Nonequilibrium Materials Engineering: Time-dependent U and beyond

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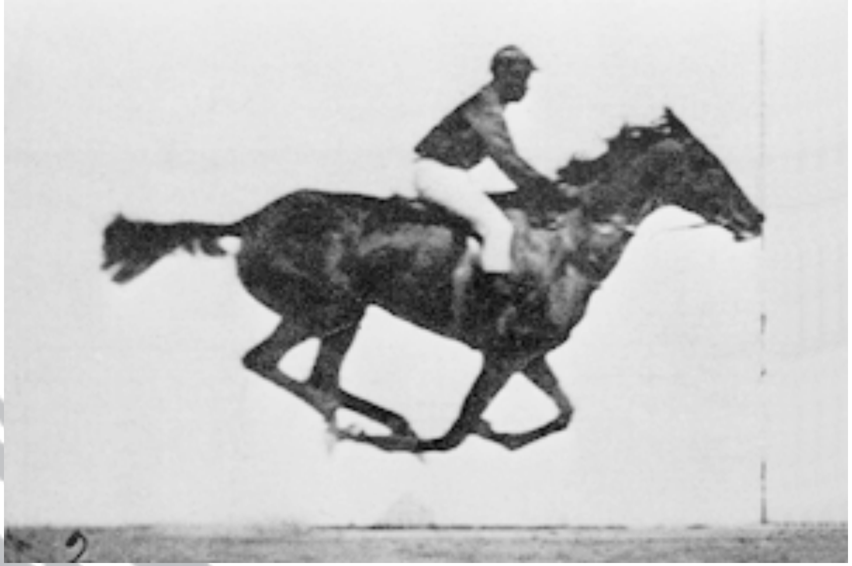
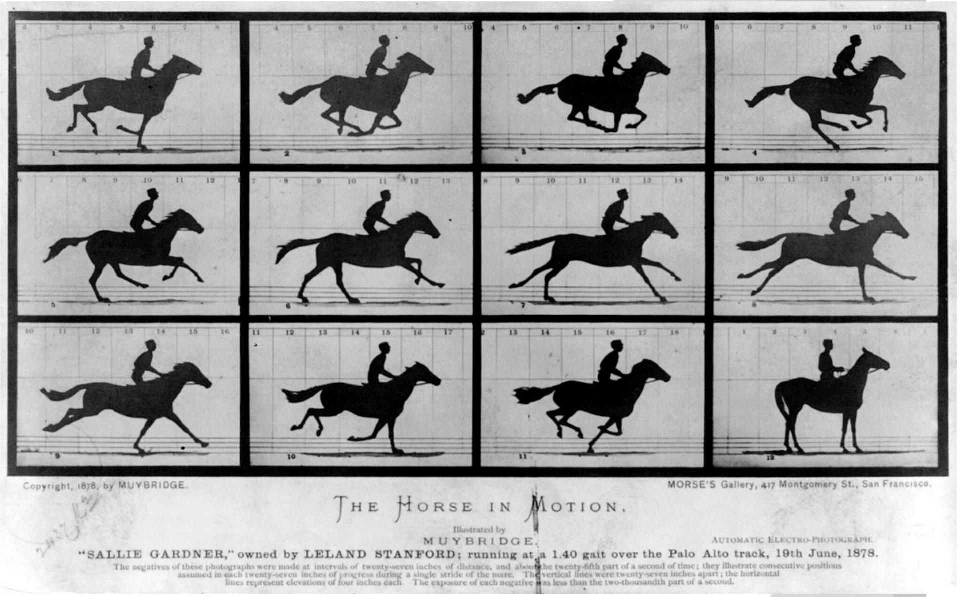
Emmy Noether Programme (SE 2558/2-1)

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
MUYBRIDGE.

AUTOMATIC ELECTRO-PHOTOGRAPH.

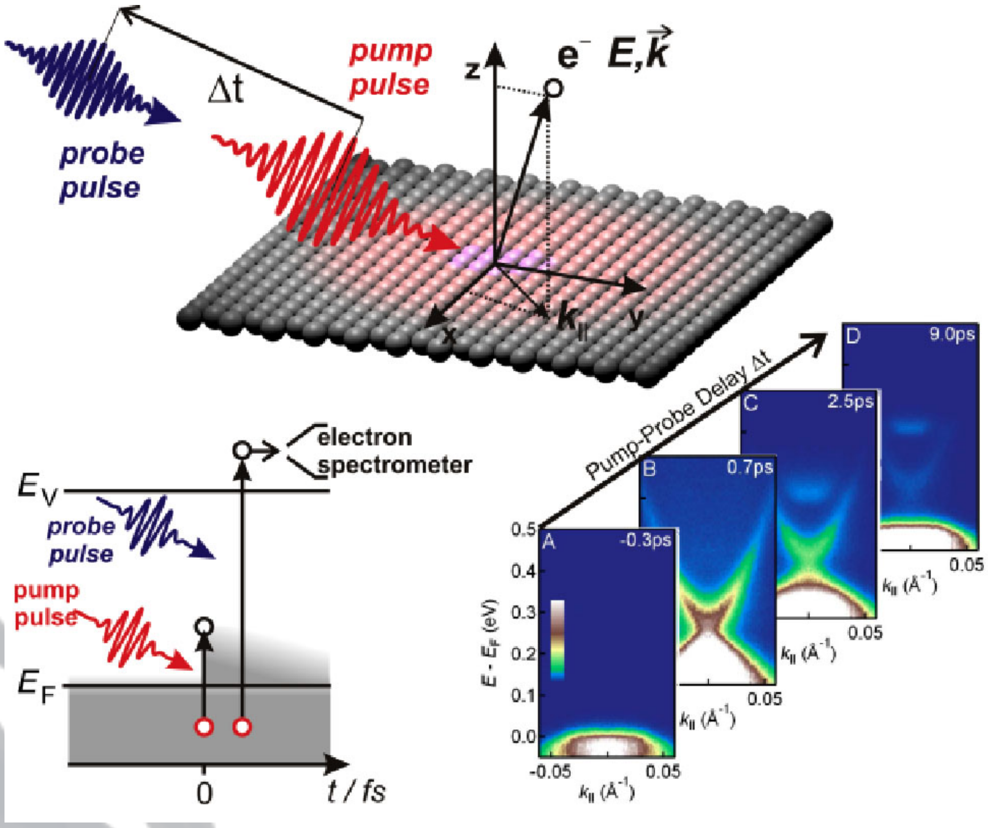
"SALLIE GARDNER," owned by LELAND STANFORD; running at a 1.40 gait over the Palo Alto track, 19th June, 1878.

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.

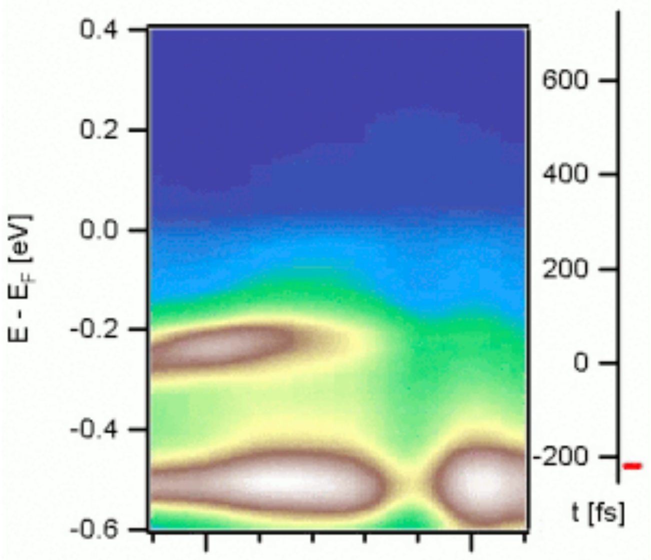
Muybridge 1887

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)

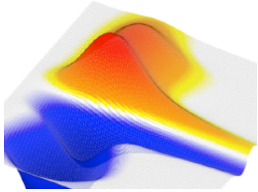
Image courtesy: J. Sobota / F. Schmitt

Ultrafast Materials Science today

Understanding the nature of quasiparticles

- Relaxation dynamics
- Control of couplings

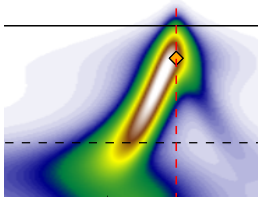
PRL 111, 077401 (2013)
PRX 3, 041033 (2013)
PRB 87, 235139 (2013)
PRB 90, 075126 (2014)
Nat. Comm. 7, 13761 (2016)
PRB 95, 024304 (2017)
PRB 95, 205111 (2017)
PRL 121, 097402 (2018)
Science Adv. 4, eaau6969 (2018)
arXiv:1808.02389



Understanding ordered phases

- Collective oscillations
- Competing orders

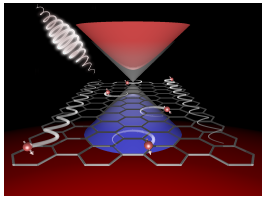
PRB 92, 224517 (2015)
PRB 93, 144506 (2016)
PRL 118, 087002 (2017)
PRB 98, 165138 (2018)
arXiv:1808.00712
arXiv:1808.04655
arXiv:1810.06536
arXiv:1810.09731



Creating new states of matter

- Floquet topological states

Nature Comm. 6, 7047 (2015)
Nature Comm. 8, 13940 (2017)
Nature Comm. 9, 4452 (2018)
arXiv:1902.05821; arXiv:1903.00339
arXiv:1905.04508



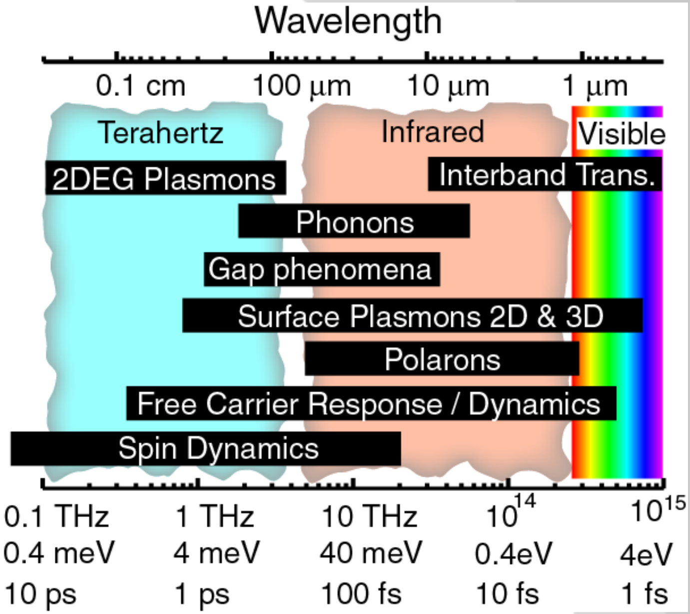
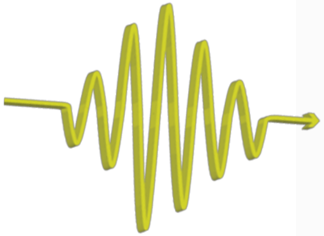


Image courtesy:
D. Basov

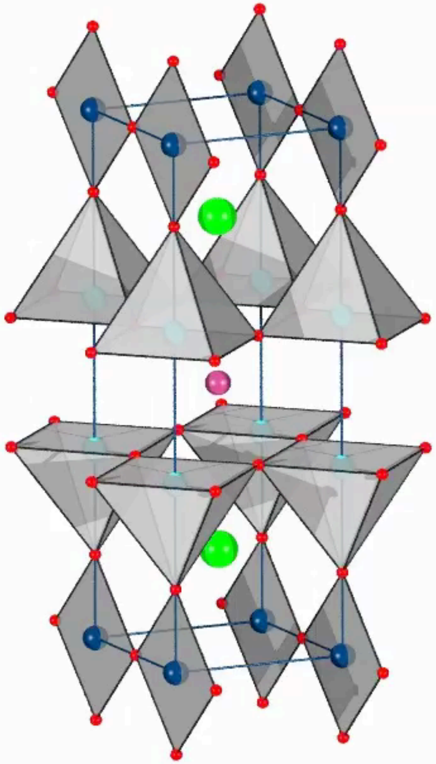
Can we use light to change materials properties?

Optically-driven superconductivity via vibrational excitation

High-T_C cuprates



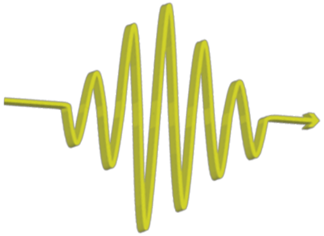
~15 μm
200 fs
5 MV/cm



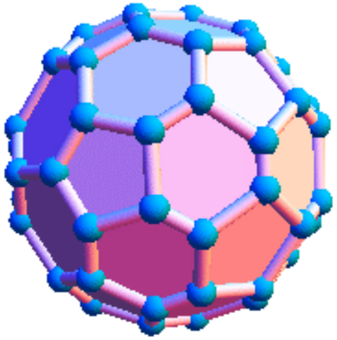
$YBa_2Cu_3O_{6+x}$

W. Hu *et al.*, Nat. Mater. **13**, 705 (2014)
R. Mankowsky *et al.*, Nature **516**, 71 (2014)

Alkali-doped Fullerides



~7-11 μm
200 fs
5 MV/cm



K_3C_{60}

M. Mitrano *et al.*, Nature **530**, 461 (2016)
A. Cantaluppi *et al.*, Nat. Phys. **14**, 837 (2018)

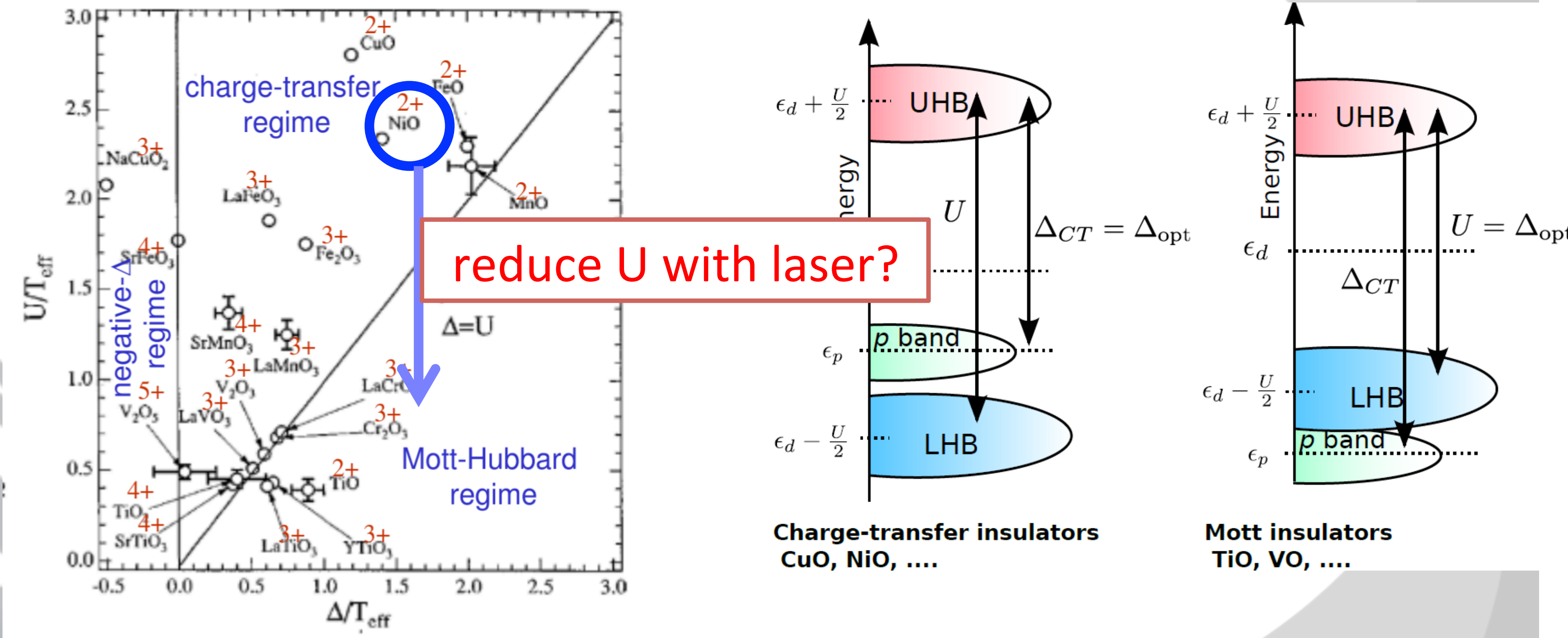
Can we use light to change materials properties?

(1) sometimes yes – but theory/understanding of light-induced superconducting optical response via „nonlinear phononics“ is difficult

(2) simpler case study for theory: modified Hubbard U in a magnetic insulator via light-induced screening (polarization)

Dynamical modification of Hubbard U

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



Charge-transfer insulators
CuO, NiO, ...

Mott insulators
TiO, VO, ...

Zaanen-Sawatzky-Allen phase diagram

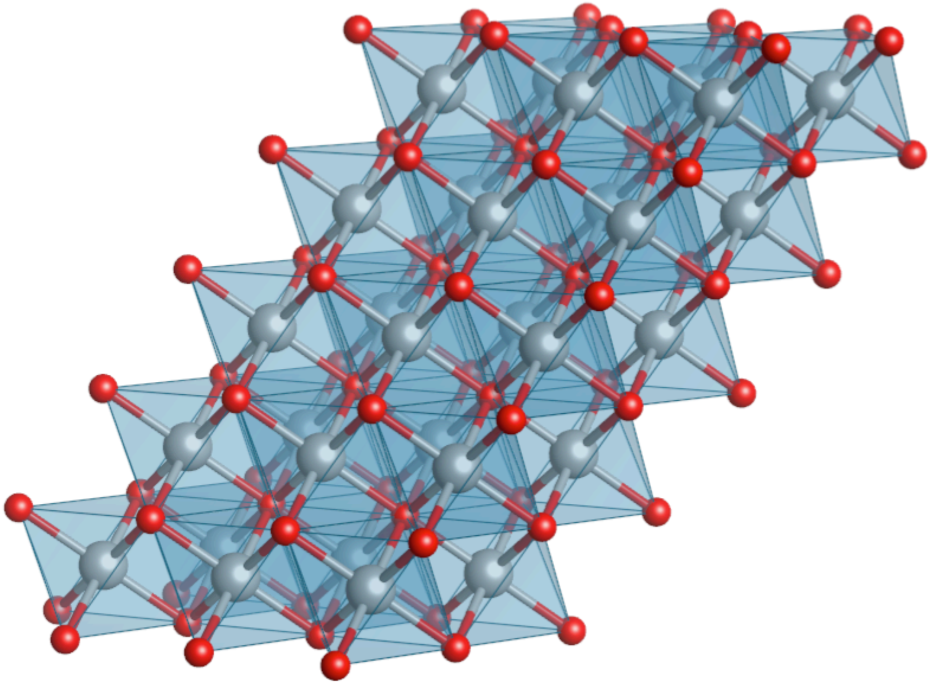
NiO as prototypical charge-transfer insulator

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

↔

occupations Coulomb integrals

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

ACBN0 functional
PRX 5,011006 (2015)

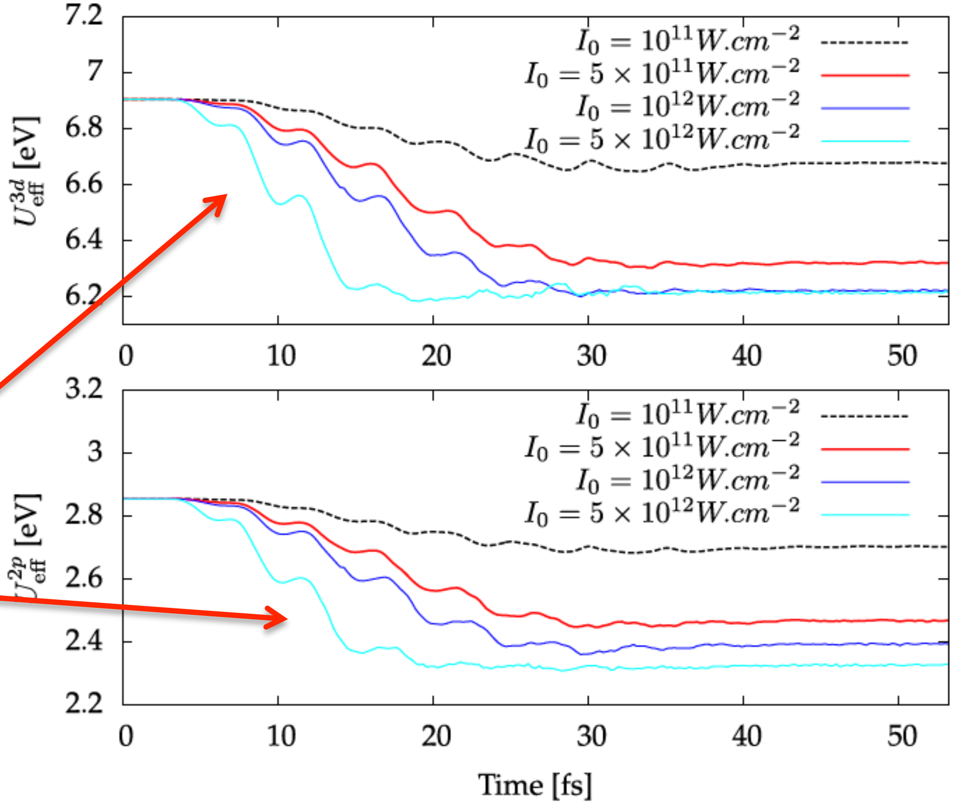
- alternative to constrained RPA
- numerically efficient
- direct extension to time-dependent case (adiabatic approximation)

Ultrafast modification of Hubbard U in NiO

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

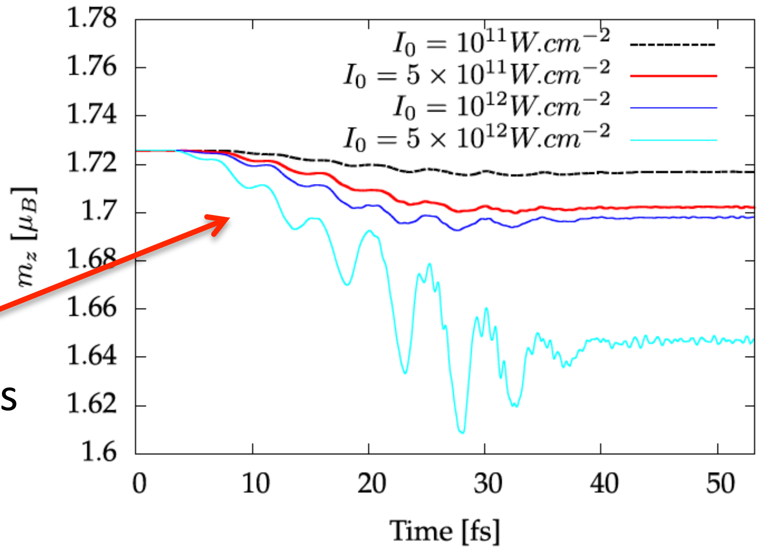
U reduces during the 25 fs laser pulse

Stronger decrease for stronger field strength



Typical intensities in strong field physics in solids

Reduction of U: mechanism

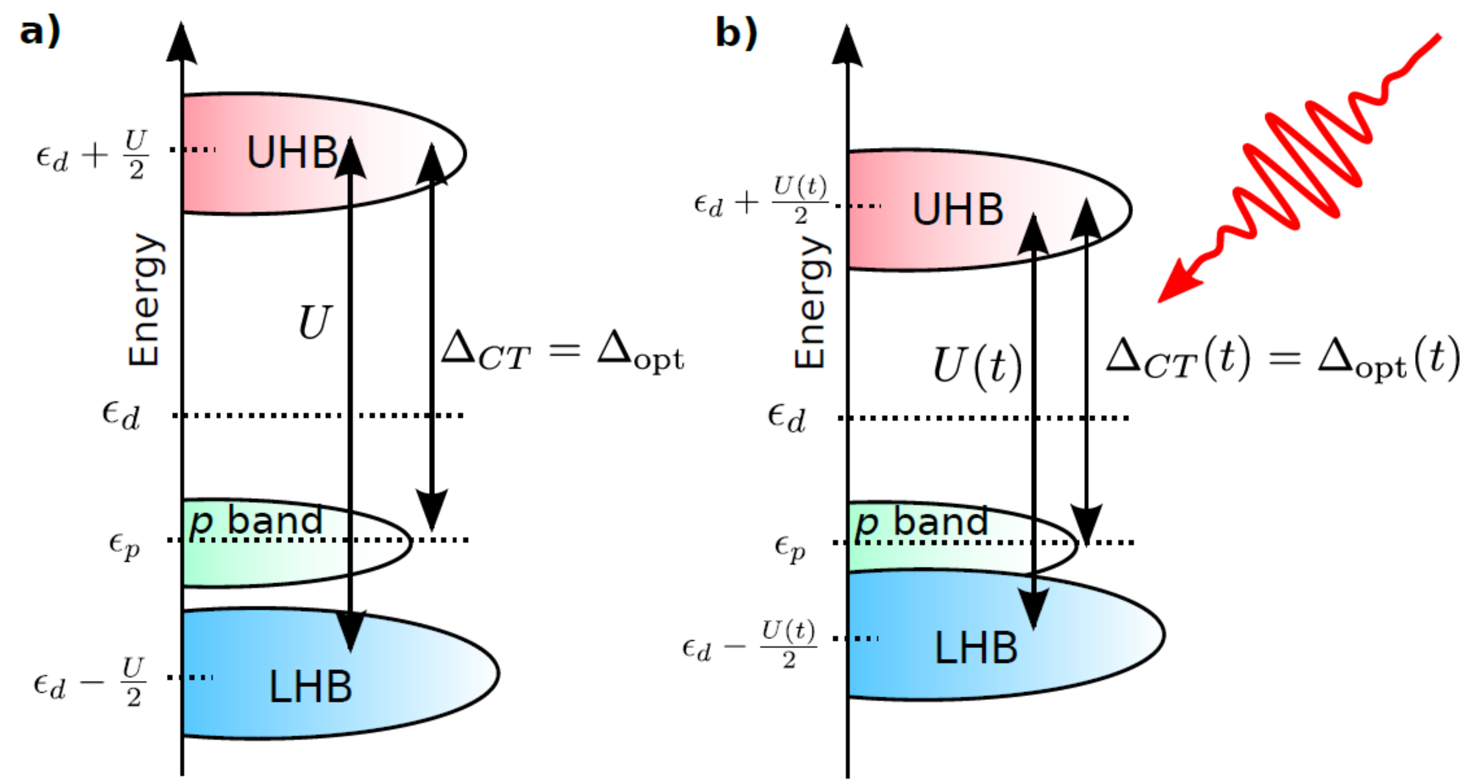


Partial demagnetization of Ni atoms

- Polarization increases
- Enhanced screening
- Decrease of U

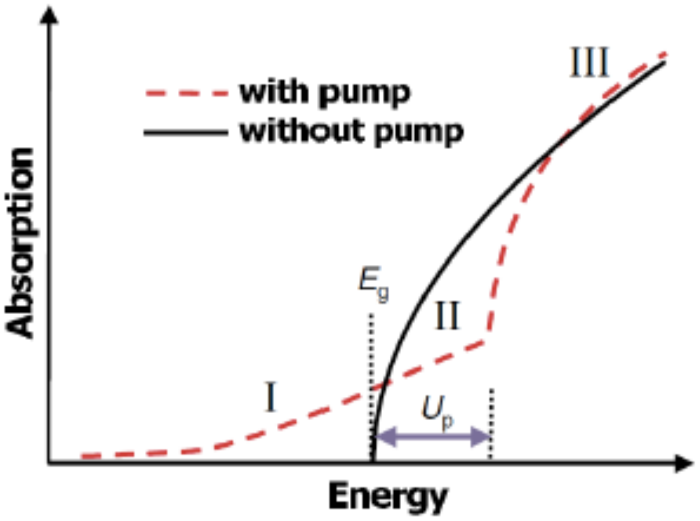
Reduction of U: effect on the optical properties

- What is the effect of time-dependent U on the optical properties ?
- Can we measure fingerprints of the time-dependent U?
- How does U changes compare to the charge transfer energy Δ ?

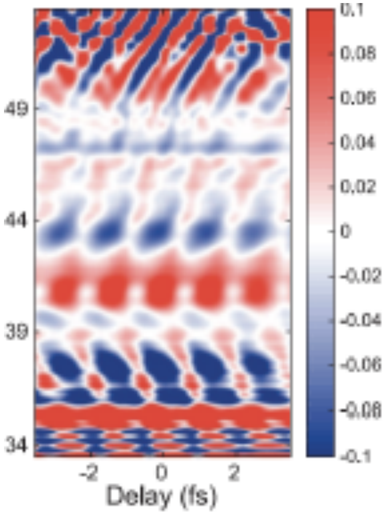


Dynamical Franz-Keldysh Effect

- „conventional“ weakly correlated semiconductors (e.g., GaAs, diamond)



[120] A. Srivastava, R. Srivastava, J. Wang, and J. Kono, "Laser-Induced Above-Band-Gap Transparency in GaAs," Phys. Rev. Lett. **93**, 157401 (2004).



[123] M. Lucchini, S. A. Sato, A. Ludwig, J. Herrmann, M. Volkov, L. Kasmi, Y. Shinohara, K. Yabana, L. Gallmann, and U. Keller, "Attosecond dynamical Franz-Keldysh effect in polycrystalline diamond," Science **353**, 916 (2016).

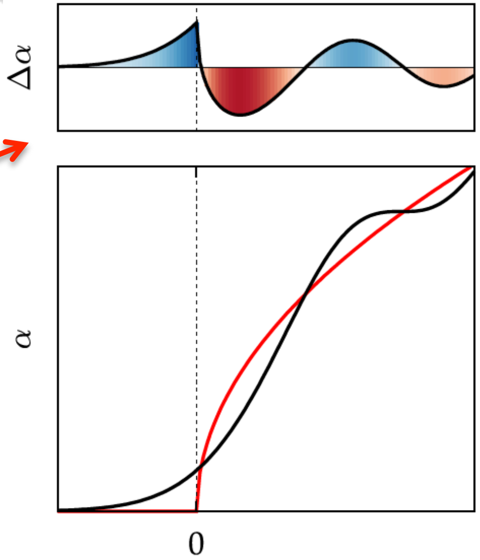
Reduction of U: effect on the optical properties

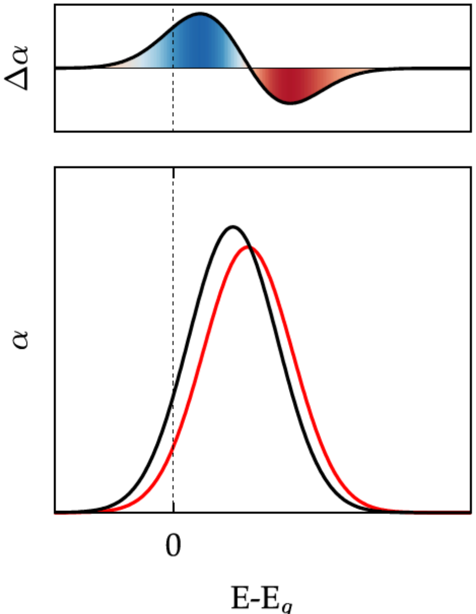
Transient absorption spectroscopy

Two effects expected:

- dynamical Franz-Keldysh effect
  (strong-field response of semiconductors)

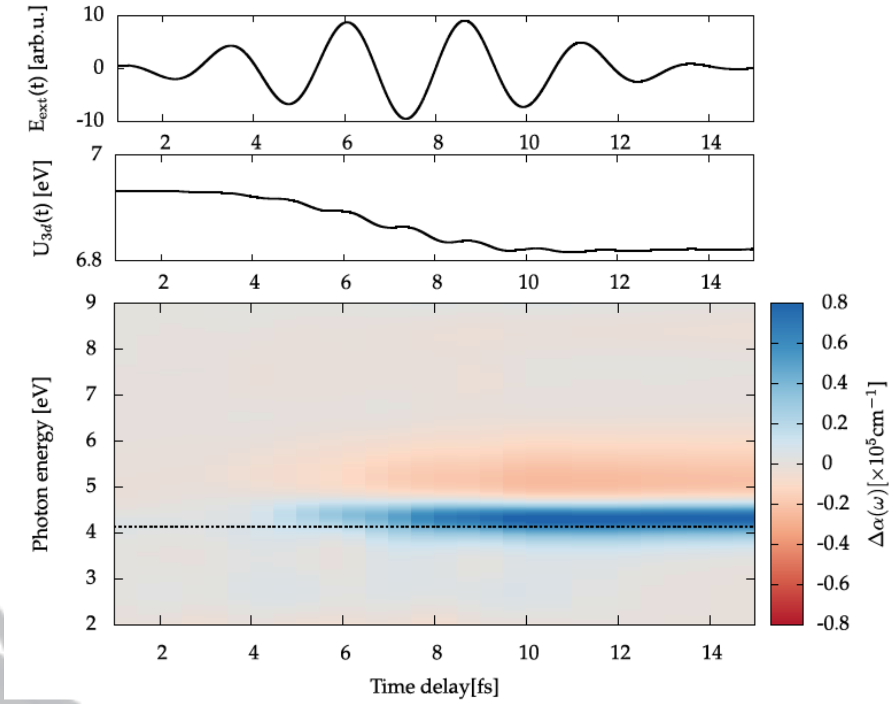
- Shift of the absorption peak
  (U decreases)

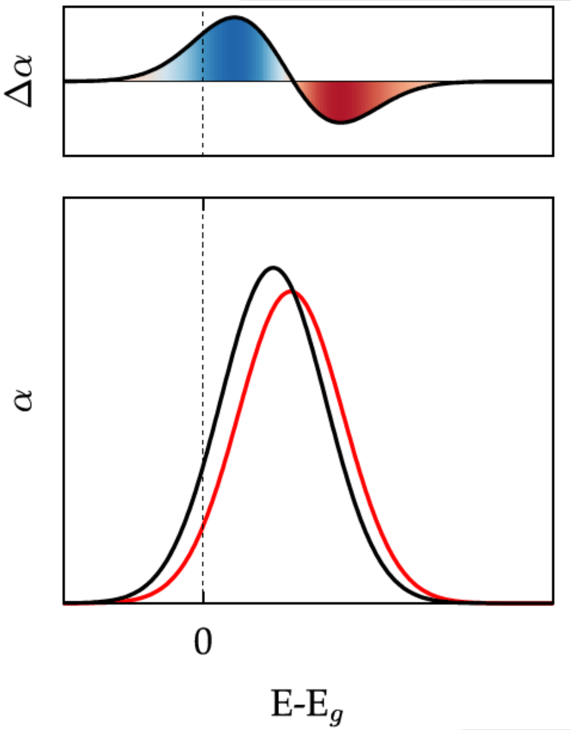




Transient absorption in NiO

Real time TDDFT calculations with dynamical U



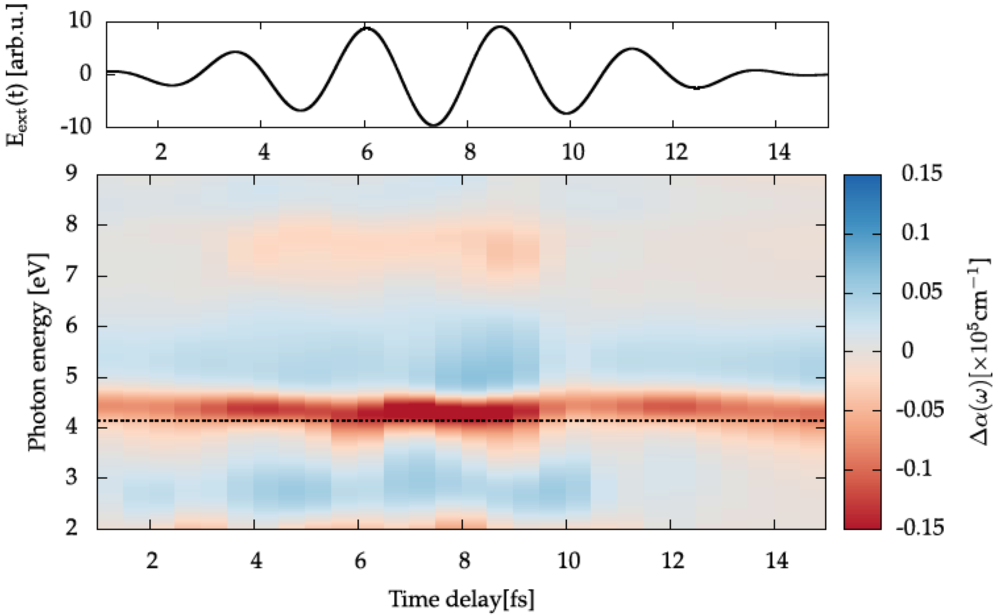


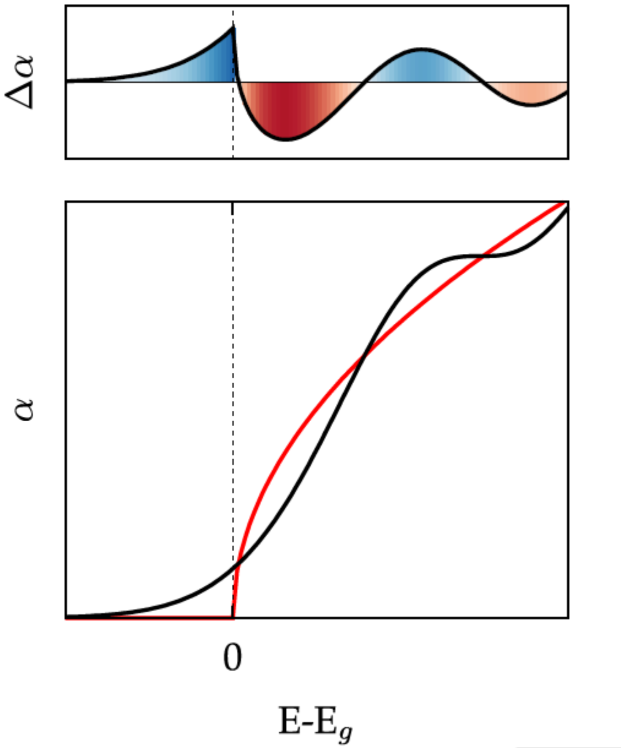
Clear shift of the absorption peak

λ=800nm
$I \sim 4.10^{12}$ W.cm^{-2}
7.5 fs FWHM

Transient absorption in NiO

Real time TDDFT calculations with frozen U

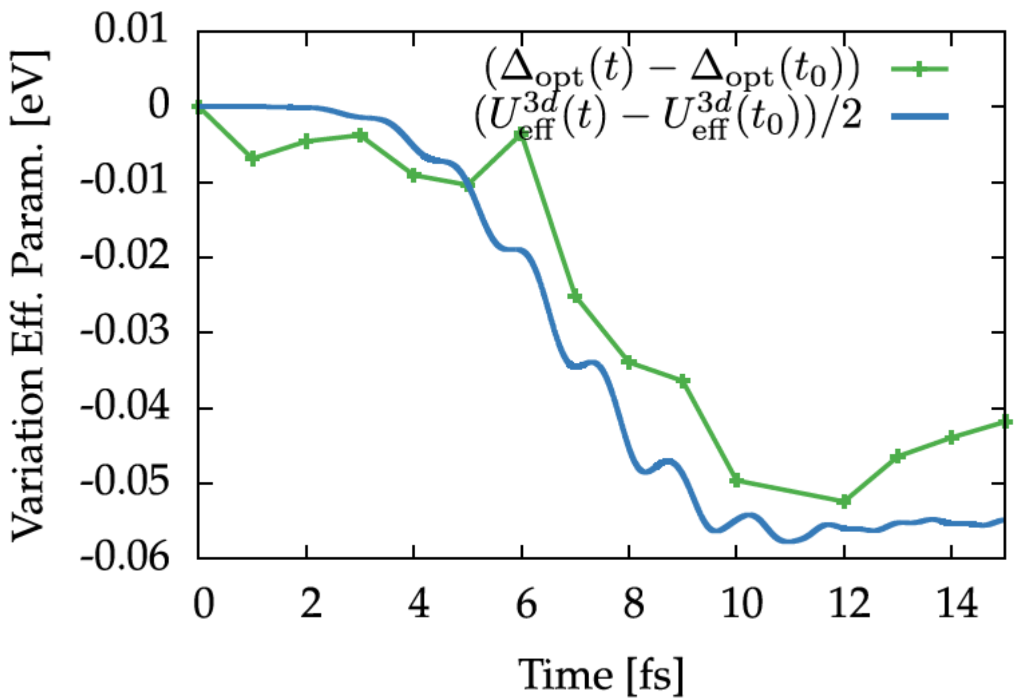


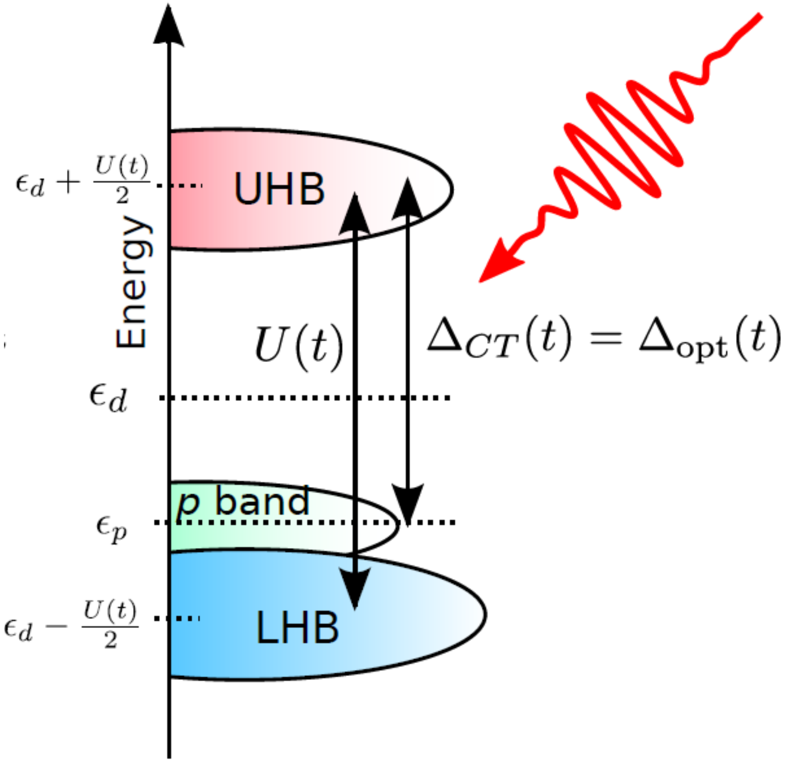


Dynamical Franz-Keldysh effect
Much smaller change
Dynamical U effects should dominate in NiO

Towards light-induced Mott insulators

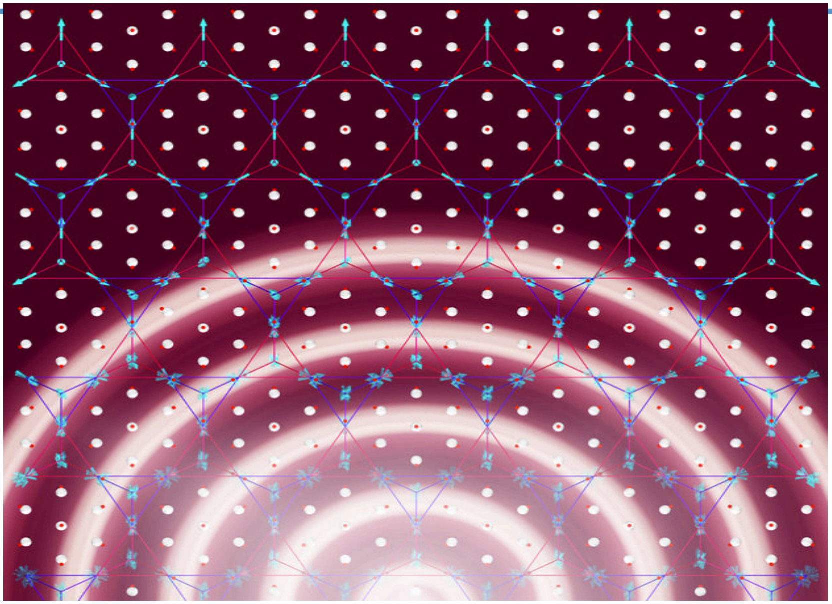
Extracted optical gap *versus* U(t)



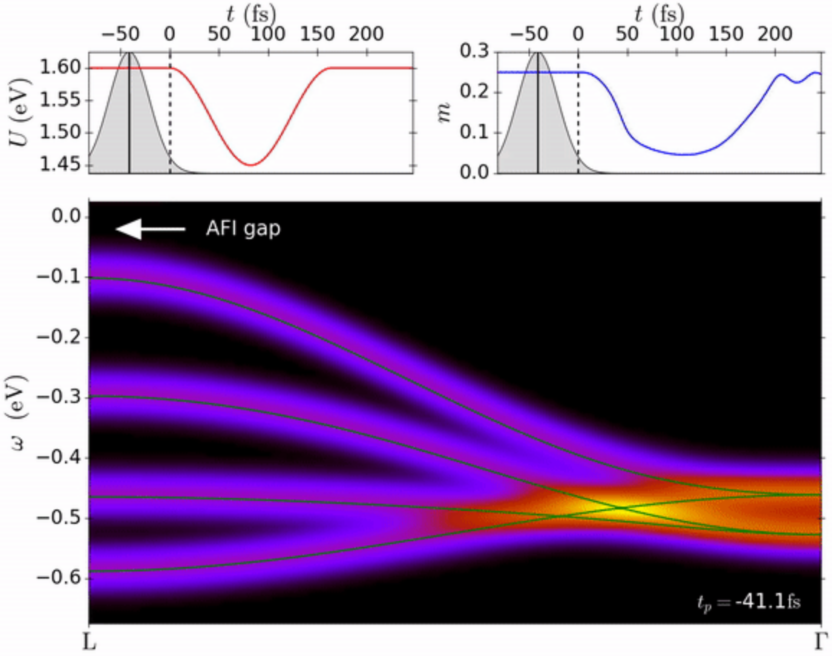


U reduces faster than the optical gap
Δ changes ~ δU/2

Engineering topology with light



G. E. Topp et al., Nature Comm. 9, 4452 (2018)



Coupling engineering

Dynamical ab initio Hubbard U

Light-induced Weyl fermions in pyrochlore iridates

Time-resolved photoemission

nonthermal effects

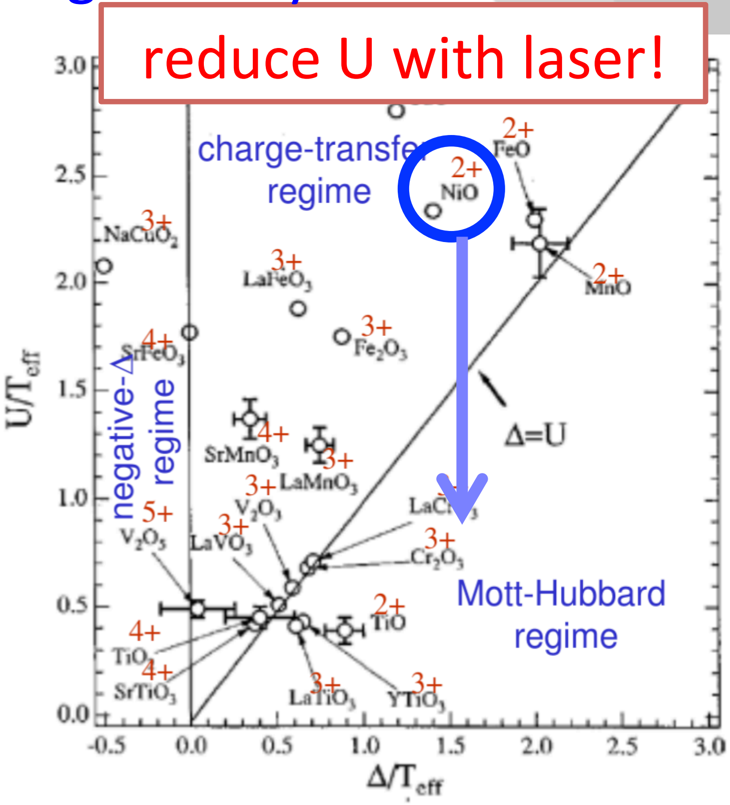
bands + distributions important

Summary

- Ultrafast reduction of Hubbard U in NiO and pyrochlore iridate
- signature: reduced charge-transfer gap in transient absorption
- Towards light-induced Mott insulators? comparison with GW+EDMFT (cf. Martin Eckstein‘s talk)?

We don‘t know what U is, but we can change it for you!

N. Tancogne-Dejean et al., PRL 121, 097402 (2018)
G. E. Topp et al., Nature Comm. 9, 4452 (2018)
N. Tancogne-Dejean et al., in preparation


reduce U with laser!


N. Tancogne-Dejean


G. Topp