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

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DFG

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Emmy Noether Programme (SE 2558/2-1)

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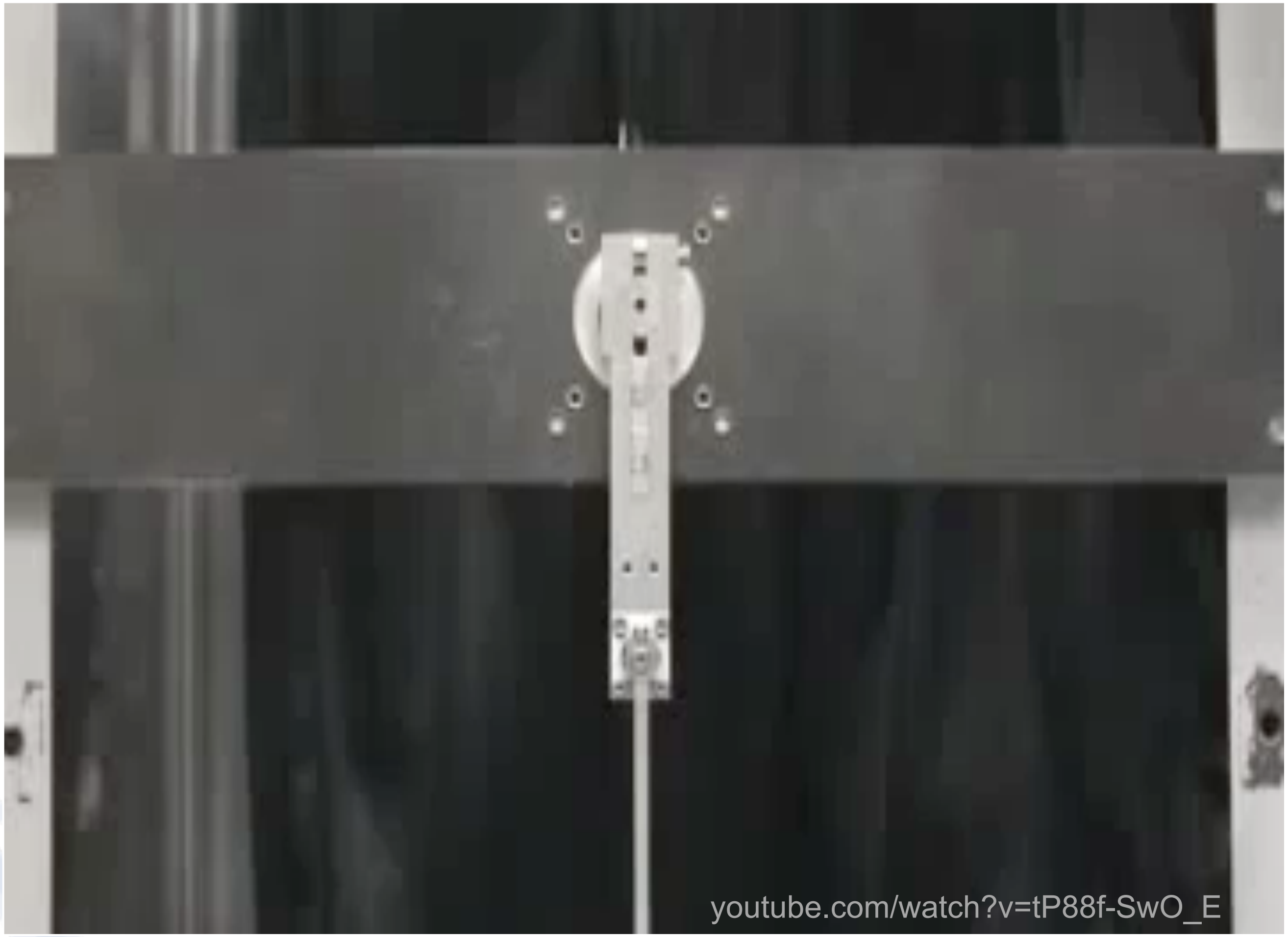
Driven is different

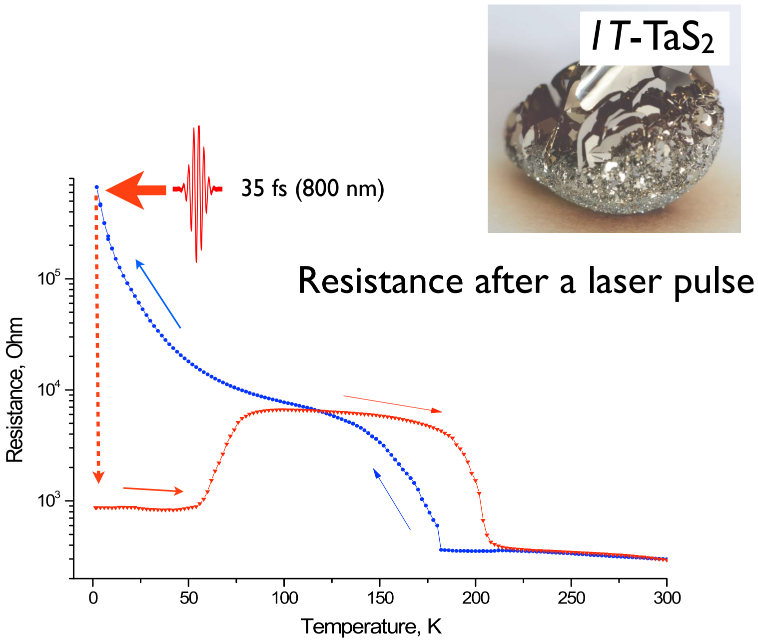
Kapitza pendulum



dynamical stabilization of a metastable state

Is driven also useful?

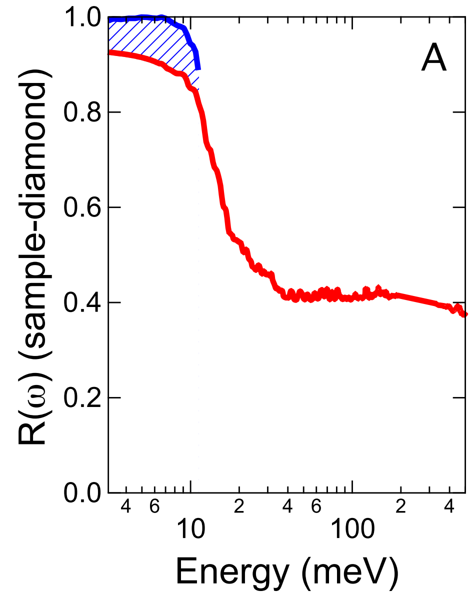
Exposing hidden states



L Stojchevska et al. Science 2014;344:177-180

... and many more.

Light-induced new states?



Possible light-induced superconductivity in K_3C_{60} at high temperature

M. Mitrano, A. Cantaluppi, D. Nicoletti, S. Kaiser, A. Perucchi, S. Lupi, P. Di Pietro, D. Pontiroli, M. Riccò, S. R. Clark, D. Jaksch & A. Cavalleri

Affiliations | Contributions | Corresponding author

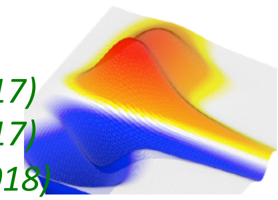
Nature **530**, 461–464 (25 February 2016) | doi:10.1038/nature16522

Ultrafast Materials Science today

Understanding the nature of quasiparticles

- Relaxation dynamics
- Control of couplings

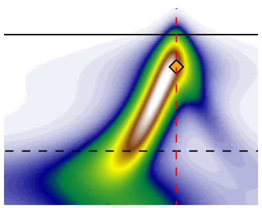
PRL 111, 077401 (2013)
PRX 3, 041033 (2013)
PRB 87, 235139 (2013)
PRB 90, 075126 (2014)
Nat. Comm. 7, 13761 (2016)
PRB 95, 024304 (2017)
PRB 95, 205111 (2017)
PRL 121, 097402 (2018)
arXiv:1802.09437
arXiv:1808.02389



Understanding ordered phases

- Collective oscillations
- Competing orders

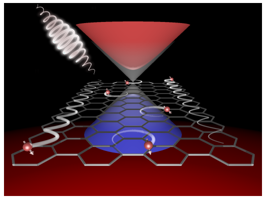
PRB 92, 224517 (2015)
PRB 93, 144506 (2016)
PRL 118, 087002 (2017)
arXiv:1806.08187
arXiv:1808.00712
arXiv:1808.04655



Creating new states of matter

- Floquet topological states

Nature Comm. 6, 7047 (2015)
Nature Comm. 8, 13940 (2017)
arXiv:1803.07447



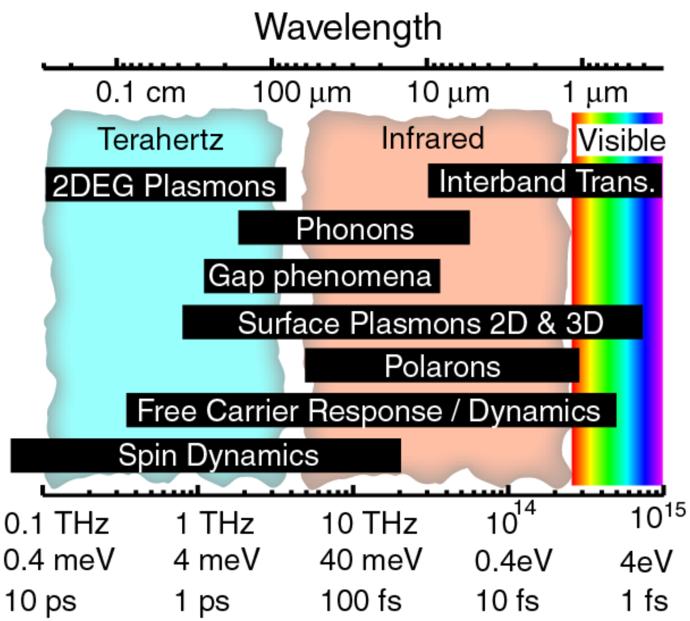


Image courtesy: D. Basov

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)

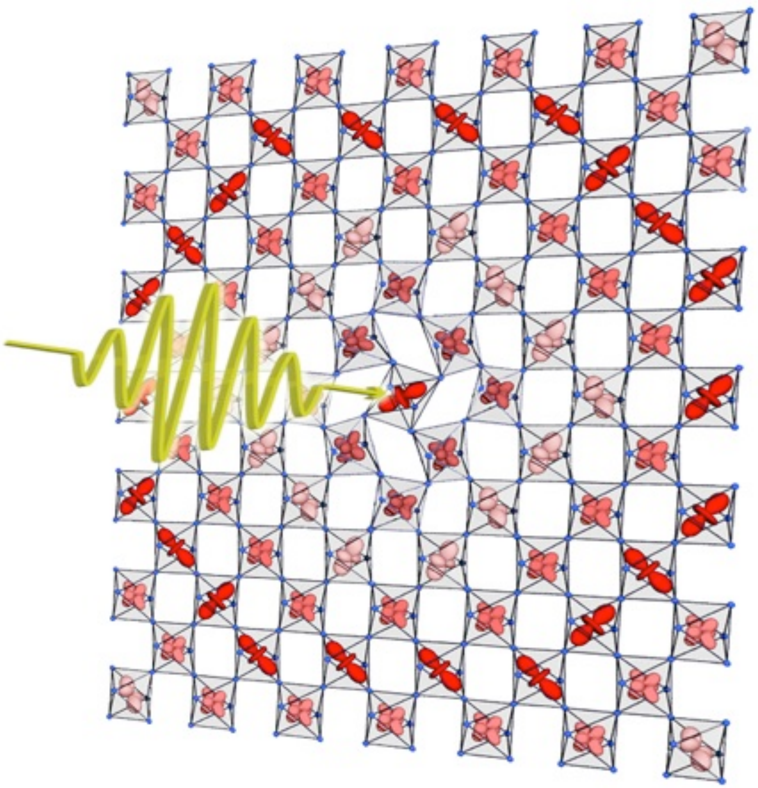
  M. A. Sentef, PRB 95, 205111 (2017) – theory

- Part II: Light-reduced Hubbard U

  Nonresonant laser driving reduces Hubbard U in NiO

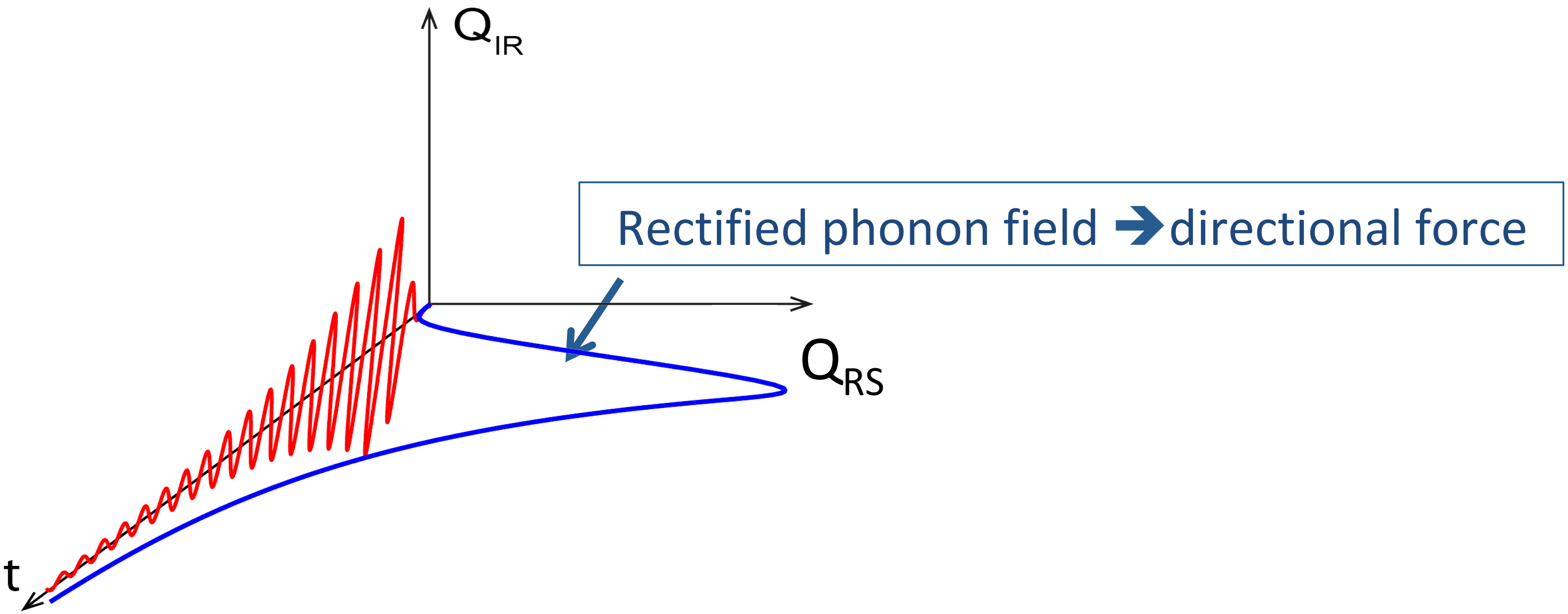
  N. Tancogne-Dejean et al., PRL 121, 097402 (2018)

I Resonant excitation of crystal lattice



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

Classical nonlinear phononics



Simplest model: classical dynamics

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

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

„nonlinear phononics“

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

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

Classical nonlinear phononics

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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 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?

Dynamically enhanced coupling

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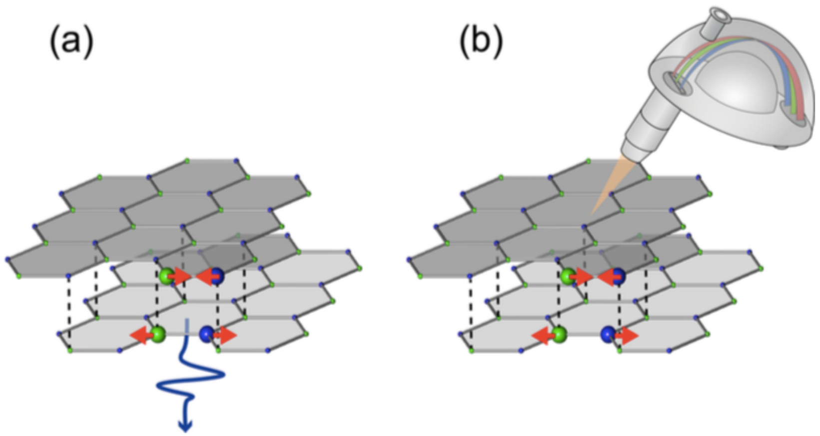
Enhanced electron-phonon coupling in graphene with periodically distorted lattice

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 coupling for pump on resonance with IR phonon

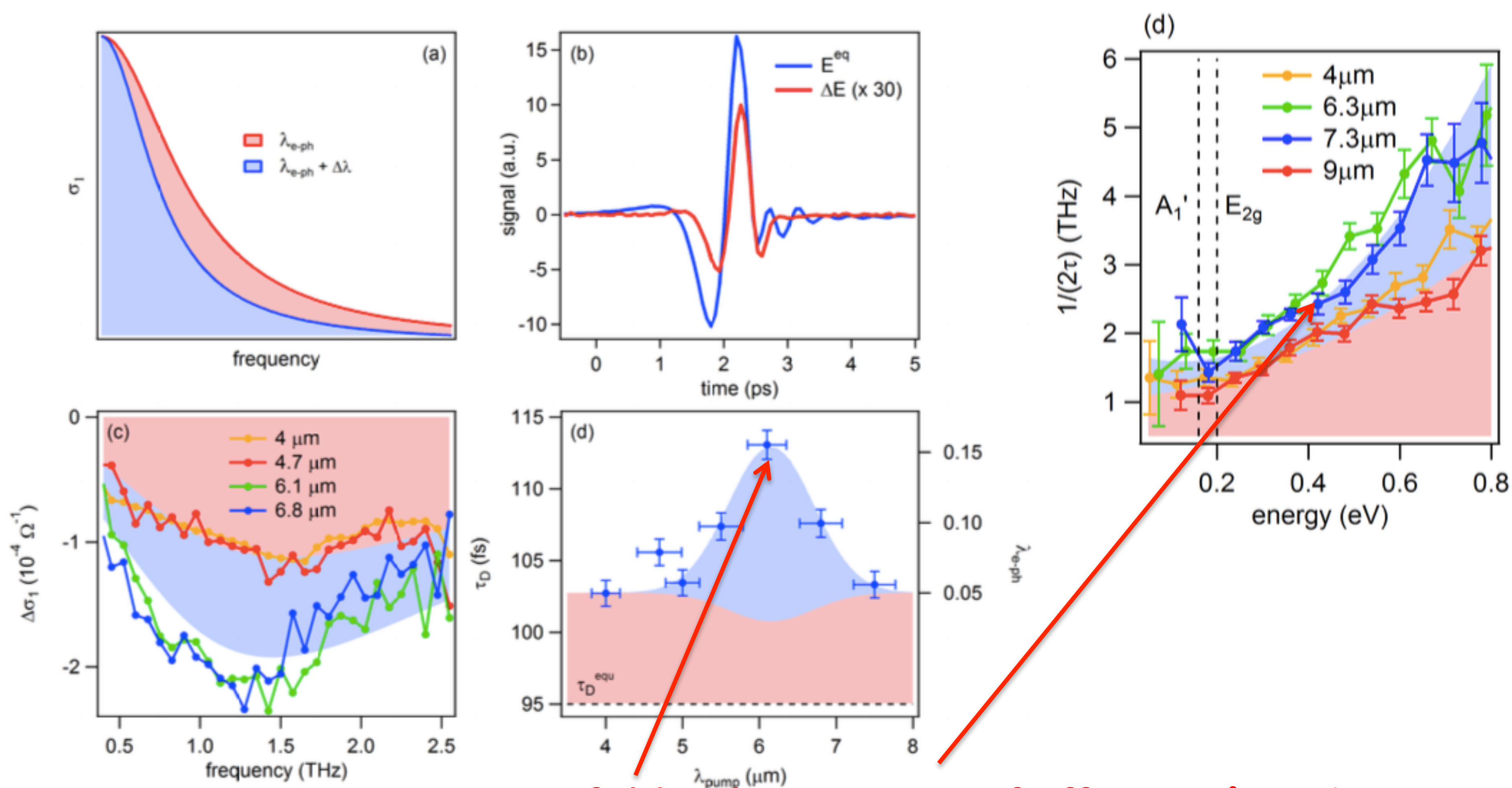


Dynamically enhanced coupling

PRB 95, 024304 (2017)

transient reduction of **THz Drude weight**

accelerated **tr-ARPES relaxation**



3-fold enhancement of effective λ_{el-ph}!

Quantum nonlinear phononics

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)

density-dependent
squeezing of phonon

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

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

Response: time-resolved spectral function

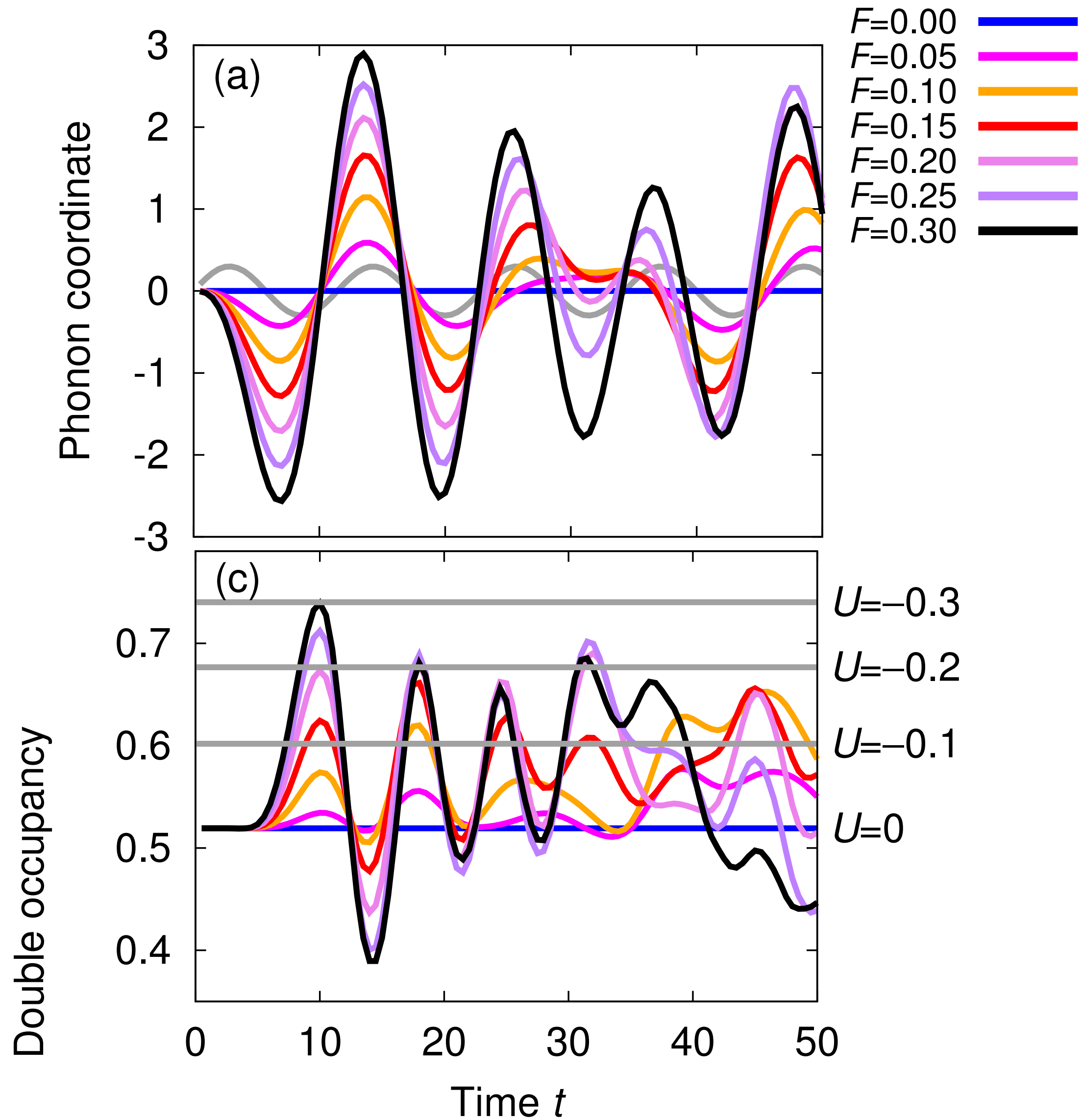
$$I(\omega, t_0) = \mathrm{Re}\int dt_1\, dt_2\, e^{i\omega(t_1-t_2)} s_{t_1,t_2,\tau}(t_0)$$
$$\times \Big[\langle\psi(t_2)|c^{\dagger}_{1,\uparrow}\mathcal{T}e^{-i\int_{t_1}^{t_2}H(t)dt}c_{1,\uparrow}|\psi(t_1)\rangle +$$
$$+ \langle\psi(t_1)|c_{1,\uparrow}\mathcal{T}e^{-i\int_{t_2}^{t_1}H(t)dt}c^{\dagger}_{1,\uparrow}|\psi(t_2)\rangle\Big],$$

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

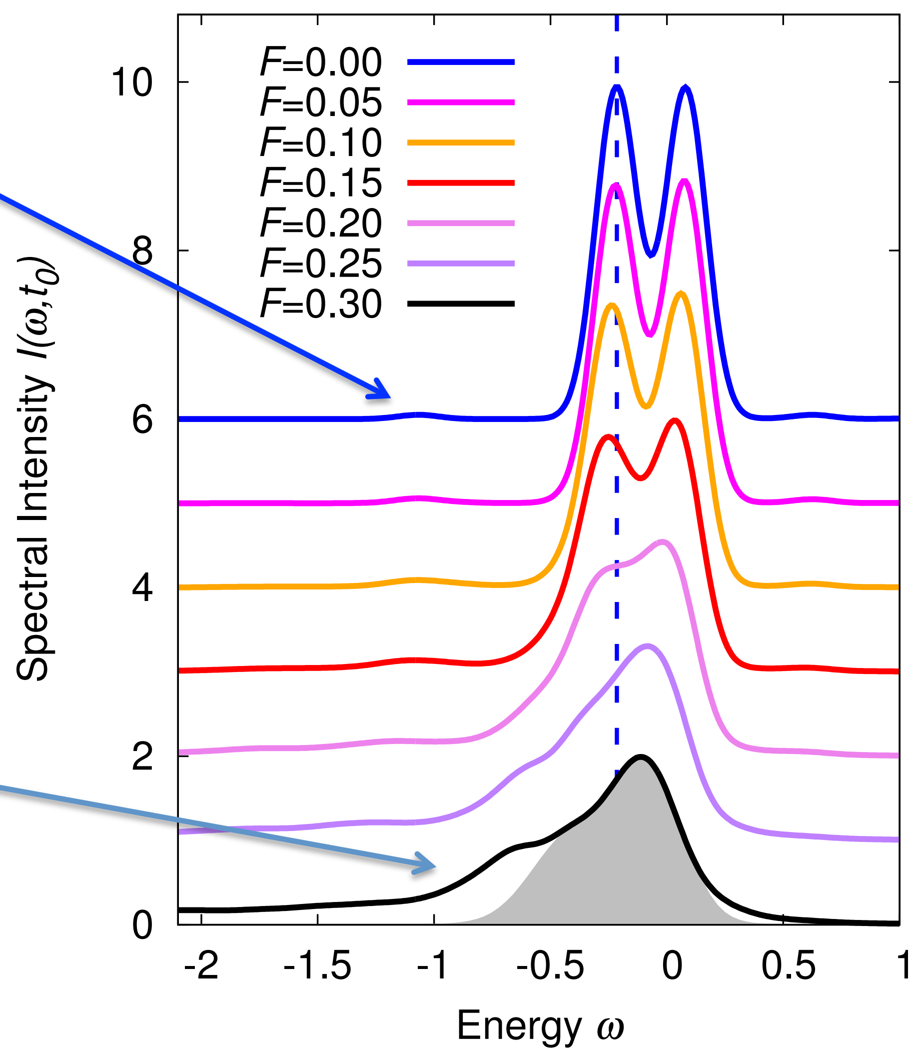
PRB 95, 205111 (2017)

Reduced coherence peaks with stronger driving

light-enhanced el-ph coupling

2-phonon shakeoff

light-induced polaron formation



Field dependence

PRB 95, 205111 (2017)

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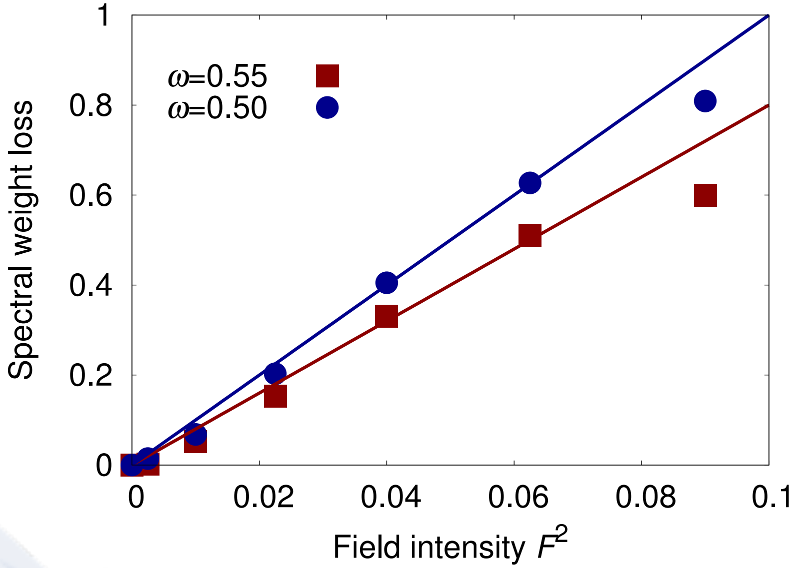
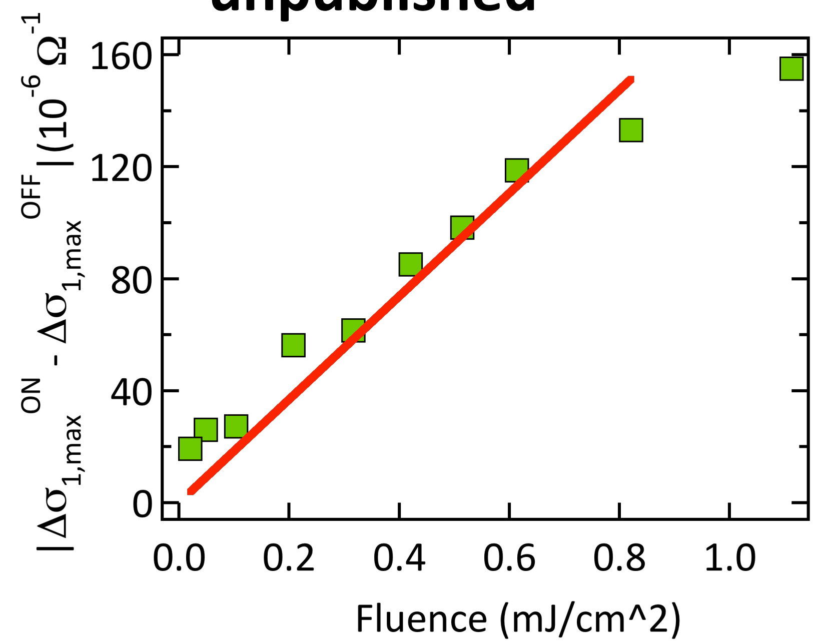
Theory



Data by E. Pomarico, unpublished



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

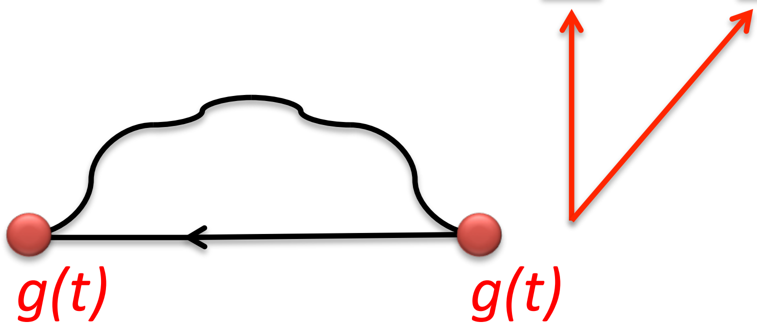
Quantum nonlinear phononics

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

$\Sigma =$



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)

II Dynamical modification of Hubbard U

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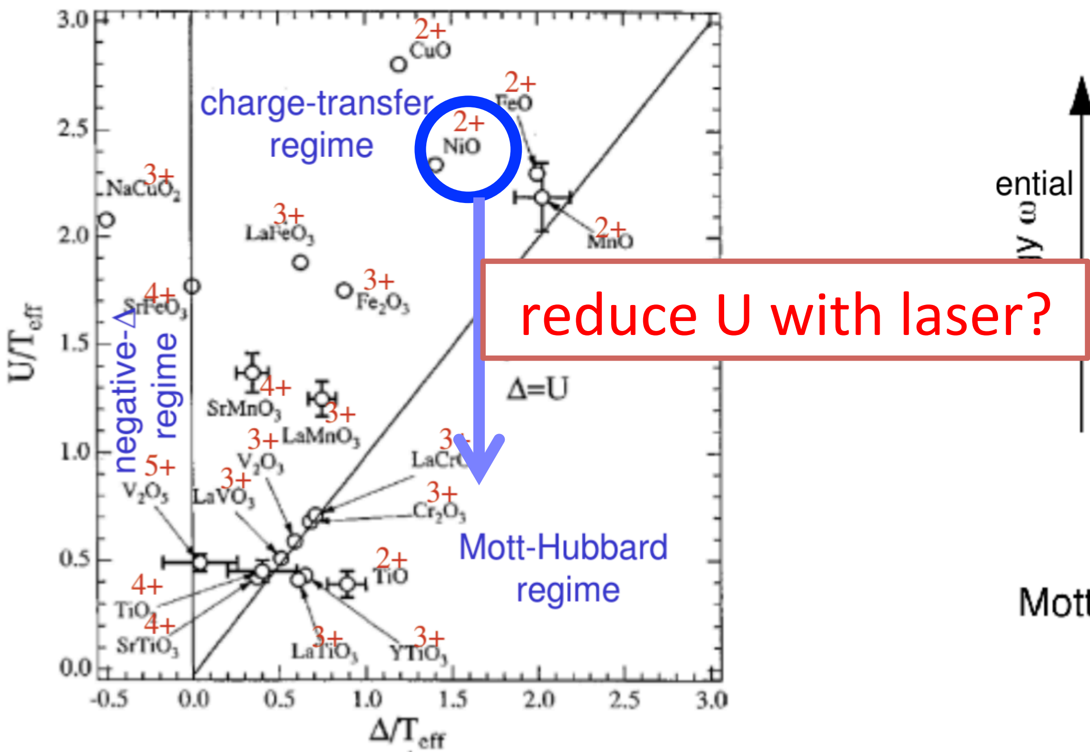
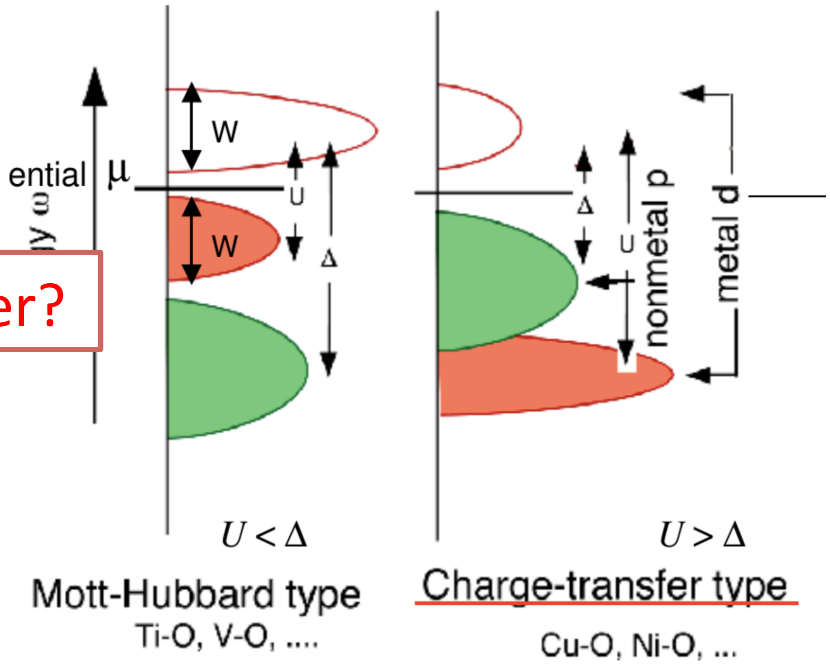
Can we drive a charge-transfer insulator towards a Mott insulator?



Zaanen-Sawatzky-Allen phase diagram



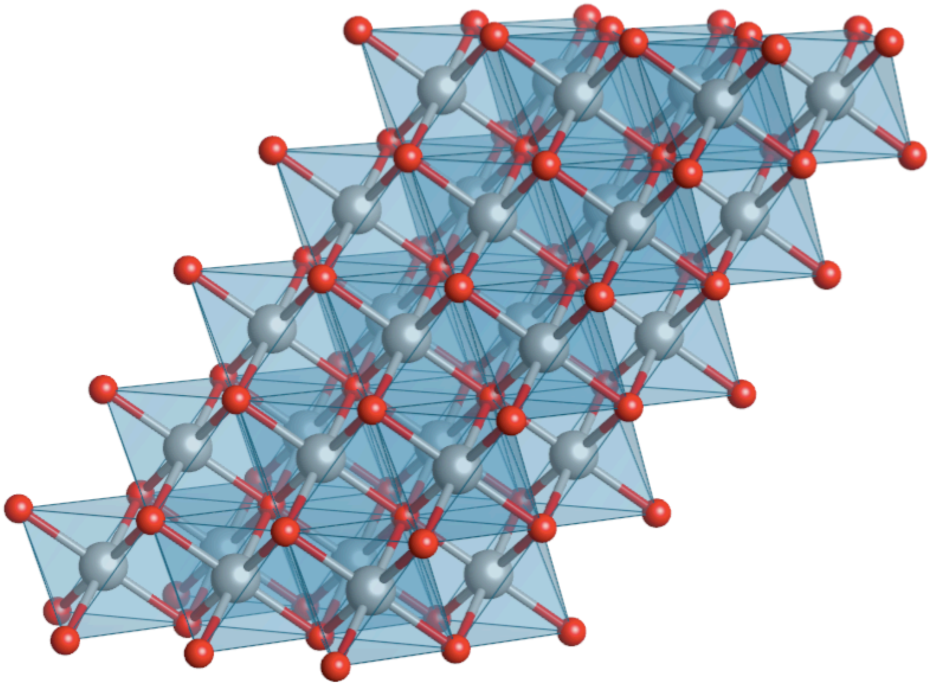
NiO as prototypical charge-transfer insulator

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 (E_{ee}) — Double counting (E_{dc})

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

Usual expression in DFT+U

↔

occupations — Coulomb integrals

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

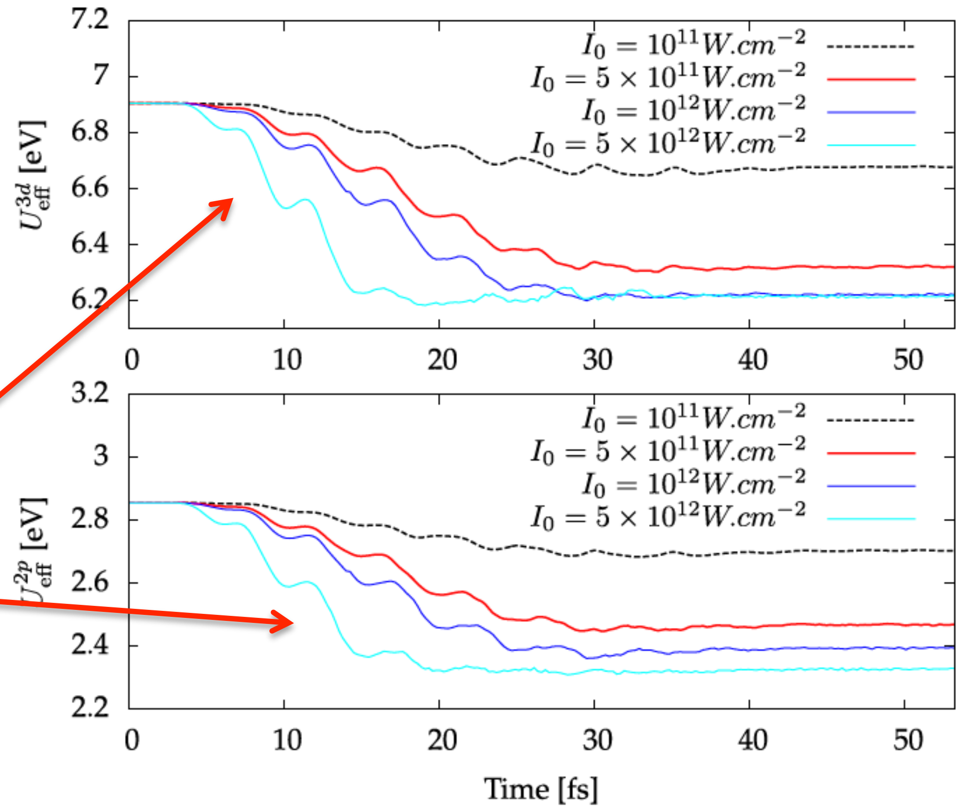
ACBN0 functional
PRX 5,011006 (2015)

- 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 reduced during the 25 fs laser pulse



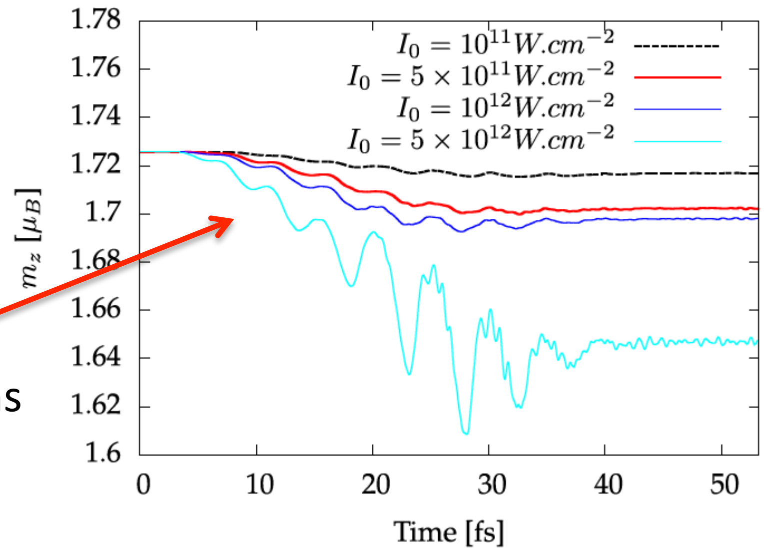
Typical intensities in strong field physics in solids

Reduction of U: mechanism

U measures the Coulomb interaction screened by itinerant electrons

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

Partial demagnetization of Ni atoms



- Light-enhanced screening
- Decrease of U

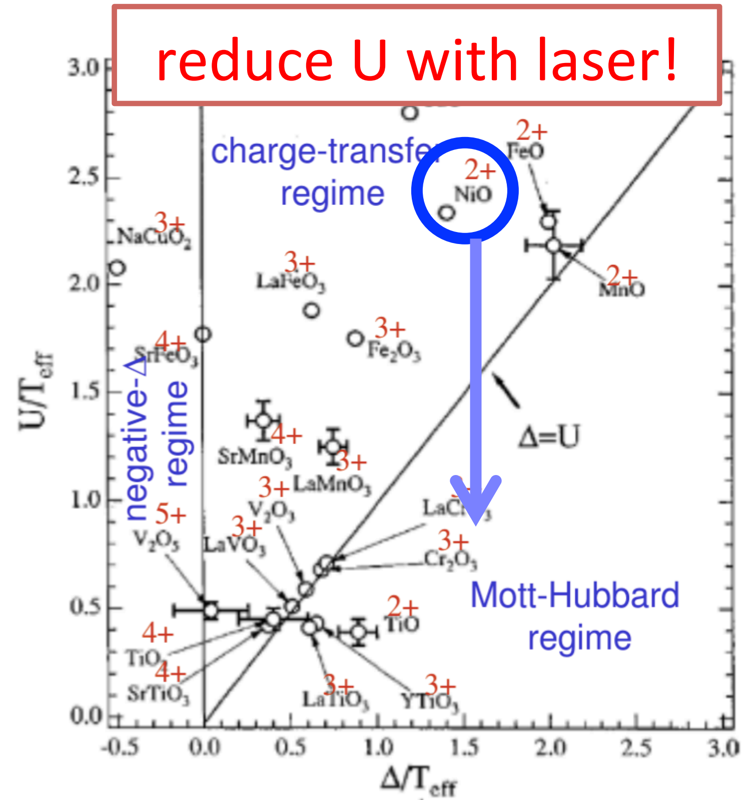
Summary II

- Ultrafast reduction of Hubbard U in NiO via induced extra screening

N. Tancogne-Dejean et al., PRL 121, 097402 (2018)

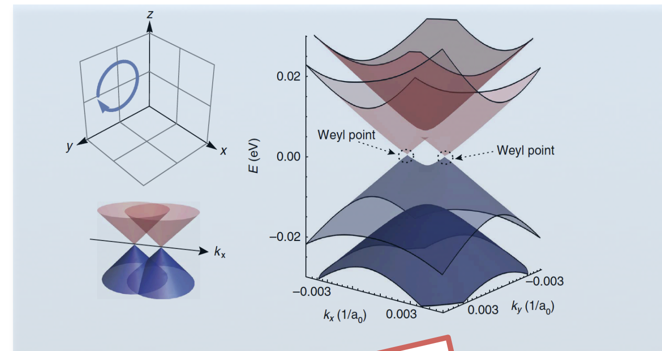
N. Tancogne-Dejean

A. Rubio

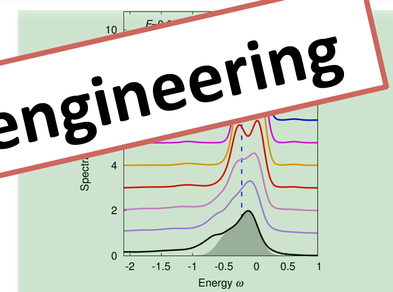


Summary

Ultrafast laser engineering of

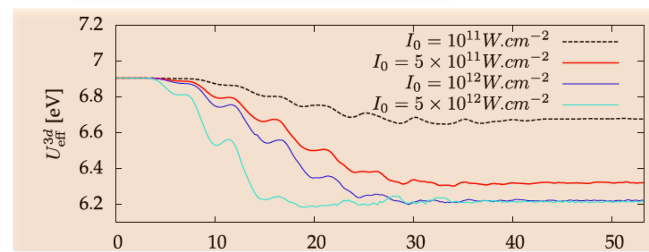


- band structure, topology (Floquet)
  Nature Commun. 6, 7047 (2015)
  Nature Commun. 8, 13940 (2017)
  arXiv:1803.07447
- electron-phonon coupling
  PRB 95, 024304 (2017)
  PRB 95, 205111 (2017)
  arXiv:1802.09437

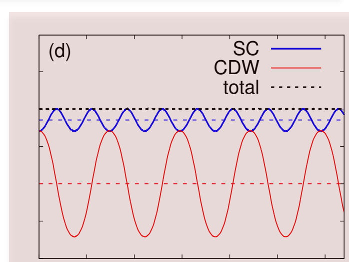


Path towards nonequilibrium materials engineering

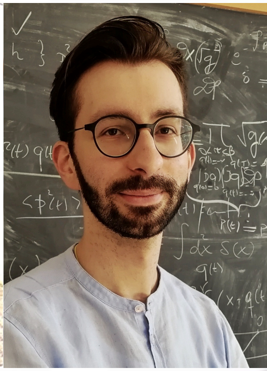
- H… …g subresonant …tions in correlated insulators)
  PRL 121, 097402 (2018)



- ordered phases
  PRB 92, 224517 (2015) *arXiv:1806.08187*
  PRB 93, 144506 (2016) *arXiv:1808.00712*
  PRL 118, 087002 (2017) *arXiv:1808.04655*



Team



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