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

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Driven is different

Kapitza pendulum

dynamical stabilization of a metastable state
Is driven also useful?

Exposing hidden states

Light-induced new states?

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

... and many more.
Ultrafast Materials Science today

Understanding the nature of quasiparticles
- Relaxation dynamics
- Control of couplings

PRX 3, 041033 (2013)  PRB 95, 205111 (2017)
PRB 90, 075126 (2014)  arXiv:1802.09437

Understanding ordered phases
- Collective oscillations
- Competing orders

PRB 93, 144506 (2016)  arXiv:1808.00712

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
  \textit{E: Pomarico et al., PRB 95, 024304 (2017)} – experiment (bilayer graphene)
  \textit{M. A. Sentef, PRB 95, 205111 (2017)} – theory

• Part II: Light-reduced Hubbard U
  Nonresonant laser driving reduces Hubbard U in NiO
  \textit{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

Rectified phonon field \( \rightarrow \) directional force

Simplest model: classical dynamics

\[
\ddot{Q}_{RS} + \Omega_{RS}^2 Q_{RS} = AQ_{IR}^2
\]

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

"nonlinear phononics"

\[
H = A Q_{IR}^2 Q_{RS}
\]

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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
• phononic rectification in YBCO
  Mankowsky et al., Nature 516, 71 (2014)
• ferroelectric switching in LiNbO$_3$

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

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 $\lambda_{el-ph}$!
Quantum nonlinear phononics

2-site toy model, solve dynamics exactly

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

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

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

Response: \( I(\omega, t_0) = \text{Re} \int dt_1 \int dt_2 e^{i\omega(t_1-t_2)} s_{t_1,t_2,\tau(t_0)} \times \left[ \langle \psi(t_2) | c_{1,\uparrow}^\dagger T e^{-i \int_{t_2}^{t_1} H(t) dt} c_{1,\uparrow} | \psi(t_1) \rangle + \langle \psi(t_1) | c_{1,\uparrow} T e^{-i \int_{t_1}^{t_2} H(t) dt} c_{1,\uparrow}^\dagger | \psi(t_2) \rangle \right] \),


density-dependent squeezing of phonon

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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)

2-phonon shakeoff

Reduced coherence peaks with stronger driving

light-enhanced el-ph coupling

light-induced polaron formation
Field dependence

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

Data by E. Pomarico, unpublished

Theory

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Quantum nonlinear phononics

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

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

Migdal-Eliashberg diagram

Effective induced linear coupling

\[ \Sigma(t,t') = ig(t)g^*(t')G(t,t')D(t,t') \]

time-dependent vertex, amplitude \( g^2 \sim F^2 \)

\( \Rightarrow \) light-induced coupling, lambda scales \( \sim F^2 \)

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Summary I

• enhanced electron-phonon coupling in phononically driven bilayer graphene

*PRB 95, 024304 (2017)*

Exact solution of electron-phonon model system:
• theoretical proposal: nonlinear el-ph coupling as mechanism behind this enhancement

*PRB 95, 205111 (2017)*
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

DFT with \textit{ab initio} and self-consistent Hubbard U (Hybrid functional)

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

- Electron-electron interaction
- Double counting

Usual expression in DFT+U

\[ E_{ee} \approx \frac{\tilde{U}}{2} \sum_{\{m\}, \sigma} N_{m}^{\sigma} N_{m'}^{-\sigma} + \frac{\tilde{U} - \tilde{J}}{2} \sum_{m \neq m', \sigma} N_{m}^{\sigma} N_{m'}^{\sigma}. \]

ACBN0 functional
PRX 5,011006 (2015)

- alternative to constrained RPA
- numerically efficient
- direct extension to \textit{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)
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

- Hubbard U (strong subresonant band structure, topology (Floquet))
  - arXiv:1808.00712
  - arXiv:1806.08187

Path towards nonequilibrium materials engineering

- ordered phases
  - PRB 93, 144506 (2016) arXiv:1808.00712

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