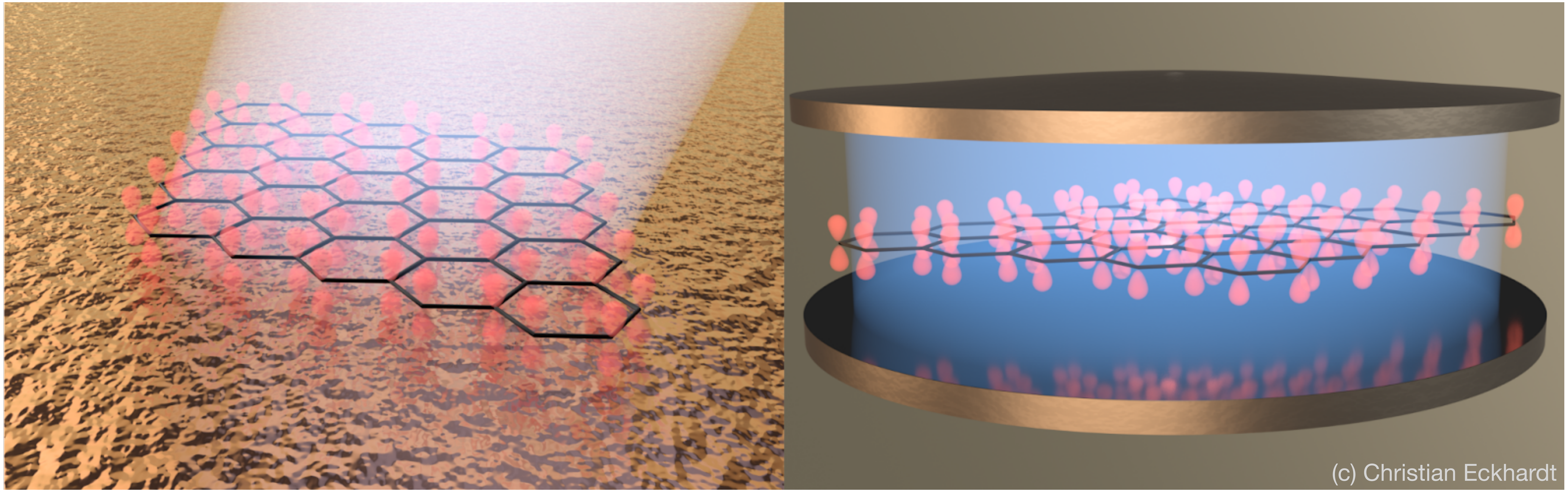
MAX PLANCK INSTITUTE
FOR THE STRUCTURE AND DYNAMICS OF MATTER

Light-matter control of quantum materials



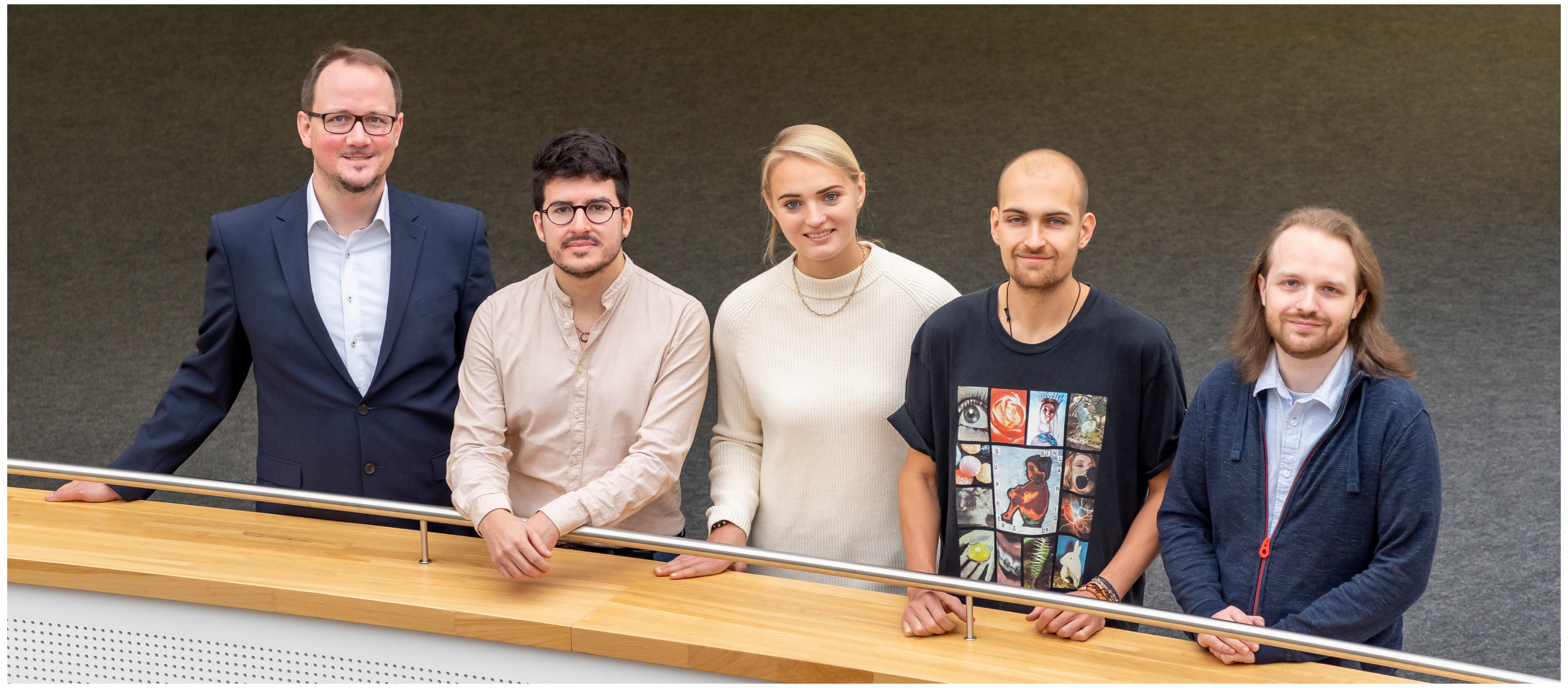
Michael A. Sentef
Theoretical Description of Pump-Probe Spectroscopies in Solids

MPSD Scientific Advisory Board Meeting
May 4, 2022

Emmy Noether-Programm
Deutsche Forschungsgemeinschaft
DFG

The group

- Funded through DFG Emmy Noether programme 2016 - 2022
- 1 postdoc, 3 PhD students
- Originally „Theoretical Description of Pump-Probe Spectroscopies in Solids“
- Now „Light-Matter Control of Quantum Materials“



REVIEWS OF MODERN PHYSICS

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Colloquium: Nonthermal pathways to ultrafast control in quantum materials

Alberto de la Torre, Dante M. Kennes, Martin Claassen, Simon Gerber, James W. McIver, and Michael A. Sentef
Rev. Mod. Phys. **93**, 041002 – Published 14 October 2021

Home > Applied Physics Reviews > Volume 9, Issue 1 > 10.1063/5.0083825

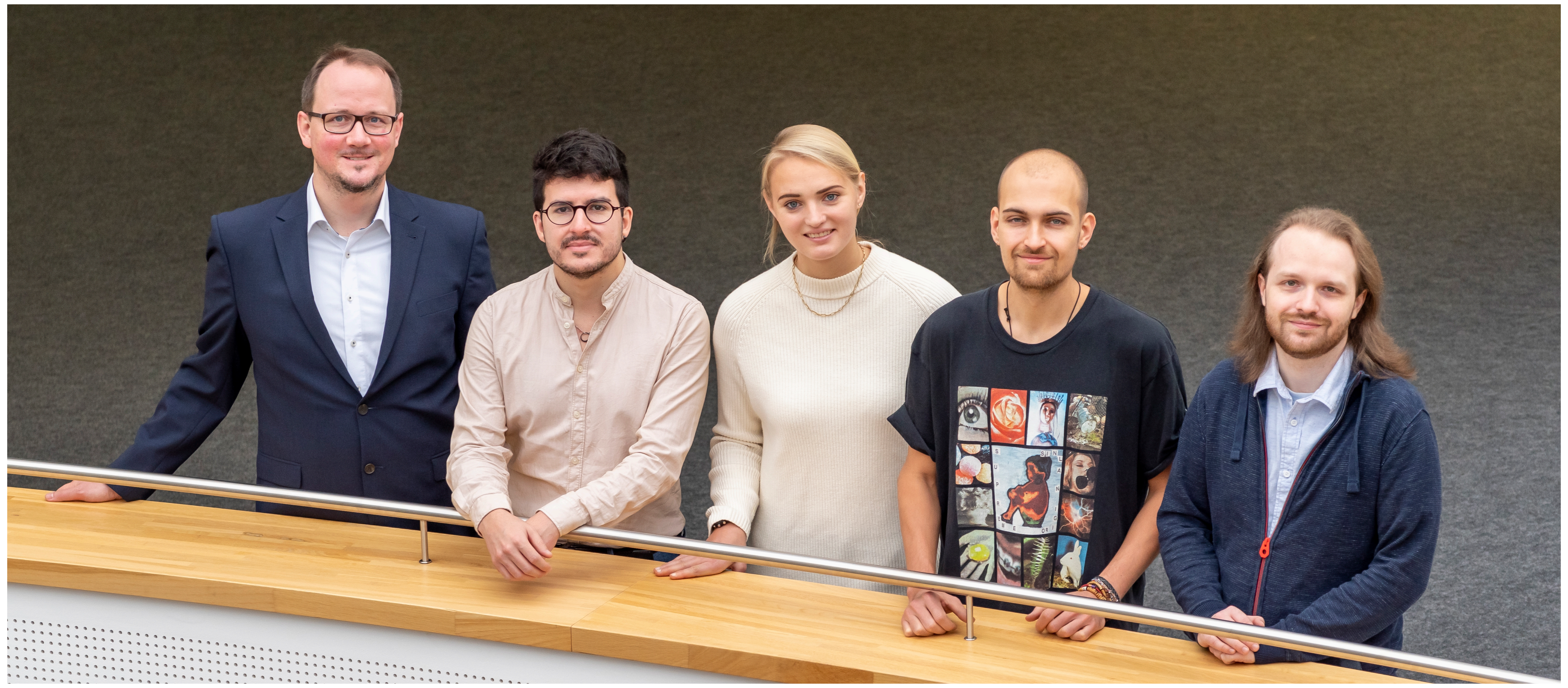
Open • Submitted: 30 December 2021 • Accepted: 31 January 2022 • Published Online: 25 February 2022

Cavity quantum materials F

Applied Physics Reviews 9, 011312 (2022); https://doi.org/10.1063/5.0083825

F. Schlawin[1,2], D. M. Kennes[1,3], *and* M. A. Sentef[1,a)]

The group



Projects:

Phd students

Mona Kalthoff Nonequilibrium phase transitions (Poster LMC1)
Damian Hofmann Neural network quantum states for dynamics (Poster LMC2)
Christian Eckhardt Cavity quantum materials (Poster LMC3)

Postdoc

José Pizarro Theory on-chip THz spectroscopy for moiré graphene (with McIver)

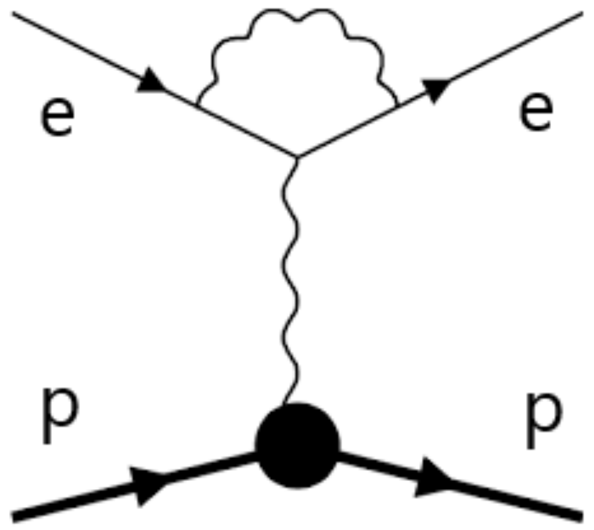
Outline

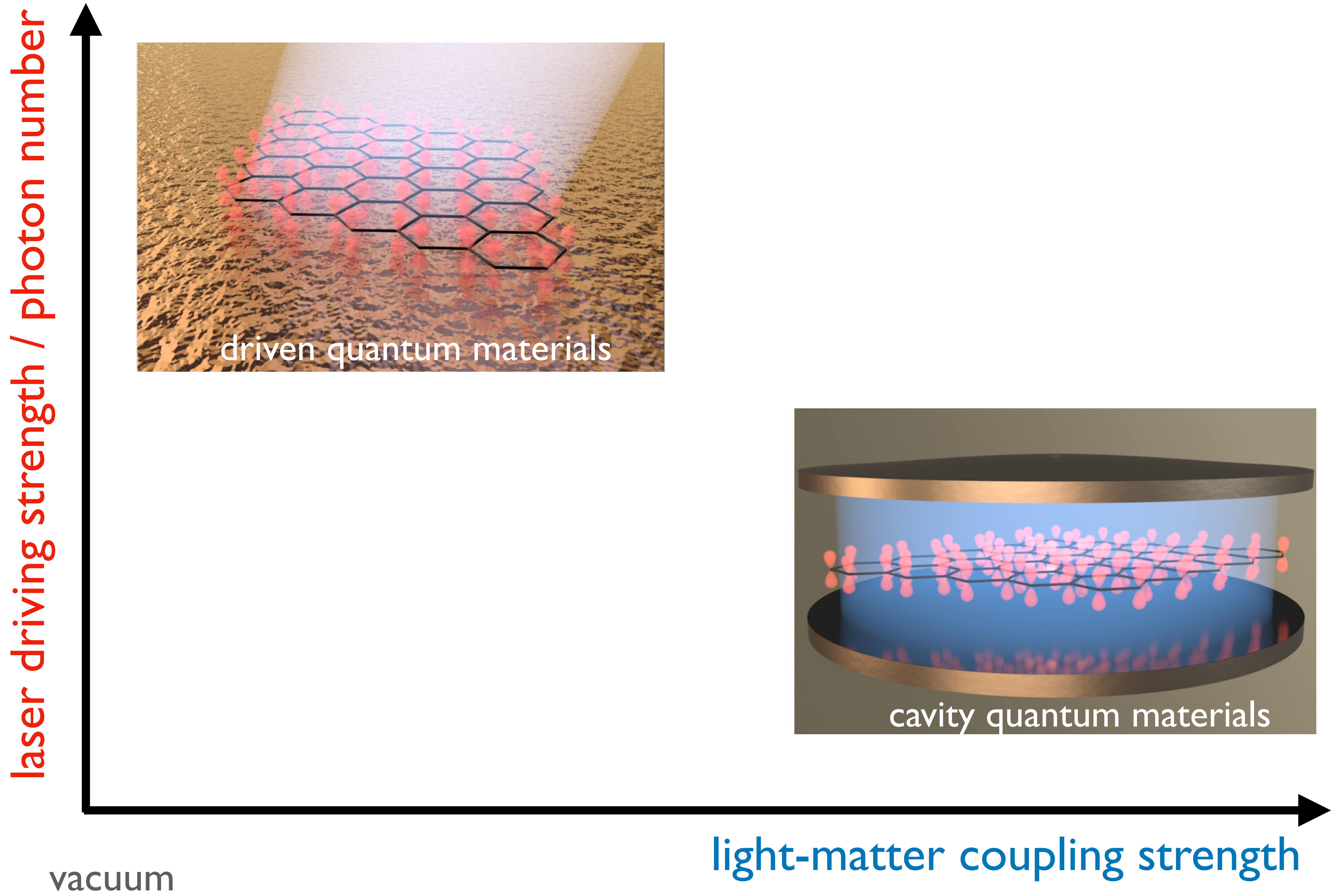
Dynamical Hubbard U

- … via optical excitation
- … via resonant phonon driving
- … via cavity embedding

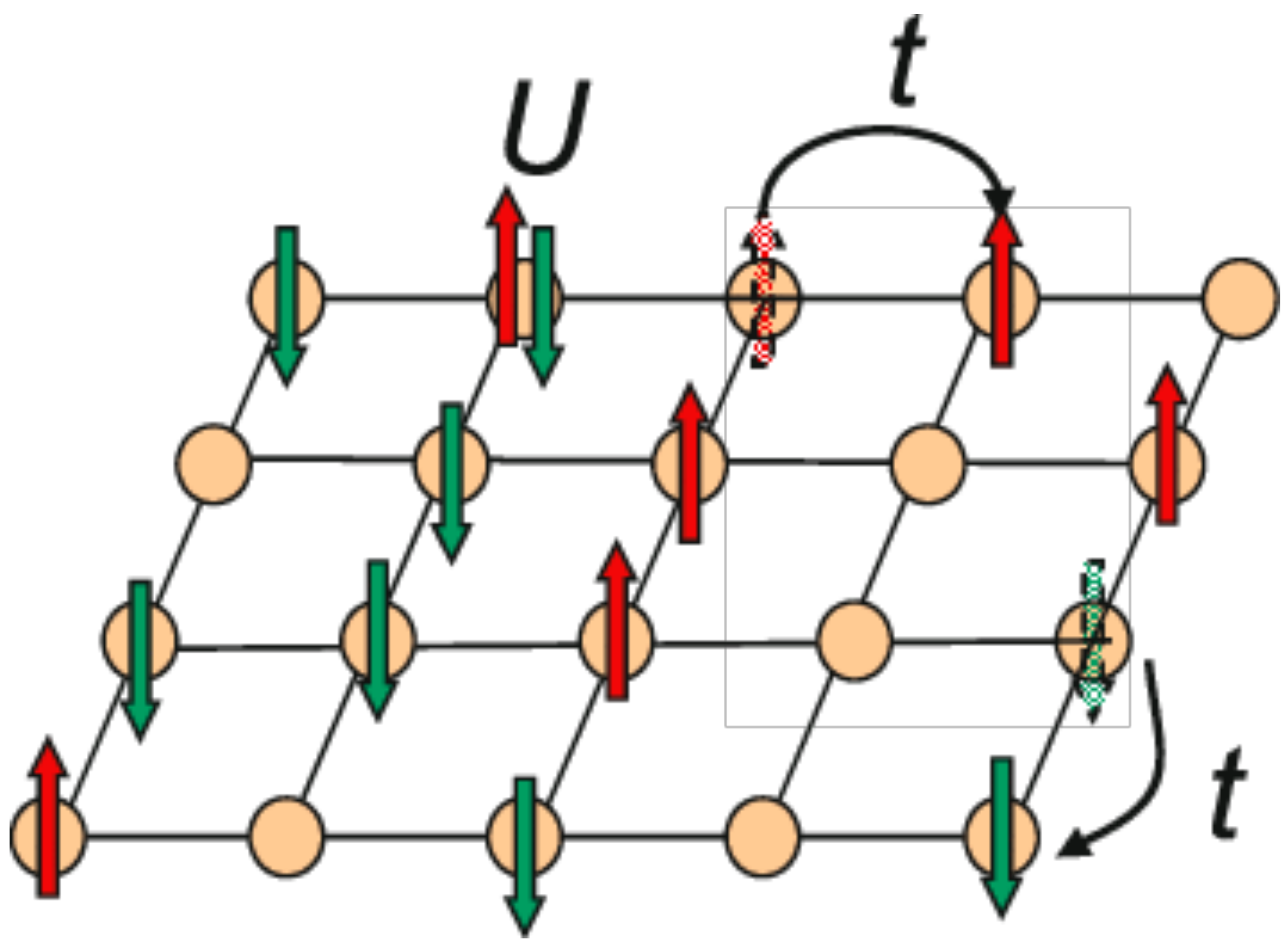
Can we employ light-matter interactions to change materials properties?

$$\alpha = \frac{1}{4\pi\varepsilon_0}\frac{e^2}{\hbar c} = \frac{\mu_0}{4\pi}\frac{e^2 c}{\hbar} = \frac{k_e e^2}{\hbar c} = \frac{e^2}{2\varepsilon_0 ch} = \frac{c\mu_0}{2R_K} = \frac{e^2 Z_0}{2h} = \frac{e^2 Z_0}{4\pi\hbar}$$

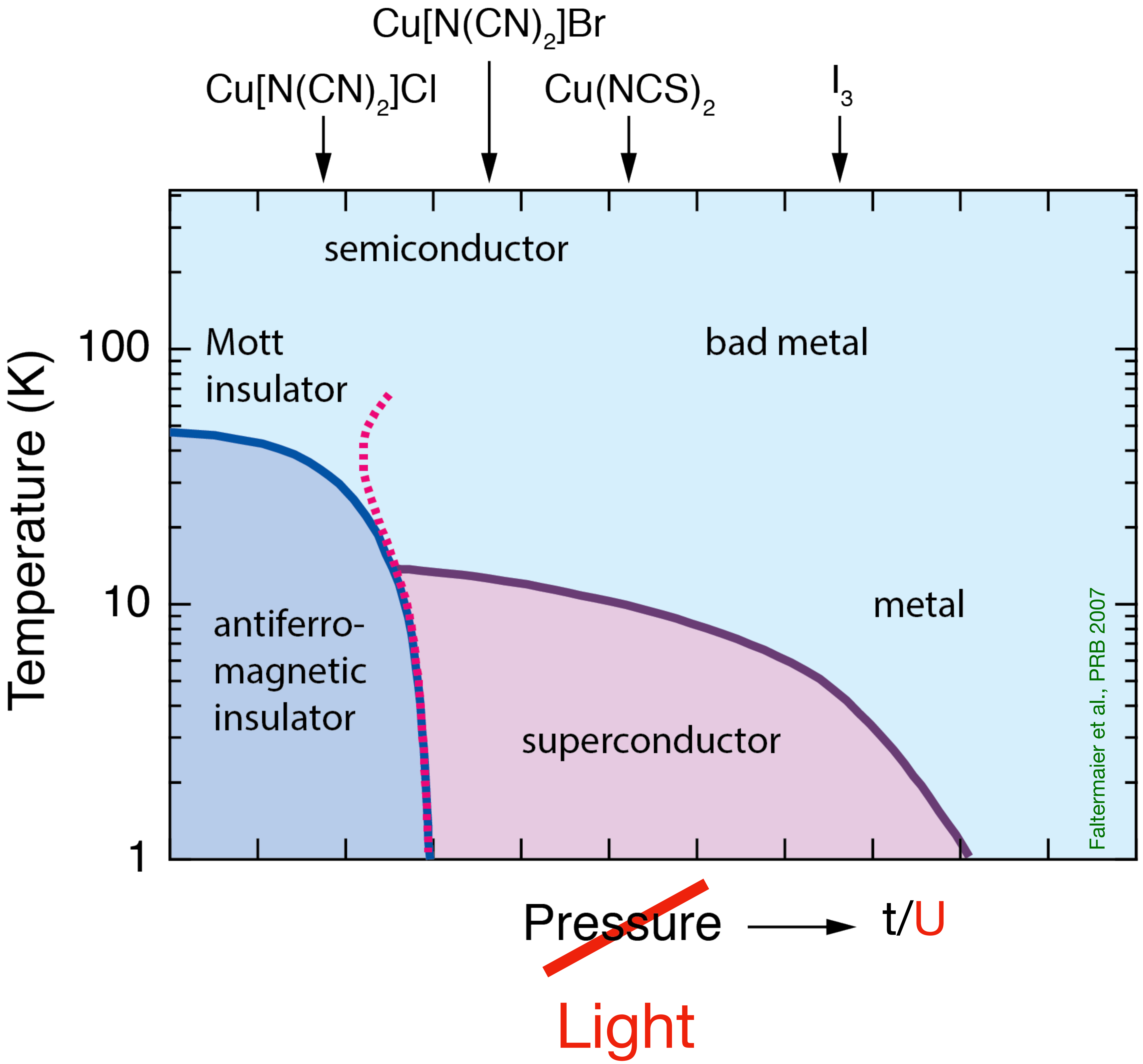




Hubbard model

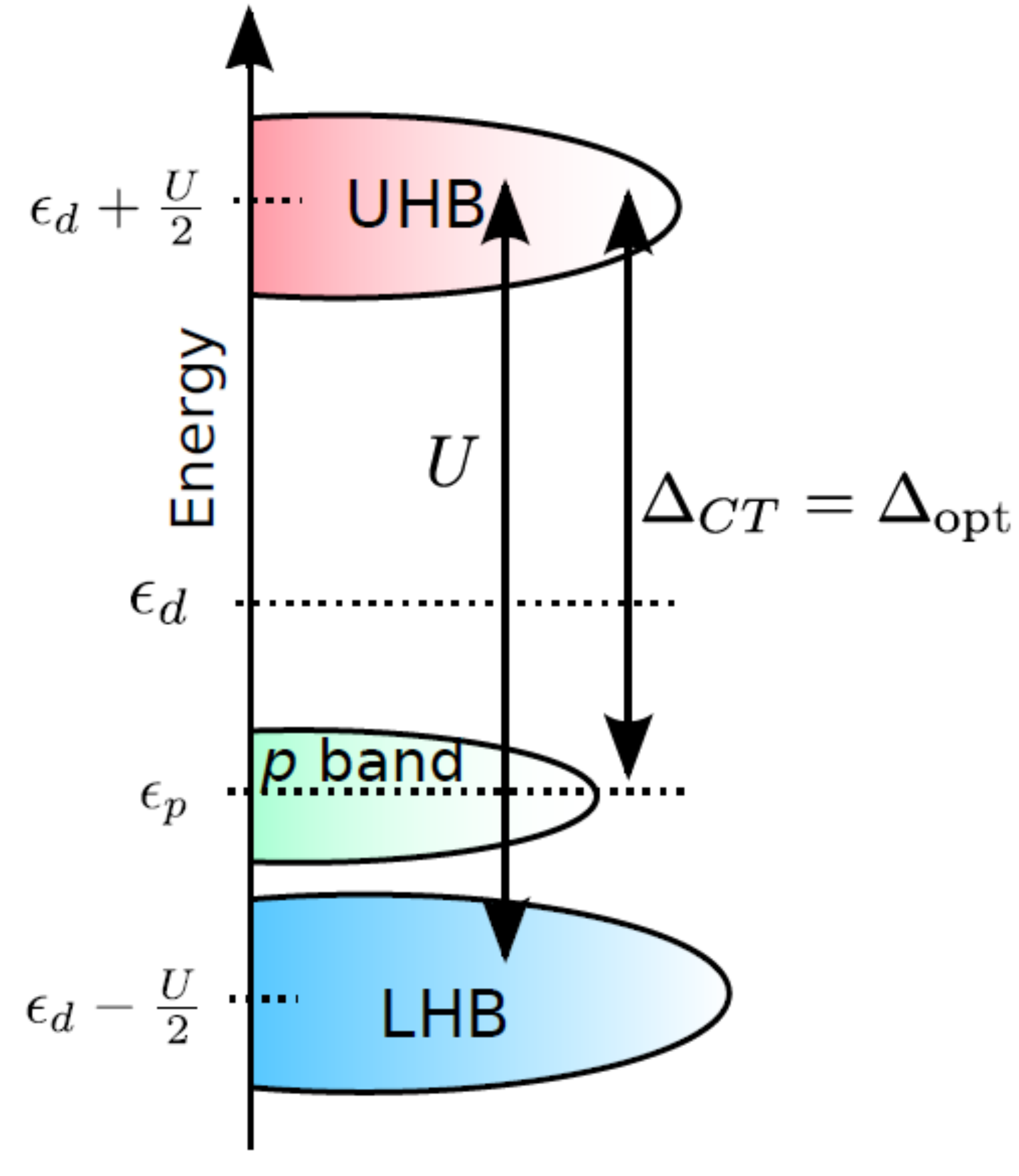


= drosophila of correlated many-electron problem



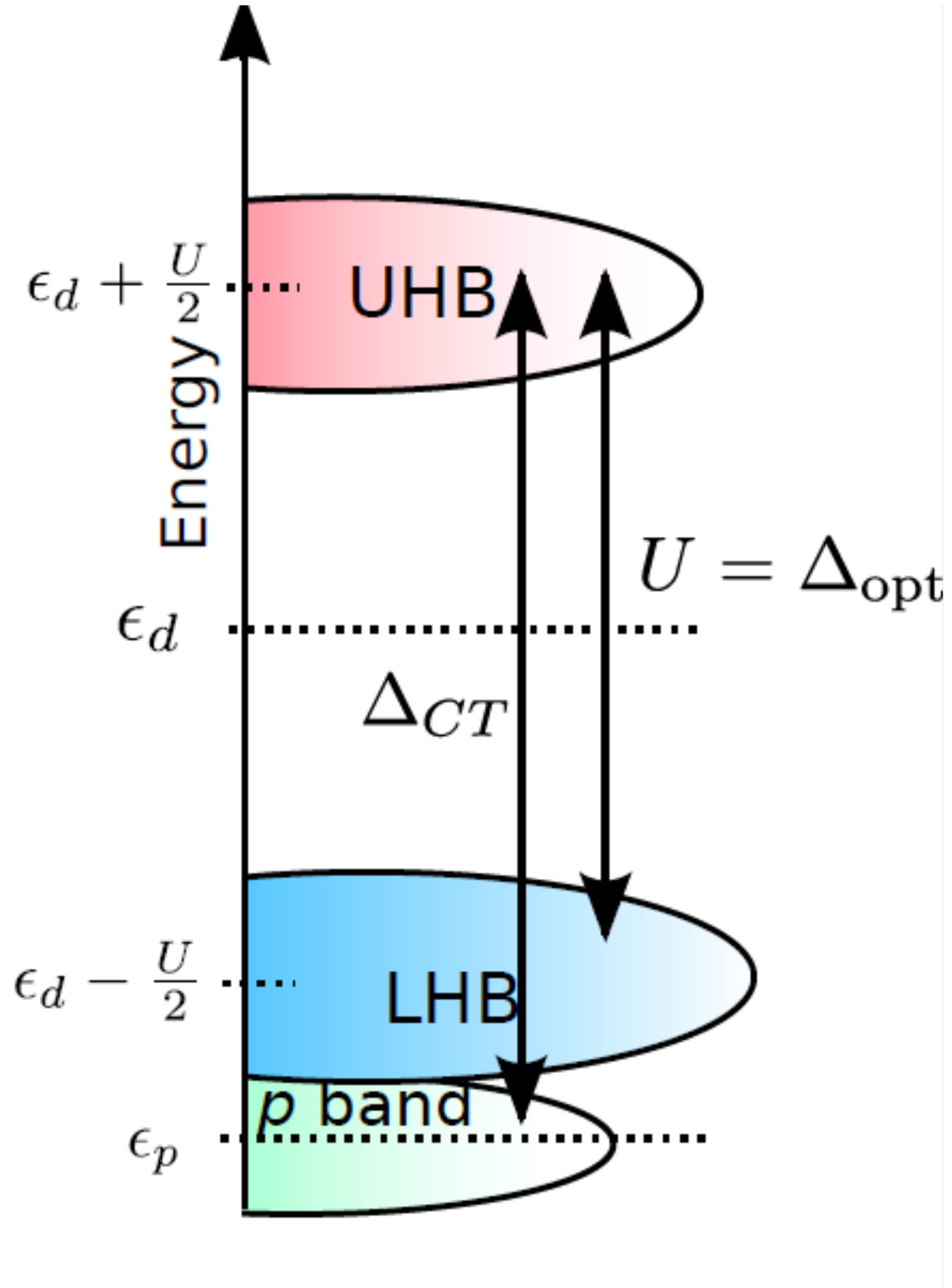
Transition-metal oxides: Mott versus charge transfer

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



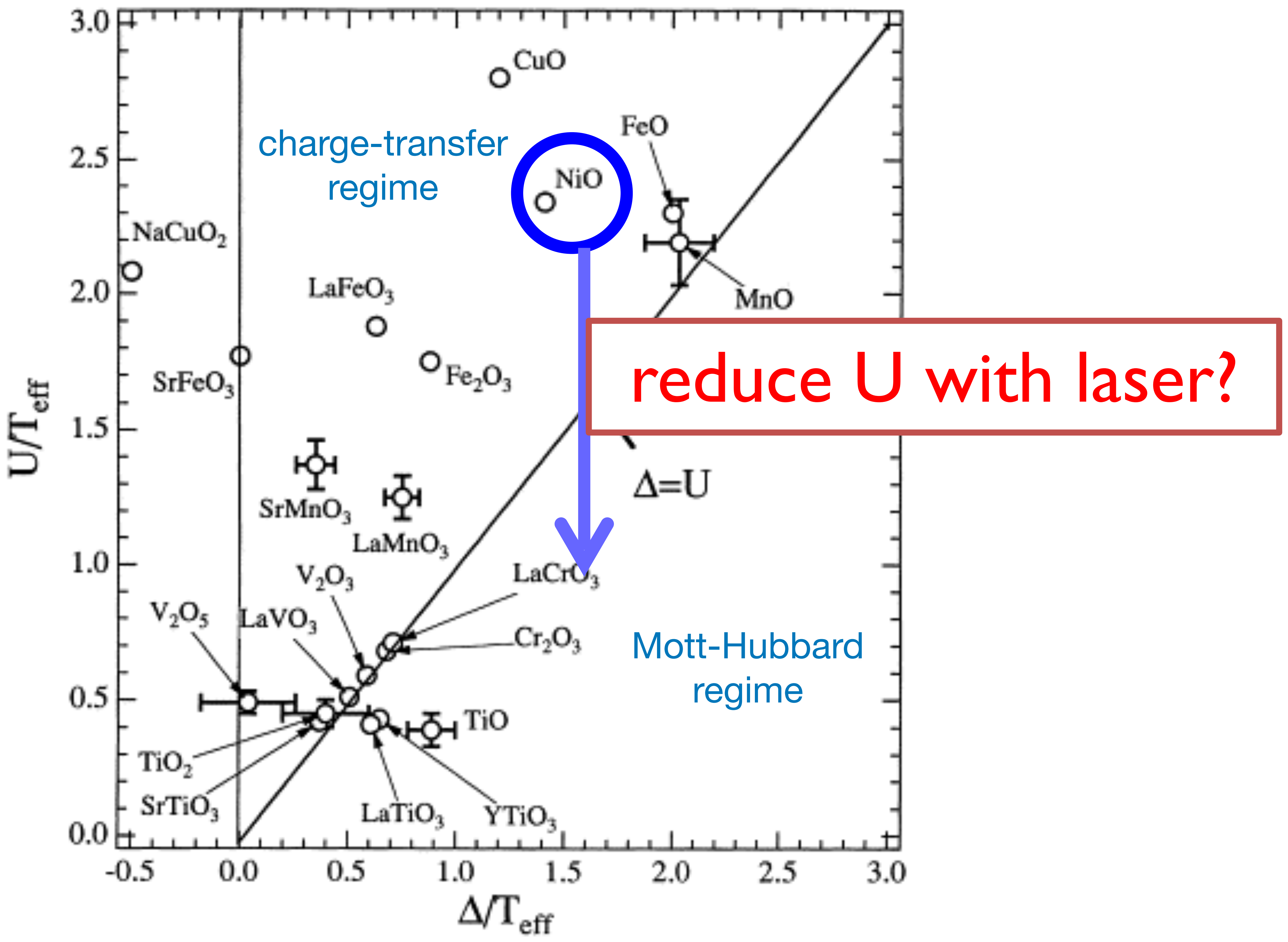
Charge-transfer insulators
CuO, NiO, ...

U>Δ



Mott insulators
TiO, VO, ...

U<Δ



Zaanen-Sawatzky-Allen classification
Fig. taken from Fujimori et al. (2001)

Time-dependent U with TDDFT+U

Nicolas Tancogne-Dejean

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)

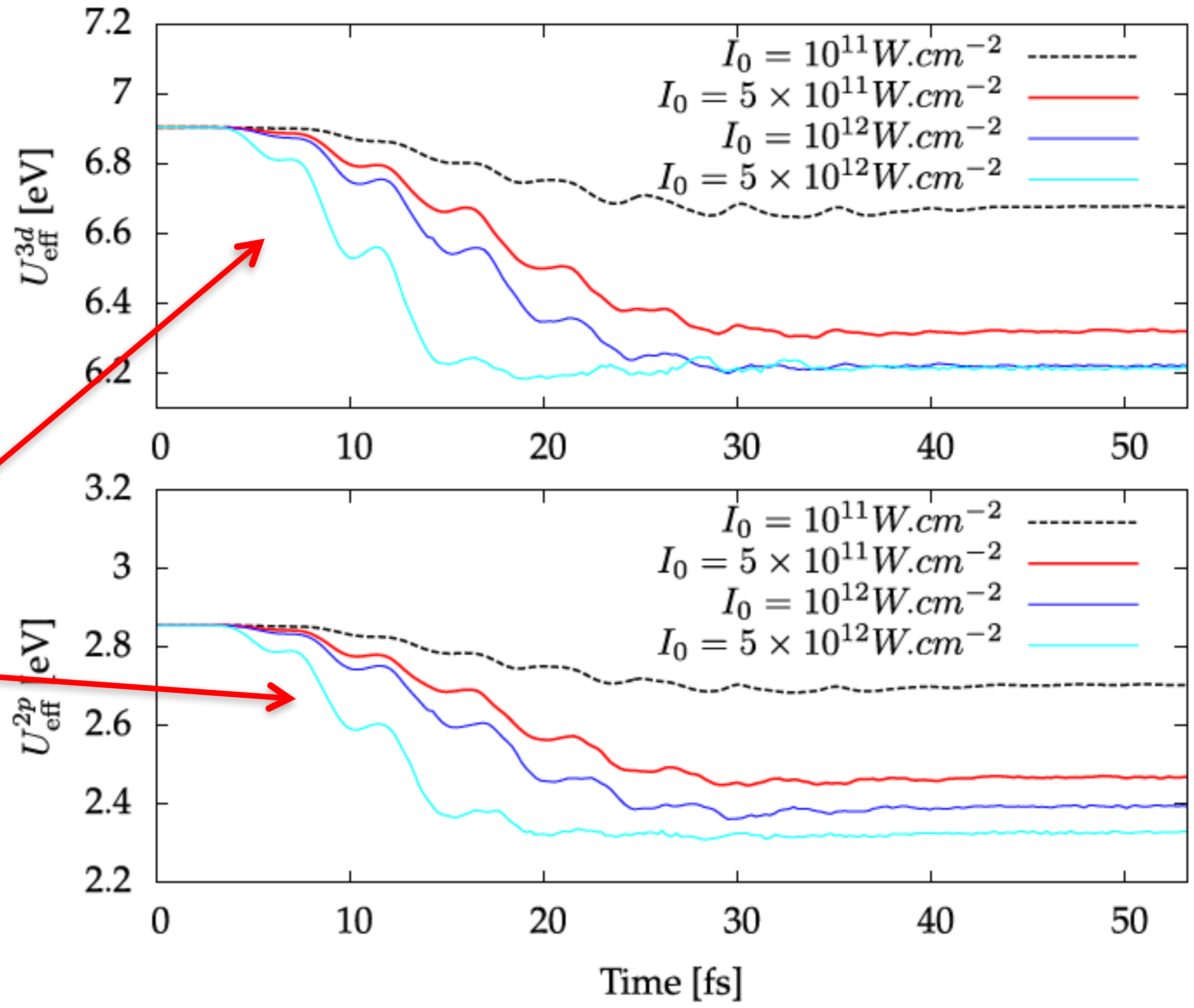
- alternative to constrained random-phase approximation
- numerically efficient
- direct extension to time-dependent case (adiabatic approximation)

Ultrafast modification of Hubbard U in NiO

Tancogne-Dejean, Sentef, Rubio, PRL 121, 097402 (2018)

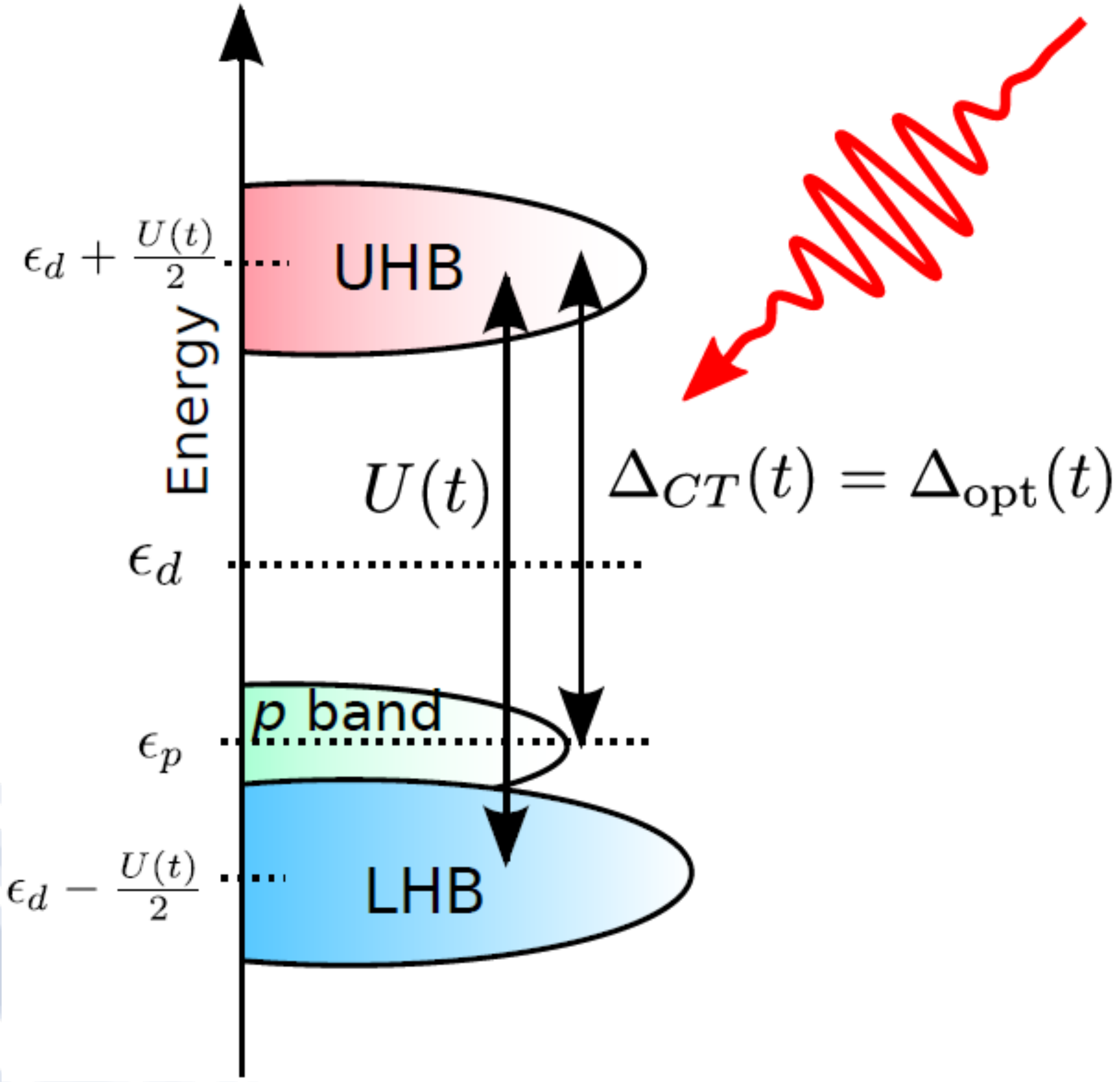
strong subresonant (0.43 eV) laser excitation:
→ high field strength without damage

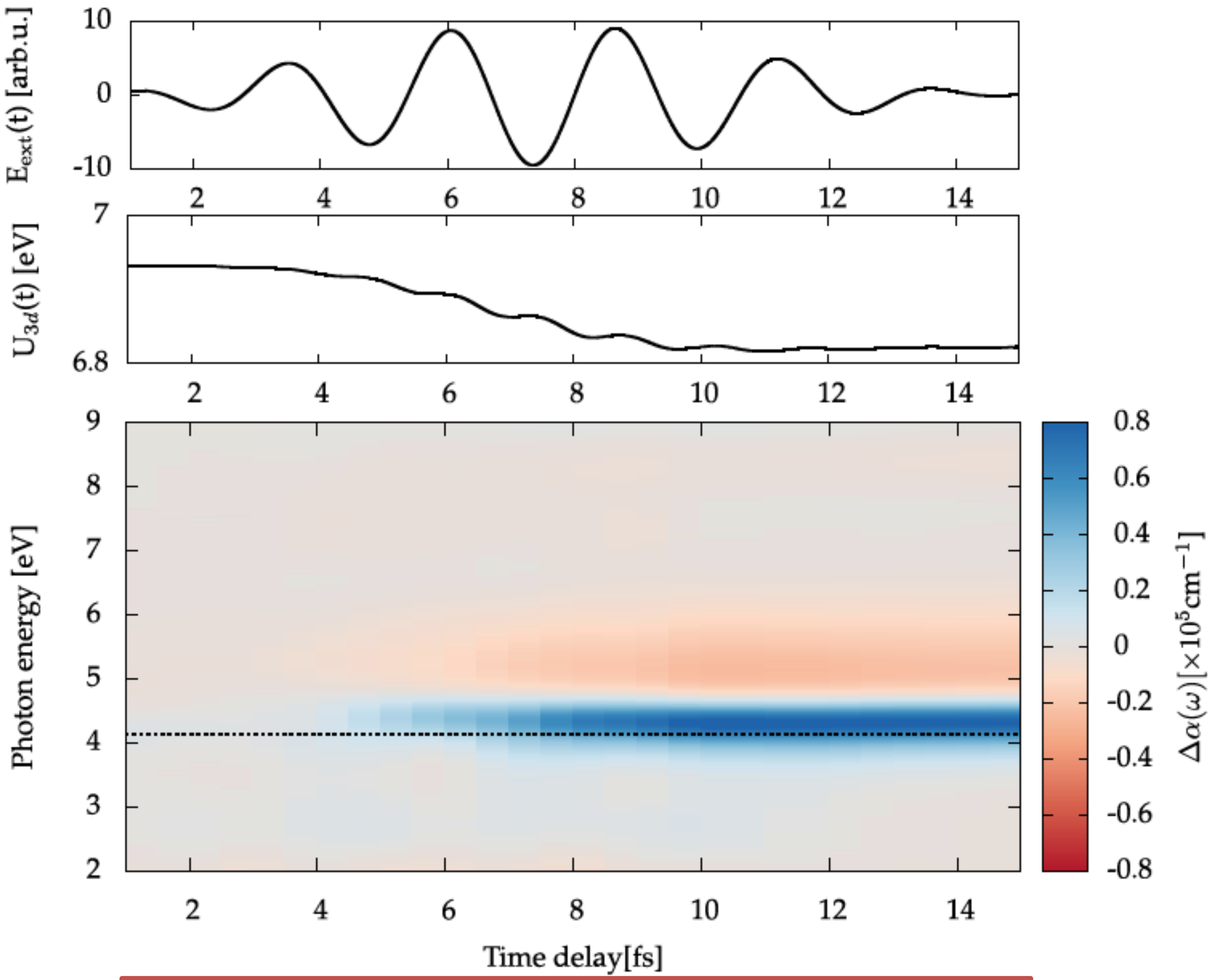
U reduced during the 25 fs laser pulse due to extra screening



Reduction of U: experimental fingerprint?

Tancogne-Dejean, Sentef, Rubio, PRB 102, 115106 (2020)



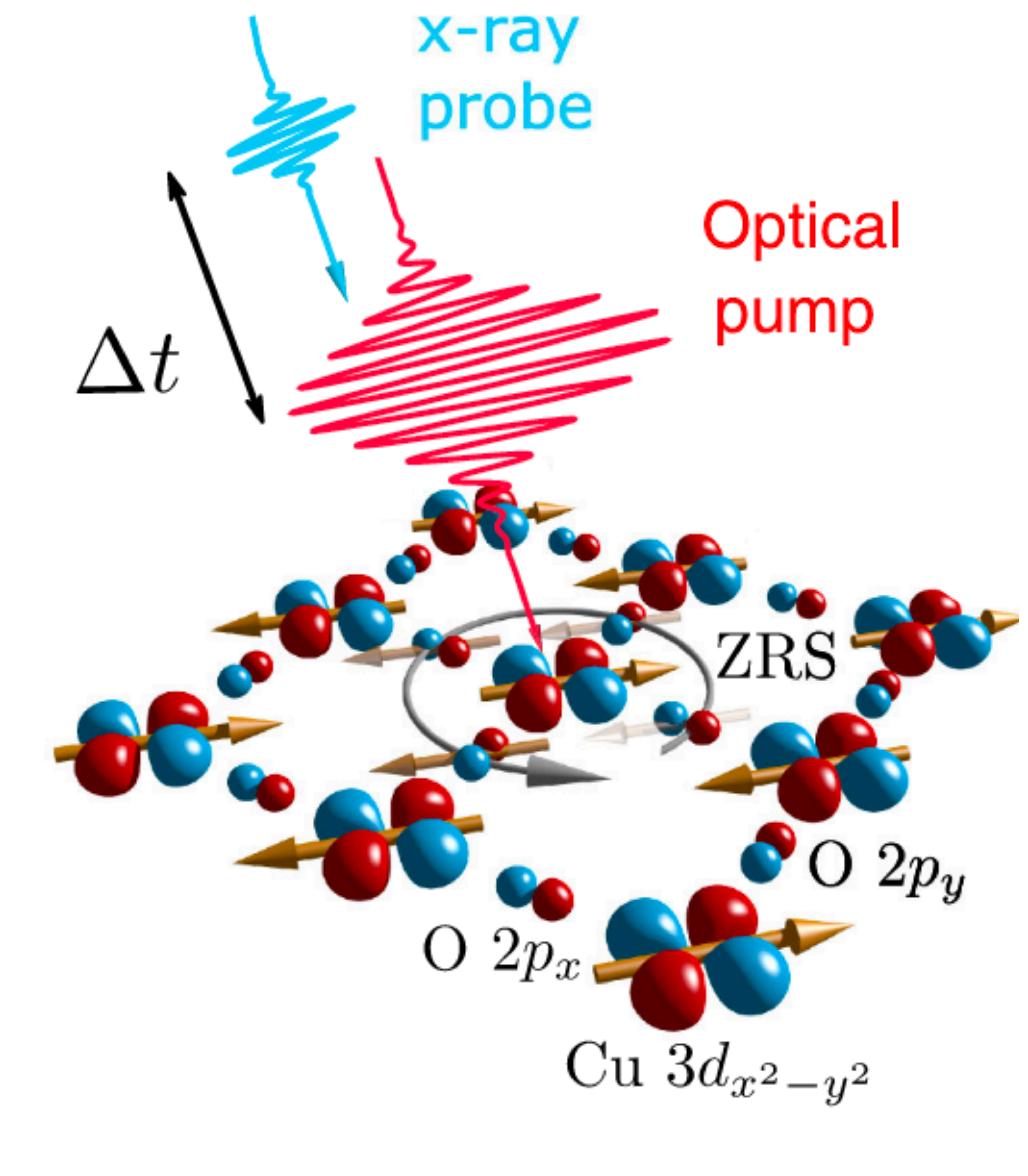


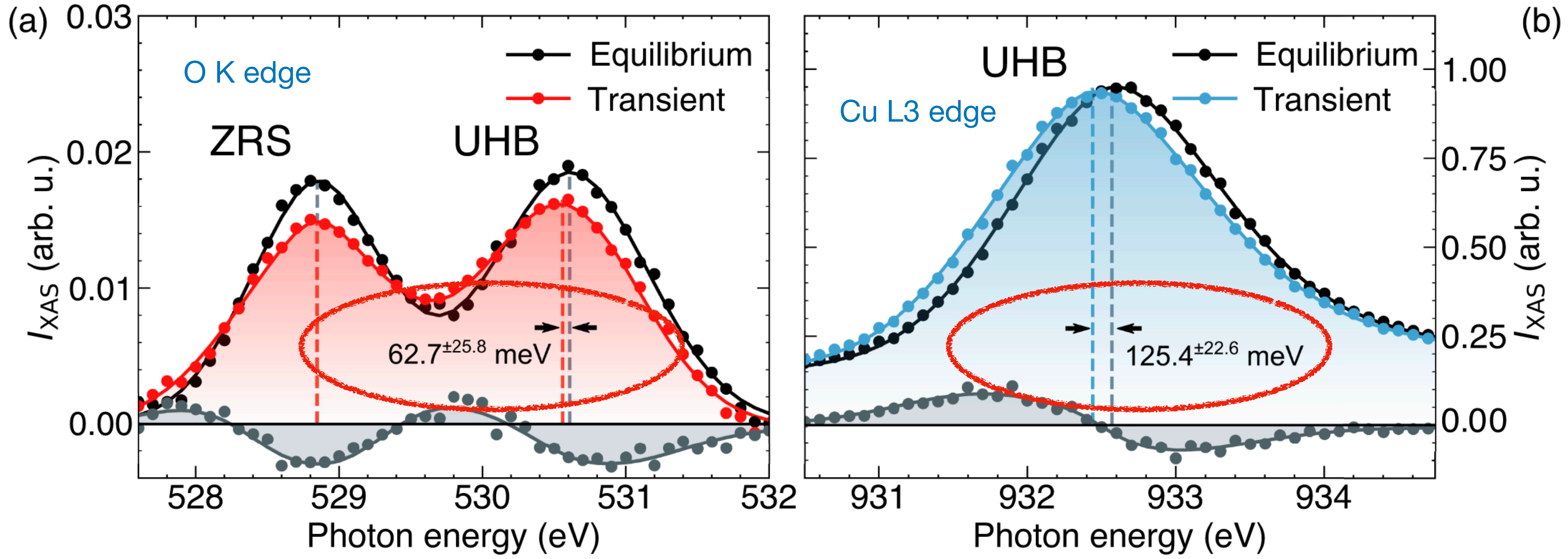
Shift of transient absorption peak

Experimental confirmation I

D. Baykusheva et al., Mitrano group, PRX 2022

Time-resolved XAS on underdoped cuprate (LBCO)

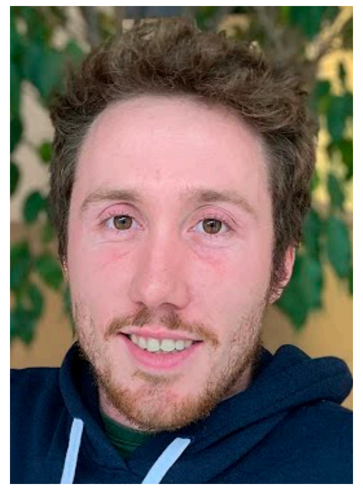




Shift of x-ray absorption peaks consistent with reduced U

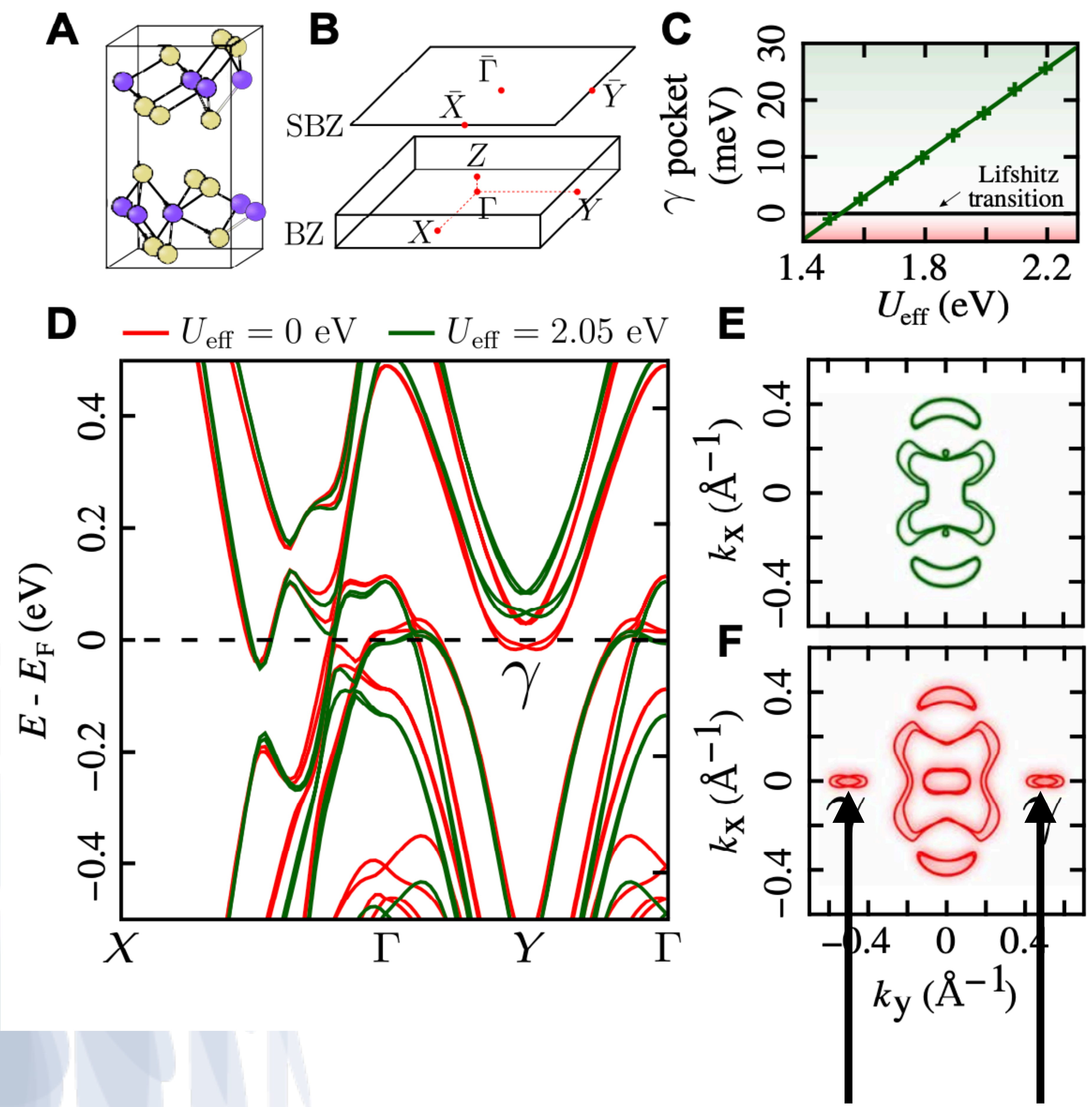
Experimental confirmation II

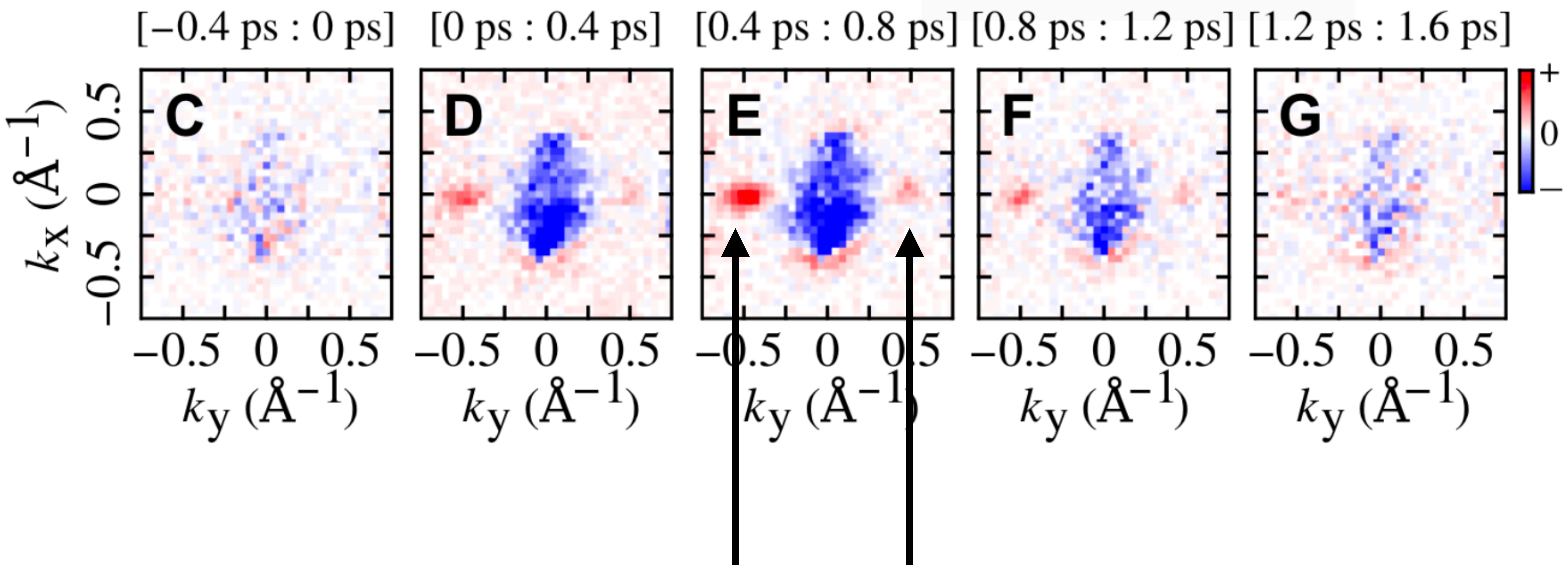
Time-resolved ARPES of Td-MoTe2



Sam Beaulieu

S. Beaulieu et int., MAS, et int., R. Ernstorfer,
Science Advances 7, eabd9275 (2021)
Collaboration with FHI Berlin





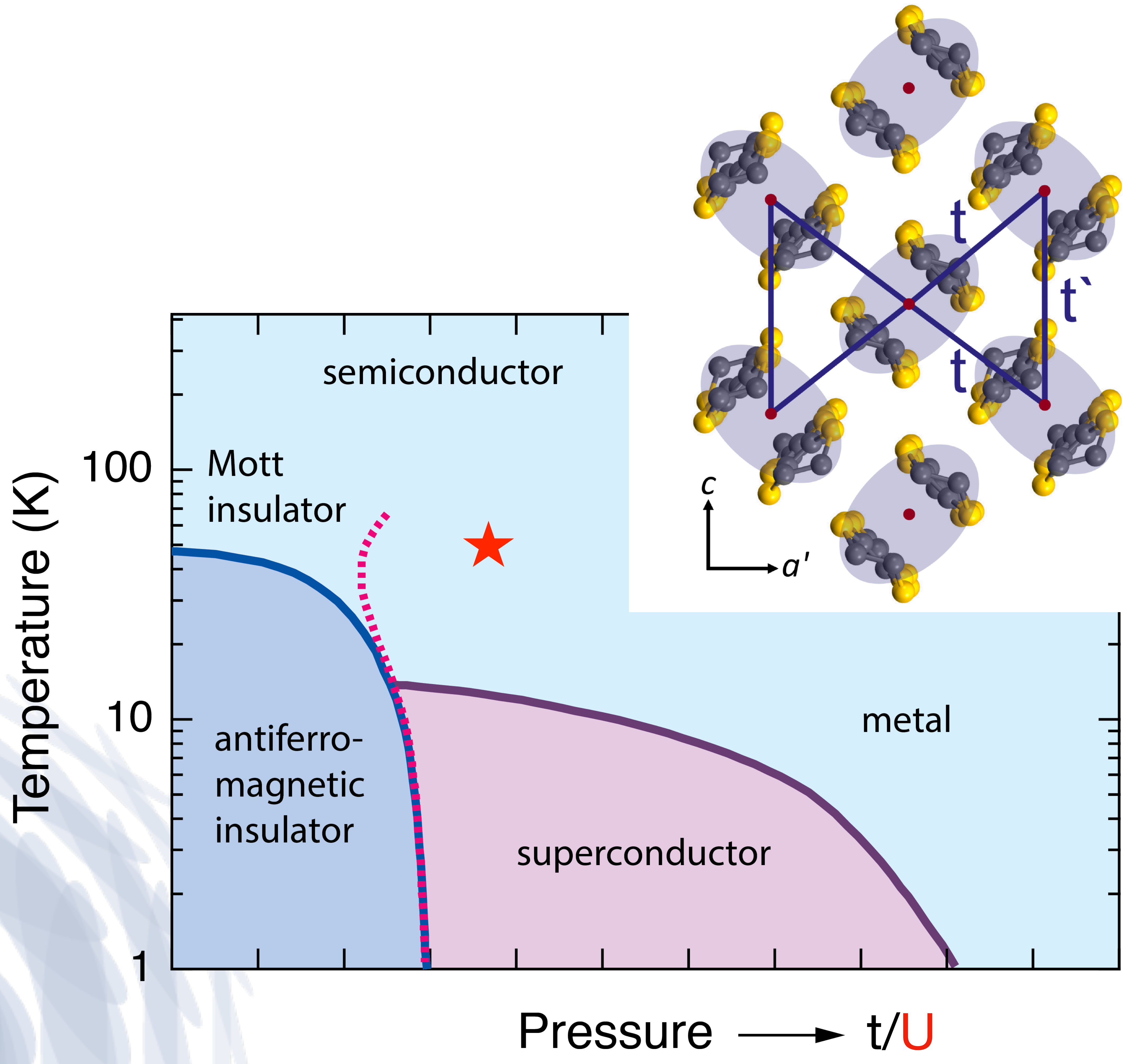
Ultrafast Lifshitz transition triggered by light-reduced U

Light-induced superconductivity

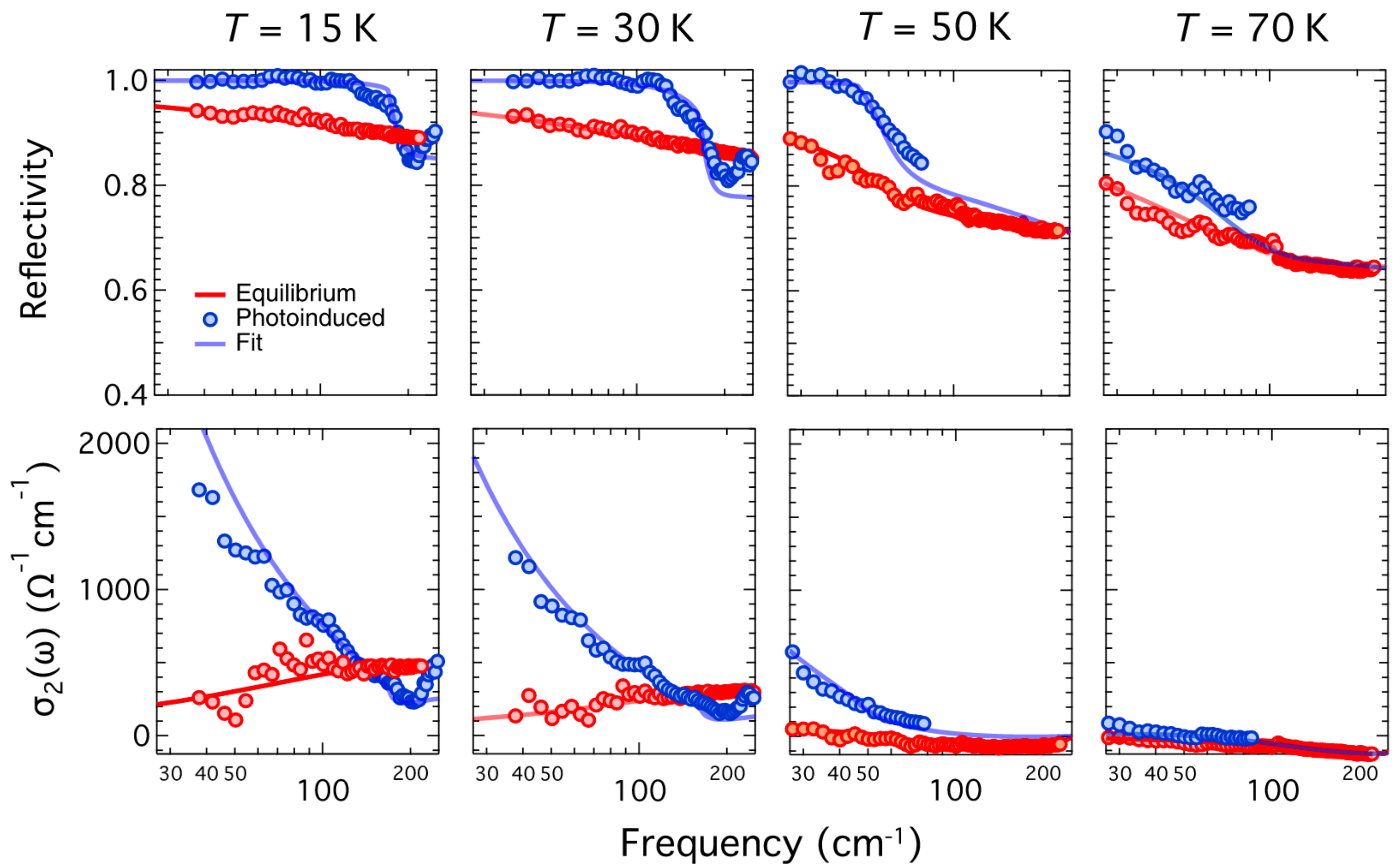
Photo-molecular high-temperature superconductivity in an organic kappa salt

Michele Buzzi Daniele Nicoletti

M. Buzzi et al., PRX 10, 031028 (2020)



Driving a molecular vibration

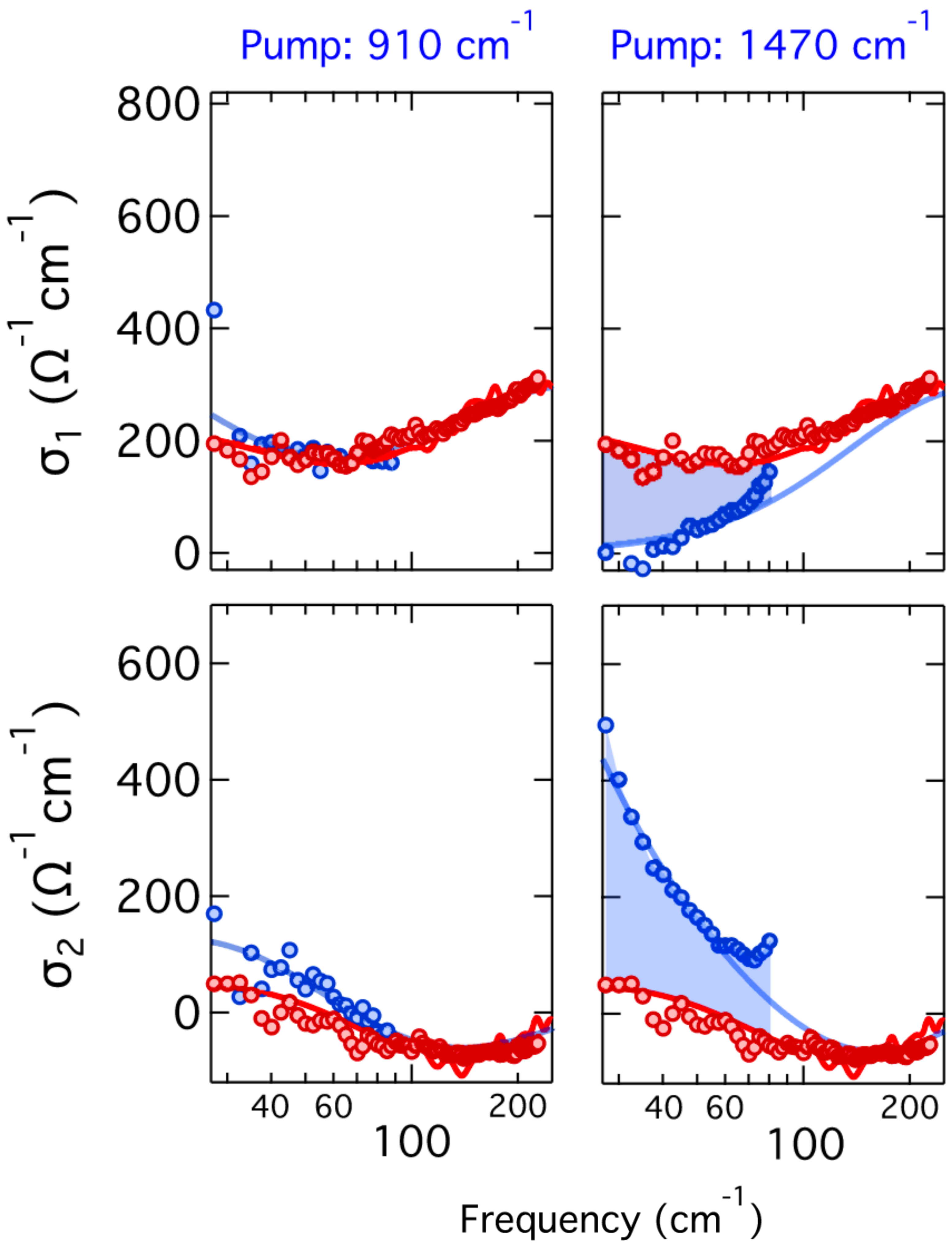


Superconducting-like optical response far above equilibrium T_c

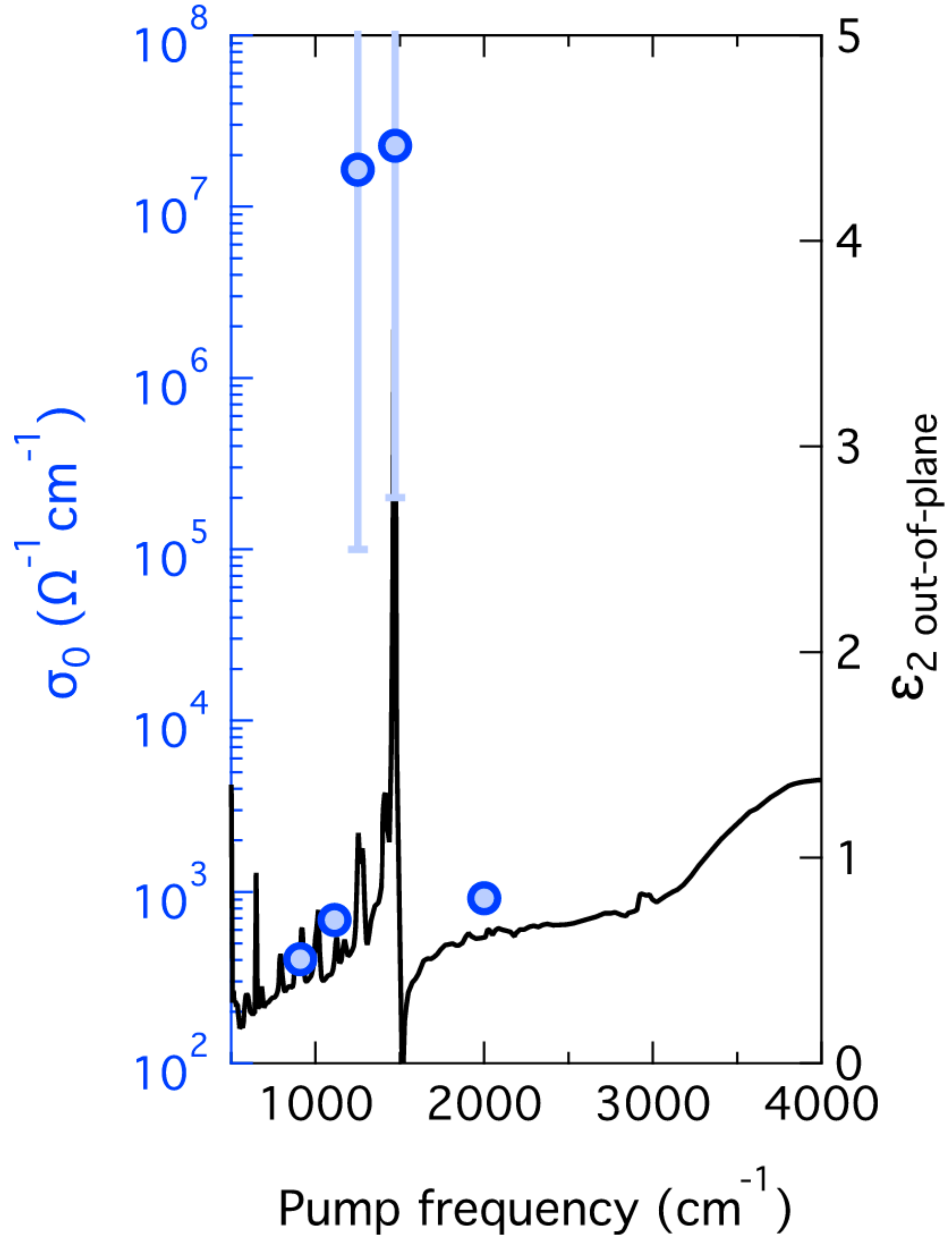
Light-induced superconductivity

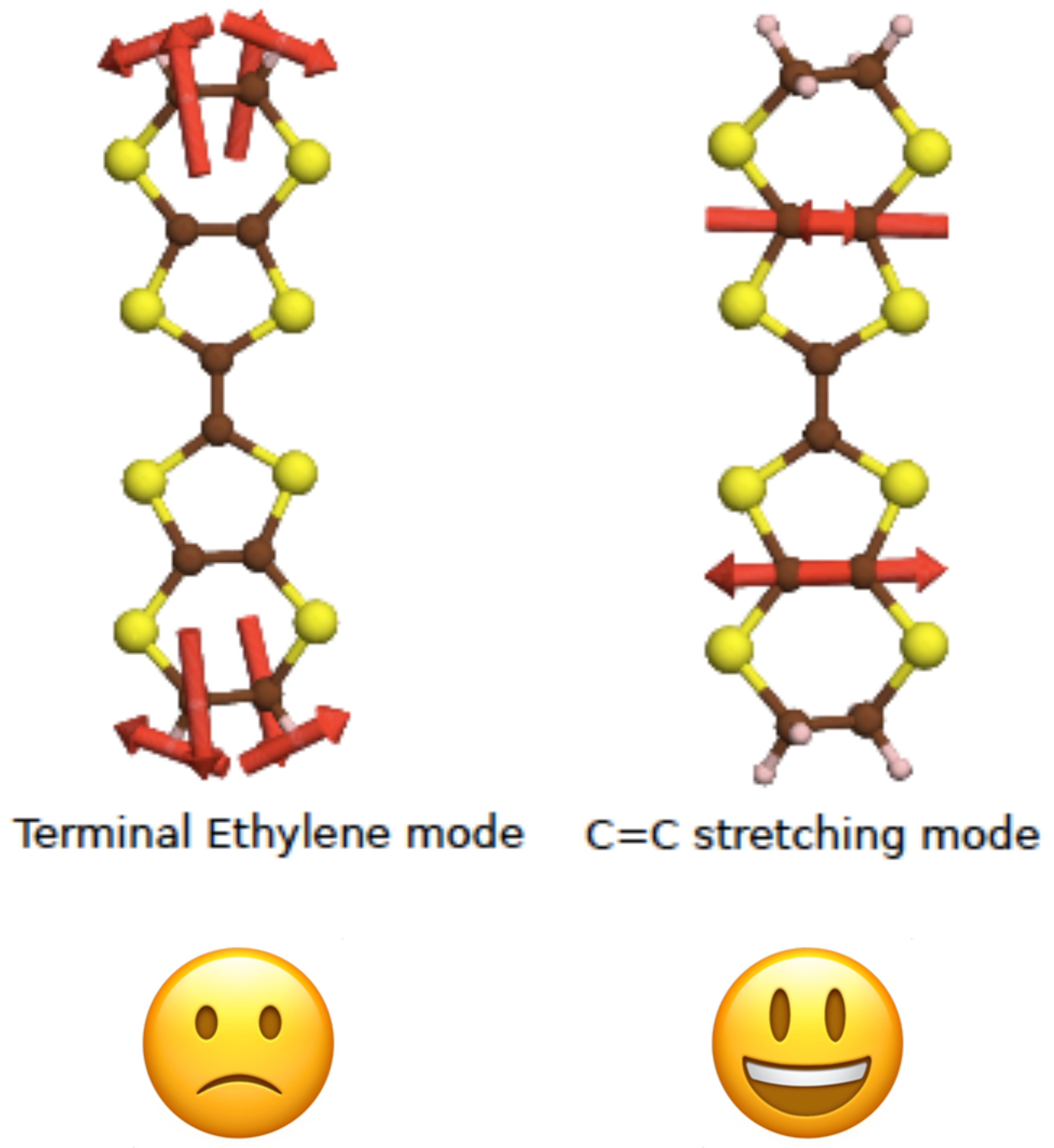
M. Buzzi et al., PRX 10, 031028 (2020)

Photo-molecular high-temperature superconductivity in an organic kappa salt



Phonon mode selectivity of light-induced superconductivity



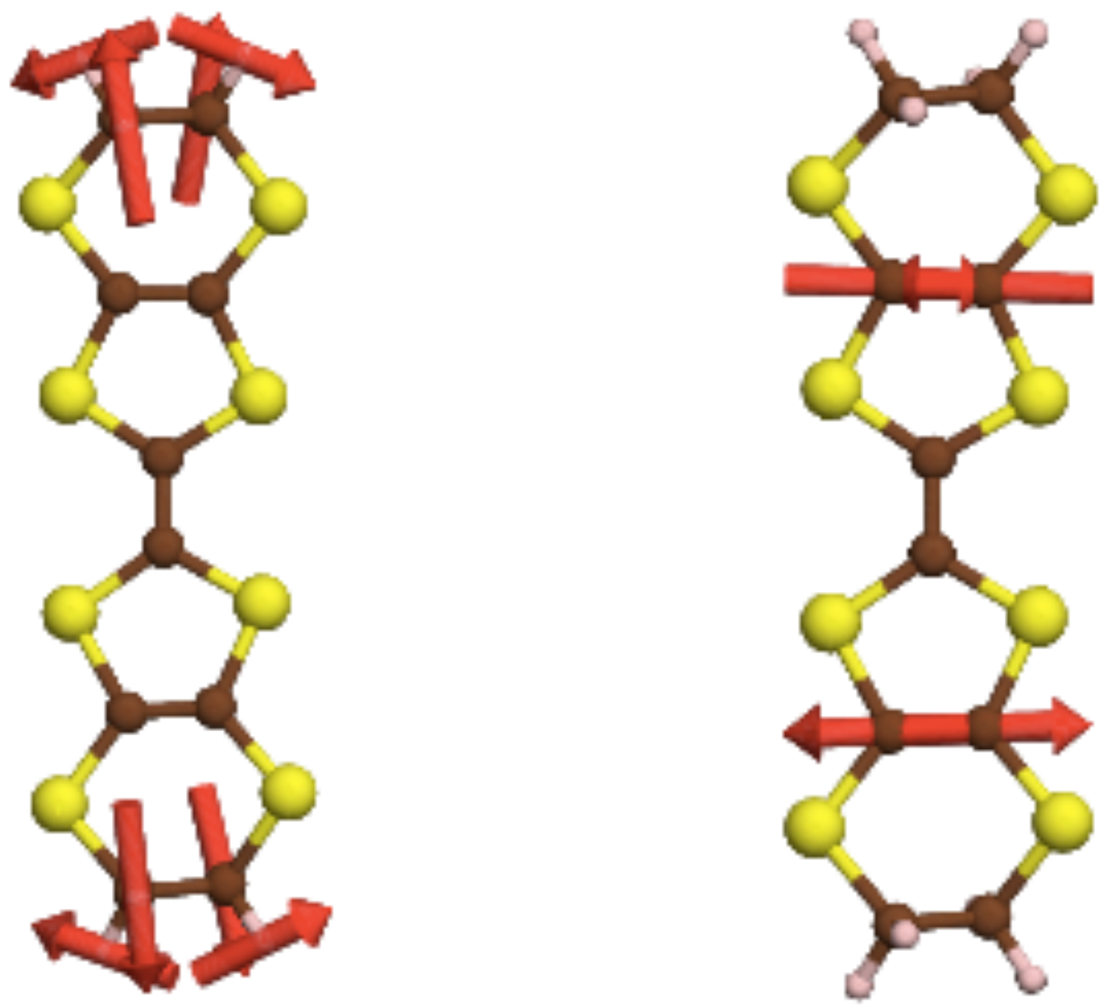


Nonlinear electron-phonon coupling

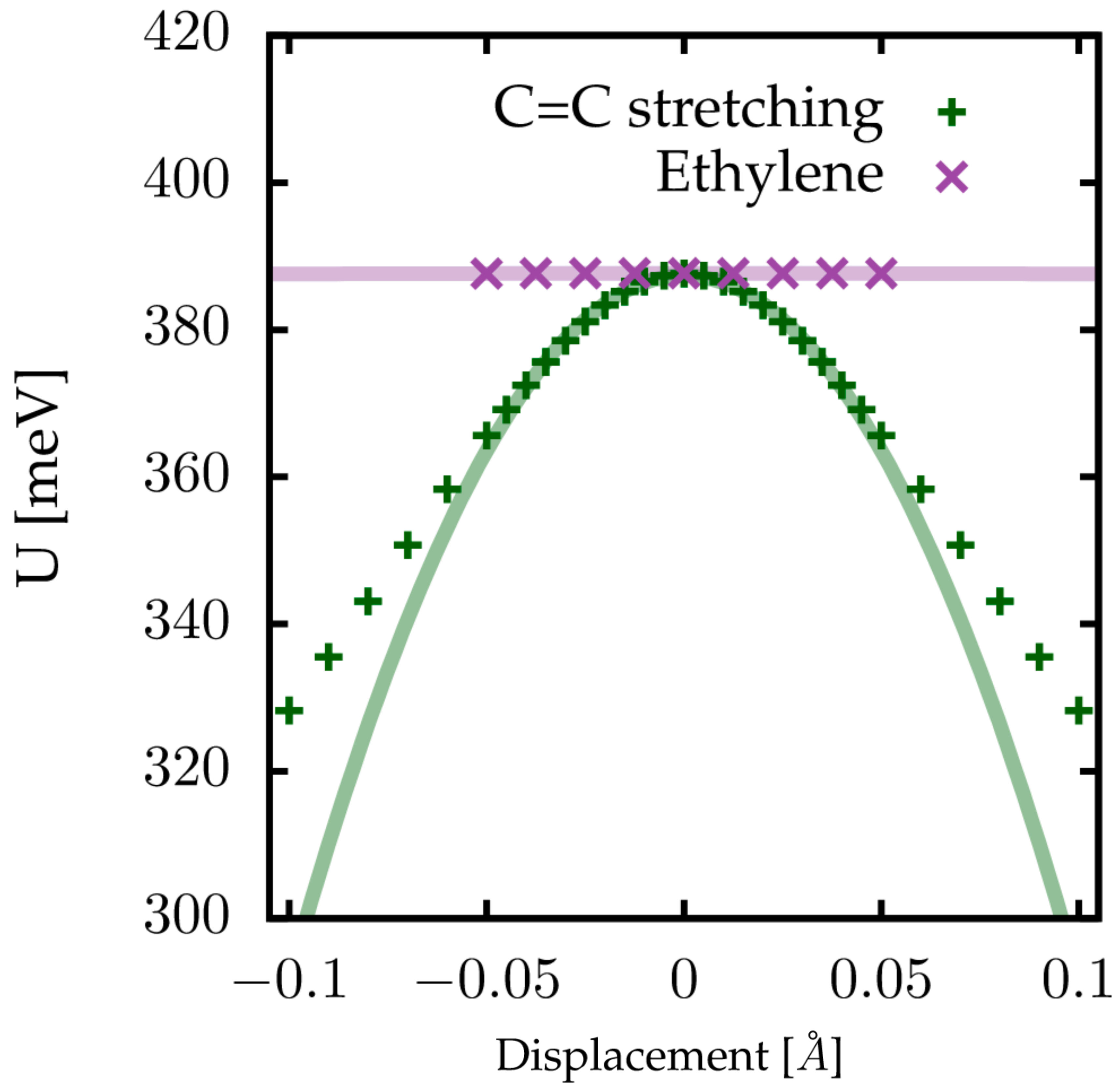
M. Buzzi et al., PRX 10, 031028 (2020)

Nicolas Tancogne-Dejean

Phonon mode selectivity of light-induced superconductivity



Terminal Ethylene mode C=C stretching mode



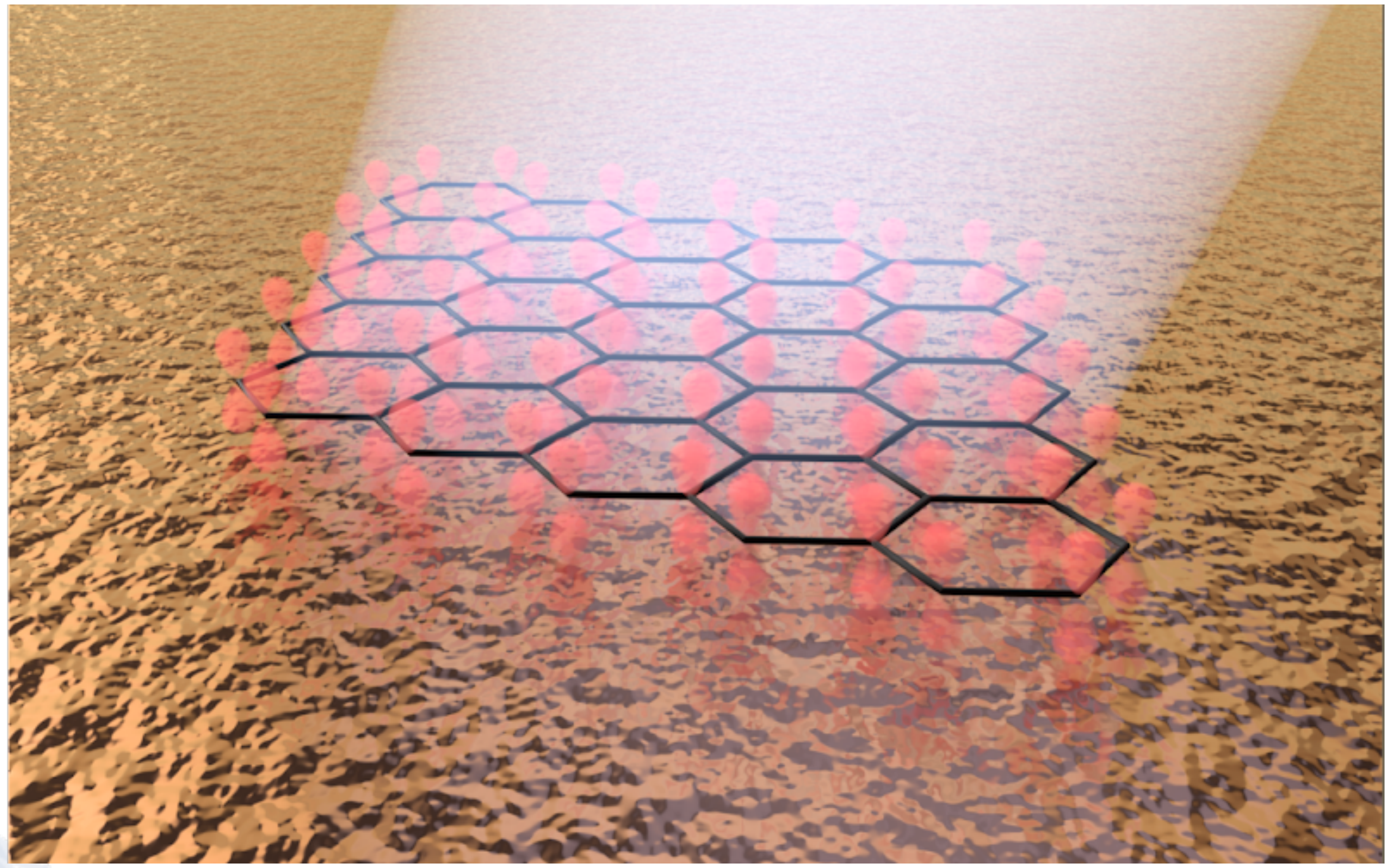
Driving C=C stretching modes leads to dynamical Hubbard U

$$H = \sum_{<ij>,\sigma} t(c^{\dagger}_{i\sigma} c_{j\sigma} + H.c.) + \sum_{[ij],\sigma} t'(c^{\dagger}_{i\sigma} c_{j\sigma} + H.c.) + U \sum_{i} (n_{i\uparrow} - \frac{1}{2})(n_{i\downarrow} - \frac{1}{2})$$

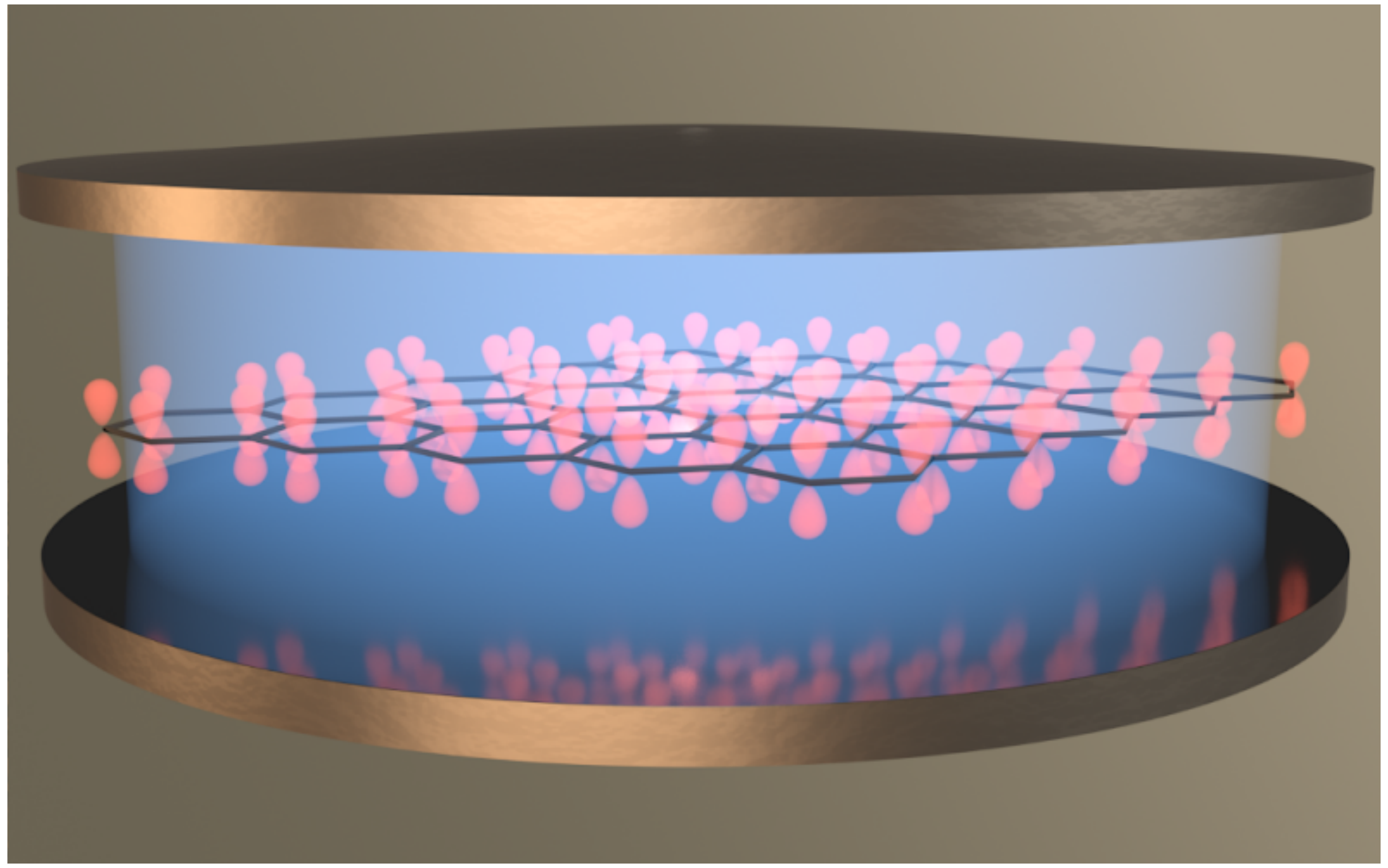
H. C. Kandpal et al., PRL 2009

Theory follow-up (J. Tindall et al., PRL 125, 137001 (2020)): Increased pairing correlation length in parametrically driven frustrated Hubbard model

Cavity quantum materials



strong laser

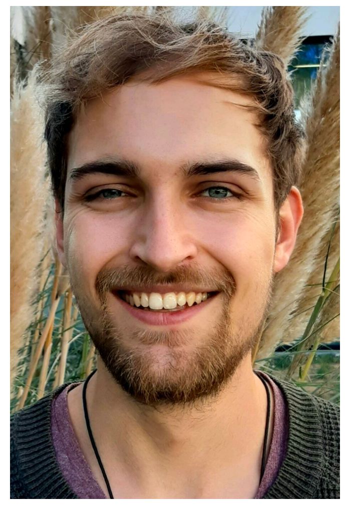


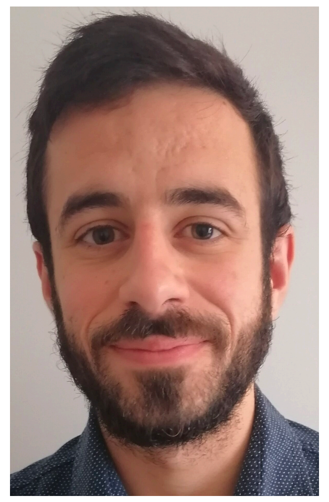
strong light-matter coupling

Applied Physics Reviews 9, 011312 (2022)

Cavity quantum materials

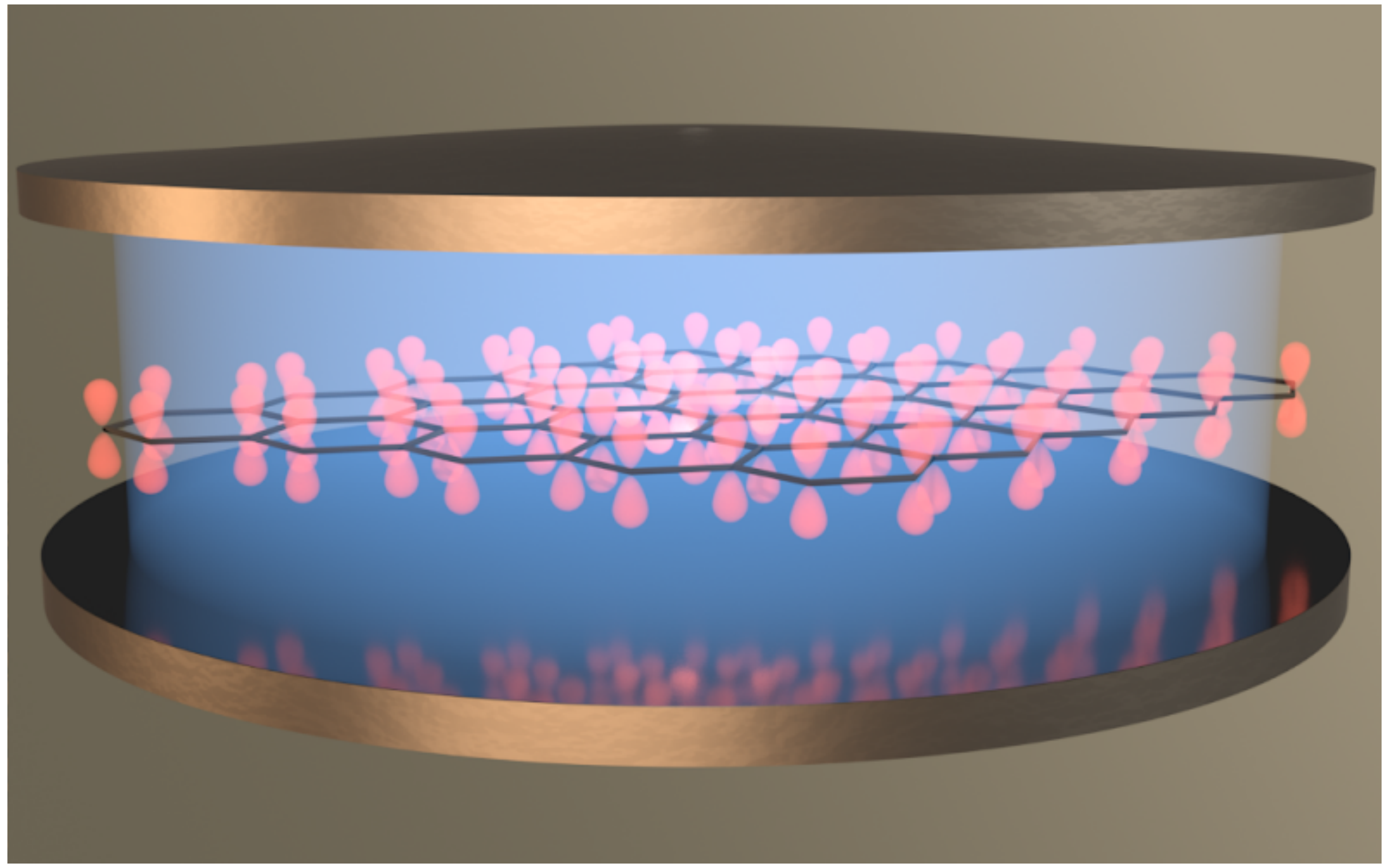
Applied Physics Reviews 9, 011312 (2022)
J. Phys. Mater. 5, 024006 (2022)



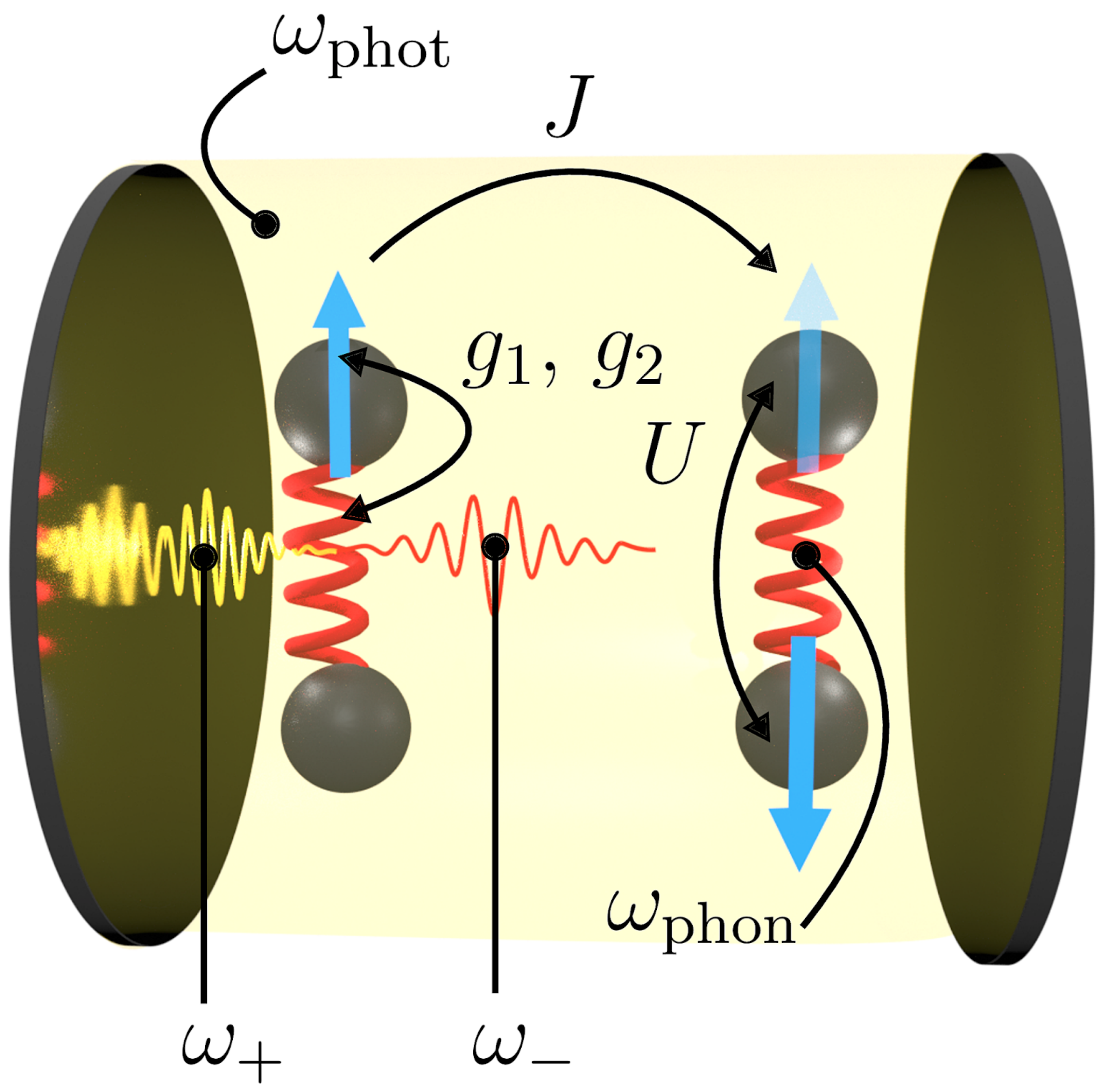


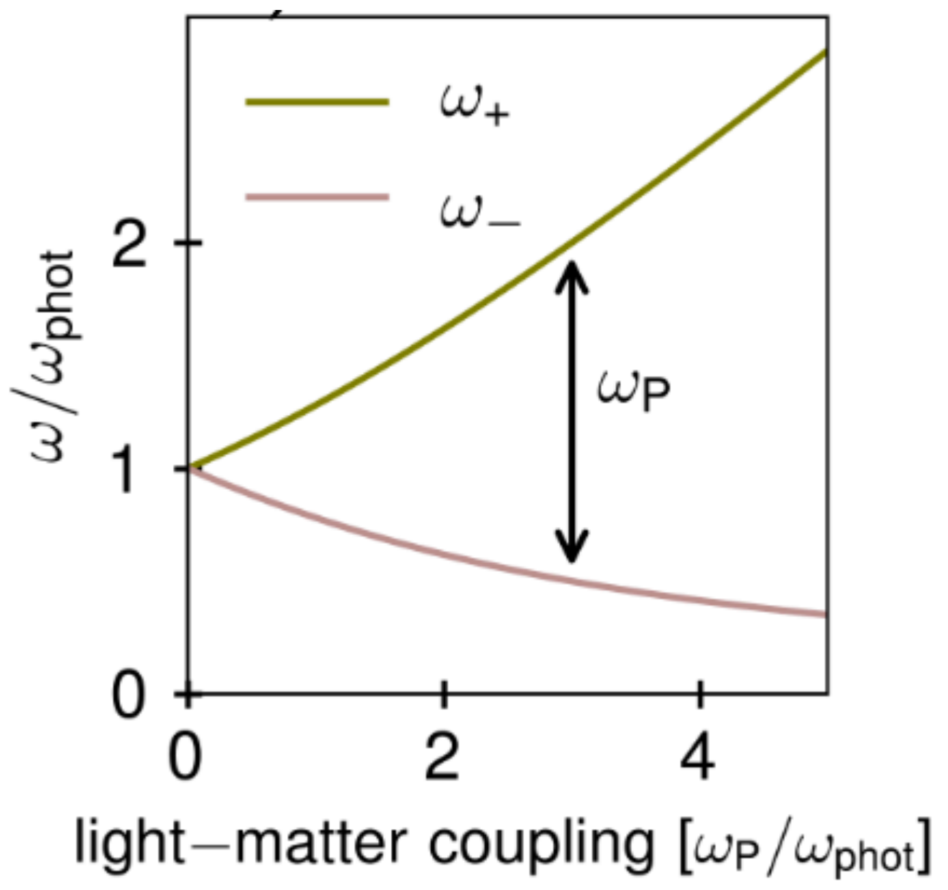
Christian Eckhardt Brieuc Le Dé

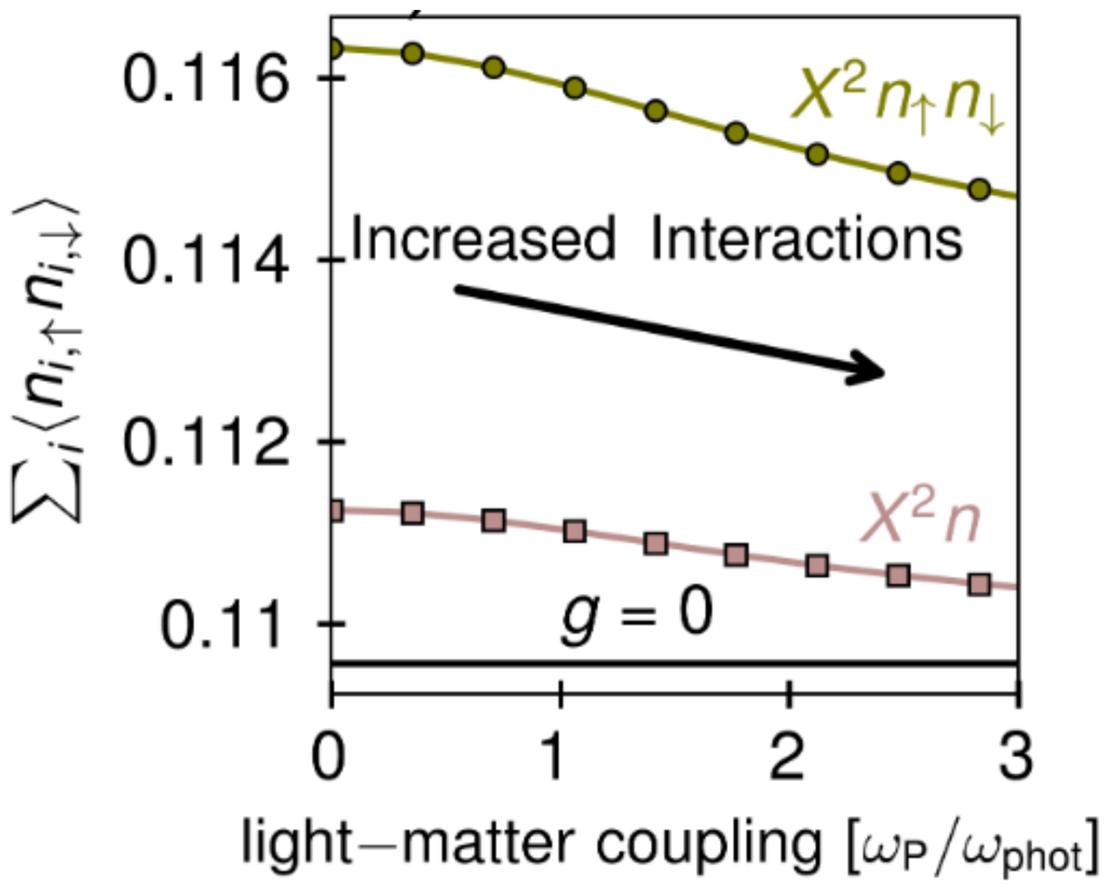
Poster LMC3



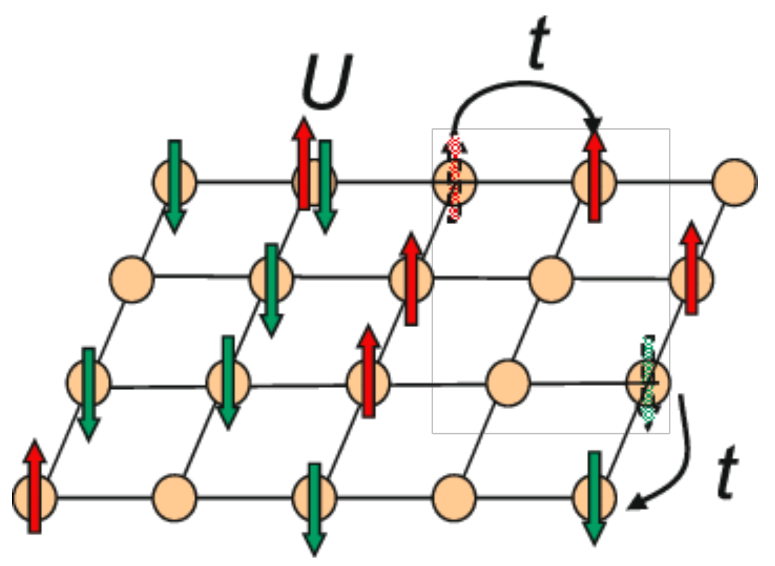
strong light-matter coupling



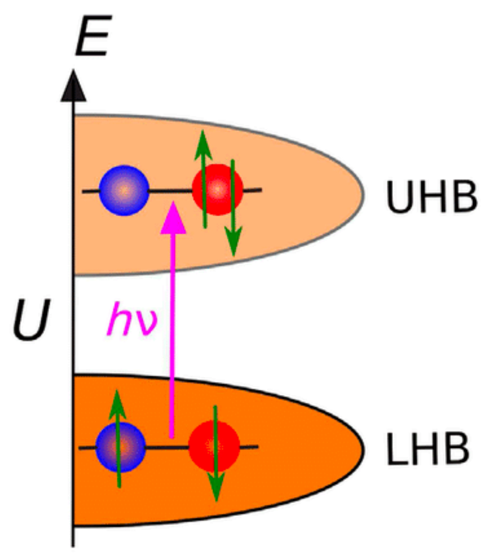




Organic molecules in cavity: enhanced effective electron-electron interactions through phonon polariton formation

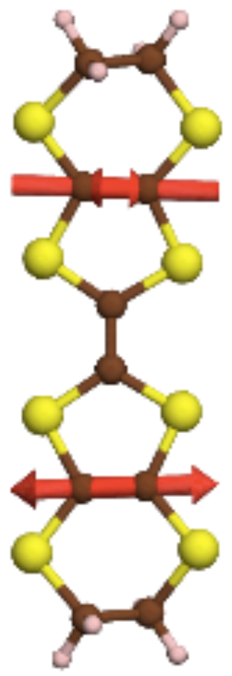


Hubbard U



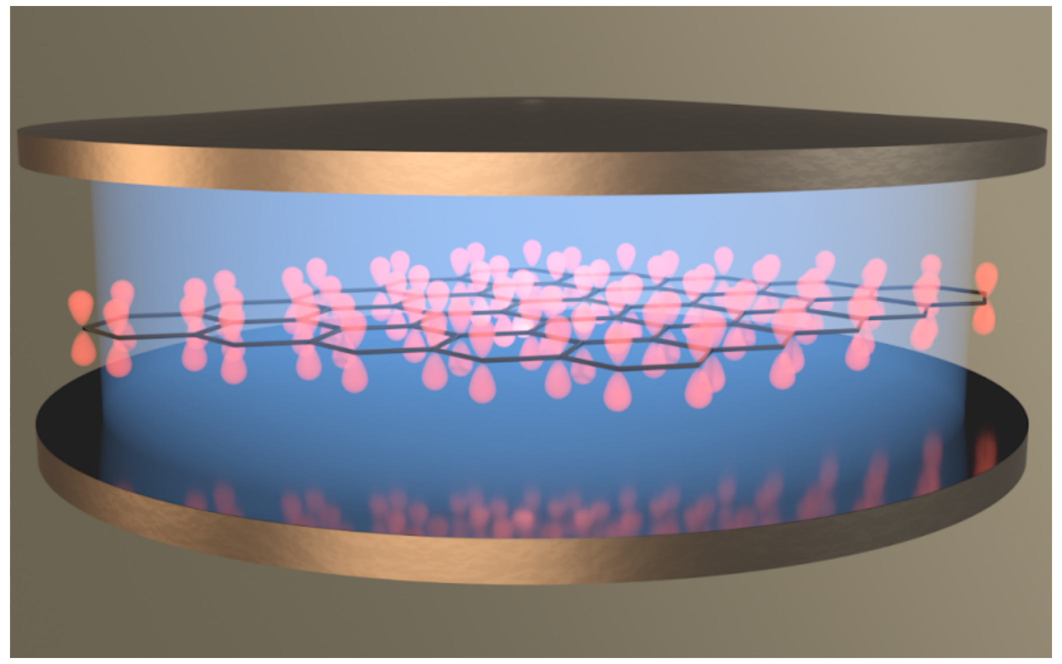
optical excitation

several %



phonon driving

~10-20 %



cavity

~1%

driven cavity

~10-20%

with lower laser intensity

Pathway towards control of quantum materials through light-matter coupling

Thank you for your attention!