Light-induced changes of couplings in materials: Enhanced electron-phonon coupling, reduced Hubbard U

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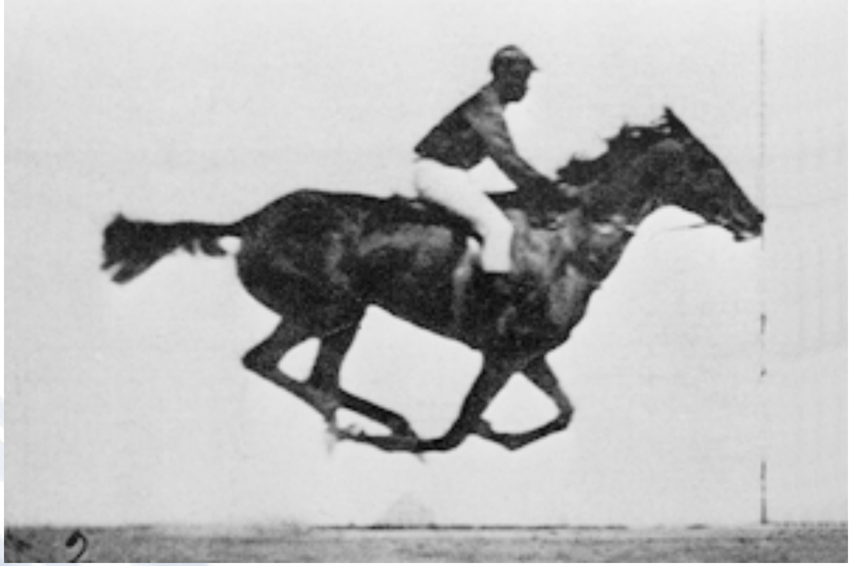
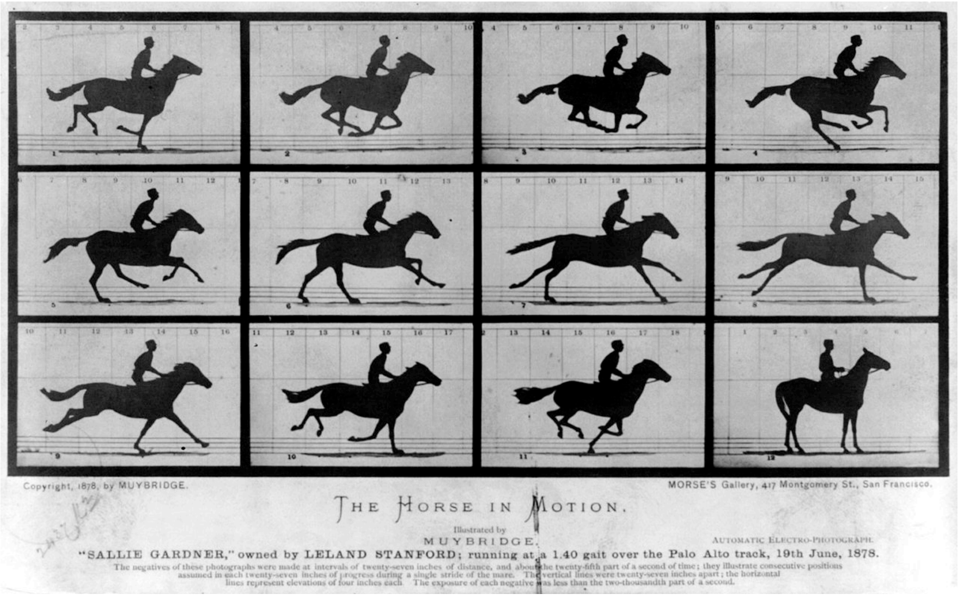
UDM 2017 Workshop, Georgetown

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

Pump-probe spectroscopy (1887)

- stroboscopic investigations of dynamic phenomena





Copyright, 1878, by MUYBRIDGE.

MORSE'S Gallery, 417 Montgomery St., San Francisco.

THE HORSE IN MOTION.

Illustrated by
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AUTOMATIC ELECTRO-PHOTOGRAPH.

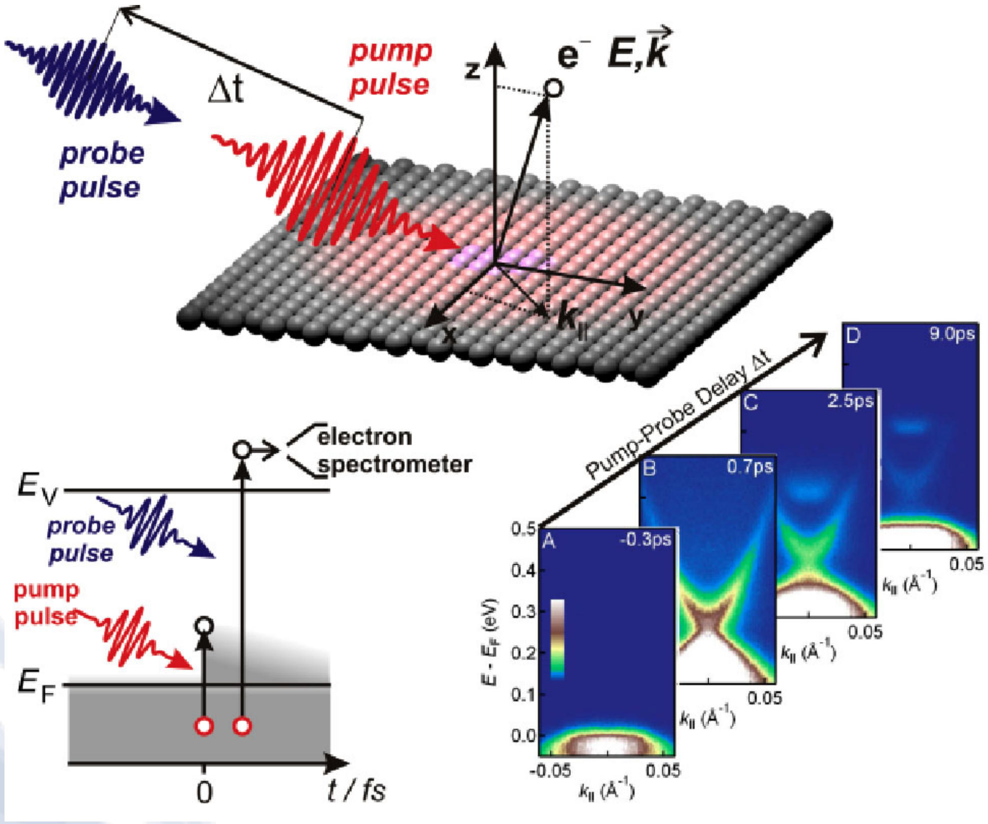
"SALLIE GARDNER," owned by LELAND STANFORD; running at a 1.40 gait over the Palo Alto track, 19th June, 1878.

The negatives of these photographs were made at intervals of twenty-seven inches of distance, and about the twenty-fifth part of a second of time; they illustrate consecutive positions assumed in each twenty-seven inches of progress during a single stride of the mare. The vertical lines were twenty-seven inches apart; the horizontal lines represent elevations of four inches each. The exposure of each negative was less than the two-thousandth part of a second.

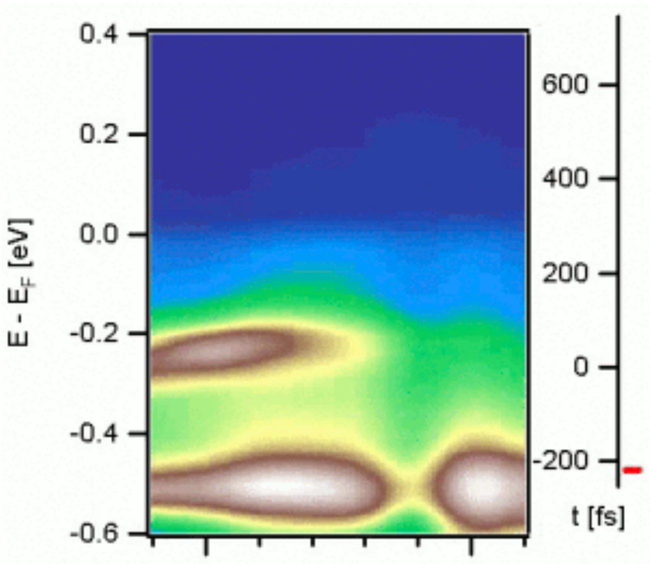
Muybridge 1887

Pump-probe spectroscopy (today)

- stroboscopic investigations of dynamic phenomena



TbTe3 CDW metal



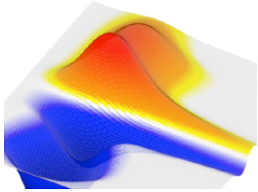
Simulations of time-resolved ARPES: PRX 3, 041033 (2013), PRB 90, 075126 (2014), PRB 92, 224517 (2015), Nature Commun. 7, 13761 (2016)

Image courtesy:
J. Sobota / F. Schmitt

Ultrafast Materials Science today

Understanding the nature of quasiparticles

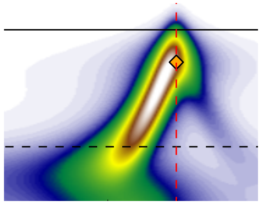
- Relaxation dynamics (Lex Kemper's talk yesterday)
- Control of couplings



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

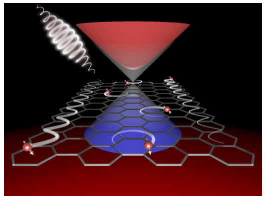
- Collective oscillations
- Competing orders



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)

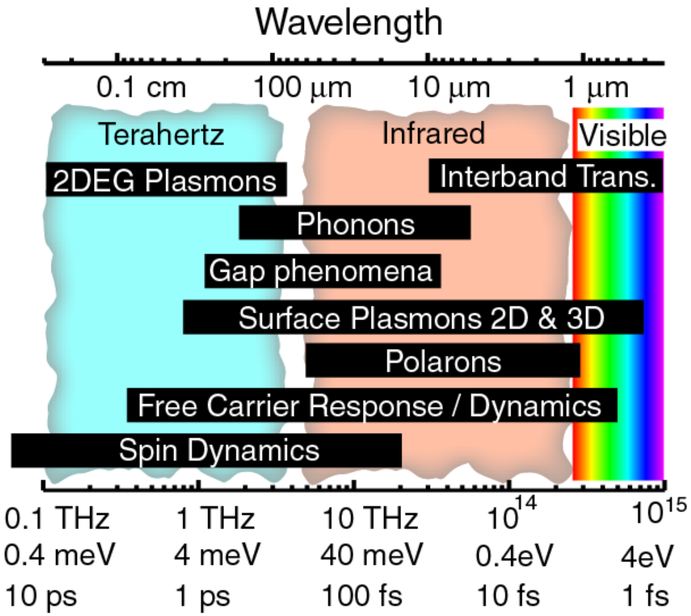


Image courtesy:
D. Basov

Outline

How to 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)

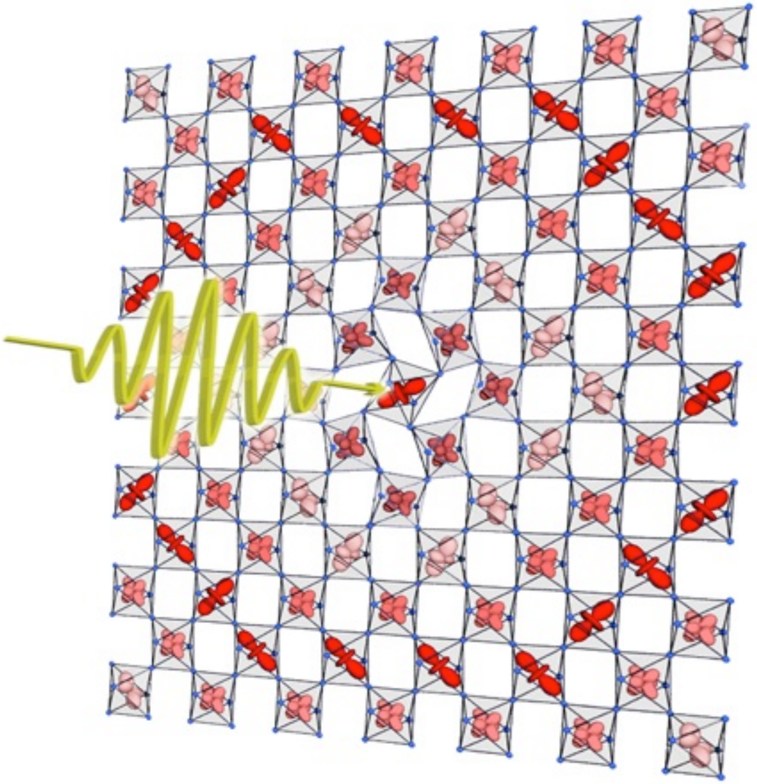
  M. A. Sentef, PRB 95, 205111 (2017) – theory

- Part II: Light-reduced Hubbard U

  Nonresonant laser driving reduces Hubbard U in NiO

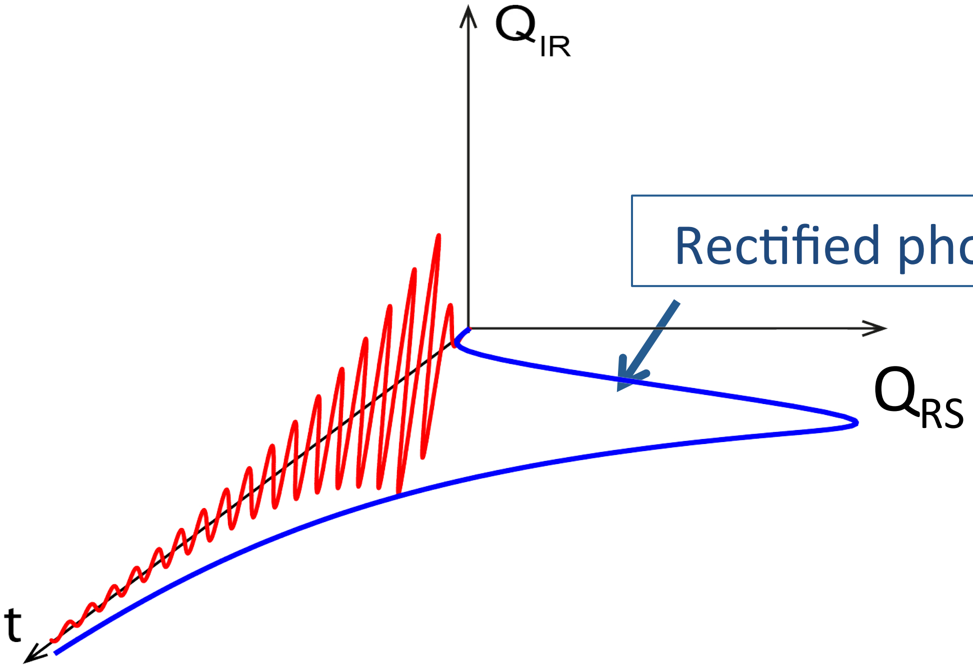
  N. Tancogne-Dejean et al., in preparation – theory

I 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

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

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

„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! Involves ordered-phase dynamics and nonequilibrium quantum many-body physics

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

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

Electron-boson coupling (bilinear)

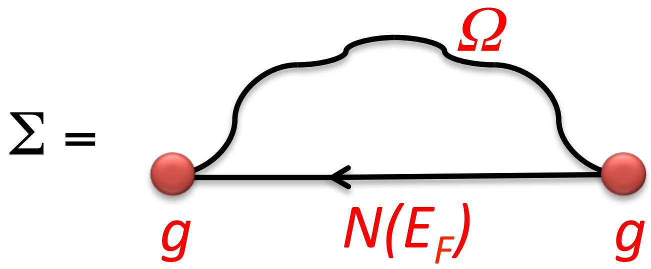
Holstein model (minimal version):

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

Electrons
(Fermi gas/liquid)

Bosons
(e.g., Einstein phonon)

Electron-boson
coupling



Migdal-Eliashberg theory
boson-mediated pairing

Quantum nonlinear phononics

PRB 95, 205111 (2017)

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

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

electron-occupation dependent squeezing of the 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 F(t):
forced coherent phonon oscillation

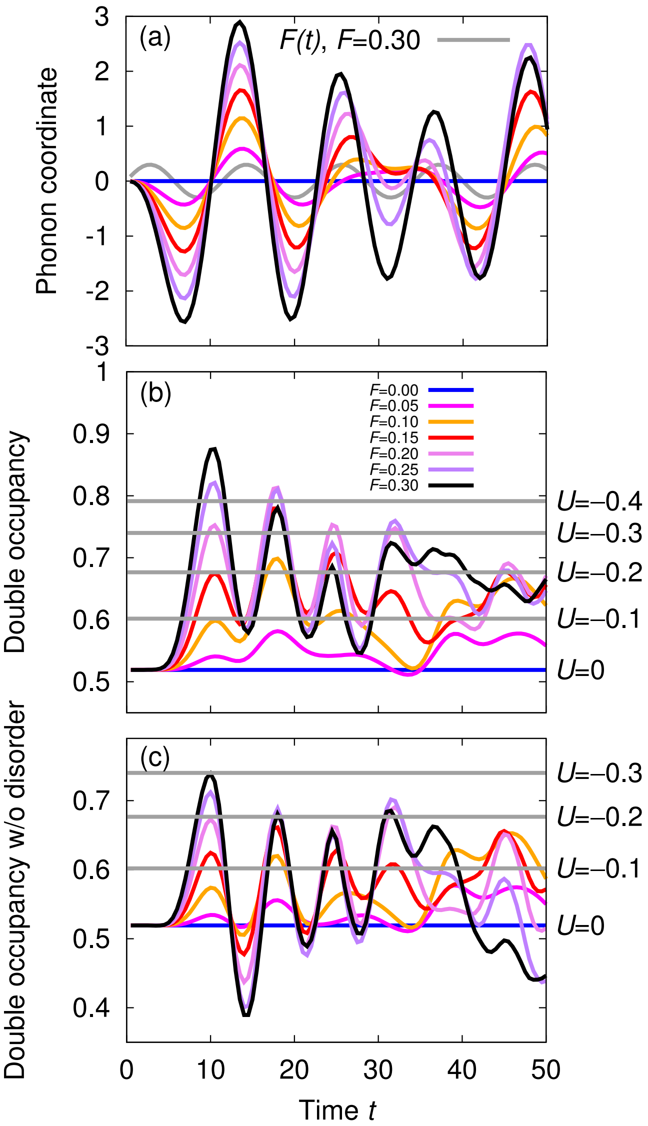
enhancement of local electronic double occupancy

<u>sum of two effects:</u>

induced disorder (cf. Kennes)

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

induced el-el attraction



Time-resolved electronic spectrum

PRB 95, 205111 (2017)

Here: $g2=-0.05 < 0$

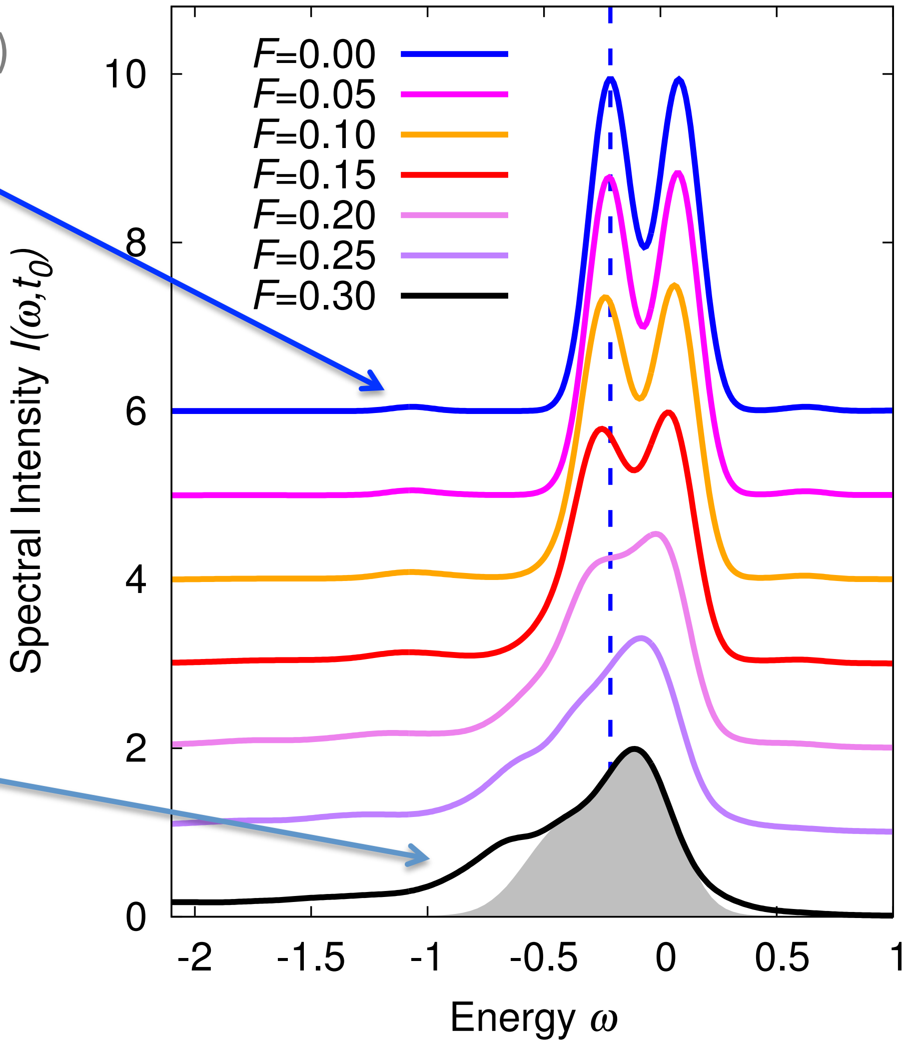
(does not matter for light-enhanced coupling)

2-phonon shakeoff

Reduced coherence peaks with stronger driving

light-enhanced el-ph coupling

light-induced polaron formation



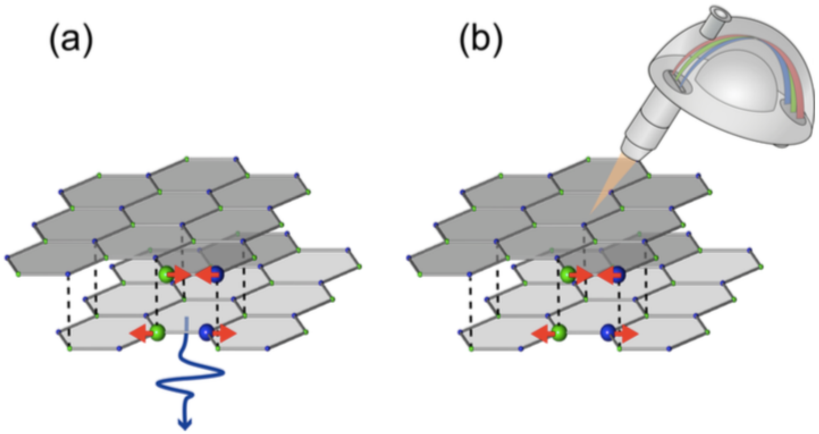
Dynamically enhanced coupling

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)

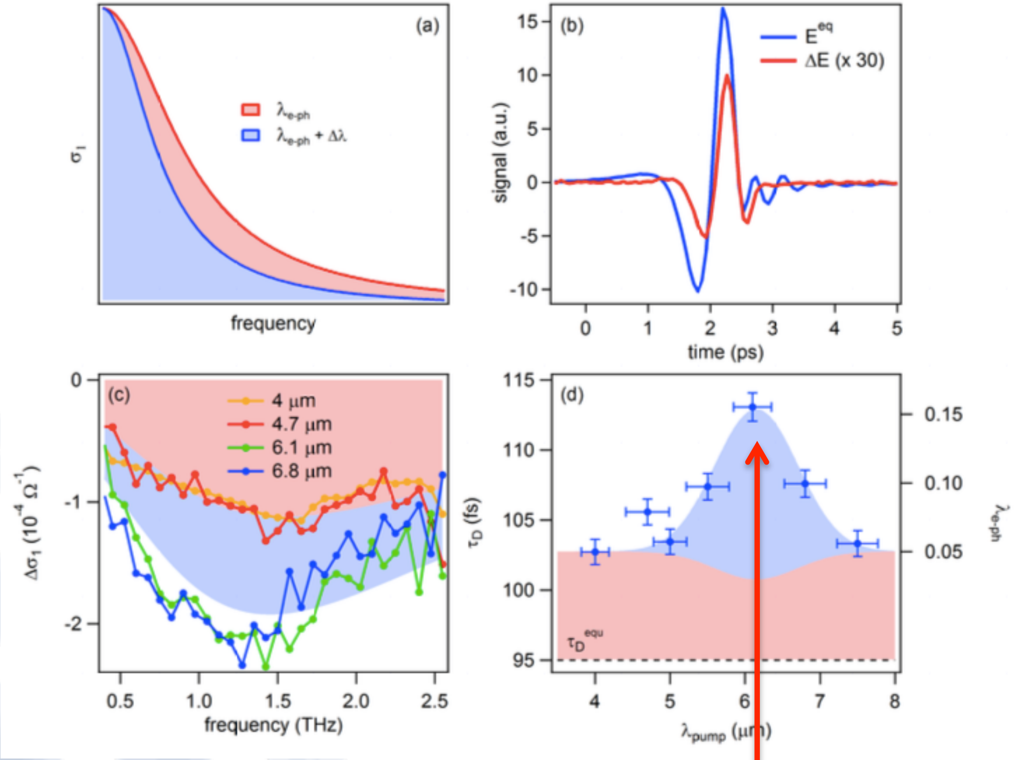
enhanced electron-phonon for pump on resonance with IR phonon



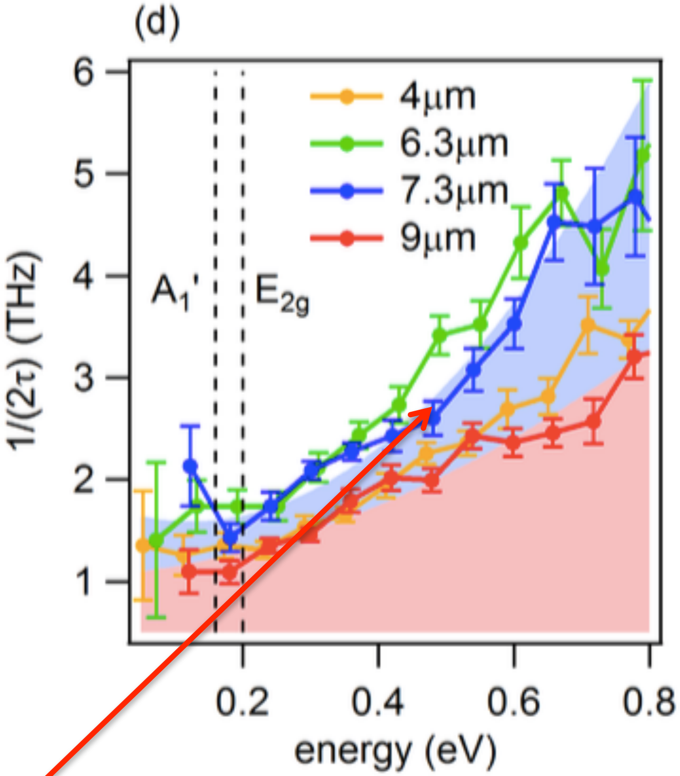
Dynamically enhanced coupling

Enhanced electron-phonon coupling in graphene with periodically distorted lattice *PRB 95, 024304 (2017)*

transient reduction of **THz Drude weight**



accelerated **tr-ARPES relaxation**

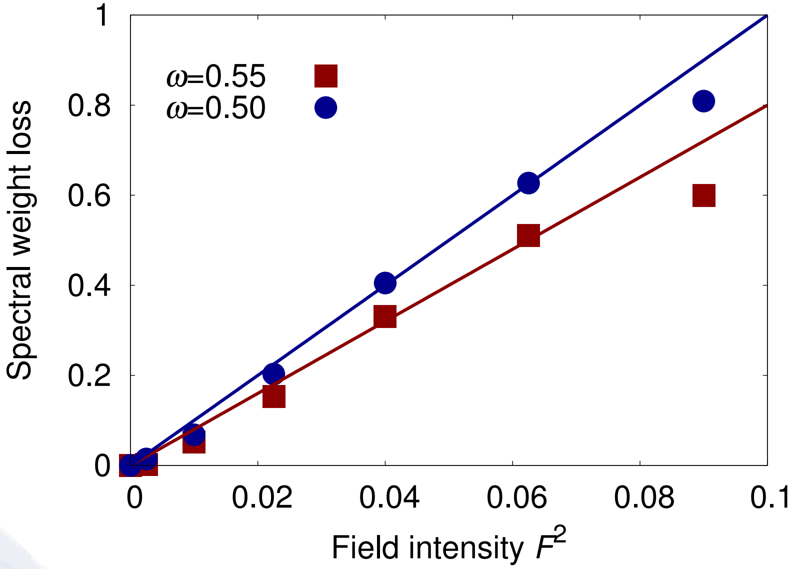


3-fold enhancement of effective $\lambda_{el\text{-}ph}$!

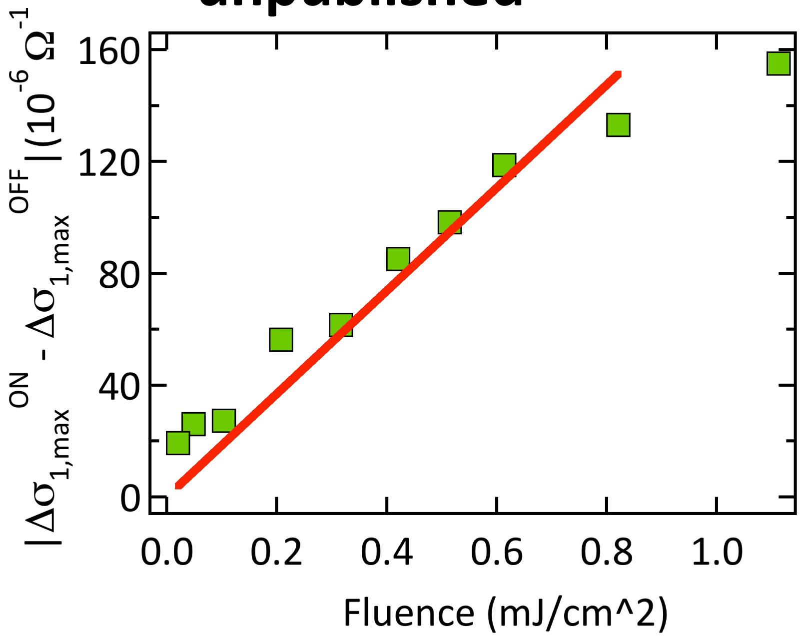
Field scaling

PRB 95, 205111 (2017)

Theory



Data by E. Pomarico, unpublished



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

Quantum nonlinear phononics

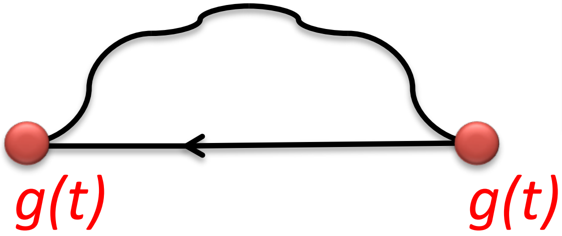
Forced coherent oscillation $\langle \hat{x}_l(t) \rangle \propto F \sin(\omega t)$

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

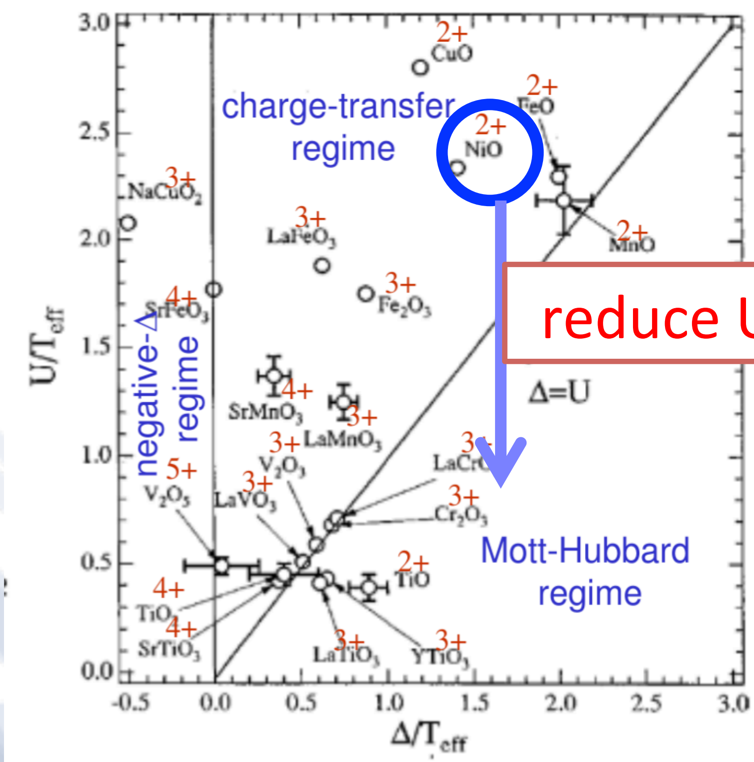
Exact solution of electron-phonon model system:

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

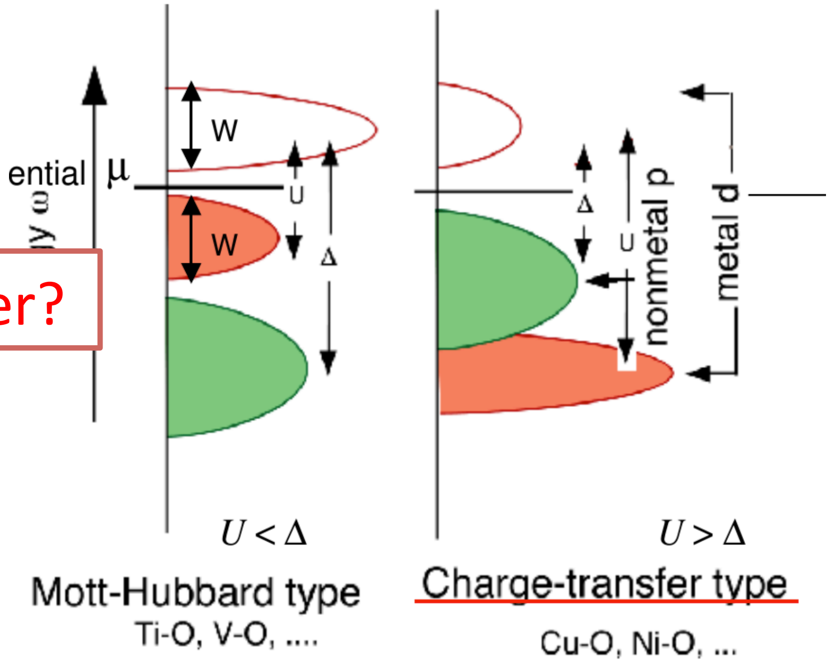
PRB 95, 205111 (2017)

II Dynamical modification of Hubbard U

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



reduce U with laser?



Mott-Hubbard type

Ti-O, V-O, ...

Charge-transfer type

Cu-O, Ni-O, ...

Zaanen-Sawatzky-Allen phase diagram

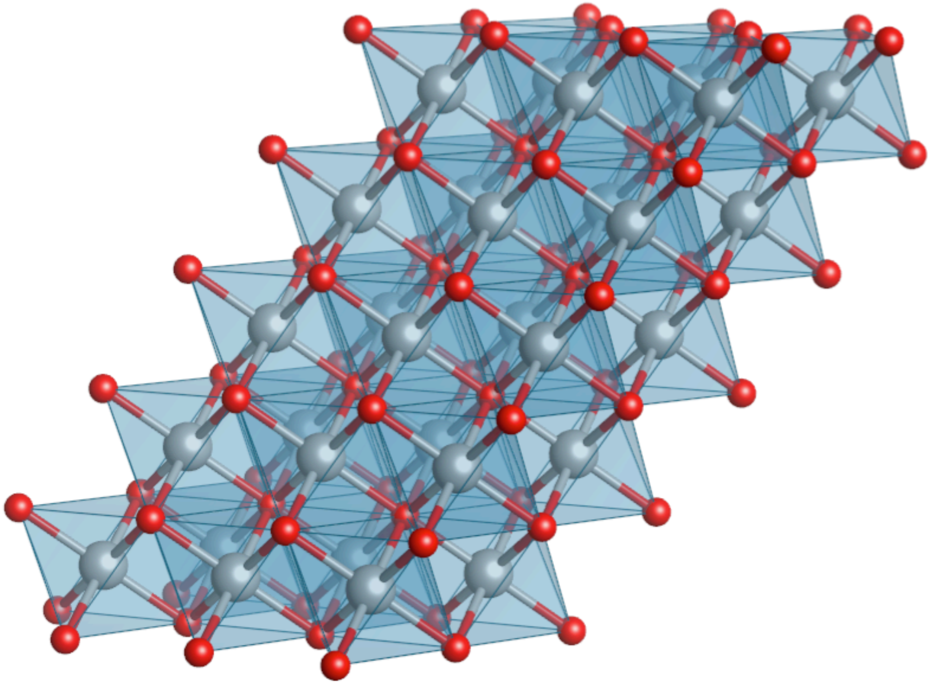
NiO as prototypical charge-transfer insulator

NiO:

Antiferromagnetic type 2

Band gap: ~4 eV (exp.)

Néel temperature: 523K



Time-dependent U with TDDFT+U

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

⟷

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

ACBN0 functional
PRX 5,011006 (2015)

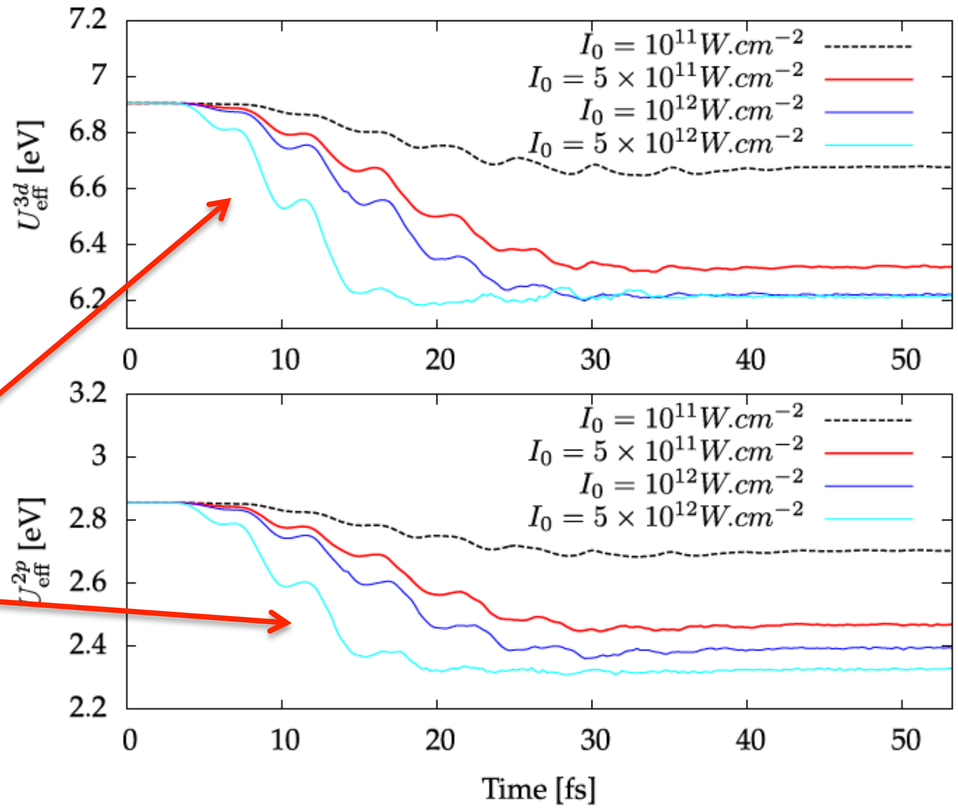
- alternative to constrained RPA
- numerically efficient
- direct extension to time-dependent case (adiabatic approximation)

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

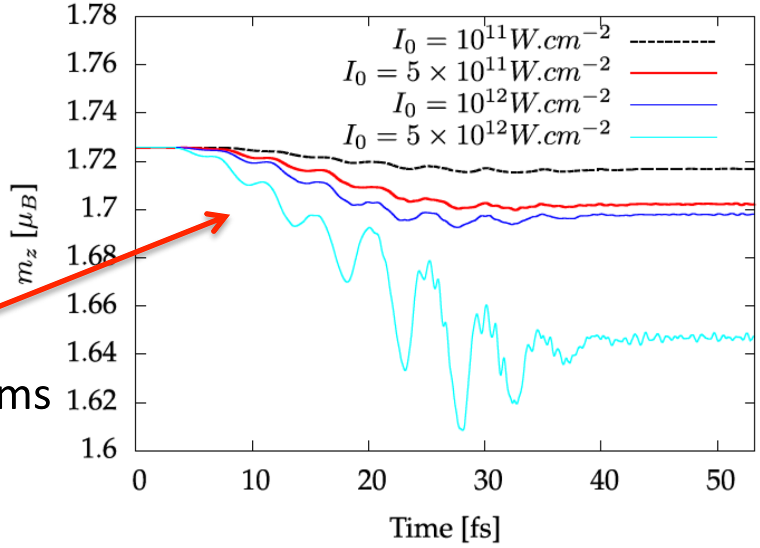


Typical intensities in strong field physics in solids

Reduction of U: mechanism

U measures the Coulomb interaction screened by itinerant electrons

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



Demagnetization of Ni atoms

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

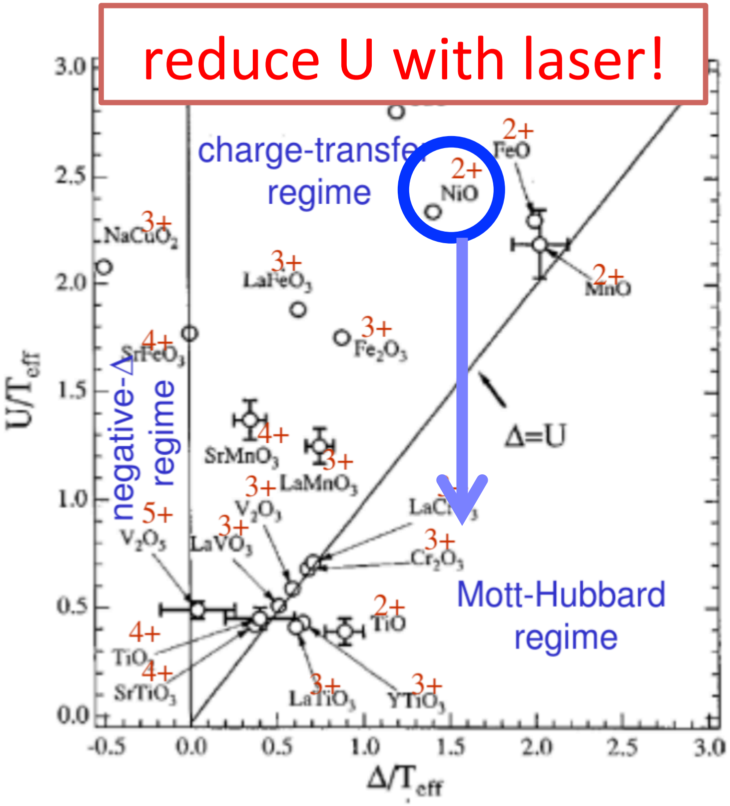
Summary II

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

N. Tancogne-Dejean et al., in preparation

N. Tancogne-Dejean

A. Rubio



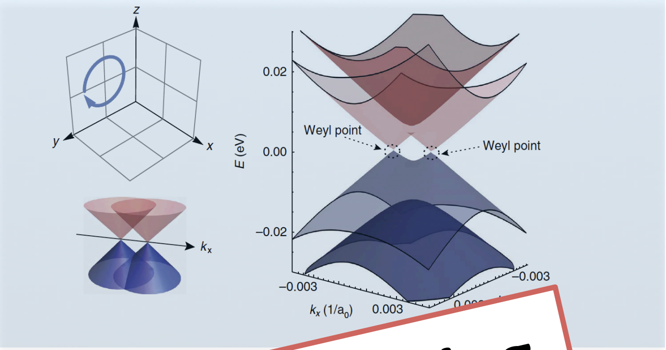
reduce U with laser!

Summary & Outlook

Ultrafast laser engineering of

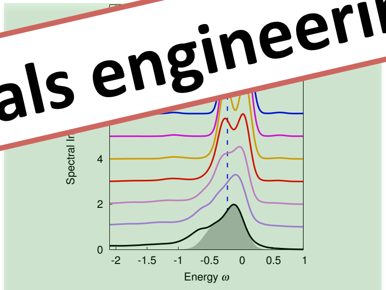
- band structure, topology (Floquet)

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



- electron-phonon coupling (quantum nonlinear phononics)

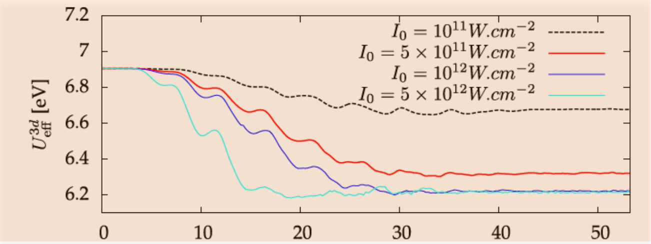
  PRB 95, 024304 (2017)
  PRB 95, 2051



Path towards nonequilibrium materials engineering

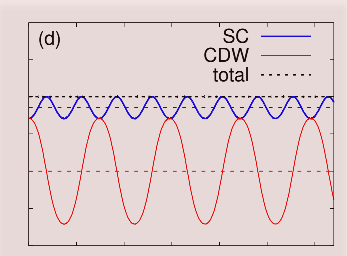
- H g subresonant ations in correlated insulators)

  forthcoming



- superconductivity

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



Summary & Outlook

Pump-probe pros and cons:

+ **ultrafast** switching (magnetism, metal-insulator, ordered phases, ...)

+ **nonthermal**: disentangling of complexity, selectivity of excitation

+ **time-resolved**: movies are more fun than static data

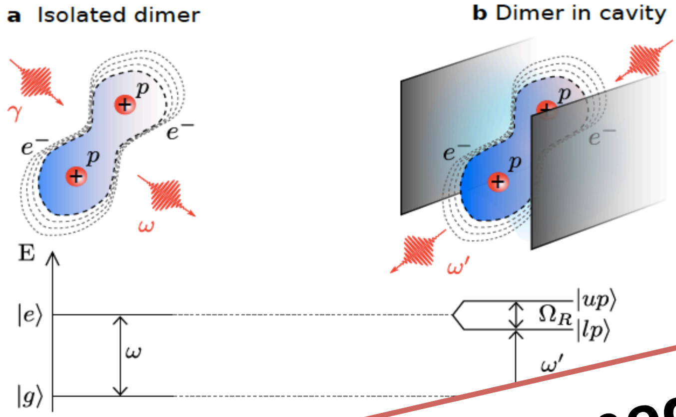
- **ultrashort**: lifetime of light-induced states (Floquet, transient SC, ...)
- **heating**: often bad for interesting states
- **strong lasers**: useful for device applications?

Interesting future direction:
Bridging quantum optics and ultrafast materials
From classical light to quantum photons

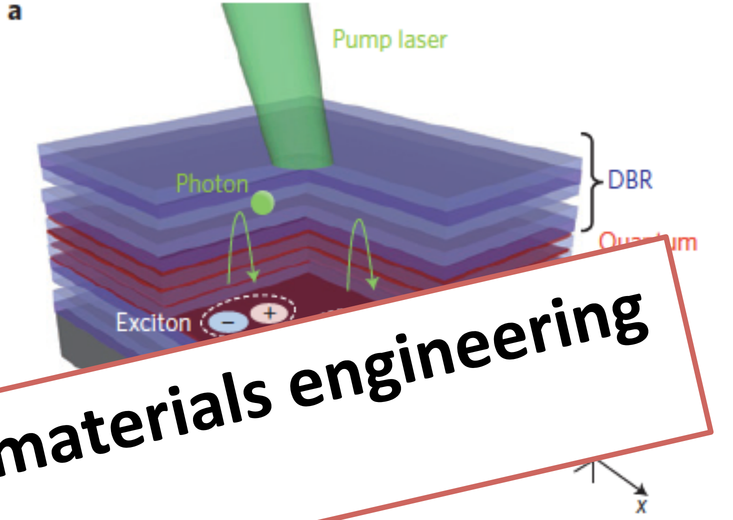
Summary & Outlook

Atoms and molecules in cavities, from weak to strong coupling in quantum-electrodynamics (QED) chemistry

Johannes Flick[a,1], Michael Ruggenthaler[a], Heiko Appel[a], and Angel Rubio[a,b,1] PNAS 114, 3026 (2017)



(c) N. Tancogne-Dejea



et al., Nature Physics 10, 803 (2014)

Polariton condensates

Path towards nonequilibrium materials engineering with quantum photons?

light-matter coupling in confined geometries

quantum photons to induce novel phases with

- less heating
- lower laser power
- longer lifetimes

Acknowledgments

Thank you for your attention!

G. Topp

R. Tuovinen

N. Tancogne-Dejean

A. Rubio

E. Pomarico

I. Gierz

A. Cavalleri

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

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