Nonequilibrium Materials Engineering beyond Floquet

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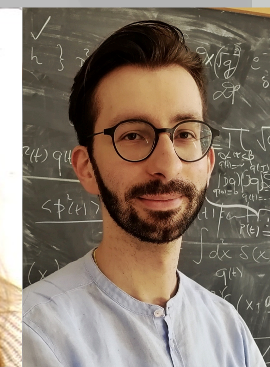
Boston University, November 2, 2018

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

Max Planck Institute for the Structure and Dynamics of Matter

Team



Mona
Kalthoff
(Dortmund
-> MPSD)

Matteo
Puviani
(Modena)

Theoretical description of pump-probe spectroscopy in solids

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Emmy Noether-Programm
Deutsche Forschungsgemeinschaft DFG

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Emmy Noether Programme (SE 2558/2-1)

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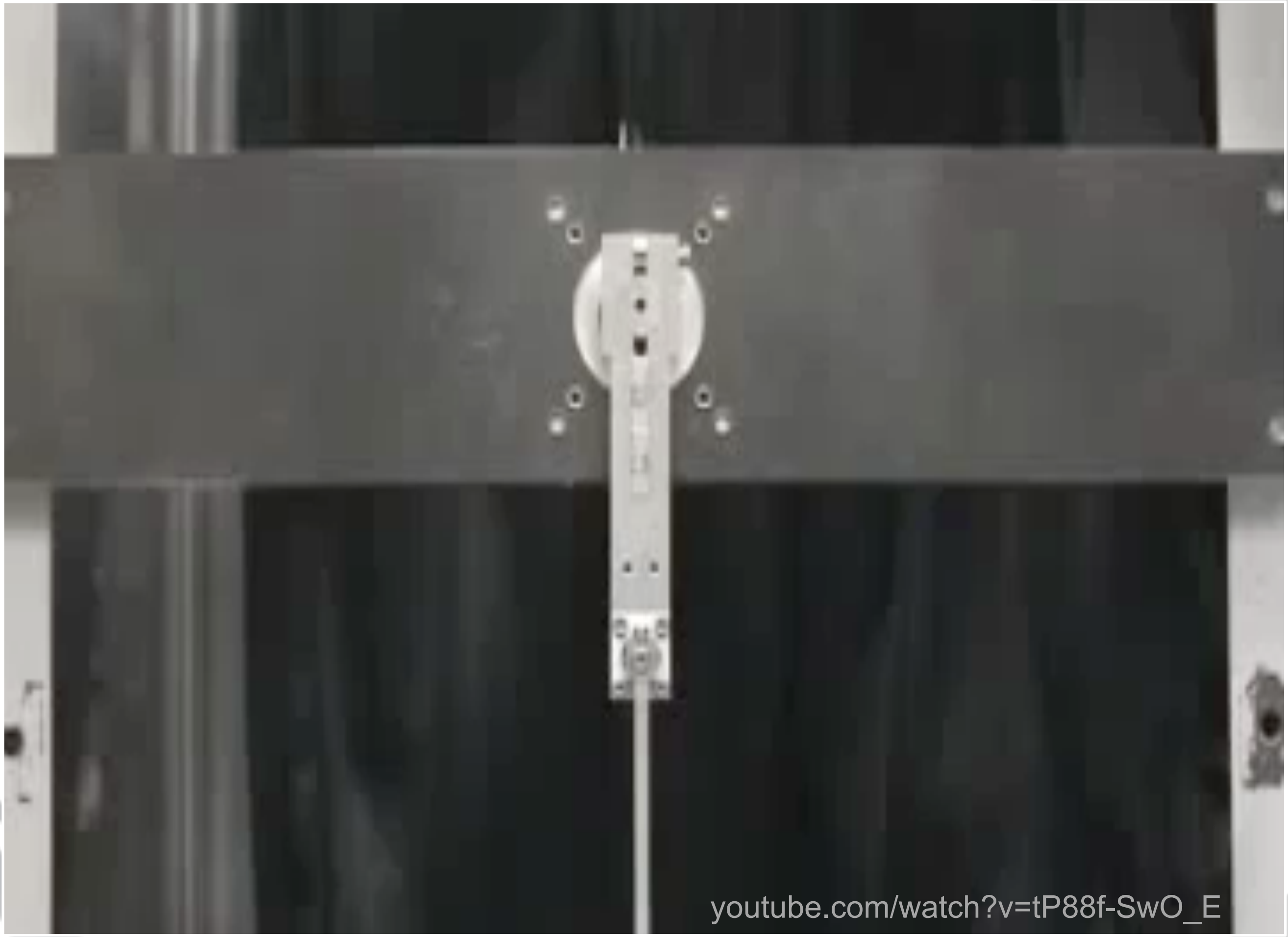
Driven is different

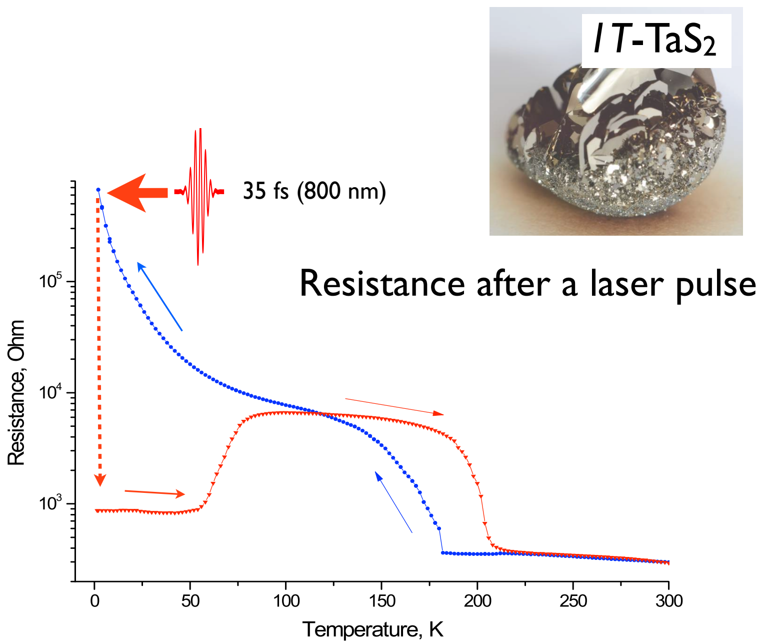
Kapitza pendulum



dynamical stabilization of a metastable state

Is driven also useful?

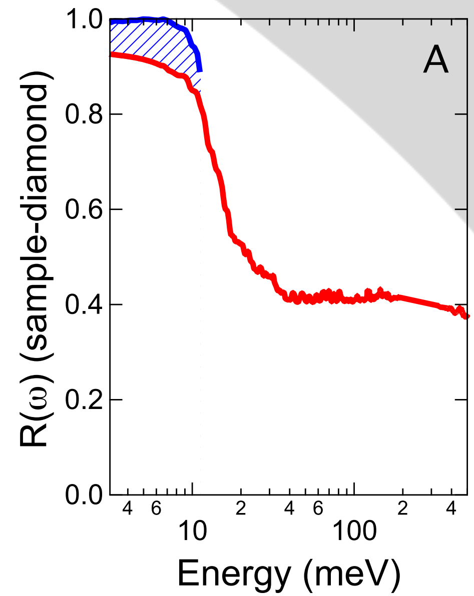
Exposing hidden states



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

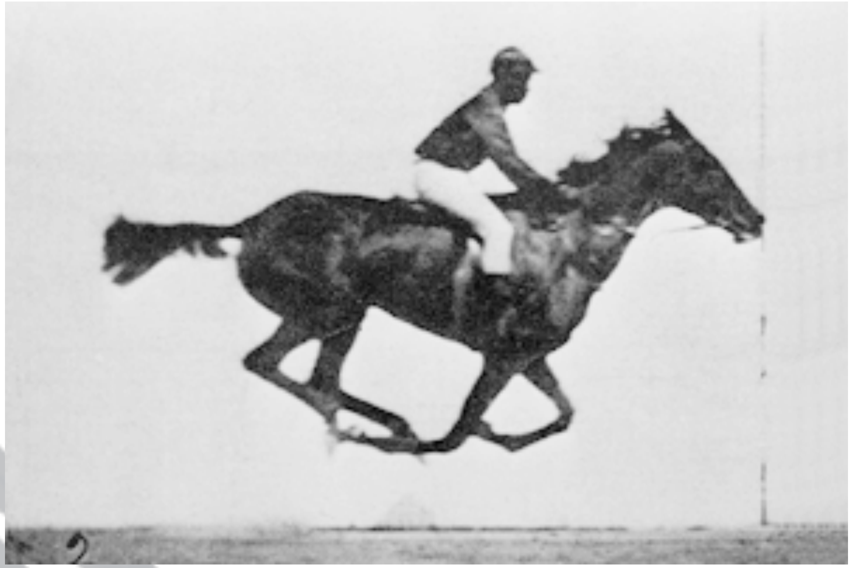
M. Mitrano, A. Cantaluppi, D. Nicoletti, S. Kaiser, A. Perucchi, S. Lupi, P. Di Pietro, D. Pontiroli, M. Riccò, S. R. Clark, D. Jaksch & A. Cavalleri

Affiliations | Contributions | Corresponding author

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

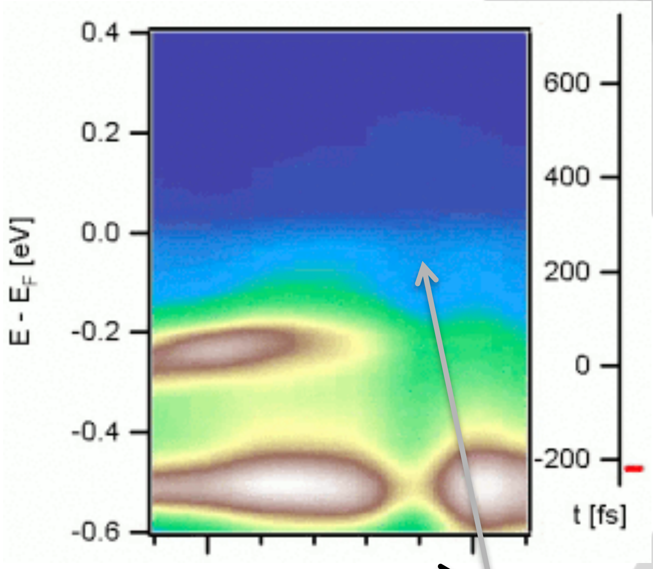
Pump-probe spectroscopy

- stroboscopic investigations of dynamic phenomena



Muybridge 1887

TbTe3 CDW metal



F. Schmitt et al., Science 321, 1649 (2008)
Image courtesy: J. Sobota / F. Schmitt

Challenge

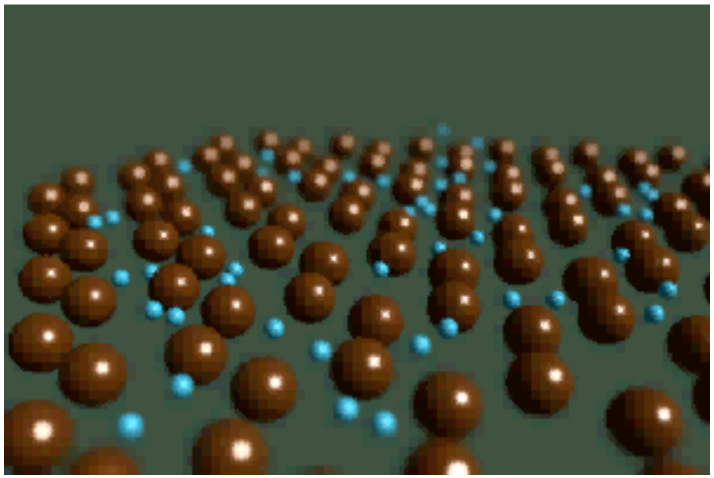
U.S. Department of Energy "Five Grand Challenges to Science and the Imagination"

- *Grand Challenge #3: How do remarkable properties of matter emerge from complex correlations of the atomic or electronic constituents and how can we control these properties?*
- *Grand Challenge #5: How do we characterize and control matter away – especially very far away – from equilibrium?*

http://science.energy.gov/bes/efrc/research/grand-challenges/

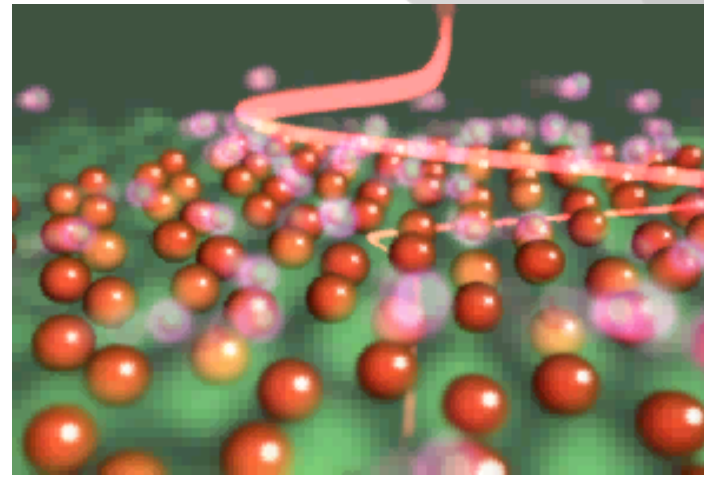
Challenge

movies by Koichiro Tanaka (Kyoto university)



many-body problem

(electrons + ions)



nonequilibrium many-body problem

(electrons + ions + photons)

Mission statement:

To understand and predict electron-ion dynamics and control of emergent nonequilibrium electronic structure

Challenge

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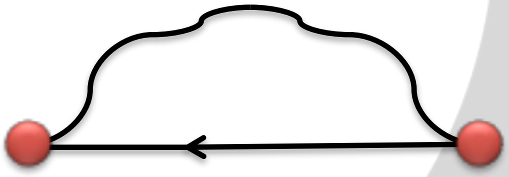
Main challenges:

- hierarchy of energy and time scales
- high laser intensities: nonperturbative/nonlinear

Possible approaches:

- first principles (time-dependent density functional theory (TDDFT))
- effective models:
  - Feynman diagrams: self-energy
  - Keldysh nonequilibrium Green's functions
  - connection with DFT: Sham-Schlüter integral equation

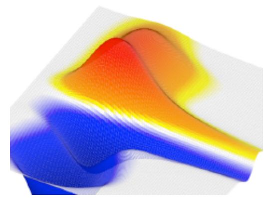


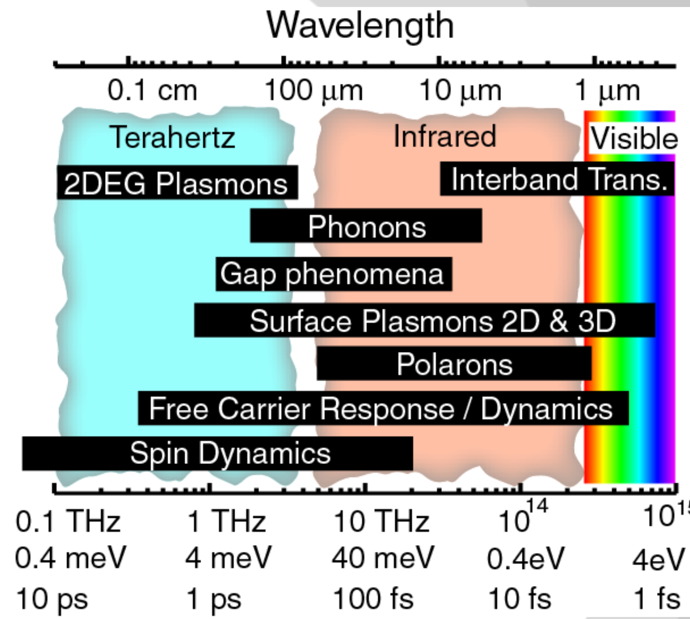
Ultrafast Materials Science today

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, Sci. Adv.
arXiv:1808.02389

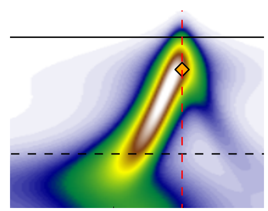




Understanding ordered phases

- 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
arXiv:1810.06536



Creating new states of matter

- nonequilibrium topological states

Nature Comm. 6, 7047 (2015)
Nature Comm. 8, 13940 (2017)
Nature Comm. 9, 4452 (2018)

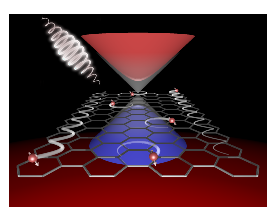


Image courtesy: D. Basov

Relaxation dynamics

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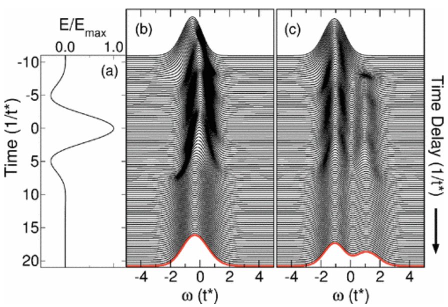
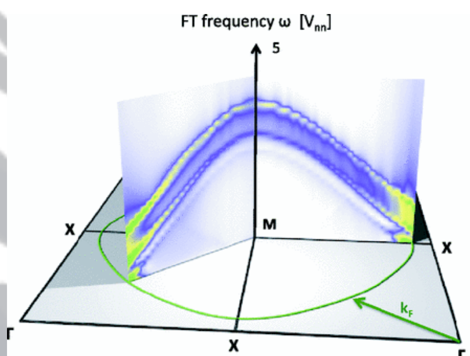
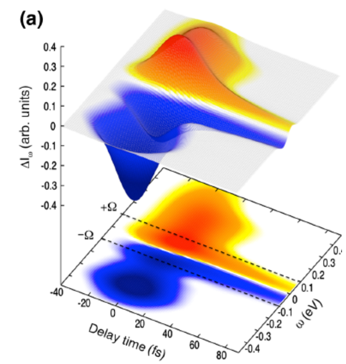


PRL 111, 077401 (2013)
nonthermal pumped states

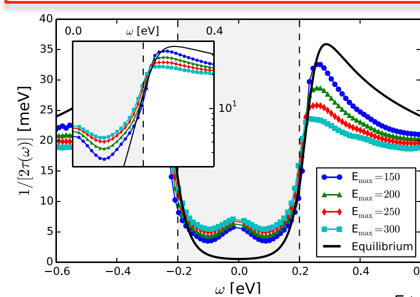


PRB 87, 235139 (2013)
extracting unoccupied electronic structure

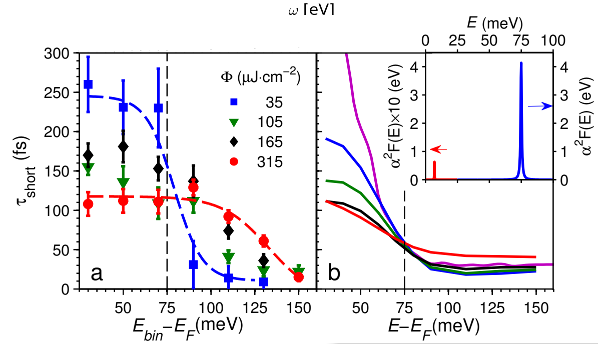
examining electron-boson coupling



PRX 3, 041033 (2013)
small fluences



PRB 90, 075126 (2014)
fluence dependence



Nat. Comm. 7, 13761 (2016)
comparison with experiment

Electron-boson coupling

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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)

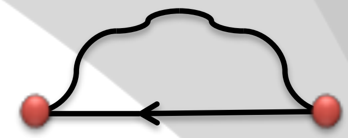
Electron-boson coupling

Pump laser:

$$\varepsilon(k) \rightarrow \varepsilon(k,t)$$

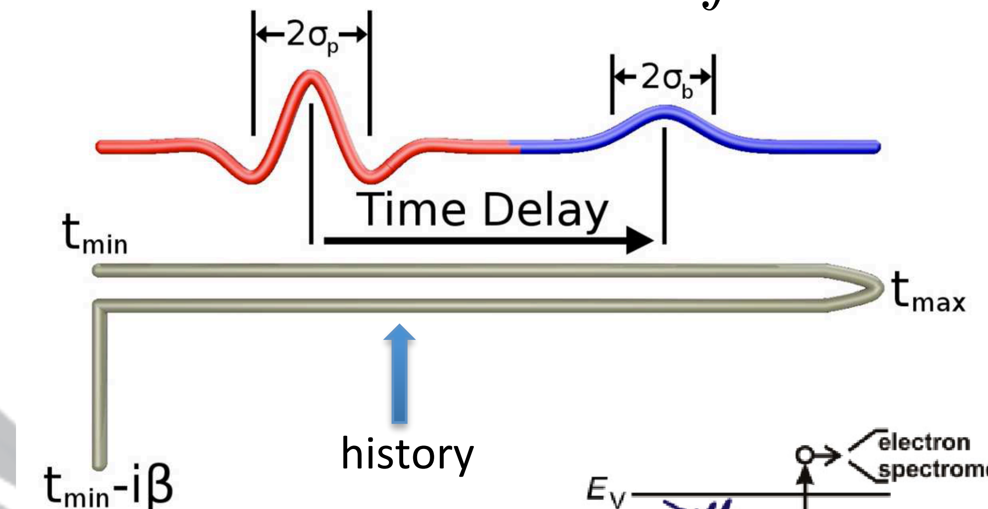
Method: Keldysh Green functions

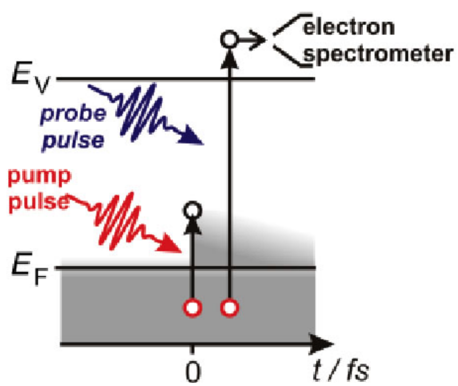
$$G_k(\omega) = G_k^0(\omega) + G_k^0(\omega)\Sigma(\omega)G_k(\omega)$$



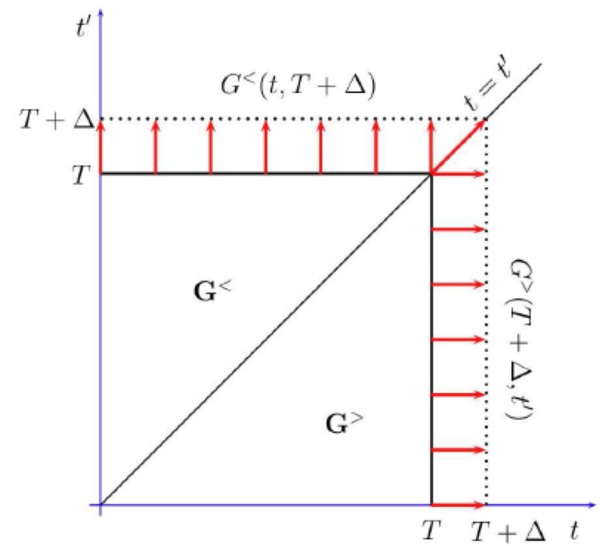
$$G_{\mathbf{k}}(t,t') = G_{\mathbf{k}}^0(t,t') + \oint dt_1 dt_2 G_{\mathbf{k}}^0(t,t_1)\underline{\Sigma(t_1,t_2)}G_{\mathbf{k}}(t_2,t')$$

self-energy Σ:
electron-electron scattering
electron-phonon scattering
...





pump-probe photoemission



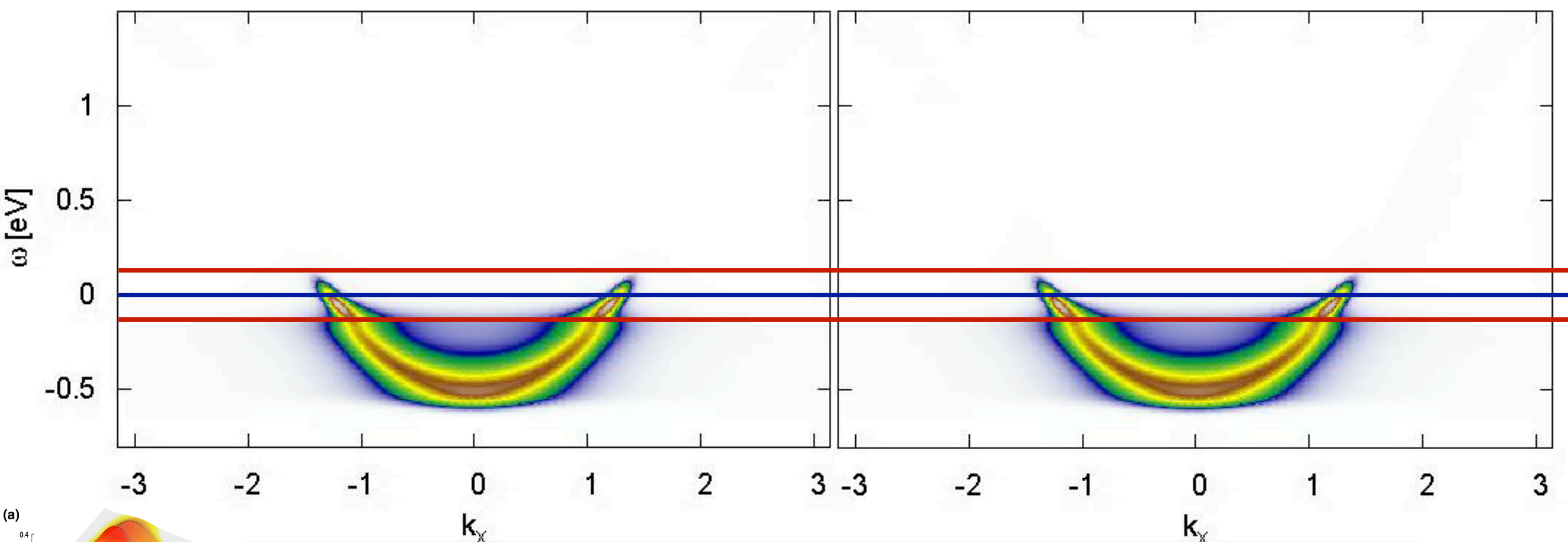
Electron-boson coupling

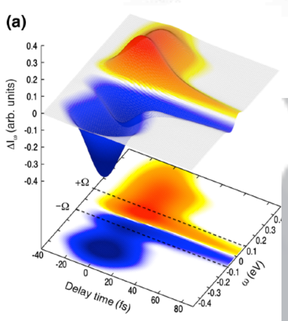
PRX 3, 041033 (2013)

Weak pump | Strong pump

time unit = 0.66 fs

t = -65.00 | t = -65.00





boson window effect for fast versus slow relaxation

nonlinear response for strong pump

Ordered phases

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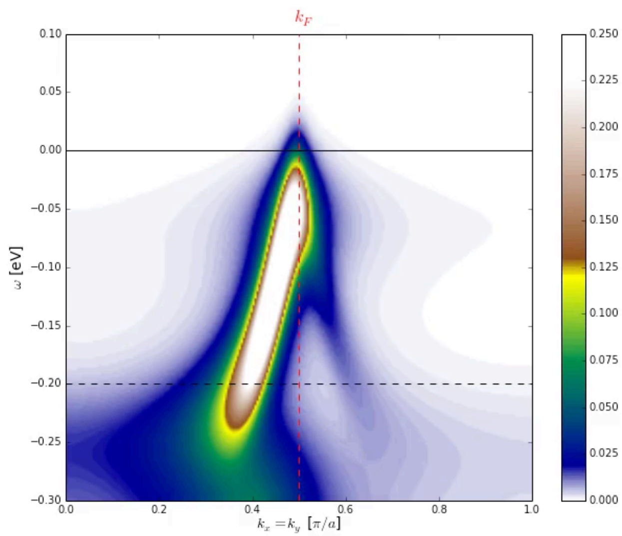

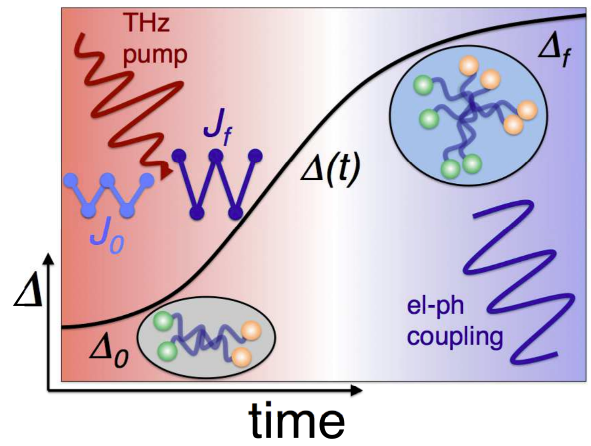

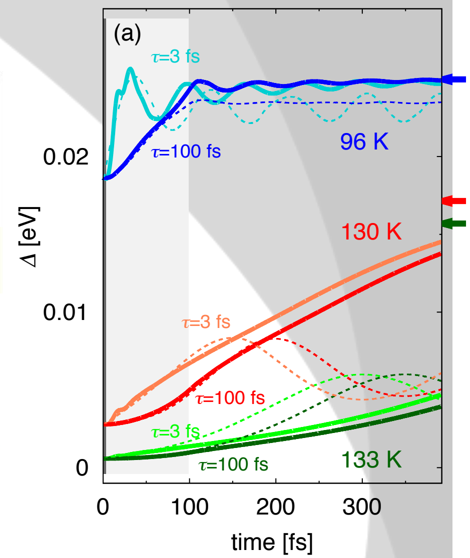

PRB 92, 224517 (2015)

Higgs amplitude mode oscillations in pump-probe photoemission spectroscopy

PRB 93, 144506 (2016)

Light-enhanced superconductivity: electron-phonon scattering versus collective order parameter dynamics

Some recent key results

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How to engineer materials away from equilibrium?

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: Optical control of chiral superconductors

Short laser pulses allow for switching of Majorana modes

M. Claassen et al., arXiv:1810.06536

Part III: From classical to quantized photon fields

Materials engineering in an optical cavity using vacuum fluctuations

M. A. Sentef et al., arXiv:1802.09437

I Resonant excitation of crystal lattice

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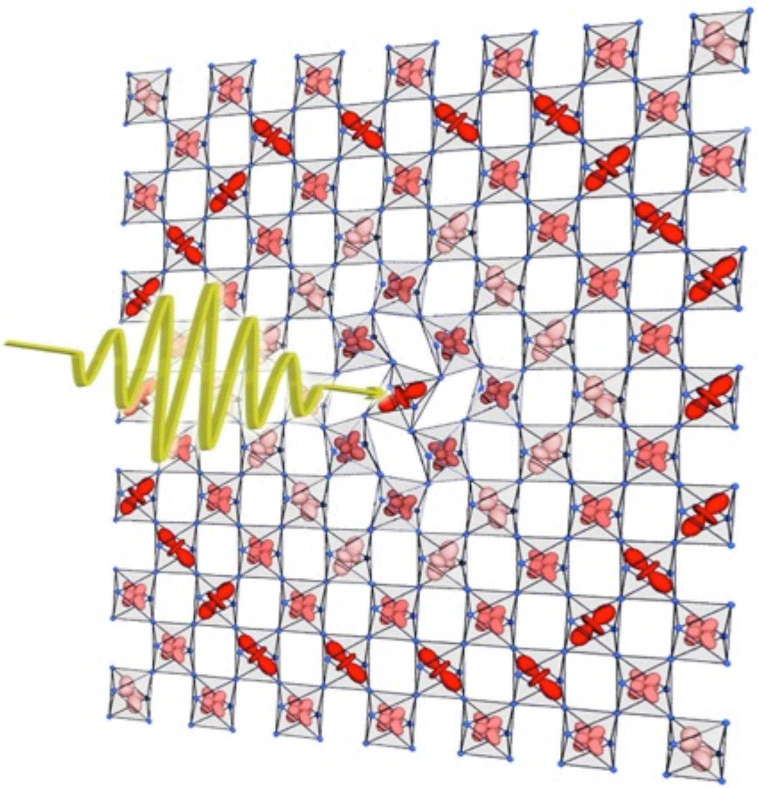

M. Först et al., Nature Physics 7, 854 (2011)

Classical nonlinear phononics

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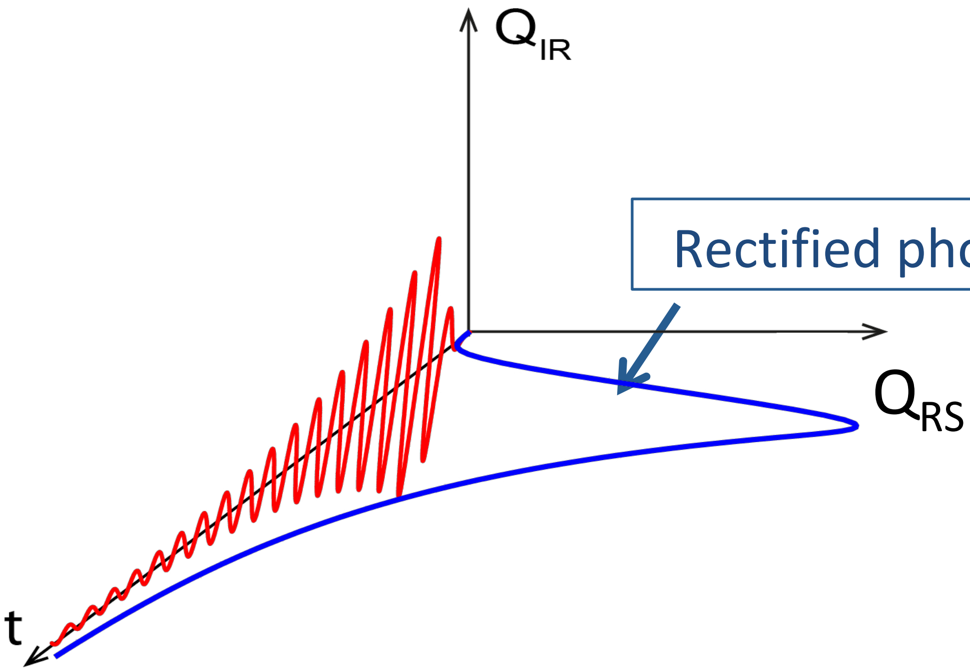

Simplest model: classical dynamics

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

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

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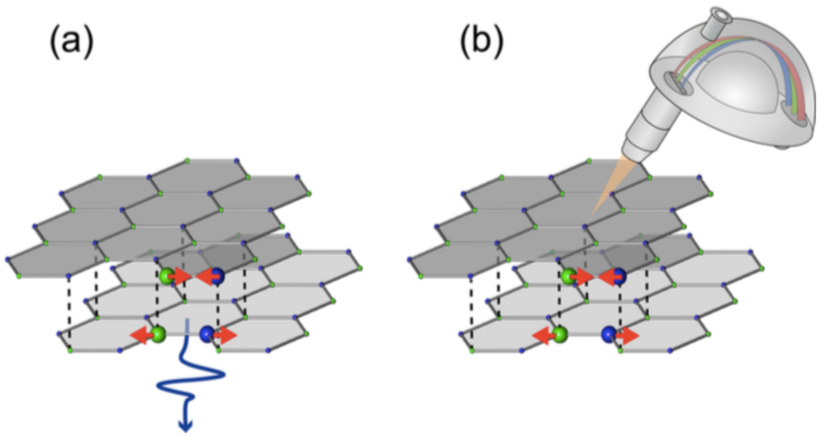
Enhanced electron-phonon coupling in graphene with periodically distorted lattice

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)

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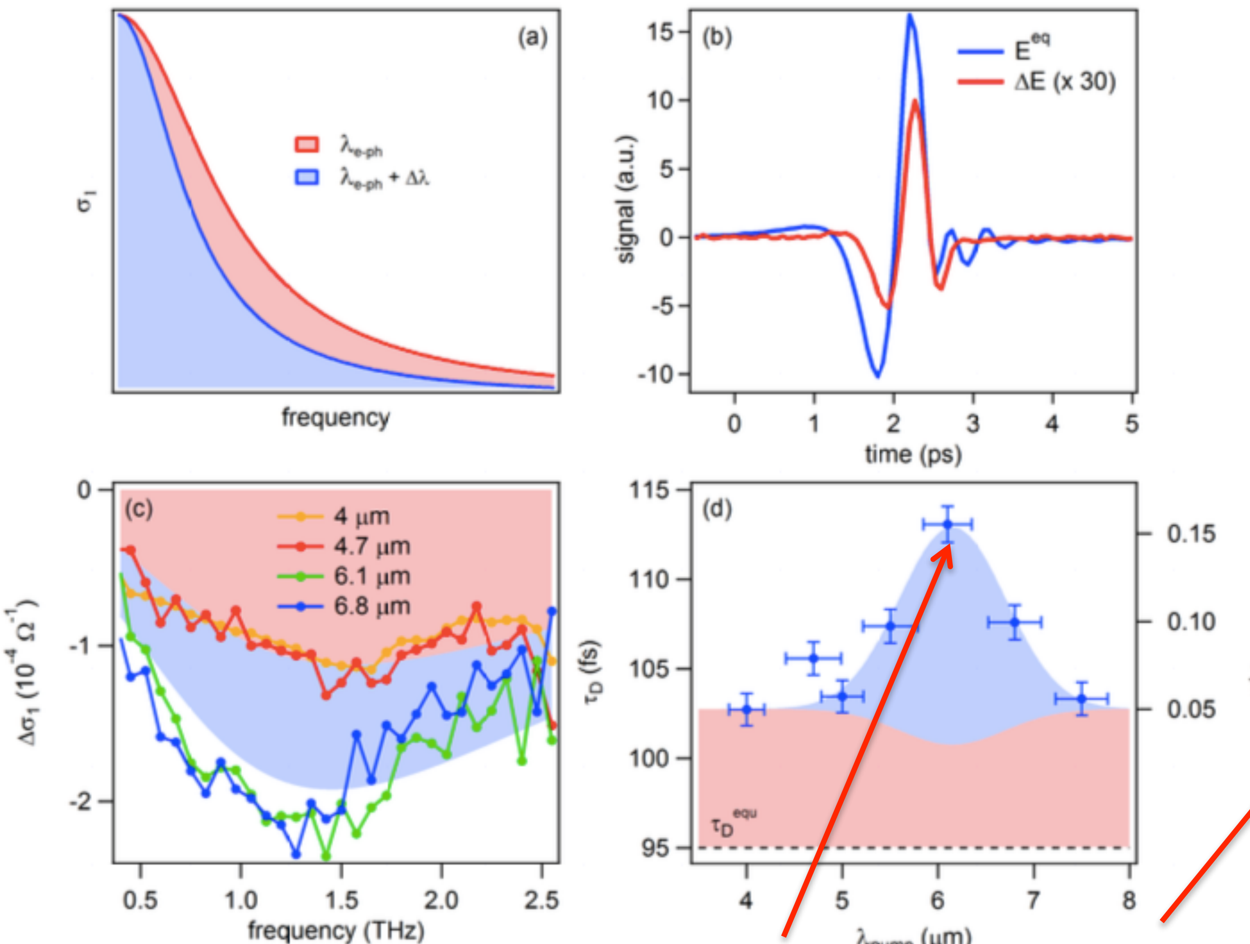
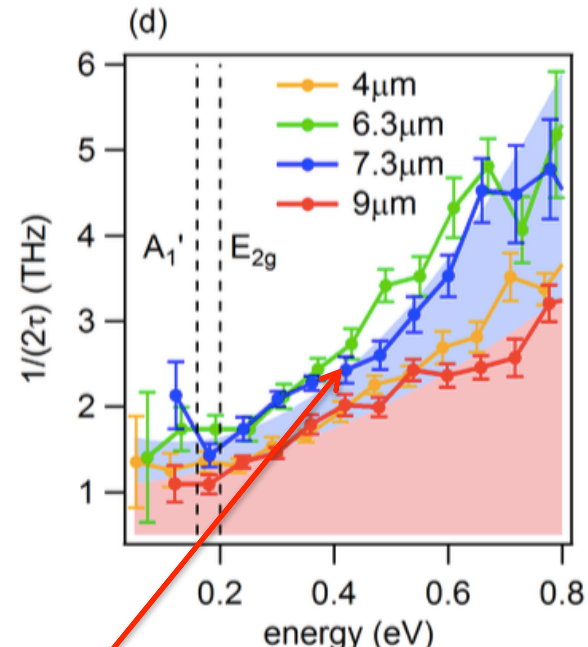
transient reduction of **THz Drude weight**

accelerated **tr-ARPES relaxation**





driving on phonon resonance: 3-fold enhancement of effective $\lambda_{el\text{-}ph}$

Quantum nonlinear phononics

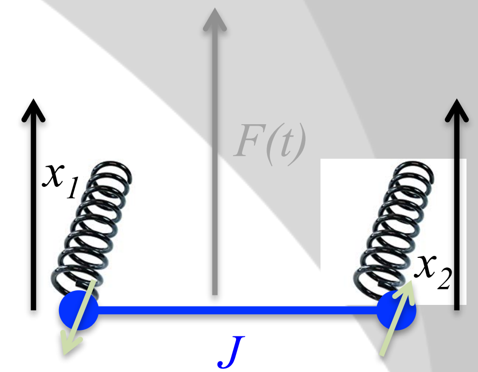
PRB 95, 205111 (2017)

2-site toy model, solve dynamics exactly

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

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

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

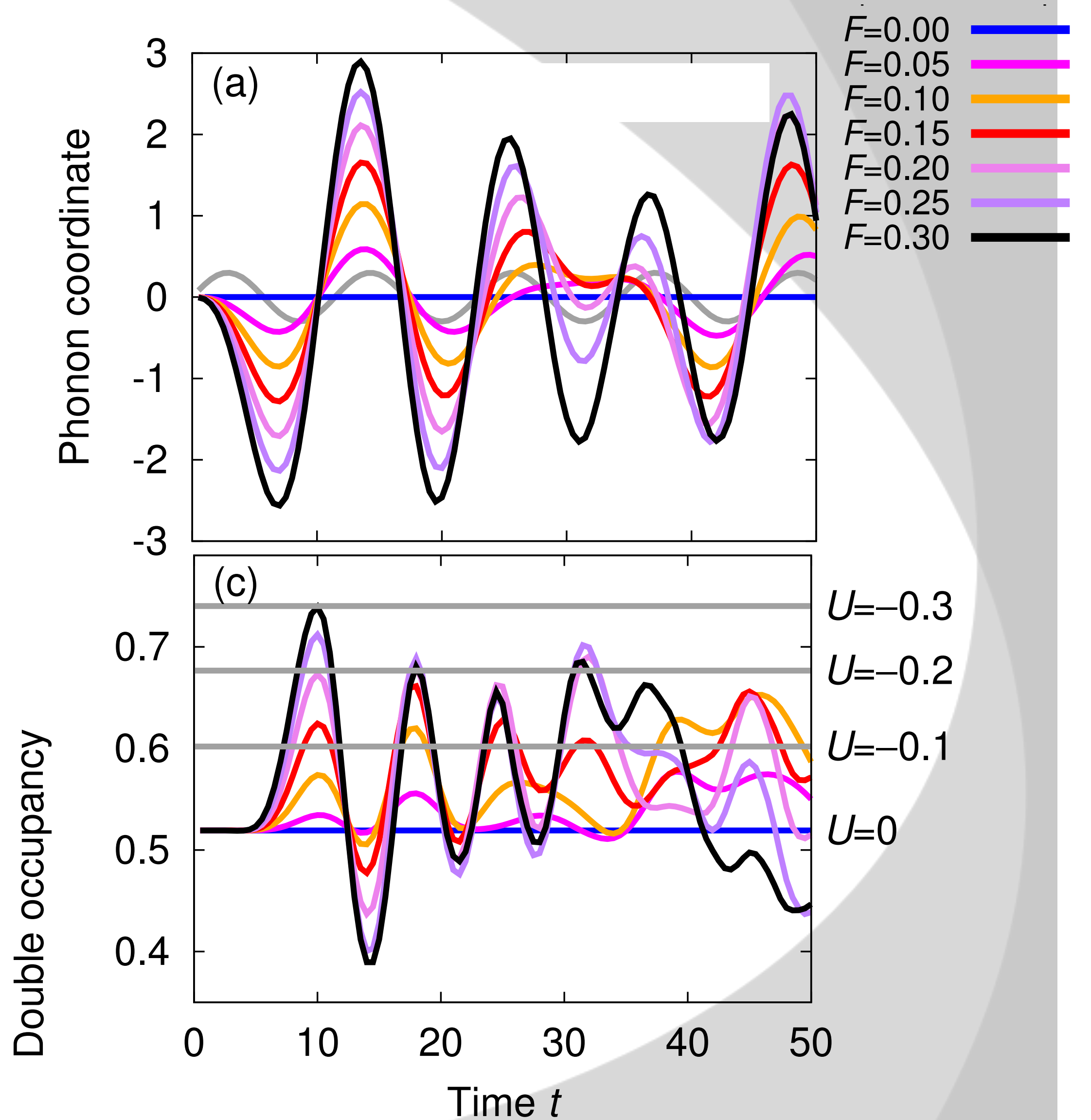
$$I(\omega, t_0) = \mathrm{Re}\int dt_1\, dt_2\, e^{i\omega(t_1-t_2)} s_{t_1,t_2,\tau}(t_0)$$
$$\times\Big[\langle\psi(t_2)|c^{\dagger}_{1,\uparrow}\mathcal{T}e^{-i\int_{t_1}^{t_2}H(t)dt}c_{1,\uparrow}|\psi(t_1)\rangle + $$
$$+ \langle\psi(t_1)|c_{1,\uparrow}\mathcal{T}e^{-i\int_{t_2}^{t_1}H(t)dt}c^{\dagger}_{1,\uparrow}|\psi(t_2)\rangle\Big],$$

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

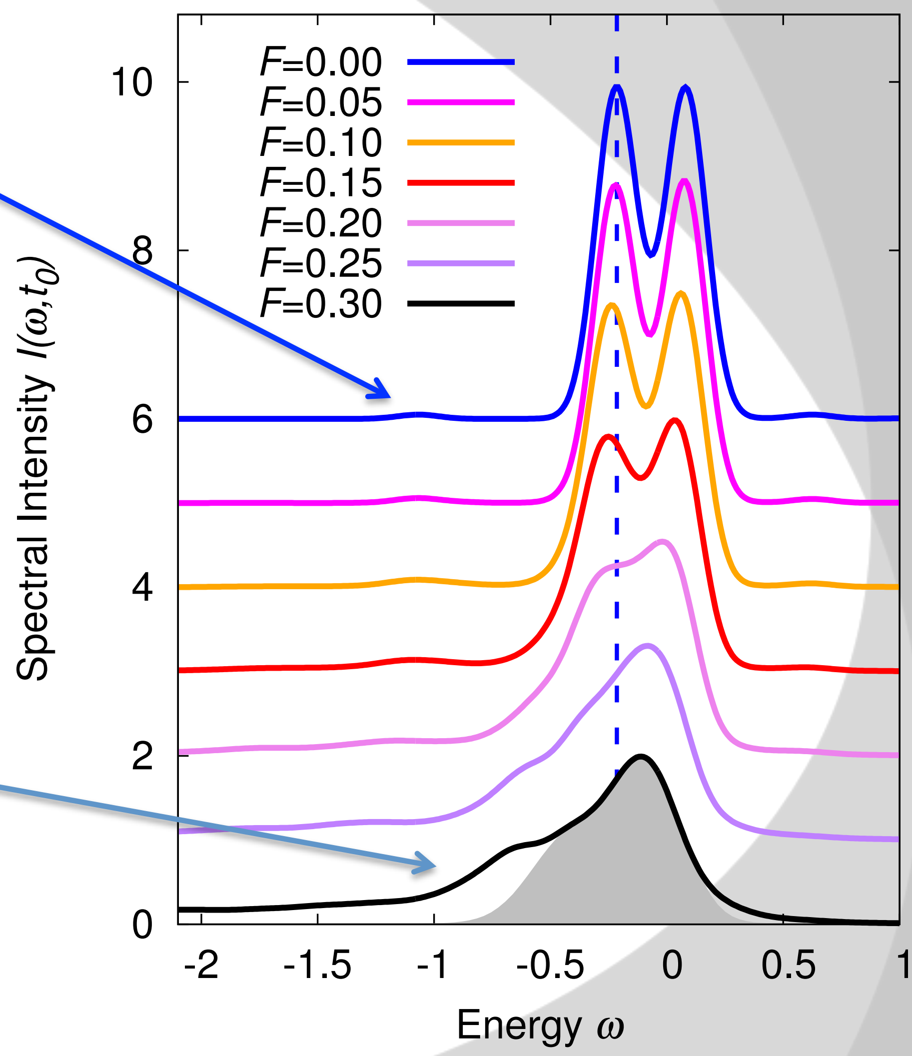
PRB 95, 205111 (2017) mpsd

2-phonon shakeoff

Reduced coherence peaks with stronger driving

light-enhanced el-ph coupling

light-induced polaron formation



Field dependence

PRB 95, 205111 (2017)

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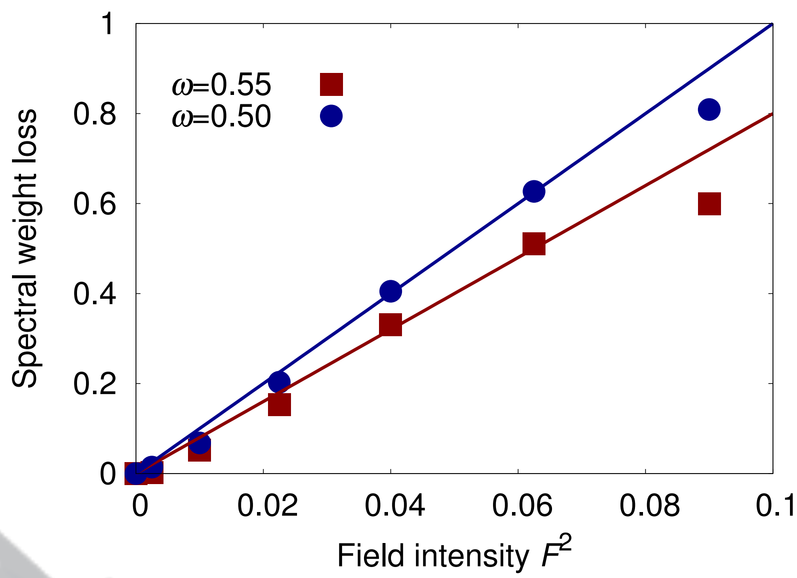
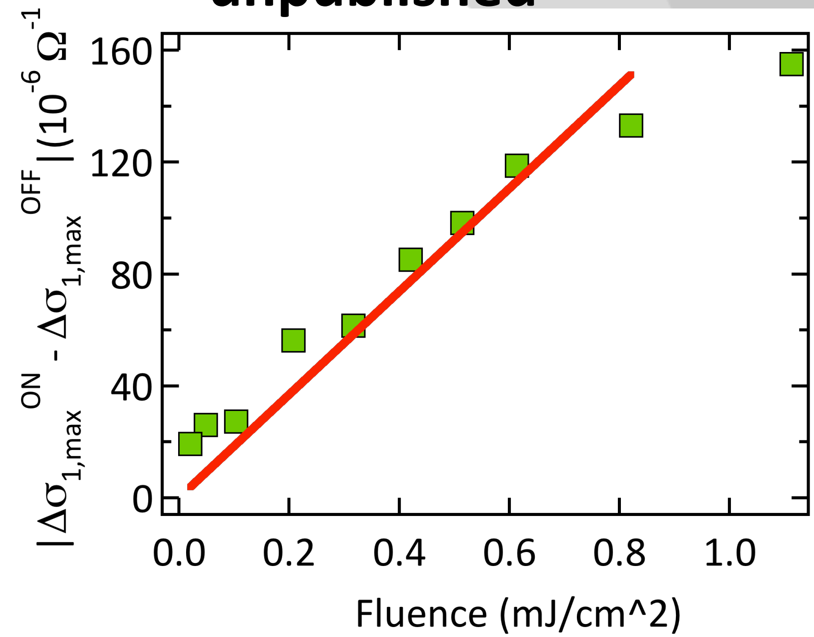
Theory



Data by E. Pomarico, unpublished



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

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)

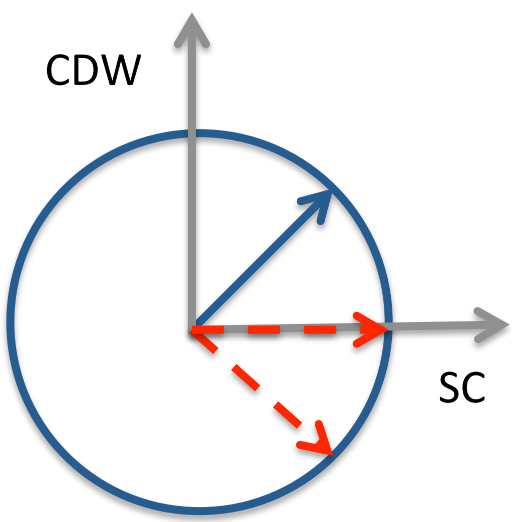
II Optical control of Majoranas

- prior work: optical control of competing orders

Theory of Laser-Controlled Competing Superconducting and Charge Orders

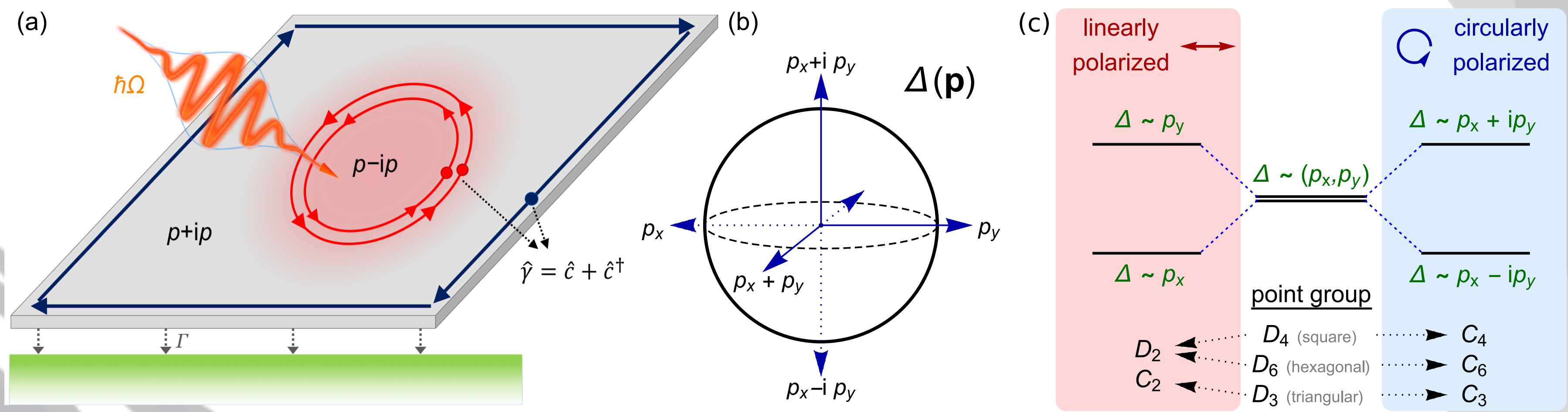
M. A. Sentef, A. Tokuno, A. Georges, and C. Kollath
Phys. Rev. Lett. **118**, 087002 – Published 21 February 2017

  – selective laser driving switches between phases



II Optical control of Majoranas

- can one switch the chirality of a 2D topological superconductor?



key idea: use two-pulse sequence with linearly and circularly polarized light

Model and Method

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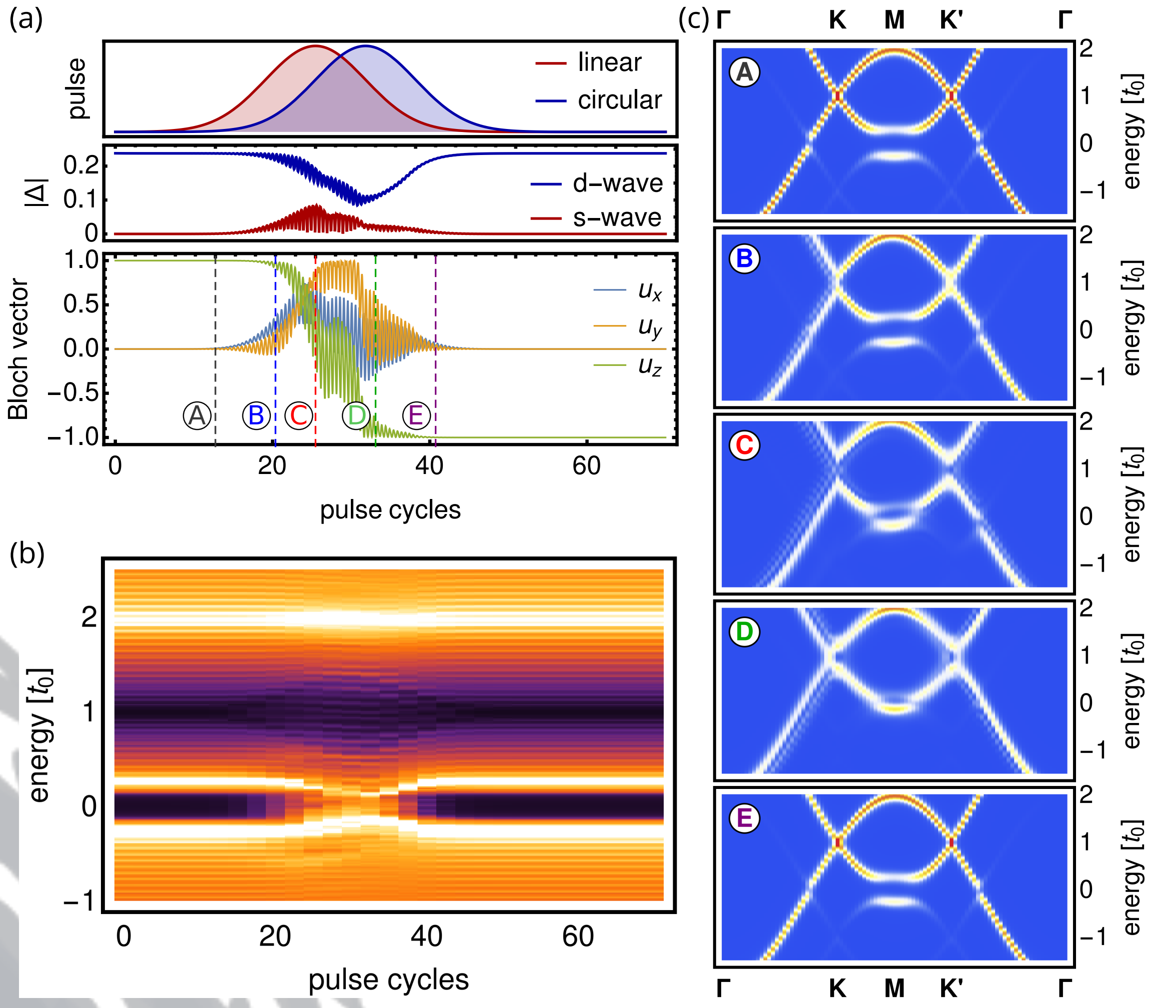
multiband Bogoliubov-de-Gennes Hamiltonians for doped graphene (d+id) and Sr2RuO4 (p+ip)
coupling to fermionic reservoir to dissipate energy
laser driving via Peierls substitution

Keldysh equations of motion for Nambu Green's functions:

$$i\partial_t \mathcal{G}_{\mathbf{k}}(t,t') = \mathcal{H}_{\mathbf{k}}(t, \mathbf{\Delta}_{\mathbf{k}}(t))\ \mathcal{G}_{\mathbf{k}}(t,t') + \int d\tau\ \hat{\Sigma}_{\mathbf{k}}(t,\tau)\ \mathcal{G}_{\mathbf{k}}(\tau,t')$$

$$\mathbf{\Delta}_{\mathbf{k}}(t) = \frac{1}{L}\sum_{j} v^{(j)} \hat{\boldsymbol{\eta}}^{(j)}_{\mathbf{k}} \sum_{\substack{\mathbf{k}' \\ \alpha\beta}} \hat{\eta}^{(j)}_{\mathbf{k}'\alpha\beta} \left\langle \hat{c}_{-\mathbf{k}',\beta\downarrow} \hat{c}_{\mathbf{k}',\alpha\uparrow} \right\rangle$$

Optical control of Majoranas



two-pulse sequence reverses d+id state in graphene

time-resolved spectroscopy tracks chirality reversal

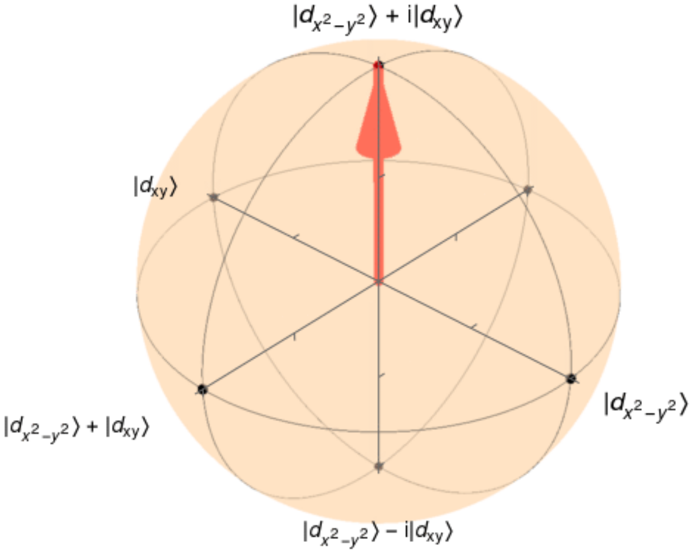
Summary II

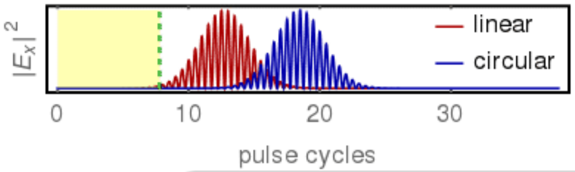
- All-optical control of chiral Majorana modes
- towards arbitrarily programmable quantum computer?

M. Claassen et al., arXiv:1810.06536, submitted to Nat. Phys.

M. Claassen

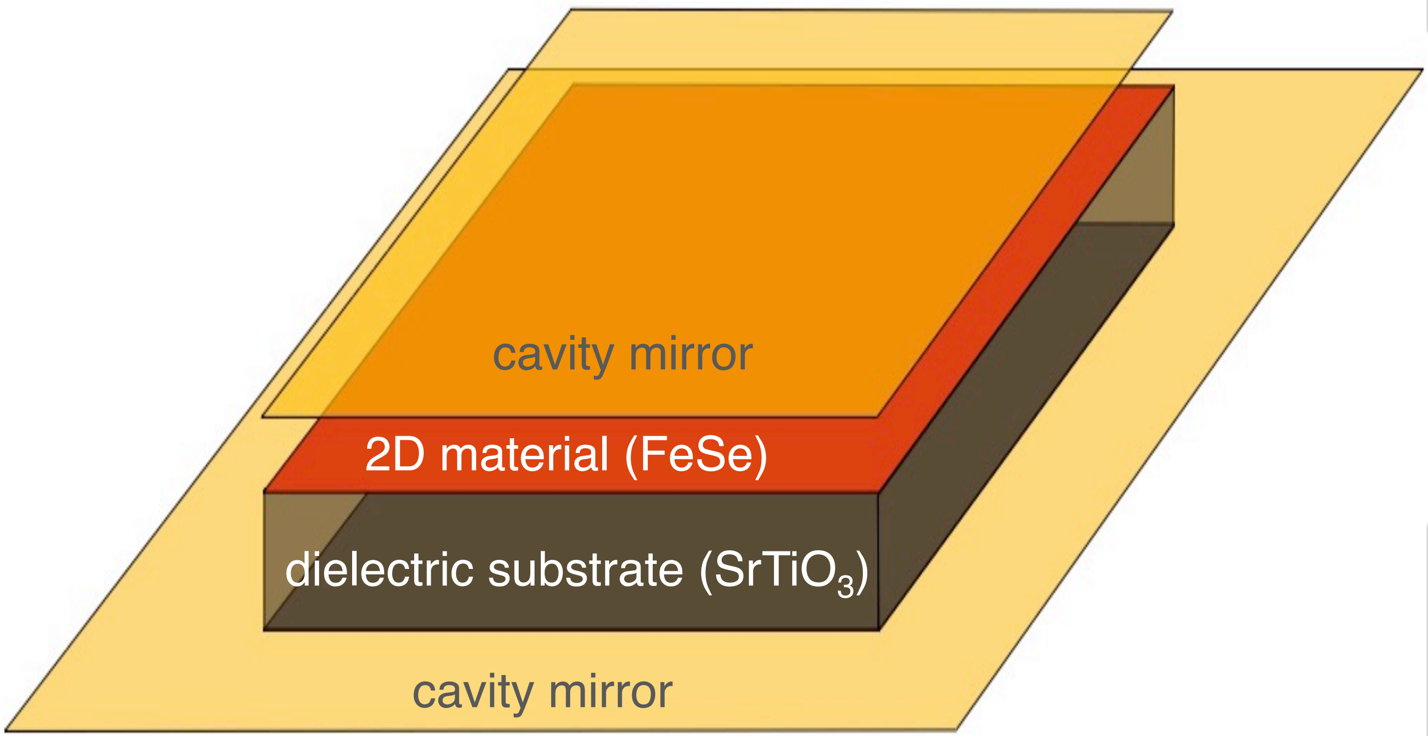
D. Kennes





III Cavity materials

- can one use enhanced vacuum fluctuations to change materials properties?



Cavity materials

mpsd

BCS superconductors: phonon-mediated superconductivity
Ginzburg, Phys. Lett. 13, 101 (1964): exciton-mediated superconductivity?
Ruvalds, Phys. Rev. B 35, 8869(R) (1987): plasmon-mediated superconductivity?

PRL 104, 106402 (2010) PHYSICAL REVIEW LETTERS week ending 12 MARCH 2010

Exciton-Polariton Mediated Superconductivity

Fabrice P. Laussy,[1] Alexey V. Kavokin,[1,2] and Ivan A. Shelykh[3,4]

PHYSICAL REVIEW B 93, 054510 (2016)

Superconductivity and other collective phenomena in a hybrid Bose-Fermi mixture formed by a polariton condensate and an electron system in two dimensions

Ovidiu Cotleţ,[1,*] Sina Zeytinoğlu,[1,2] Manfred Sigrist,[2] Eugene Demler,[3] and Ataç Imamoğlu[1]

Cavity-assisted mesoscopic transport of fermions: Coherent and dissipative dynamics.

Hagenmüller et al., 1801.09876

Cavity quantum-electrodynamical polaritonically enhanced electron-phonon coupling and its influence on superconductivity

M. A. Sentef,[1,*] M. Ruggenthaler,[1] and A. Rubio[1,2,3]

1802.09437

Cavity-mediated electron-photon superconductivity

Frank Schlawin[1], Andrea Cavalleri[1,2] and Dieter Jaksch[1]

1804.07142

Superradiant Quantum Materials

Giacomo Mazza[1,2,*] and Antoine Georges[2,3,1,4]

1804.08534

Cavity Quantum Eliashberg Enhancement of Superconductivity

Jonathan B. Curtis,[1,2,*] Zachary M. Raines,[1,2] Andrew A. Allocca,[1,2] Mohammad Hafezi,[1] and Victor M. Galitski[1,2]

1805.01482

Manipulating quantum materials with quantum light

Martin Kiffner[1,2], Jonathan Coulthard[2], Frank Schlawin[2], Arzhang Ardavan[2], and Dieter Jaksch[2,1]

1806.06752

Cavity superconductor-polaritons

Andrew A. Allocca,* Zachary M. Raines, Jonathan B. Curtis, and Victor M. Galitski

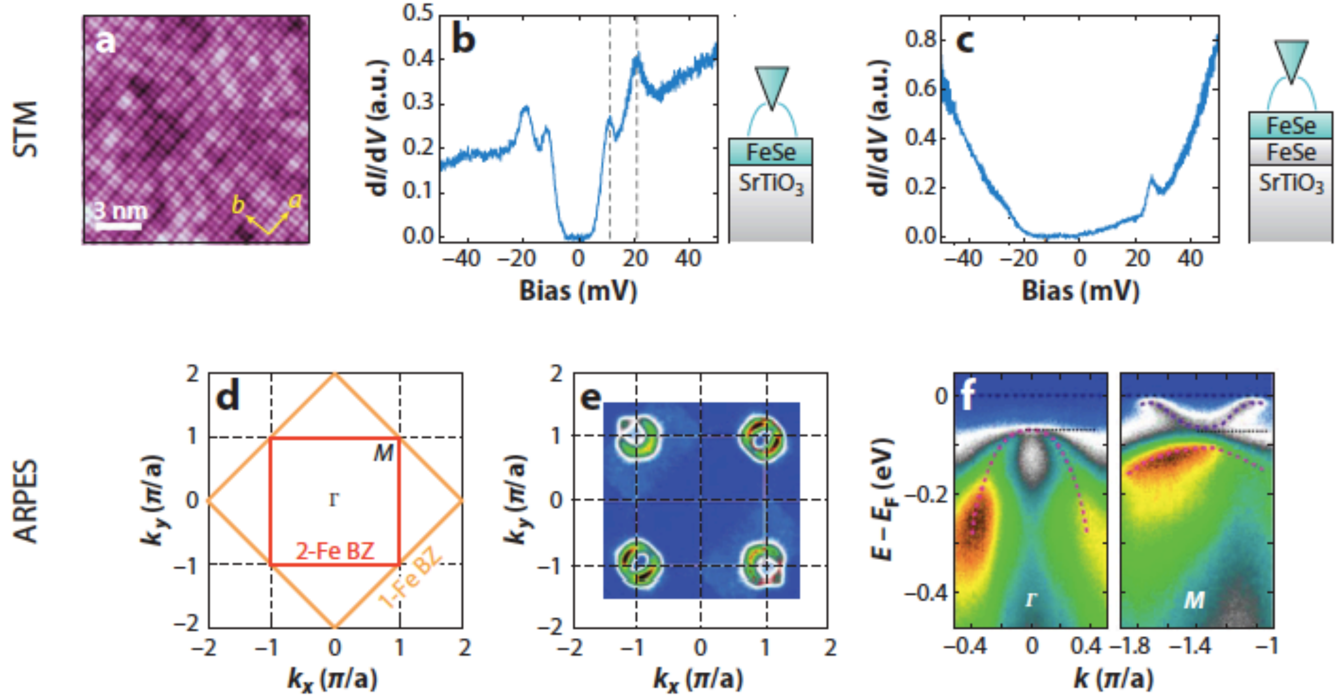
1807.06601

Ab-initio Exciton-polaritons:
Cavity control of Dark Excitons in two dimensional Materials

Simone Latini,[1,*] Enrico Ronca,[1,†] Umberto De Giovannini,[1,2,‡] Hannes Hübener,[1,§] and Angel Rubio[1,3,¶]

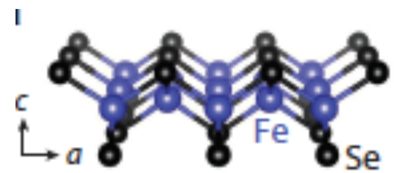
1810.02672

monolayer FeSe/STO



monolayer FeSe/STO: Tc > 65 K

bulk FeSe: Tc = 9 K



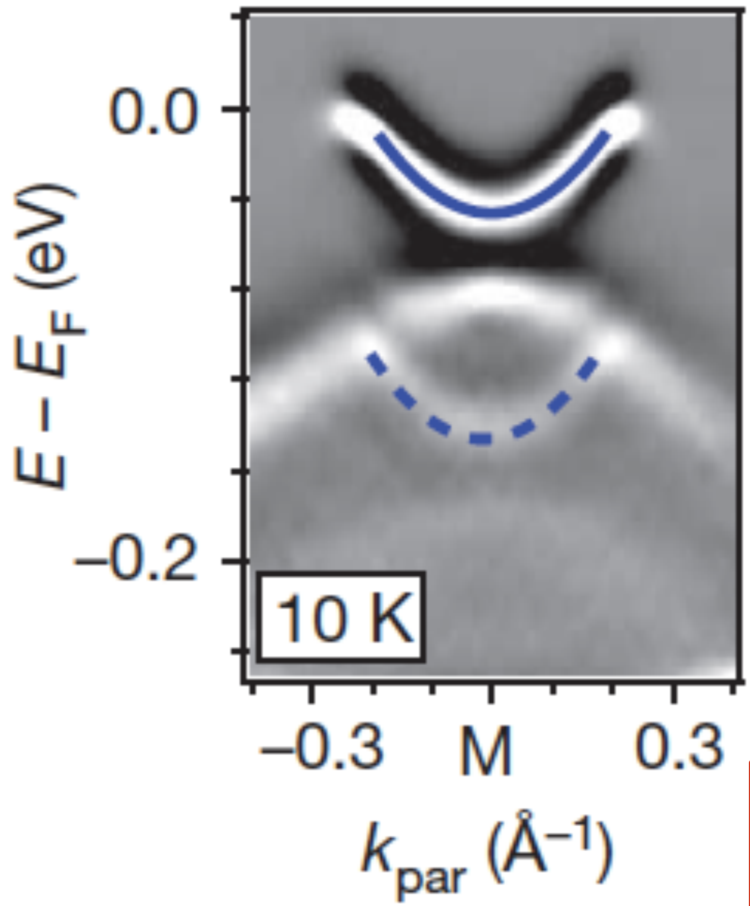
Wang QY, Li Z, Zhang WH, Zhang ZC, Zhang JS, et al. 2012. *Chin. Phys. Lett.* 29:037402

Liu D, Zhang W, Mou D, He J, Ou YB, et al. 2012. *Nat. Commun.* 3:931

Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)

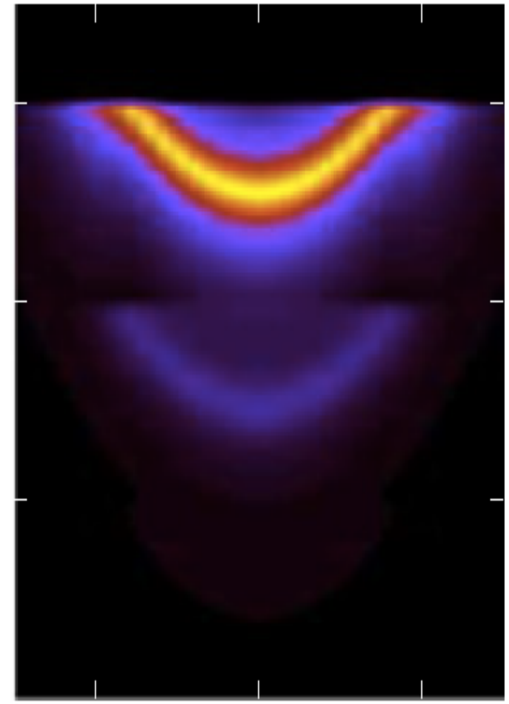
monolayer FeSe/STO: ARPES

experiment



Lee et al., Nature 515, 245 (2014)

theory



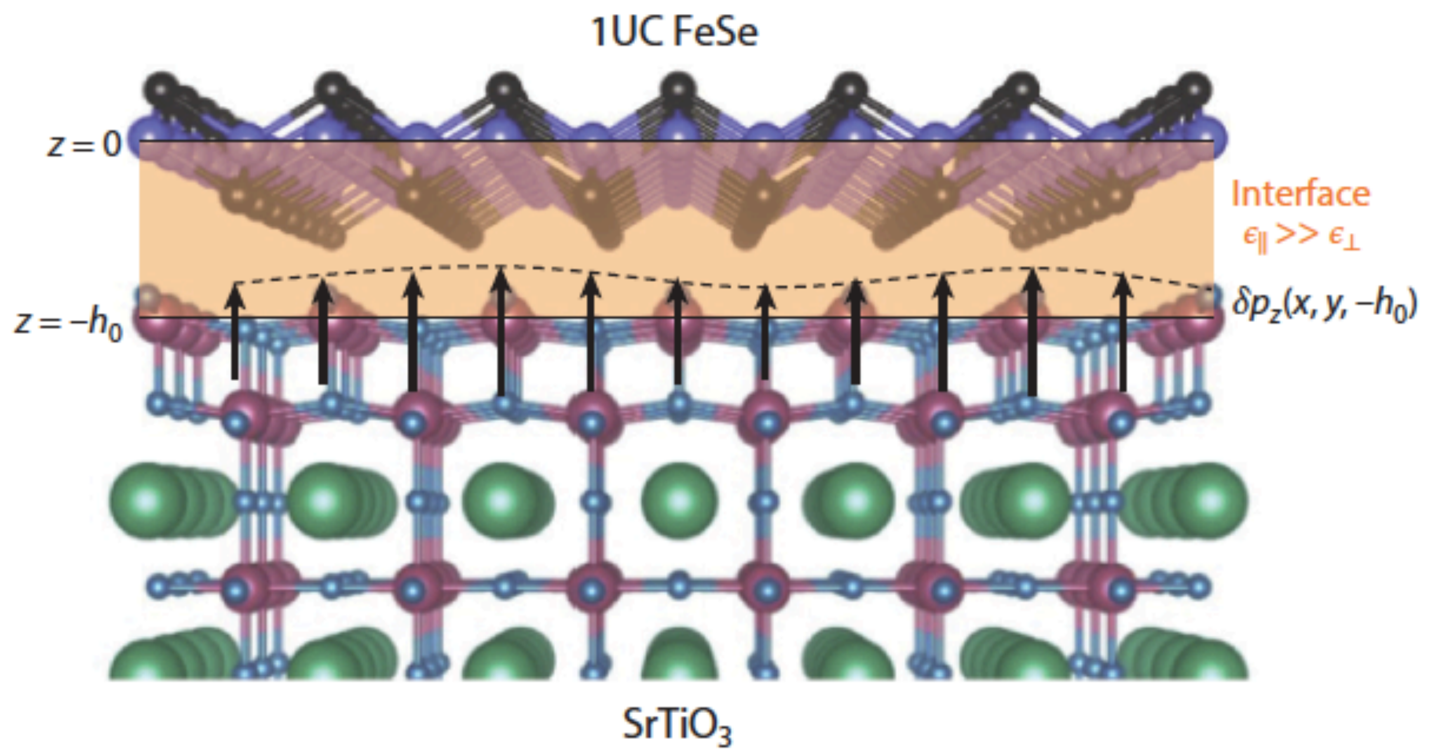
replica bands: forward (small-q) electron-phonon scattering

Rademaker et al., New J. Phys. 18, 022001 (2016)

monolayer FeSe/STO: interfacial phonon

bare el-phonon vertex $g(\vec{q}) = g_0 \exp(-|\vec{q}|/q_0)$ *Lee et al., Nature 515, 245 (2014)*

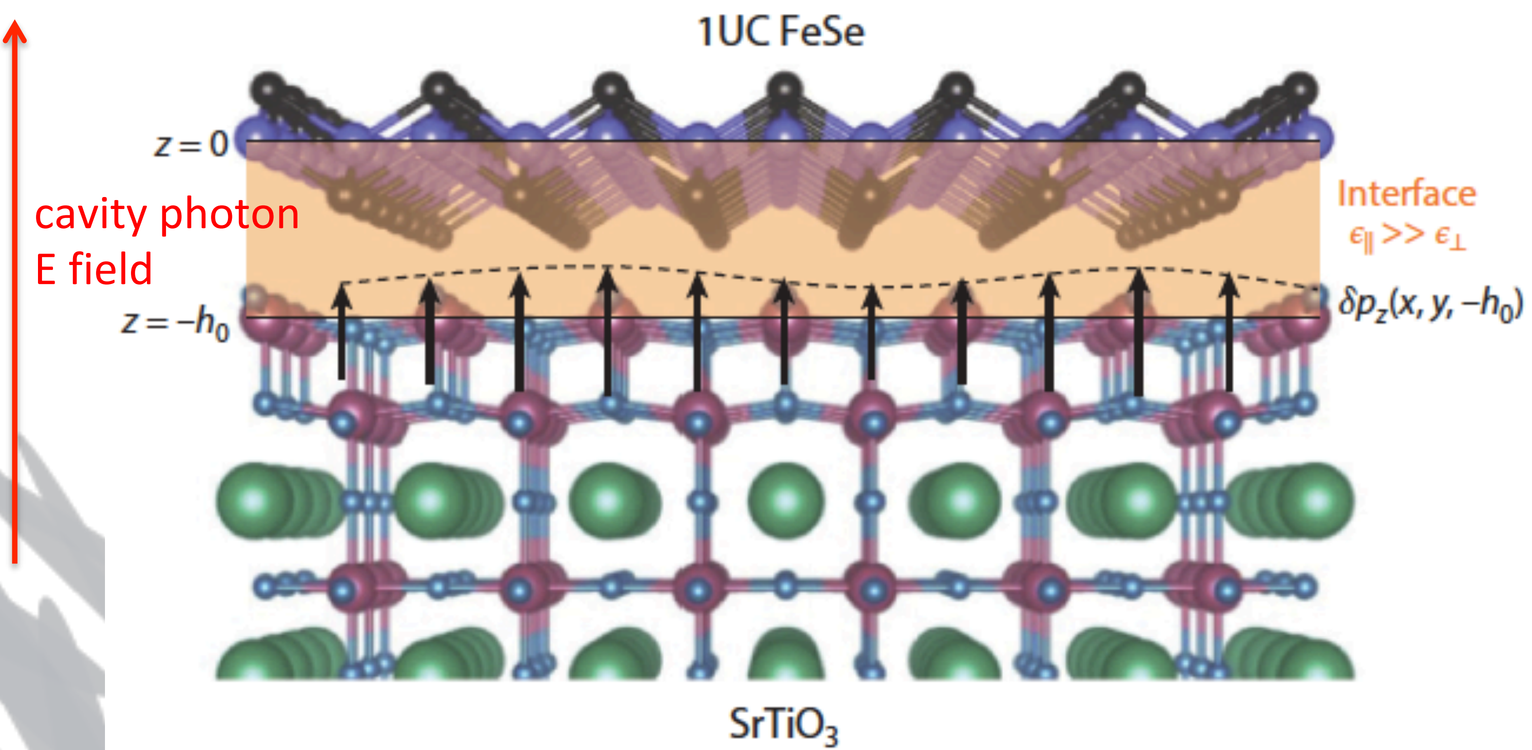
$$q_0^{-1} = h_0\sqrt{\epsilon_{\|}/\epsilon_{\perp}} \quad \epsilon_{\|}/\epsilon_{\perp} \approx 100$$



Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)

Cavity engineering

- idea: use phonon polaritons to enhance electron-phonon coupling



Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)

Model and Method

mpsd

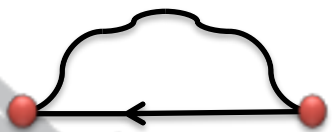
electrons — el-polariton coupling — polaritons

$$H = \sum_{\vec{k},\sigma} \epsilon_{\vec{k}} c^\dagger_{\vec{k},\sigma} c_{\vec{k},\sigma} + \frac{1}{\sqrt{N}} \sum_{\vec{k},\vec{q},\sigma,\lambda=\pm} c^\dagger_{\vec{k}+\vec{q},\sigma} c_{\vec{k},\sigma} (g^*_\lambda(\vec{q}) \alpha^\dagger_{-\vec{q},\lambda} + g_\lambda(\vec{q}) \alpha_{\vec{q},\lambda}) + \sum_{\vec{q},\lambda=\pm} \omega_\lambda(\vec{q}) \alpha^\dagger_{\vec{q},\lambda} \alpha_{\vec{q},\lambda}$$

bare el-phonon vertex $g(\vec{q}) = g_0 \exp(-|\vec{q}|/q_0)$ $\quad q_0^{-1} = h_0 \sqrt{\epsilon_\| / \epsilon_\perp}$

G-self-consistent Migdal-Eliashberg diagram

$$\hat{\Sigma}(\vec{k}, i\omega_n) = \frac{-1}{N\beta} \sum_{\vec{q},m,\lambda=\pm} |g_\lambda(\vec{q})|^2 D^{(0)}_\lambda(\vec{q}, i\omega_n - i\omega_m) \hat{\tau}_3 \hat{G}(\vec{k}+\vec{q}, i\omega_m) \hat{\tau}_3$$

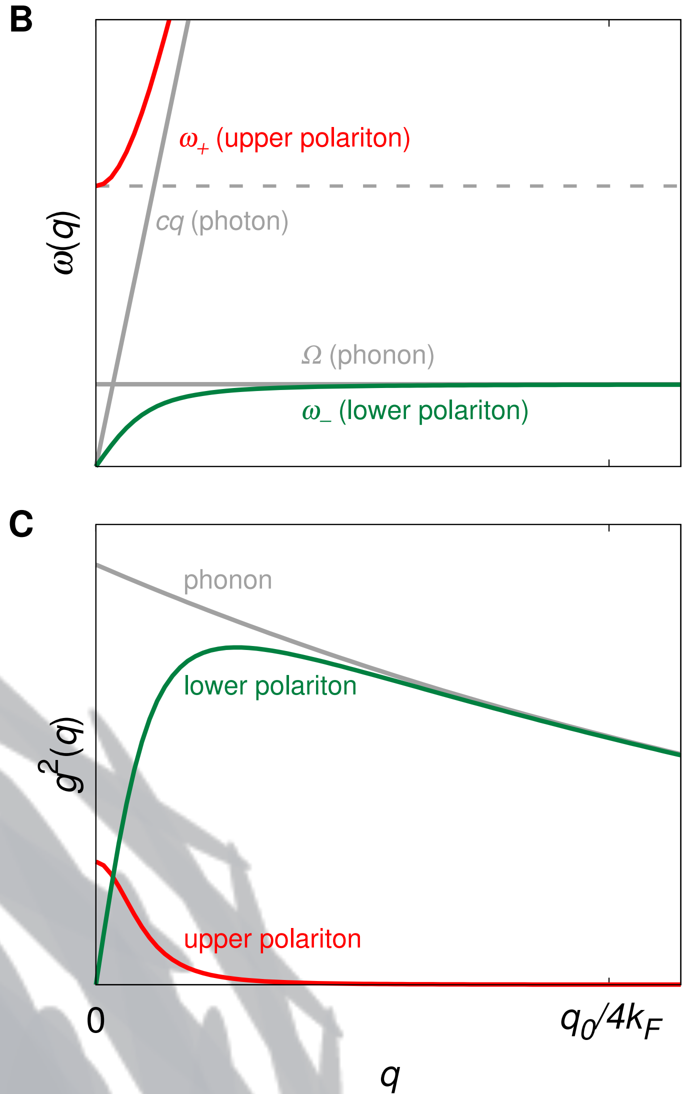


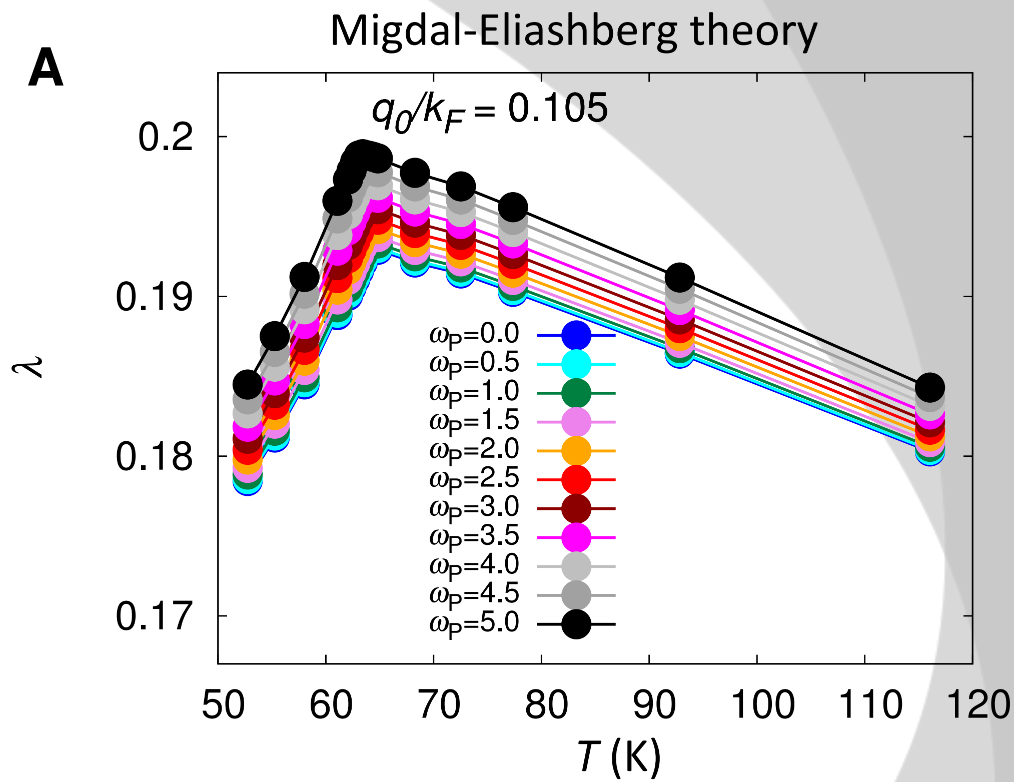
$$\hat{\Sigma}(\vec{k}, i\omega_n) = i\omega_n [1 - Z(\vec{k}, i\omega_n)] \hat{\tau}_0 + \chi(\vec{k}, i\omega_n) \hat{\tau}_3 + \phi(\vec{k}, i\omega_n) \hat{\tau}_1$$

$$\lambda \equiv Z(\vec{k}_F, i\pi/\beta) - 1$$

Mass enhancement: $m^*/m = 1 + \lambda$

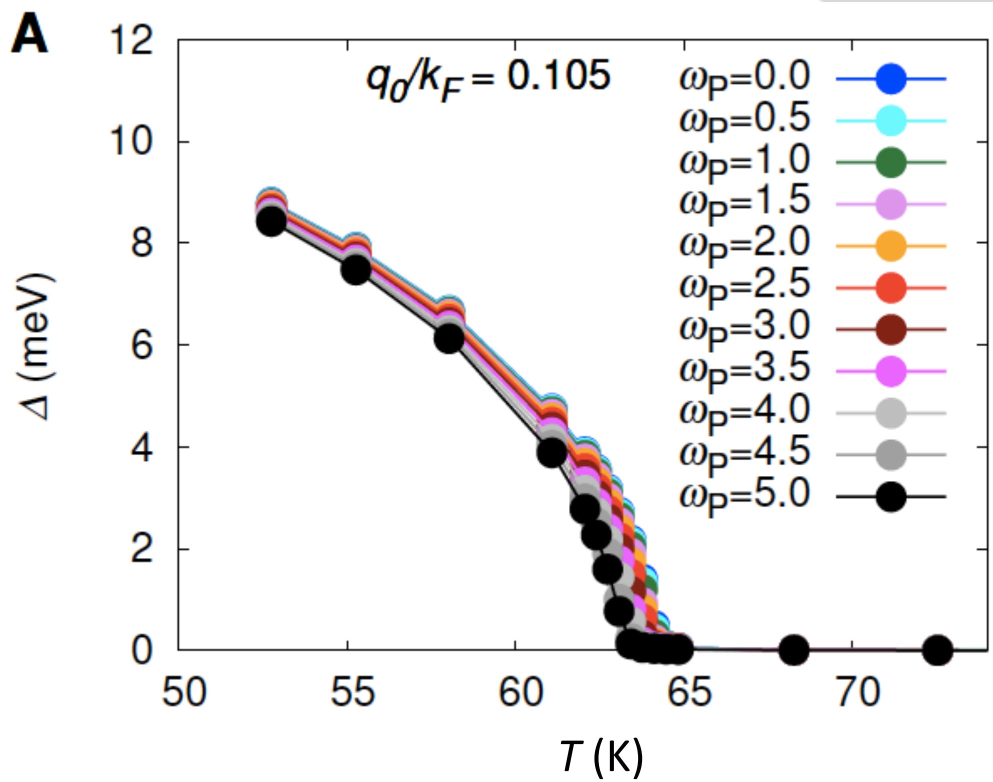
Cavity materials: Phonon polaritons





enhanced electron-phonon coupling, controlled by cavity volume

Superconductivity



suppressed superconductivity despite enhanced el-ph coupling

forward scattering

$$T_C \approx \frac{\lambda\Omega}{2 + 3\lambda}$$

vs.

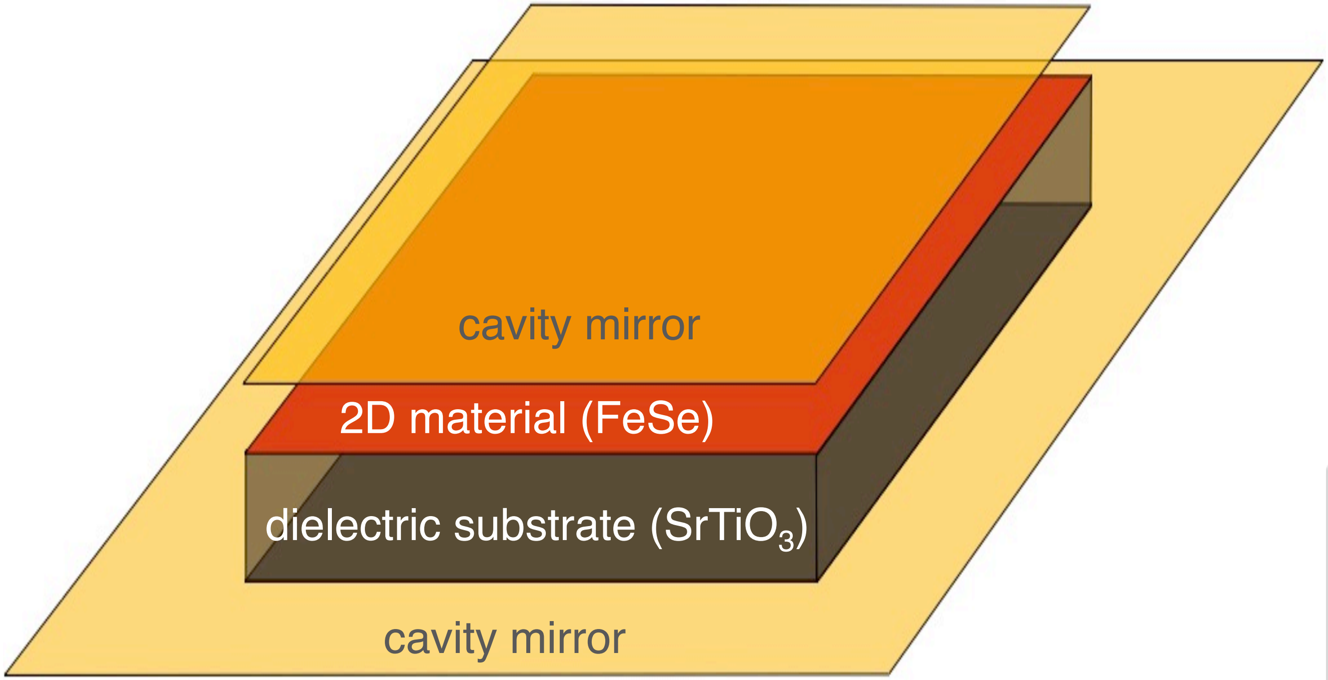
$$T_{C,\mathrm{BCS}} \approx 1.13\Omega \exp(-\tfrac{1}{\lambda})$$

q-independent scattering

Summary III

- cavity leads to enhanced electron-phonon coupling
- can one also enhance superconductivity?

M. A. Sentef, M. Ruggenthaler, A. Rubio, arXiv:1802.09437 (to appear in Science Adv.)

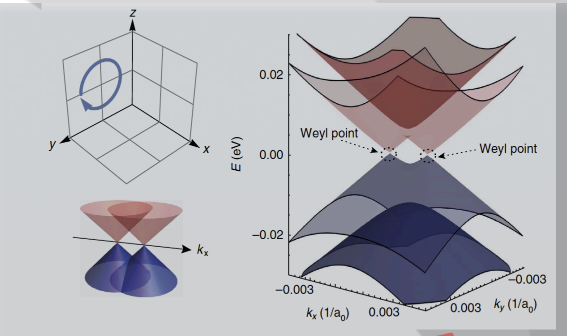


Summary

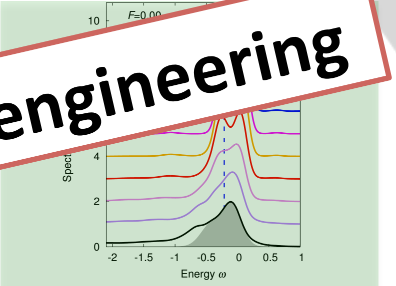
mpsd

Ultrafast laser engineering of

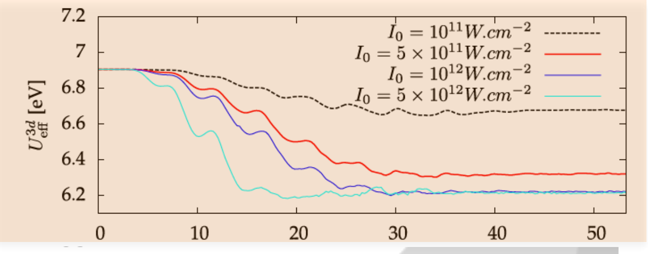
- band structure, topology (Floquet)
  Nature Commun. 6, 7047 (2015)
  Nature Commun. 8, 13940 (2017)
  arXiv:1803.07447



- electron-phonon coupling
  PRB 95, 024304 (2017)
  PRB 95, 205111 (2017)
  arXiv:1802.09437

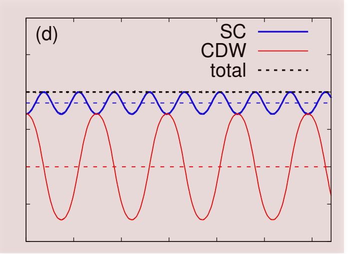


- Hubb … subresonant … tions in correlated insulators)
  PRL 121, 097402 (2018)



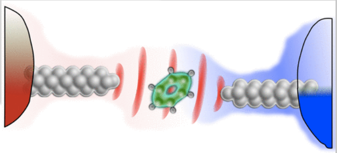
Towards nonequilibrium materials engineering

- ordered phases
  PRB 92, 224517 (2015)
  PRB 93, 144506 (2016)
  PRL 118, 087002 (2017)
  arXiv:1806.08187
  arXiv:1808.00712
  arXiv:1808.04655
  arXiv:1810.06536

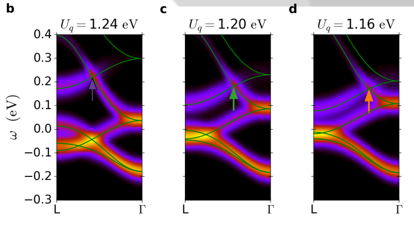


Outlook: Group projects

R. Tuovinen (postdoc): nonequilibrium Green‘s functions (GKBA) for time-resolved transport and excitonic condensates *(JCTC 14, 2495 (2018); arXiv:1808.00712)*



G. Topp (PhD student): modeling time-resolved spectroscopy, electron-lattice dynamics *(arXiv:1803.07447, Nature Comm.)*



S. Ramirez (PhD student) light-induced Majoranas

M. Kalthoff (PhD student) time-dependent matrix product states (t-DMRG) for Floquet engineering of correlated systems (w/ D. Kennes, FU Berlin)

D. Hofmann (master student) topological exciton polaritons (master), machine learning for time-dependent variational wave functions (w/ G. Carleo, CCQ NYC)

X. Wang (student, Tsinghua) Green‘s functions for cavity 2D materials with focus on topology

M. Puviani (PhD st., Modena) quantum nonlinear phononics, ultrafast melting of ferrielectric charge-density wave *(arXiv:1806.08187, PRB)*