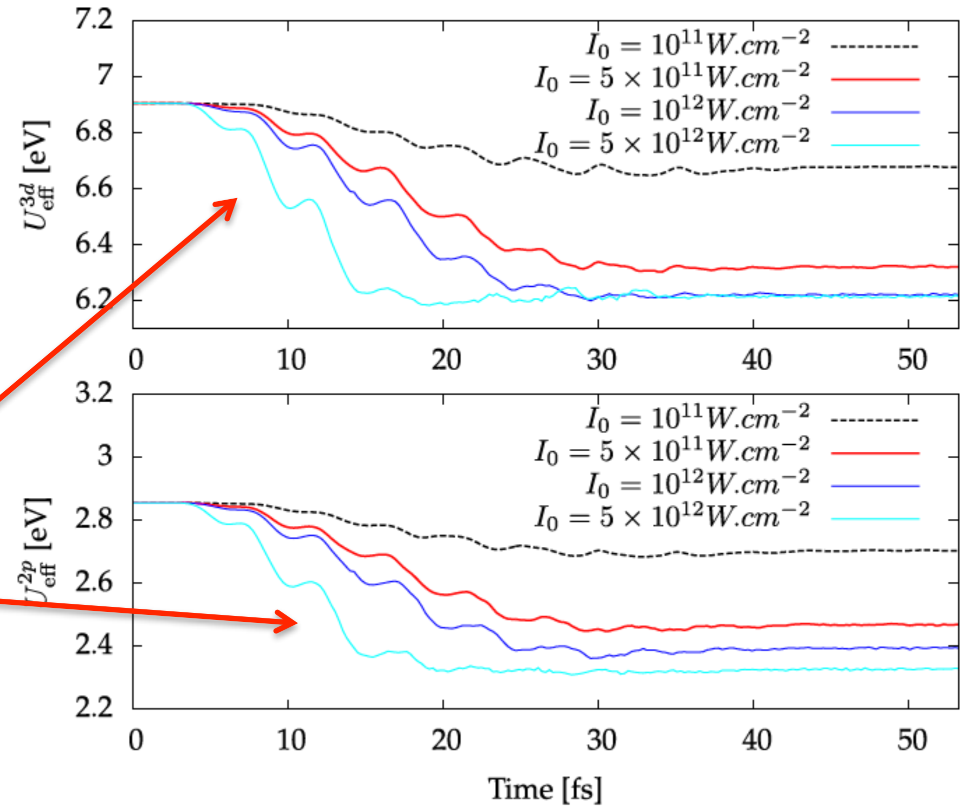
- alternative to constrained RPA
- numerically efficient
- direct extension to time-dependent case (adiabatic approximation)

Ultrafast modification of Hubbard U in NiO

strong subresonant (0.43 eV) laser excitation:
 -> high field strength without damage

U reduces during the 25 fs laser pulse

Stronger decrease for stronger field strength



Typical intensities in strong field physics in solids

Reduction of U: mechanism

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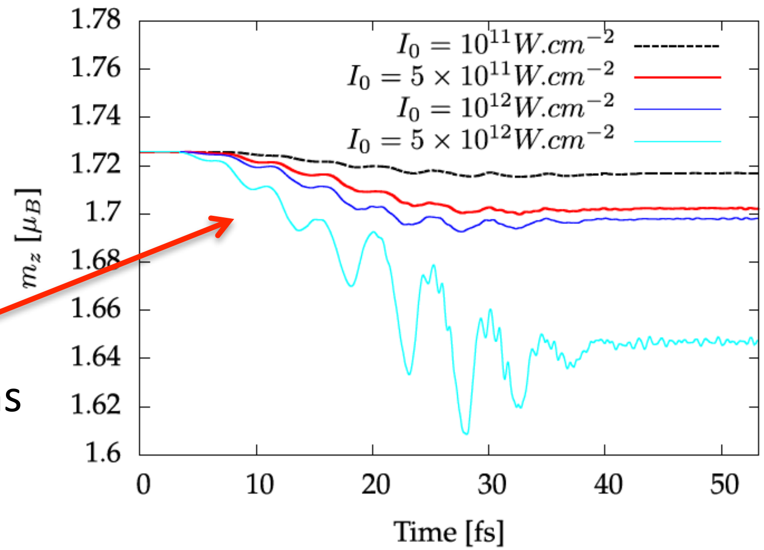
U measures the Coulomb interaction screened by itinerant electrons

Laser excites electrons from occupied localized orbitals (3d of Ni and 2p of O)

Partial demagnetization of Ni atoms



- Polarization of itinerant electrons increases
- Enhanced screening
- Decrease of U

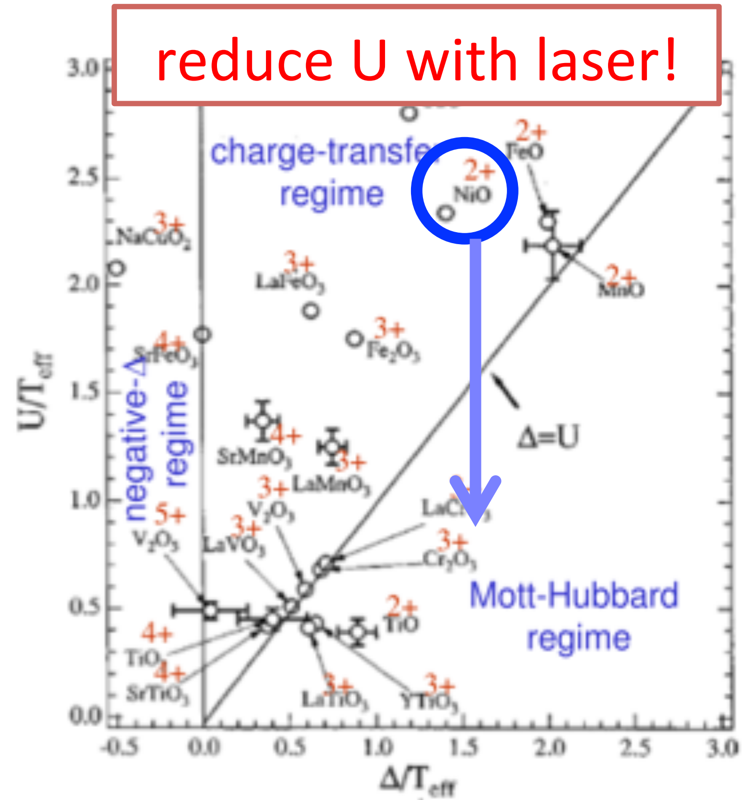
Summary II

- Ultrafast reduction of Hubbard U in NiO via induced extra screening
- Towards light-induced Mott insulators?

N. Tancogne-Dejean et al., 1712.01067

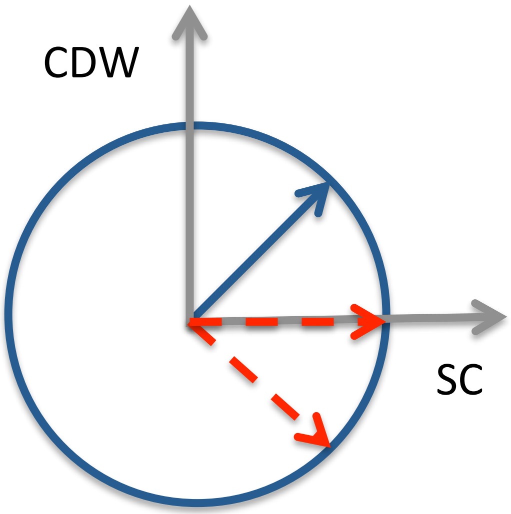
N. Tancogne-Dejean

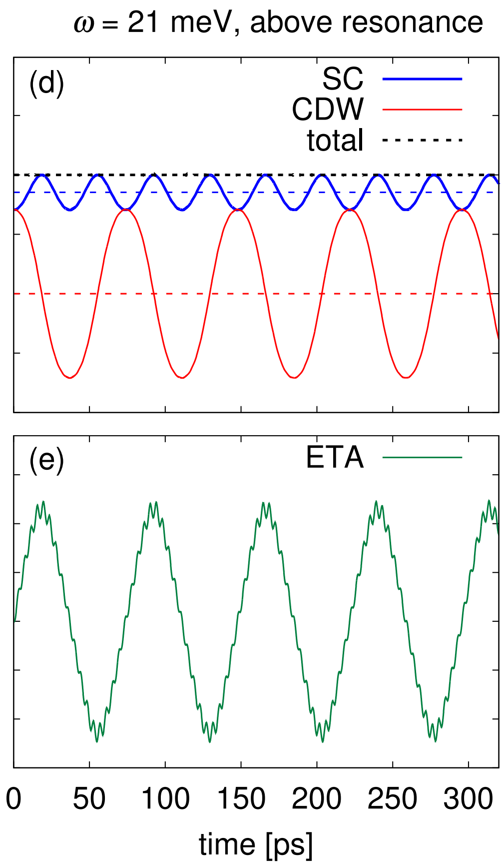
A. Rubio



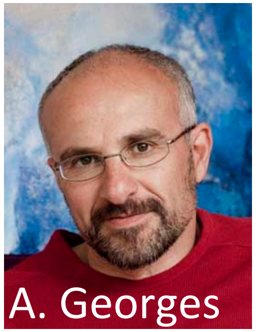
Laser-controlled competing orders

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



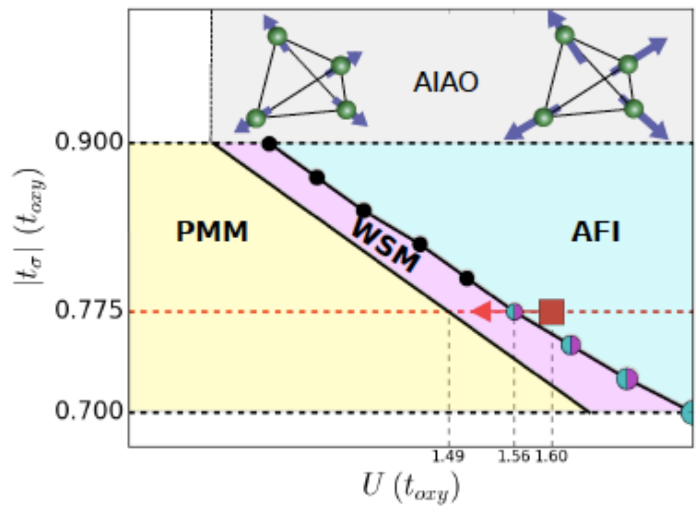


Controlling competing orders by driving near gap resonance



Nonthermal magnetic Weyl semimetal

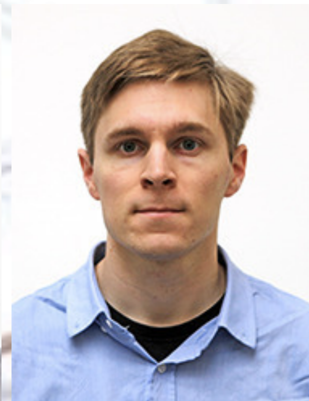
in preparation



Nonthermal pathway to magnetic Weyl semimetal in pyrochlore iridates

Acknowledgments


G. Topp


R. Tuovinen


N. Tancogne-Dejean


M. Ruggenthaler


A. Rubio

J. K. Freericks – Georgetown
T. P. Devereaux – Stanford
A. Georges – Paris/Flatiron NYC
C. Kollath – Bonn
... and many more


A. Kemper


E. Pomarico


I. Gierz


A. Cavalleri

Emmy Noether-Programm
Deutsche Forschungsgemeinschaft DFG

DFG

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Funded through Deutsche Forschungsgemeinschaft
Emmy Noether Programme (SE 2558/2-1)

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Summary

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- band structure, topology (Floquet)

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

- electron-phonon coupling (quantum nonlinear phononics)

  PRB 95, 024304 (2017)
  PRB 95, 205…

- … subresonant …tions in correlated insulators)

  arXiv:1712.01067

- superconductivity

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  PRB 93, 144506 (2016)
  PRL 118, 087002 (2017)
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Path towards nonequilibrium materials engineering