Photodoped charge transfer insulators

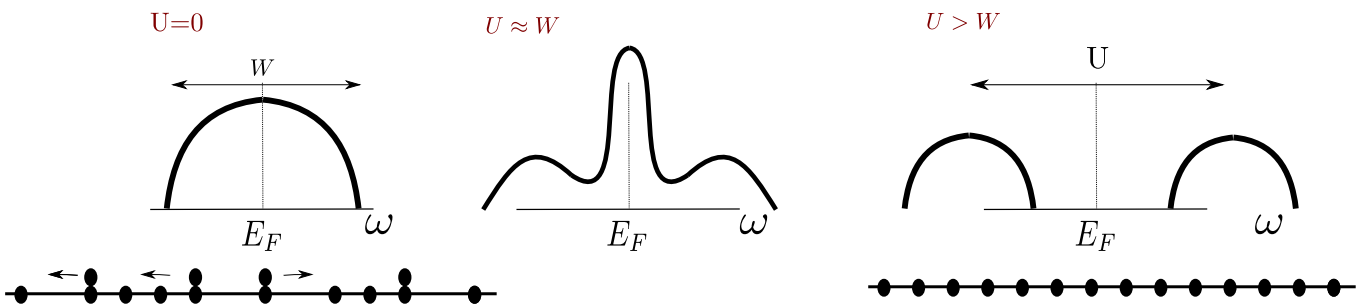
Denis Golež
CCQ, Flatiron Institute

Taming Non-Equilibrium Systems: from Quantum Fluctuation to Decoherence, July 2019

Mott insulators

- Failure of band theory
- Strong electron-electron interaction
- Hubbard model and Mott gap
- Metal-insulator transition

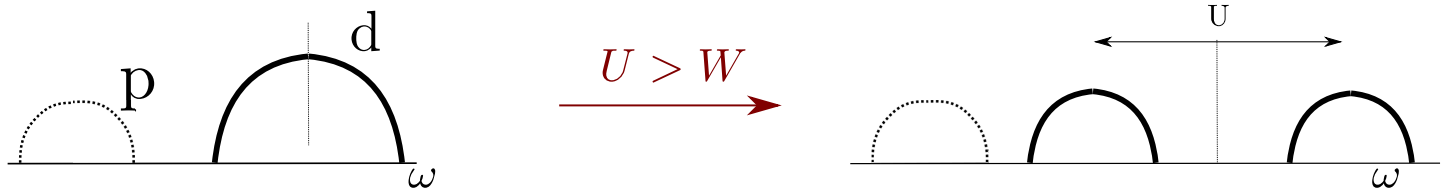
$$H = -t \sum_{<i,j>\sigma} c^{\dagger}_{i,\sigma} c_{j,\sigma} + U \sum_{i} n_{i\downarrow} n_{i\uparrow}$$



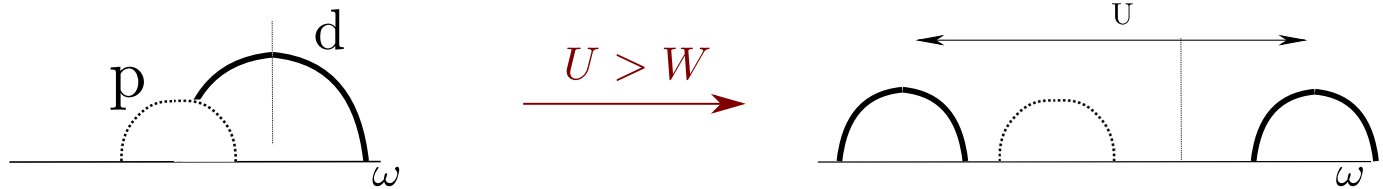
Charge transfer insulators

- Multi band physics

Mott

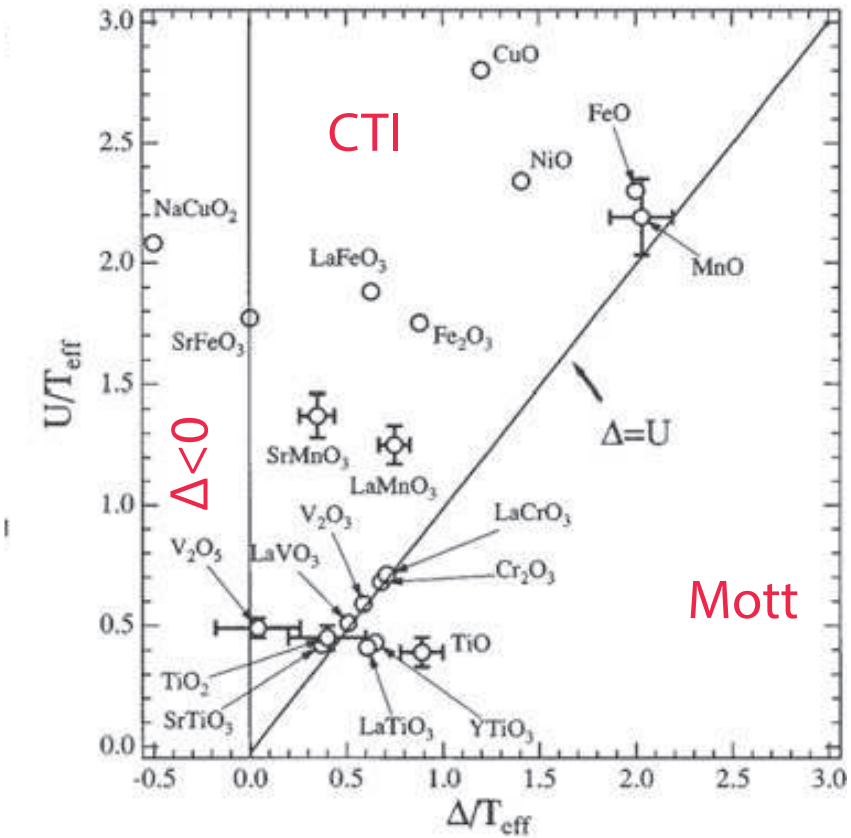


CTI



Charge transfer insulators

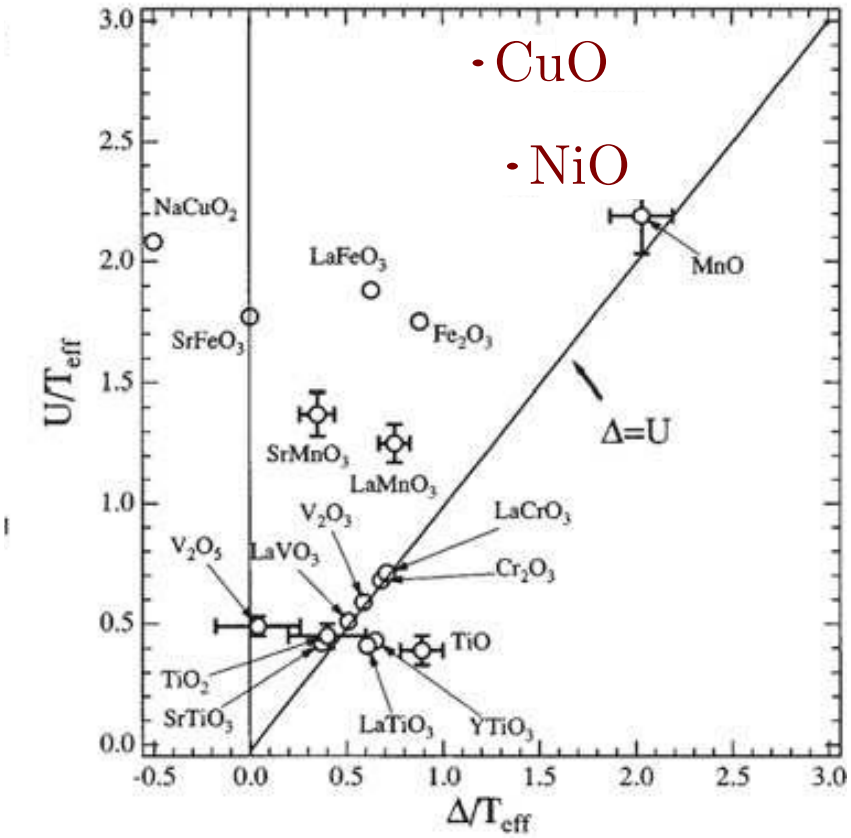
- Multi band physics
- Zaanen-Sawatzky-Allen diagram



F. Gebhard, The Mott Metal-Insulator Transition

Charge transfer insulators

- Multi band physics
- Zaanen-Sawatzky-Allen diagram



F. Gebhard, The Mott Metal-Insulator Transition

Pump-probe on Mott insulators

- Use strong laser pulses to photo-excite charge carriers
- Delayed probe pulse (optics, photo-emission, RIXS, . . .)

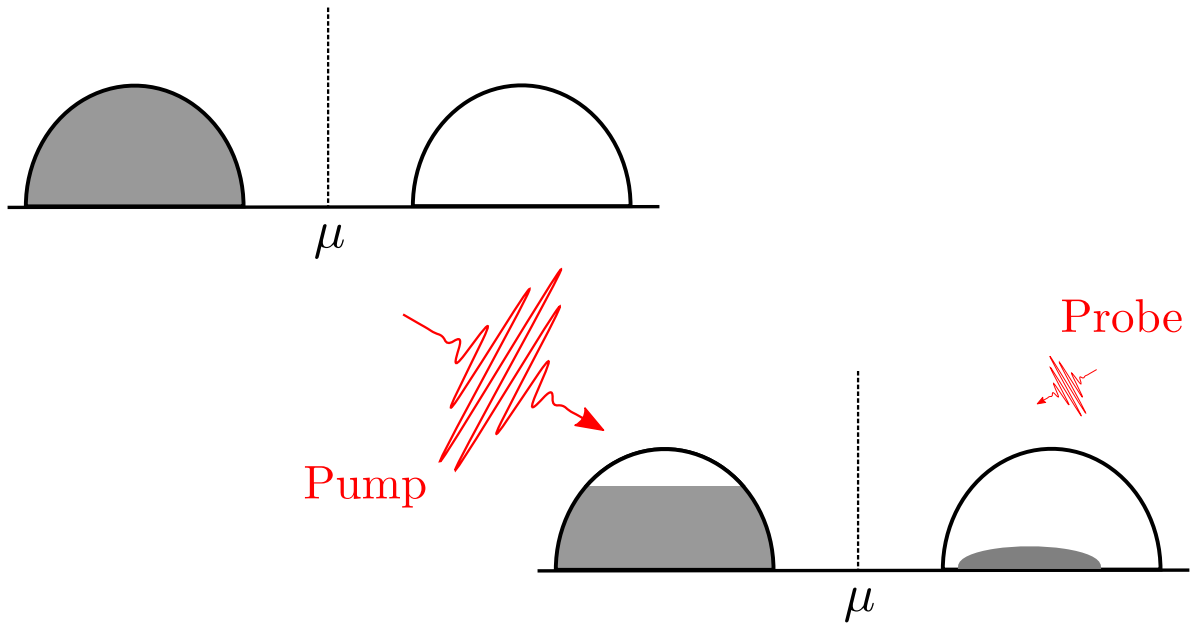
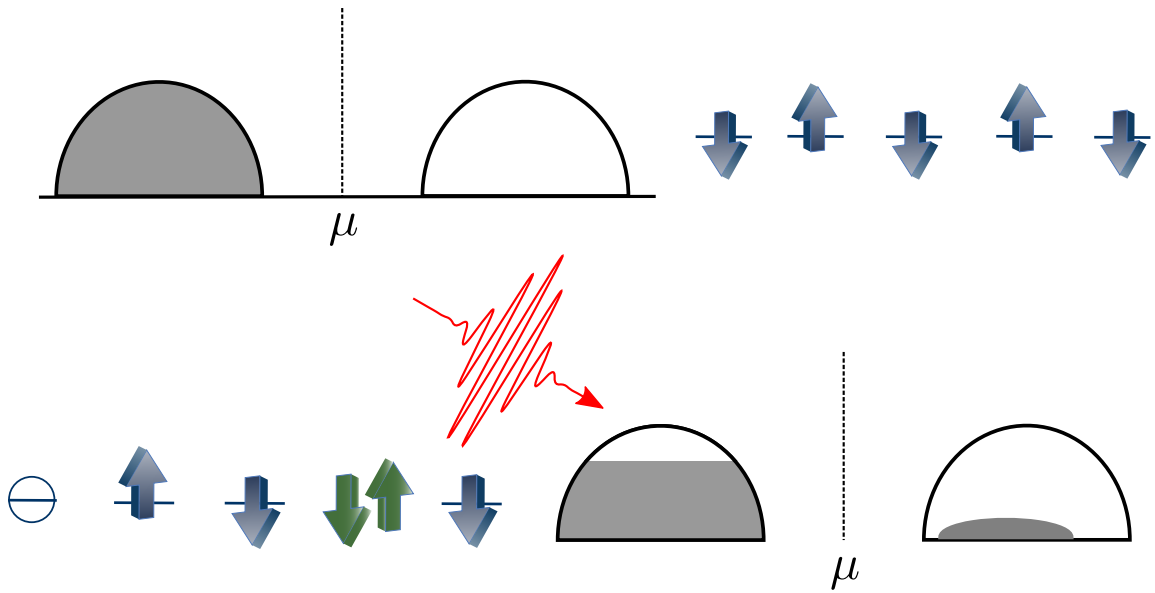


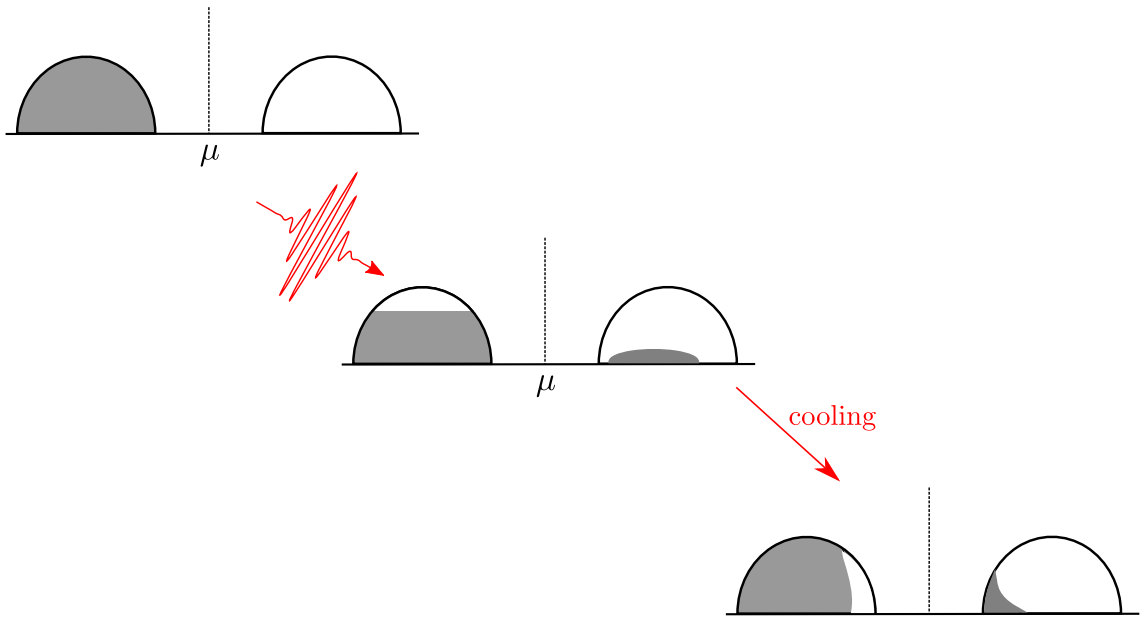
Photo-excitation of Mott insulators - II

- Use strong laser pulses to photo-excite charge carriers
- Mobile doublons and holons



Relaxation

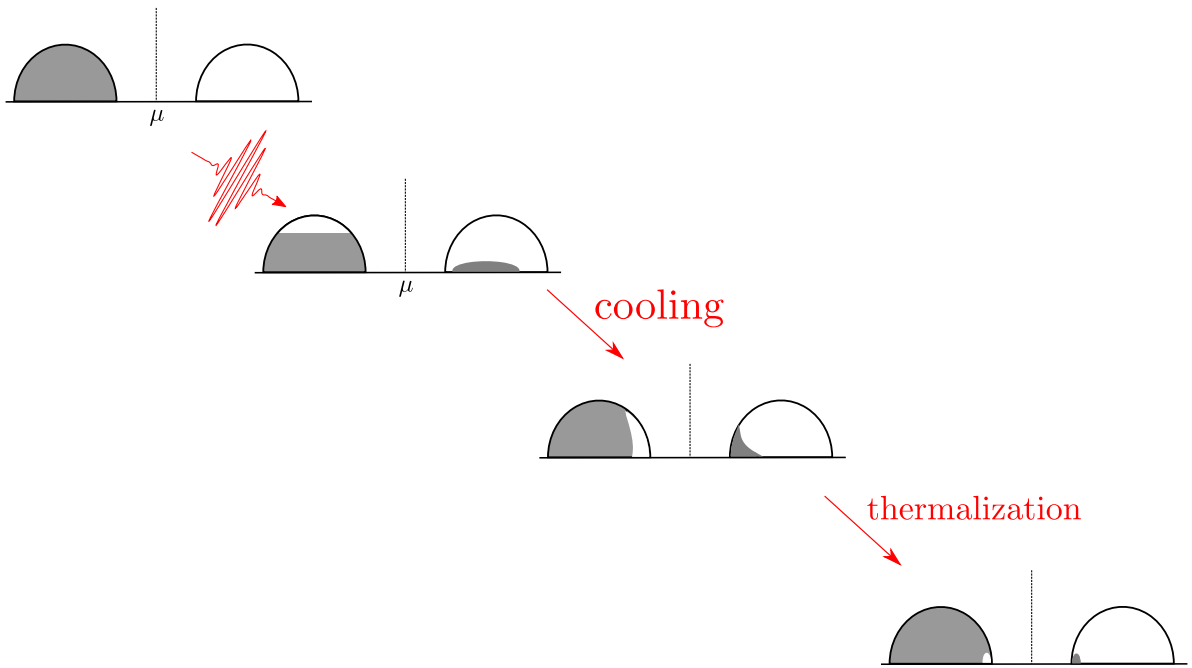
- Holon and doublon number conserved
- Role of bosonic modes (spins, phonons, plasmons)
- Kinetic processes



Semsarna, et.al. PRB 82,224302(2010)
Eckstein, et.al. PRB 035122 (2011)
Lenarčič, et.al. PRL 111,016401 (2013)

Thermalization

- Holon doublon recombination
- Exponentially suppressed - energy conservation
- Time scale separation between cooling and thermalization



Semsarna, et.al. PRB 82,224302(2010)
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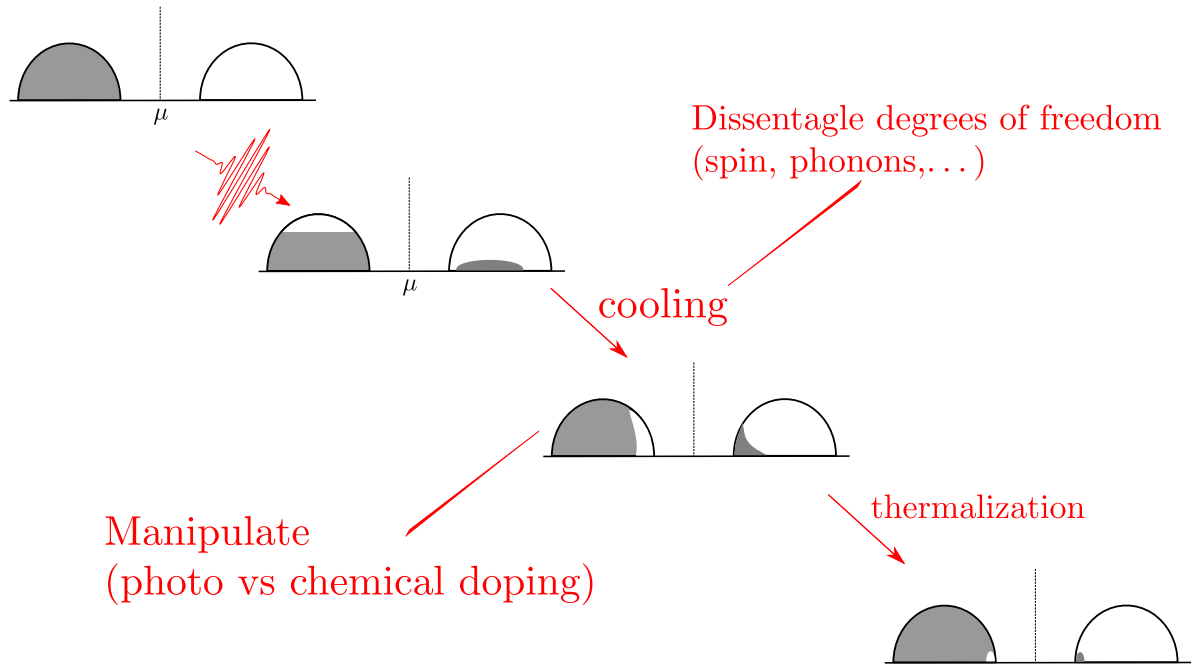
Goals

- Is multiband picture essential ?
- Properties of trapped states
- Role of collective modes: charge and spin screening



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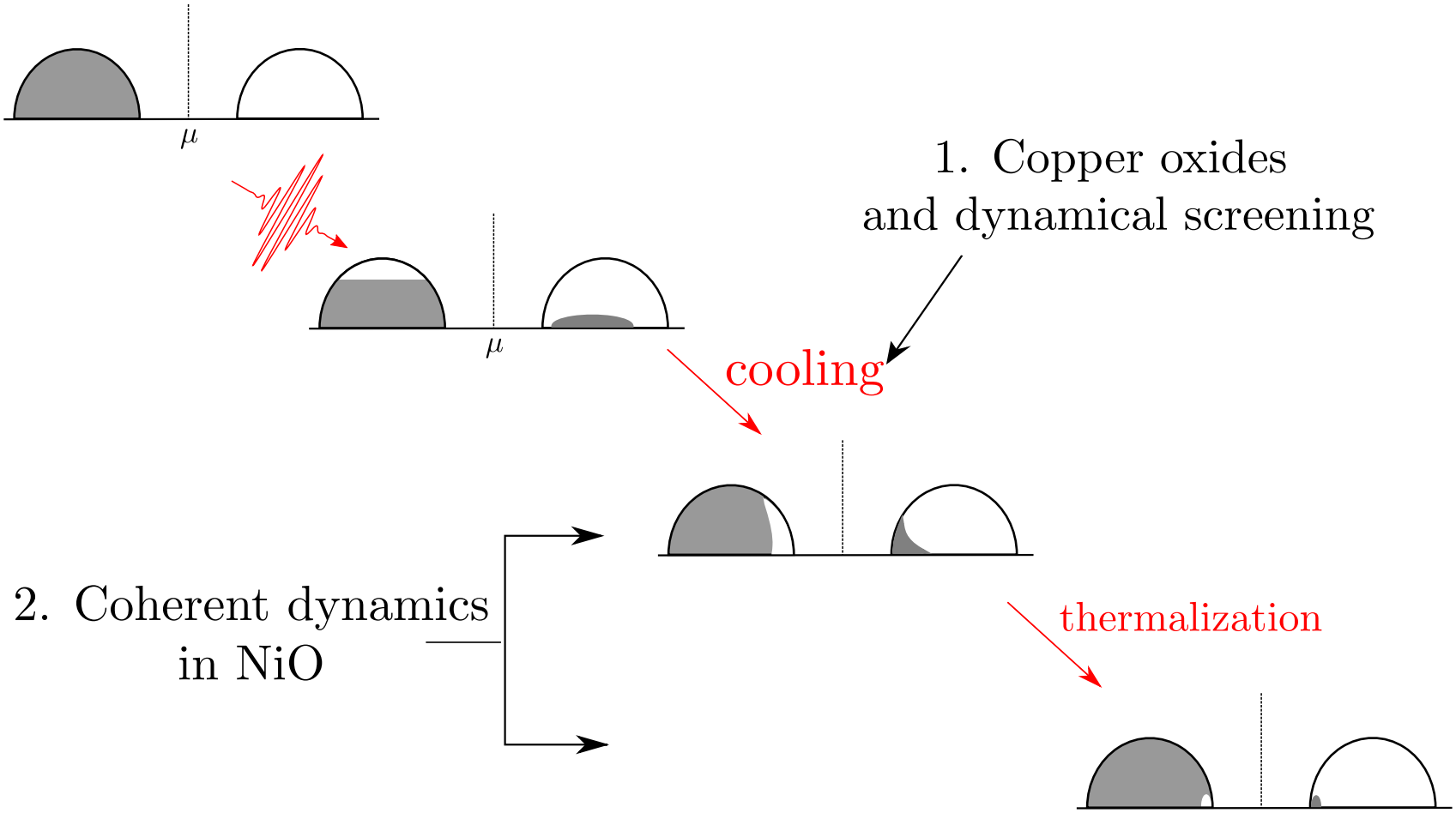
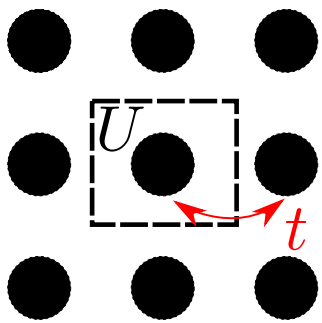
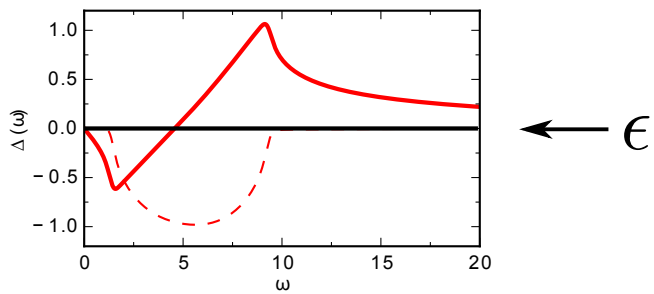


DMFT

$$H = -t \sum_{<i,j>} [c_j^\dagger c_i + h.c.] + U \sum_i n_{i\downarrow} n_{i\uparrow}$$

- Hybridization function $\Delta(t, t')$
- Local self-energy





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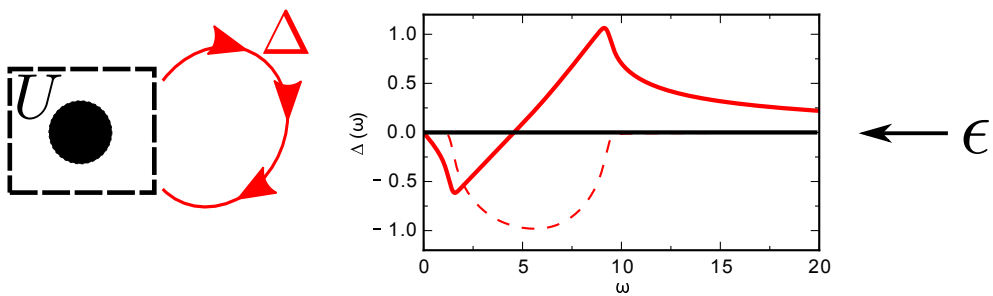
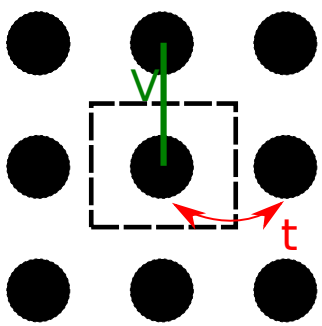
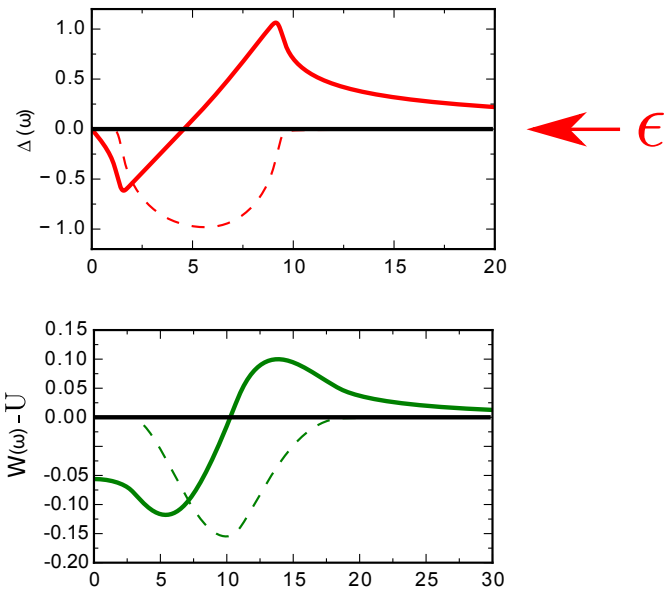


DMFT and screening

$$H = -t \sum_{<i,j>} [c_j^\dagger c_i + h.c.] + U \sum_i n_{i\downarrow} n_{i\uparrow} + V \sum_{<i,j>} n_i n_j$$

- Hybridization function $\Delta(t, t')$
- Effective interaction $W(t, t')$
- Local self-energy and polarization





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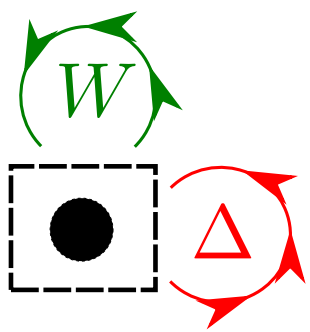
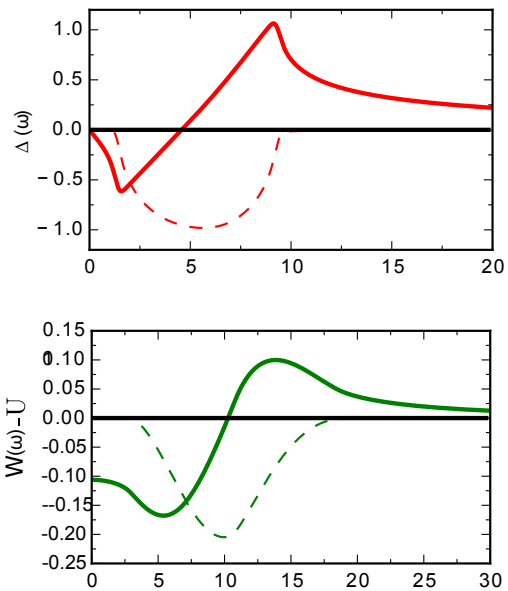
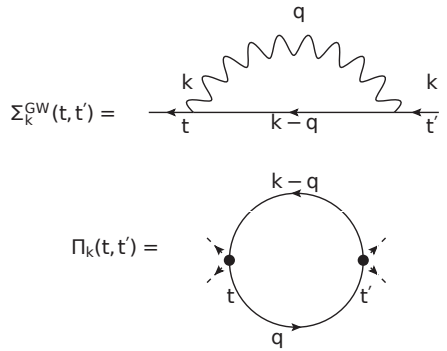
- Hybridization function $\Delta(t, t')$
- Effective interaction $W(t, t')$
- Local self-energy and polarization - EDMFT





Non-local fluctuations

$$\Sigma_k = \Sigma^{EDMFT} + \Sigma^{GW}_k - \Sigma^{GW}_{loc}$$
$$\Pi_k = \Pi^{EDMFT} + \Pi^{GW}_k - \Pi^{GW}_{loc}$$



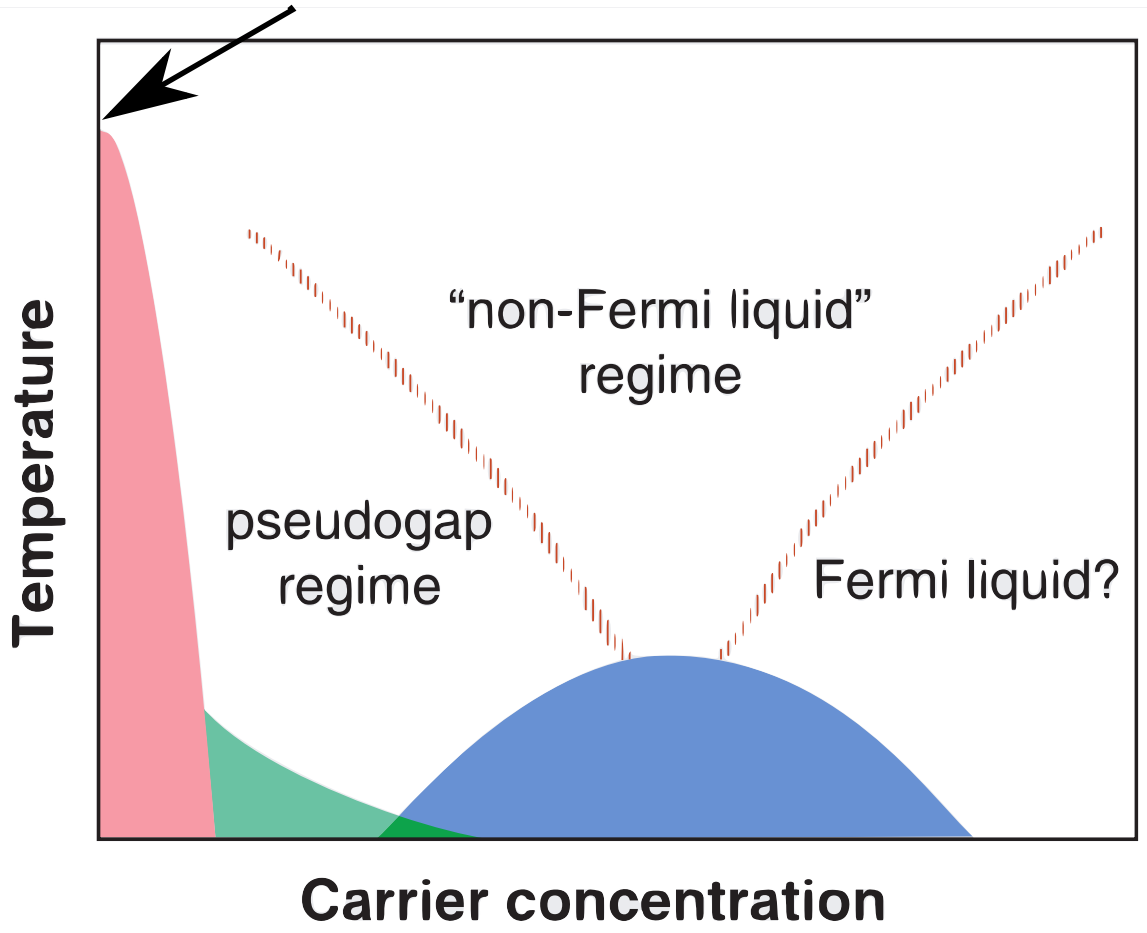
1. Effect of non-local fluctuations using GW+EDMFT

References:

- Eq. implementation (Sun et.al. PRB 66,085120 (2002))
- Full implementation (Ayral et.al. PRL 109, 226401 (2012))
- Non-equilibrium implementation (DG et.al. PRL 118,246402(2017))
- Ab-initio for $SrVO_3$(Boehnke et.al. PRB 94,201106(2016))

Phase diagram

- Role of multiband and screening
- Half-filled and high-temperatures



J. Orenstein et.al. Science 228, 468(2000)

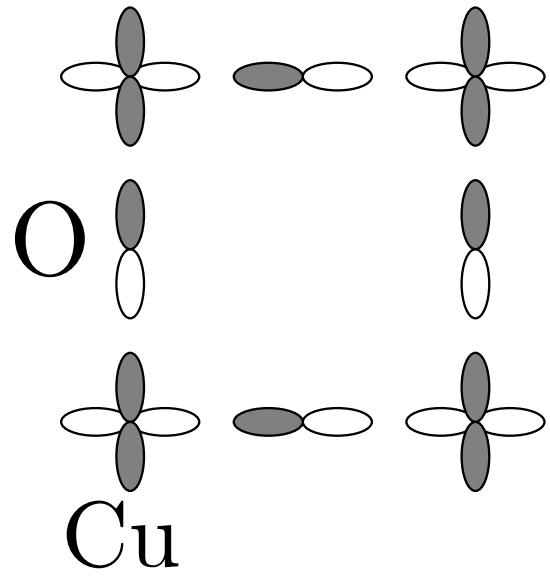
Emery model

$$H = H_{\mathrm{e}} + H_{\mathrm{kin}} + H_{\mathrm{int}}$$

$$H_{\mathrm{e}} = \epsilon_d \sum_i n_i^d + (\epsilon_d + \Delta_{pd}) \sum_{i,\delta} n_i^p,$$

$$H_{\mathrm{kin}} = \sum_{ij\sigma} \sum_{(\alpha,\beta)\in(d,p_x,p_y)} t_{ij}^{\alpha\beta} c_{i\alpha\sigma}^{\dagger} c_{j\beta\sigma},$$

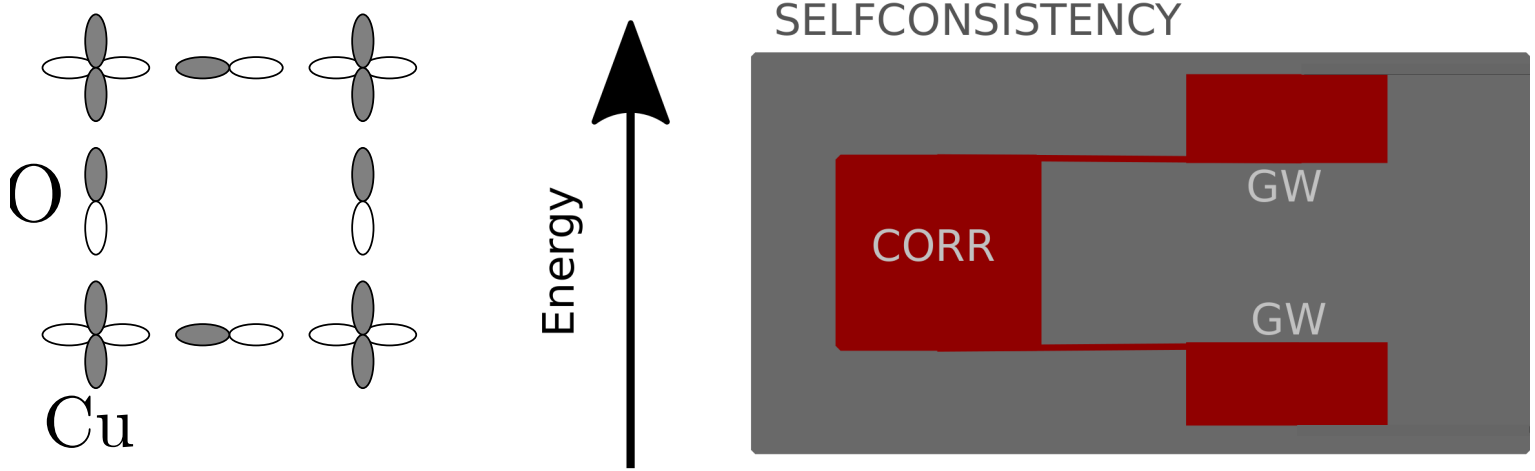
$$H_{\mathrm{int}} = \sum_{ij} \sum_{(\alpha,\beta)\in(d,p_x,p_y)} U_{ij}^{\alpha\beta} n_i^{\alpha} n_j^{\beta},$$



La_2CuO_4: $U^{dd} = 5.0$ eV, $U^{dp} = 2.0$ eV, $t^{dp} = 0.5$ eV, $t^{dd} = -0.1$ eV, $t^{pp} = 0.15$ eV, $\Delta_{pd} = -3.5$ eV

Multiscale description

- Downfolding for Emery model
- d-orbital within DMFT and p-orbitals with computational cheaper approaches (HF,GW)



Equilibrium spectrum

- Antibonding band - Zhang-Rice singlet
- Bonding band
- Upper Hubbard band

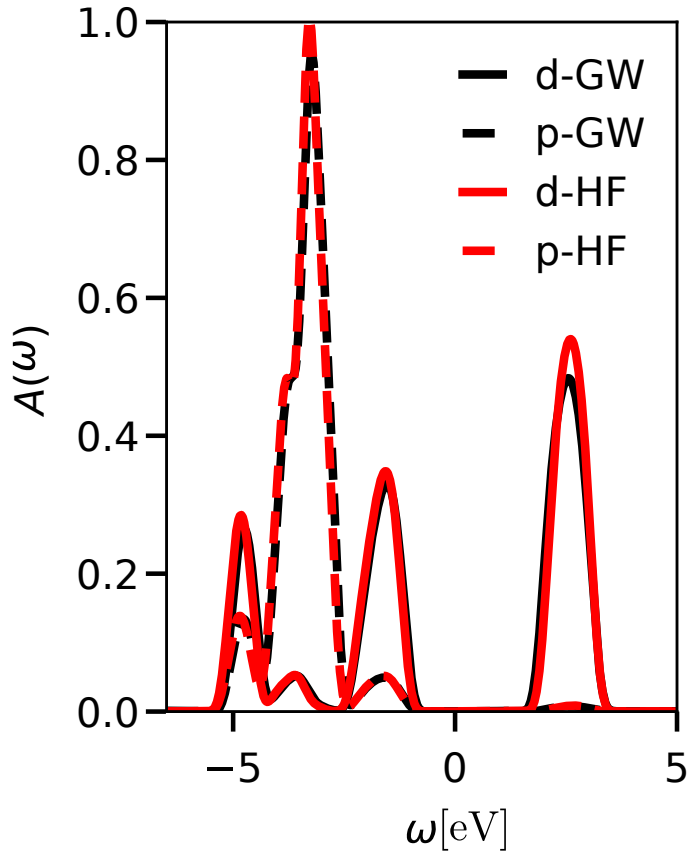
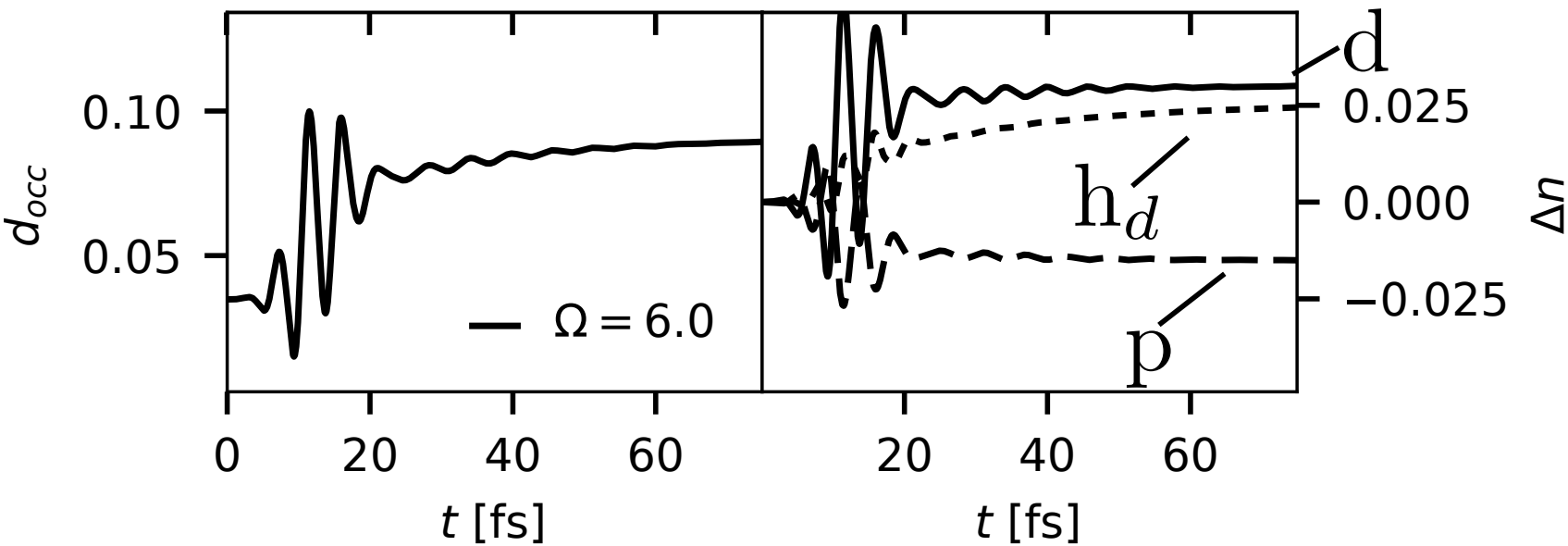


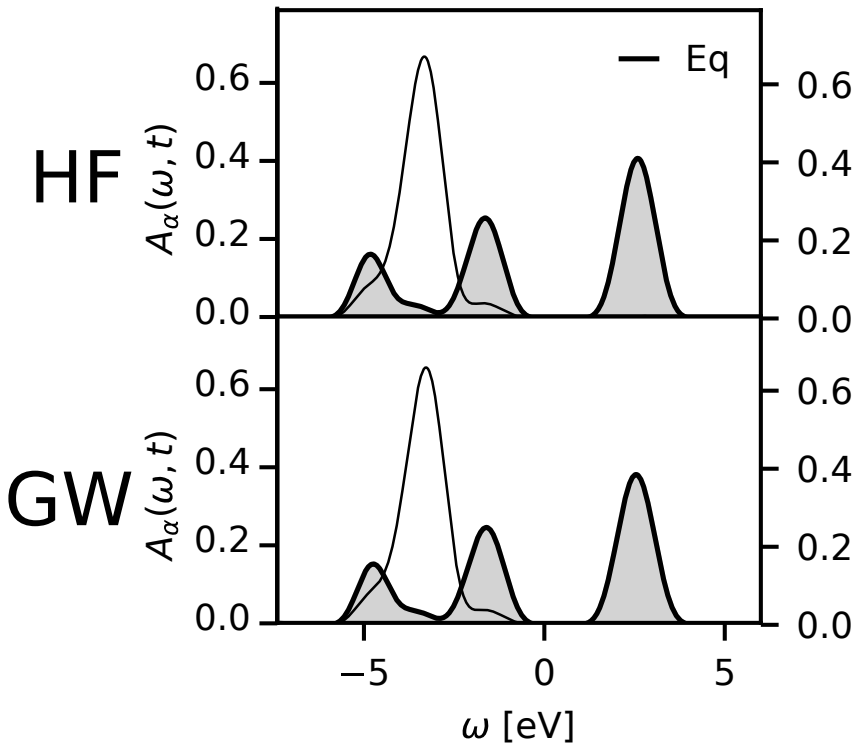
Photo-excitation

- Transfer from p to d electrons
- Photo-induced double occupancy
- Number of holes on d orbital $h_d = \Delta d_{occ} - 2\Delta n_p$



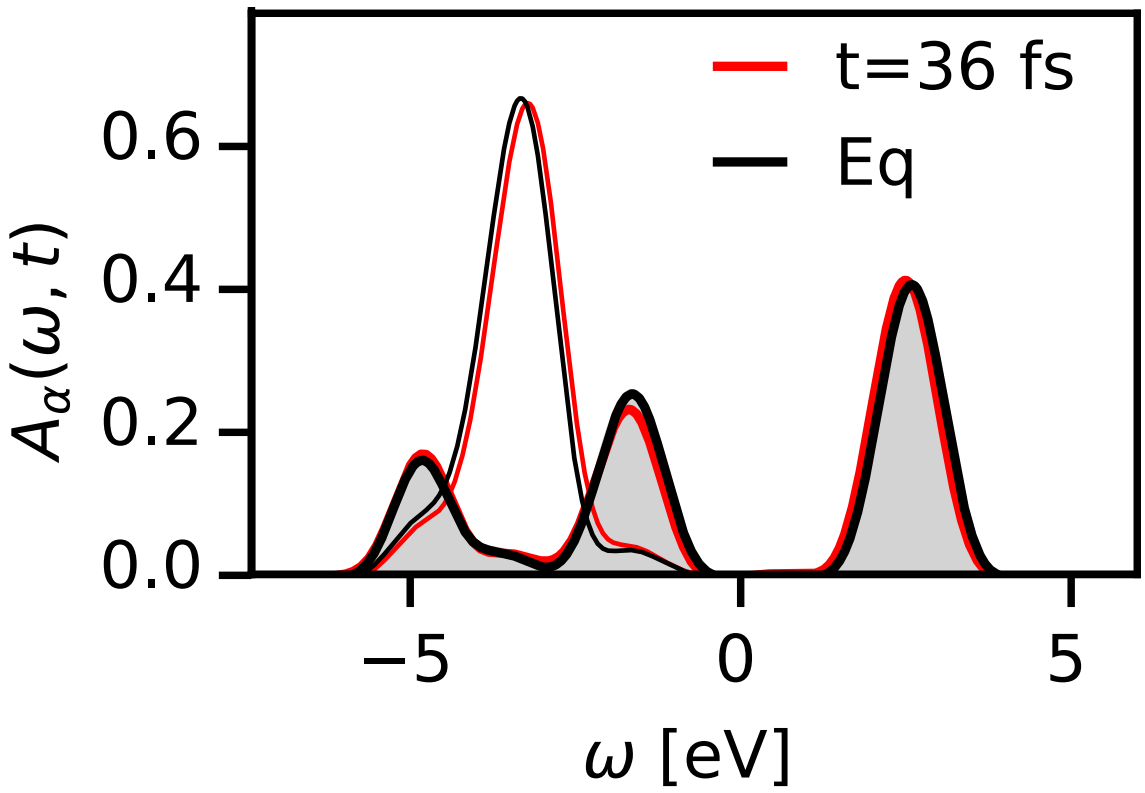
PES

- Dynamical screening without importance

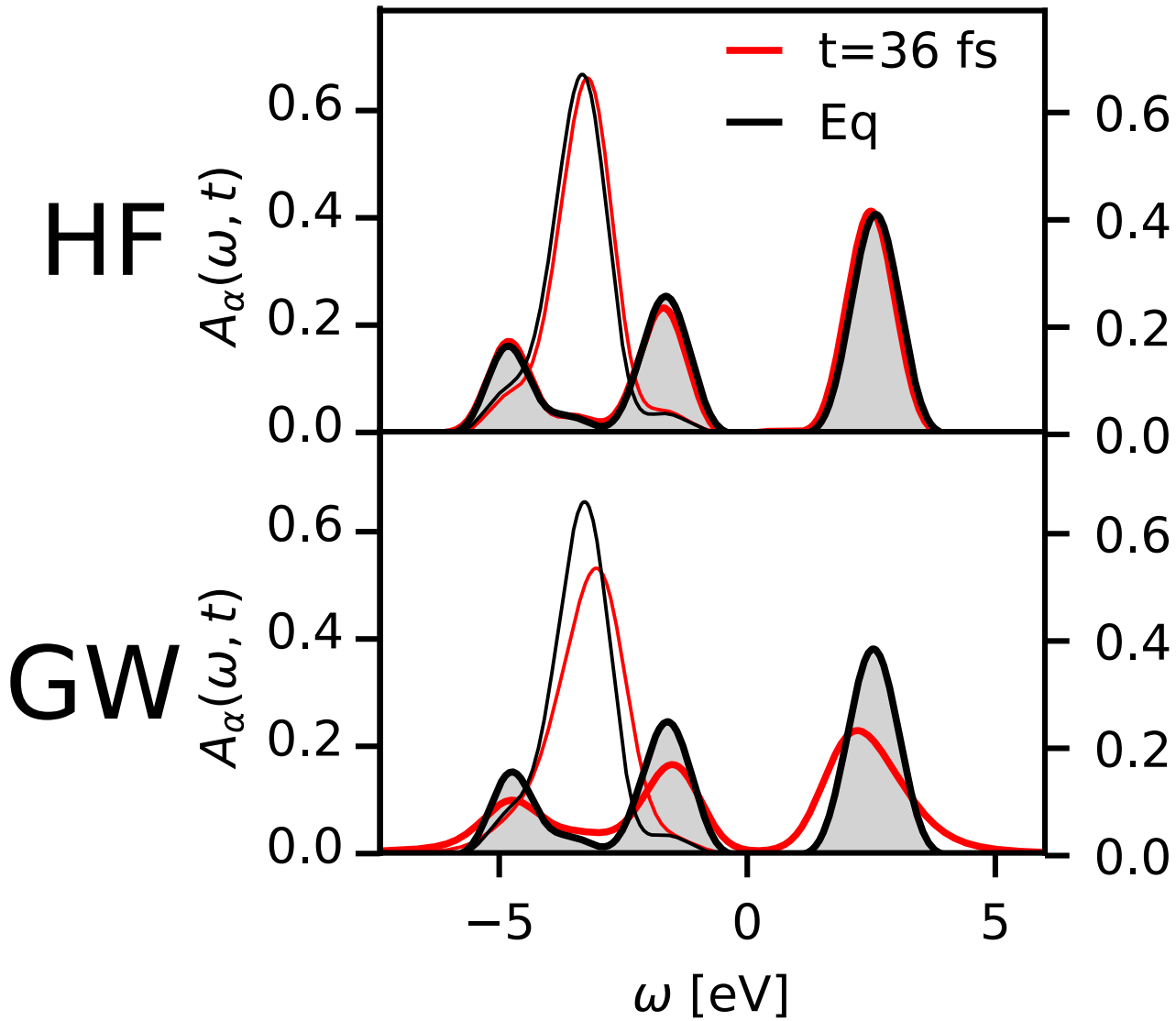


- Hartree shift due to electron-hole attraction

$$\Delta\Sigma^{\mathrm{H}}_{dd} = (U_{dd} - 2U_{dp})\Delta n_d$$

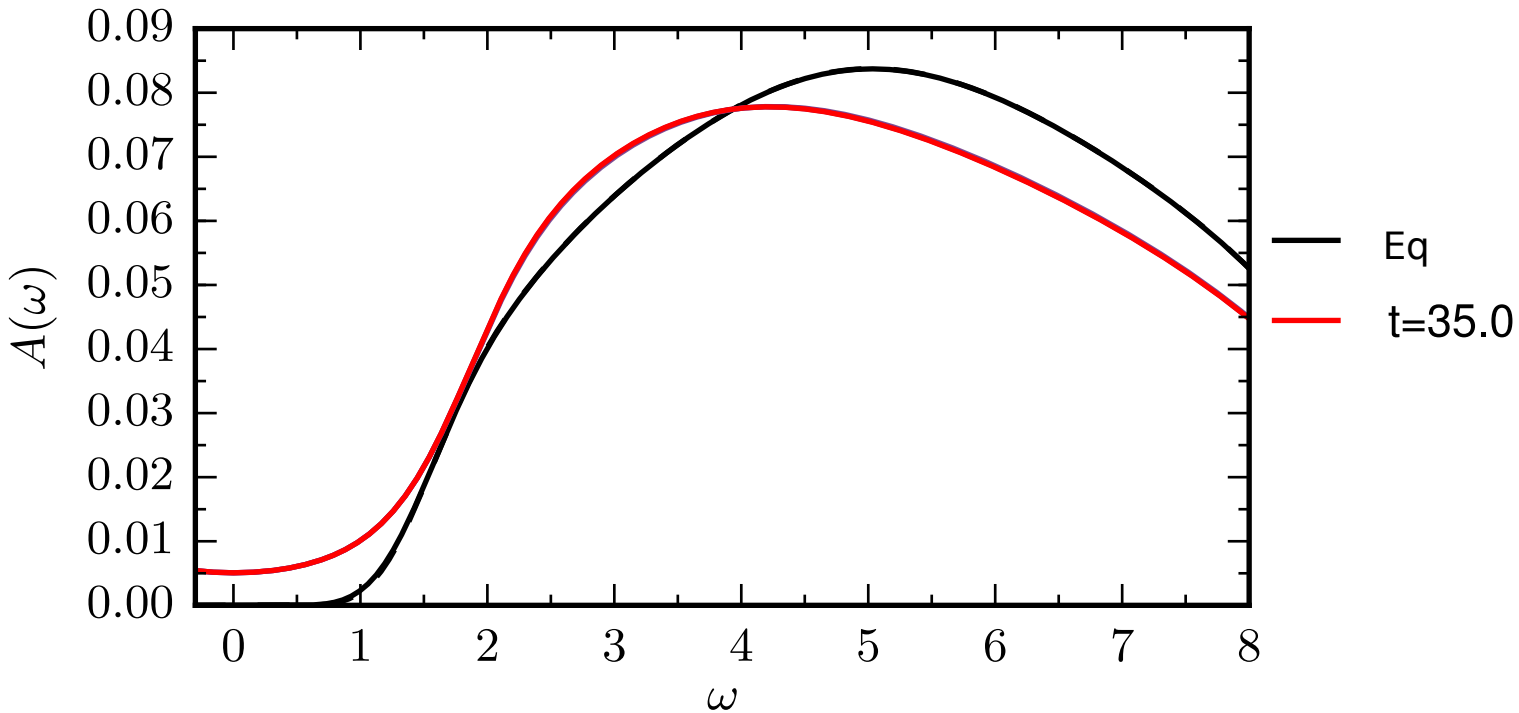


t-PES: dynamical screening



Single band Mott insulator

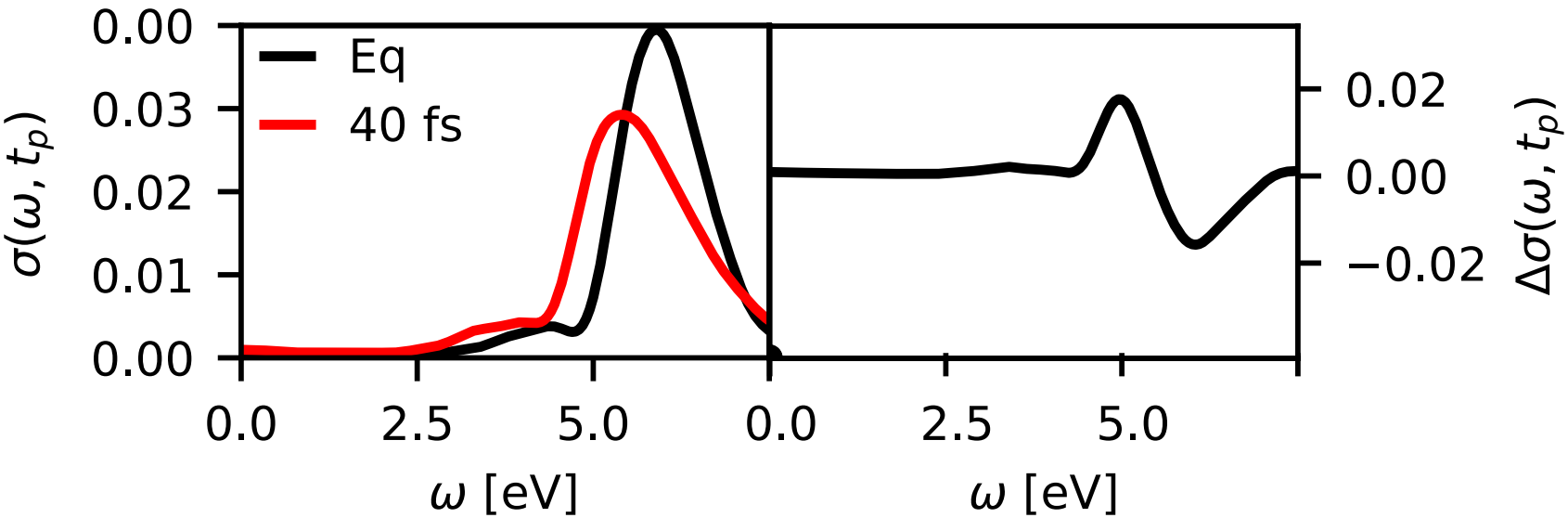
- Minor reduction and broadening of the Hubbard gap
- Dynamical screening enhanced in multiband case



DG et.al. PRB 92,195123(2015)

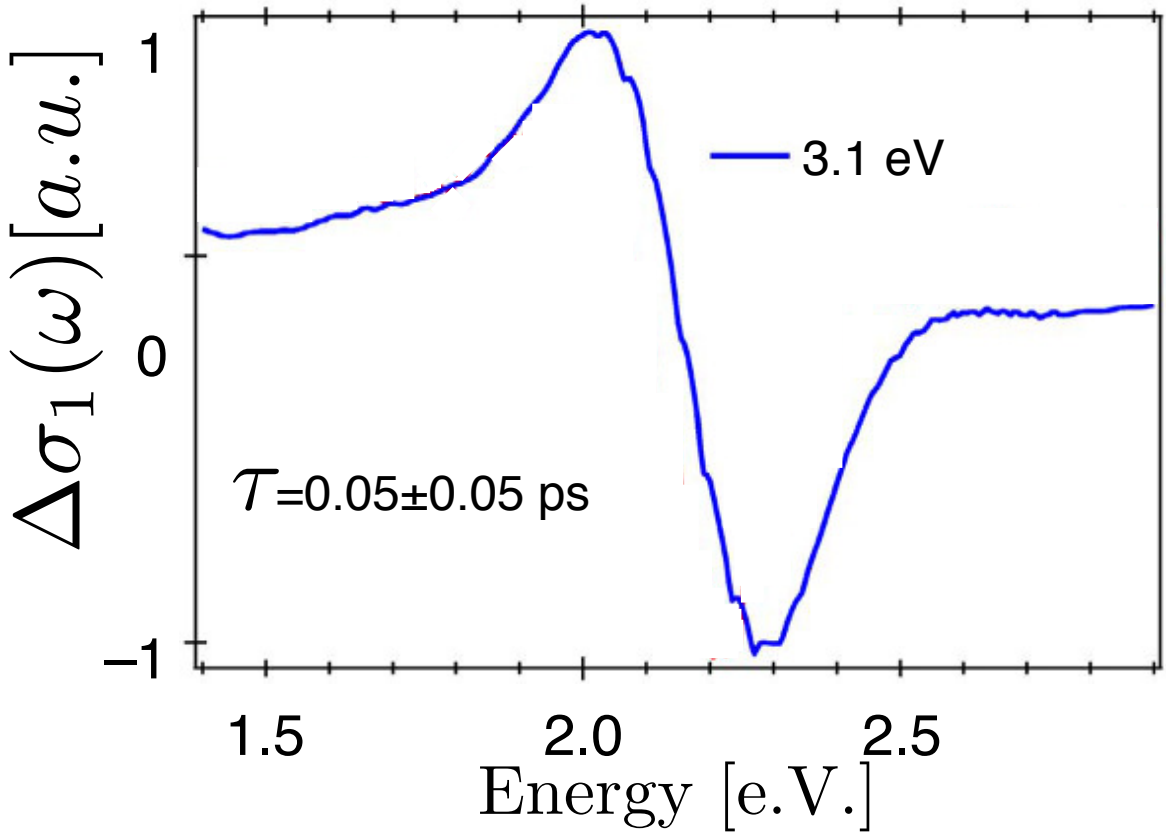
Optical conductivity

- Red shift
- Enhancement by dynamical screening



Optical conductivity - experiment

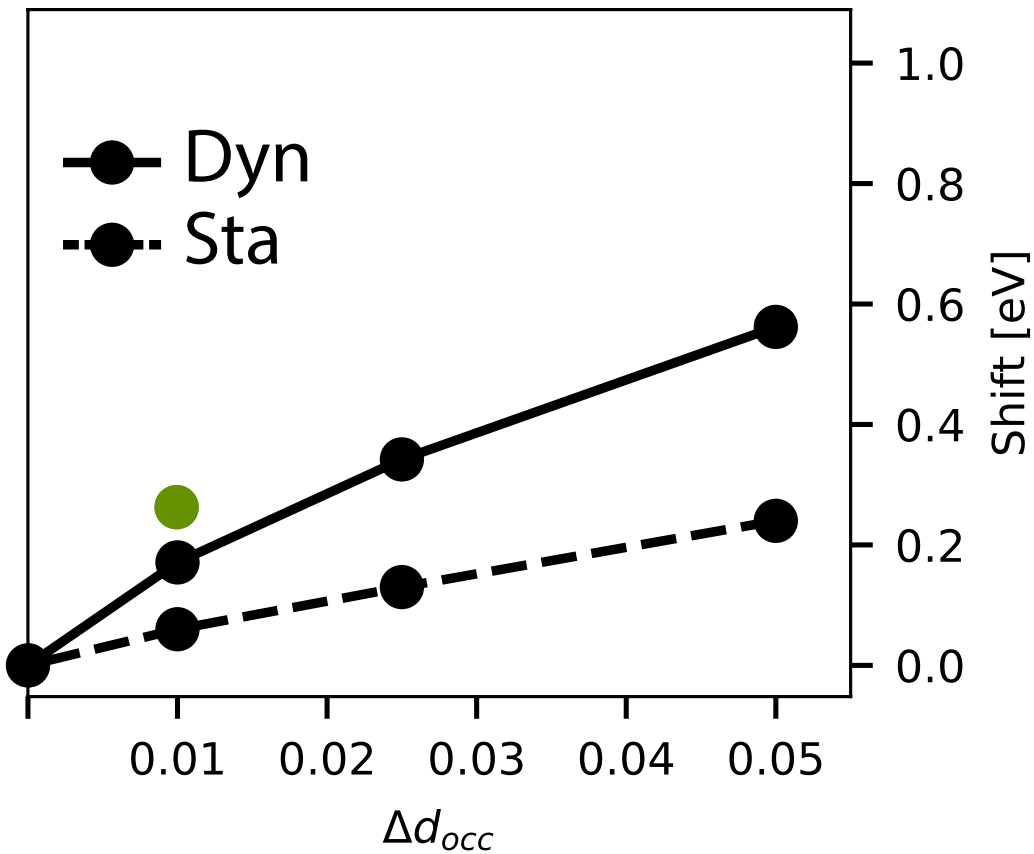
- Pump probe on La_2CuO_4 -transient reflectivity
- Above gap (3.5 eV) excitation



Novelli et.al. Nat. Comm. 6112(2014)

Optical conductivity

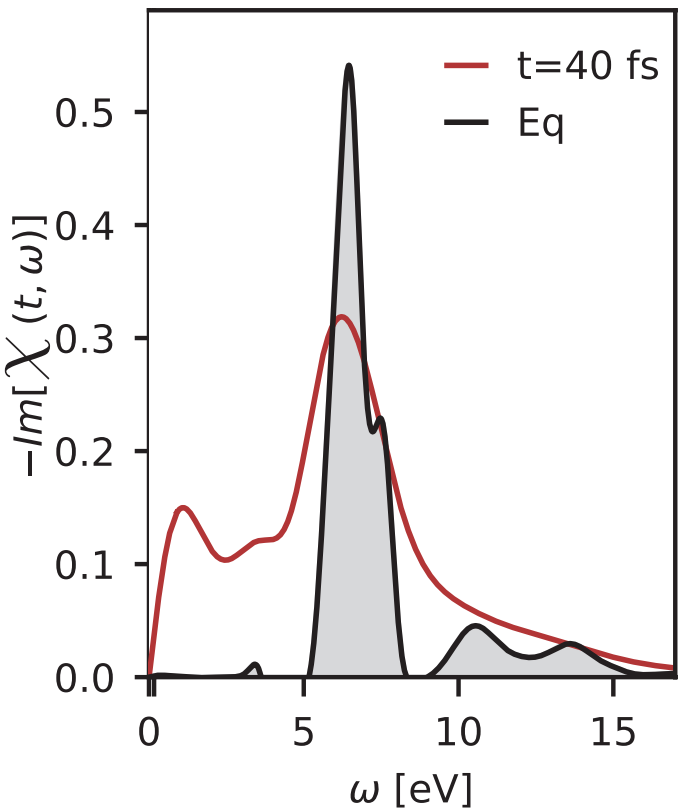
- Red shift
- Enhancement by dynamical screening
- Larger renormalization in experiment
- Effect of AFM



Screening

Charge susceptibility $Im[\chi(t,\omega)]$

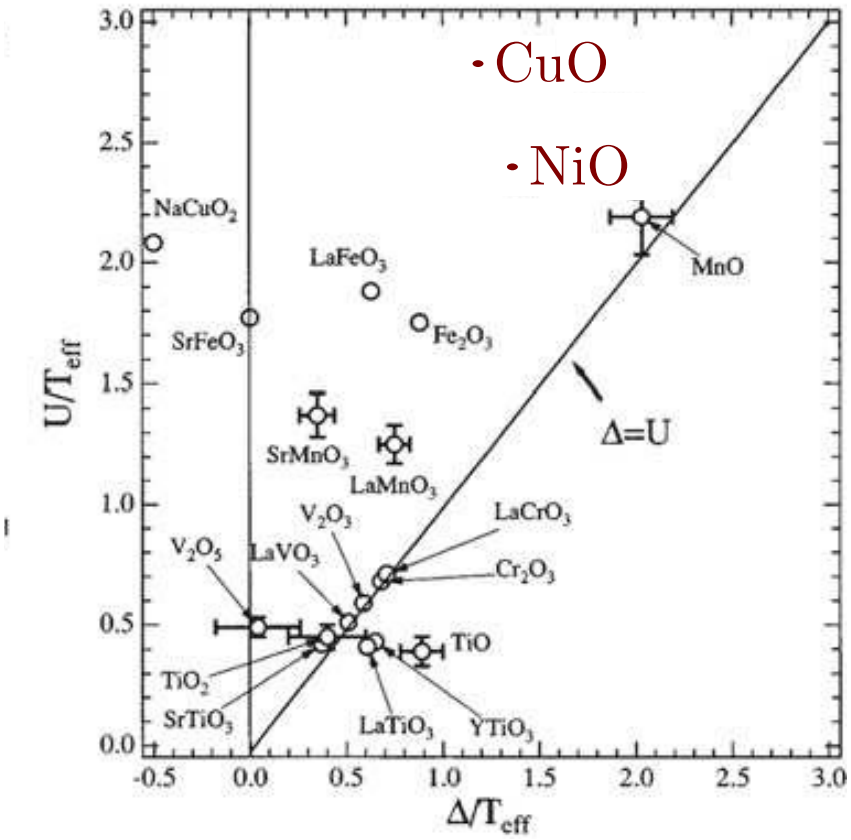
1. Photo-induced screening channel
2. Strong scattering with plasmons $\rightarrow$ broadening of spectrum



Message I

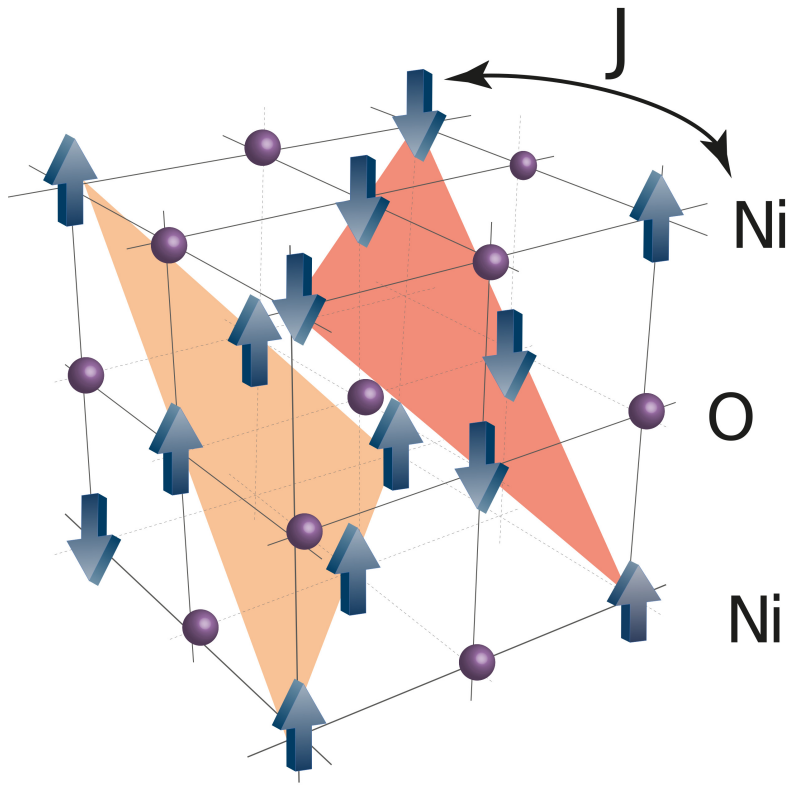
- Strong band gap renormalization in charge-transfer insulators
- Importance of non-local fluctuations (dynamical screening)
- Effect of incoherent dynamics on experimental probes
- Similar results by hybrid time-dependent DFT:
  N. Tancogne-Dejean, et.al. PRL 121, 097402 (2018)
  N. Tancogne-Dejean, et.al. arXiv:1906.11316

NiO


· CuO
· NiO
NaCuO2
LaFeO3
MnO
SrFeO3
Fe2O3
Δ=U
SrMnO3
LaMnO3
V2O3
LaCrO3
V2O5
LaVO3
Cr2O3
TiO
TiO2
SrTiO3
LaTiO3
YTiO3
U/Teff
Δ/Teff
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
0.0
0.5
1.0
1.5
2.0
2.5
3.0

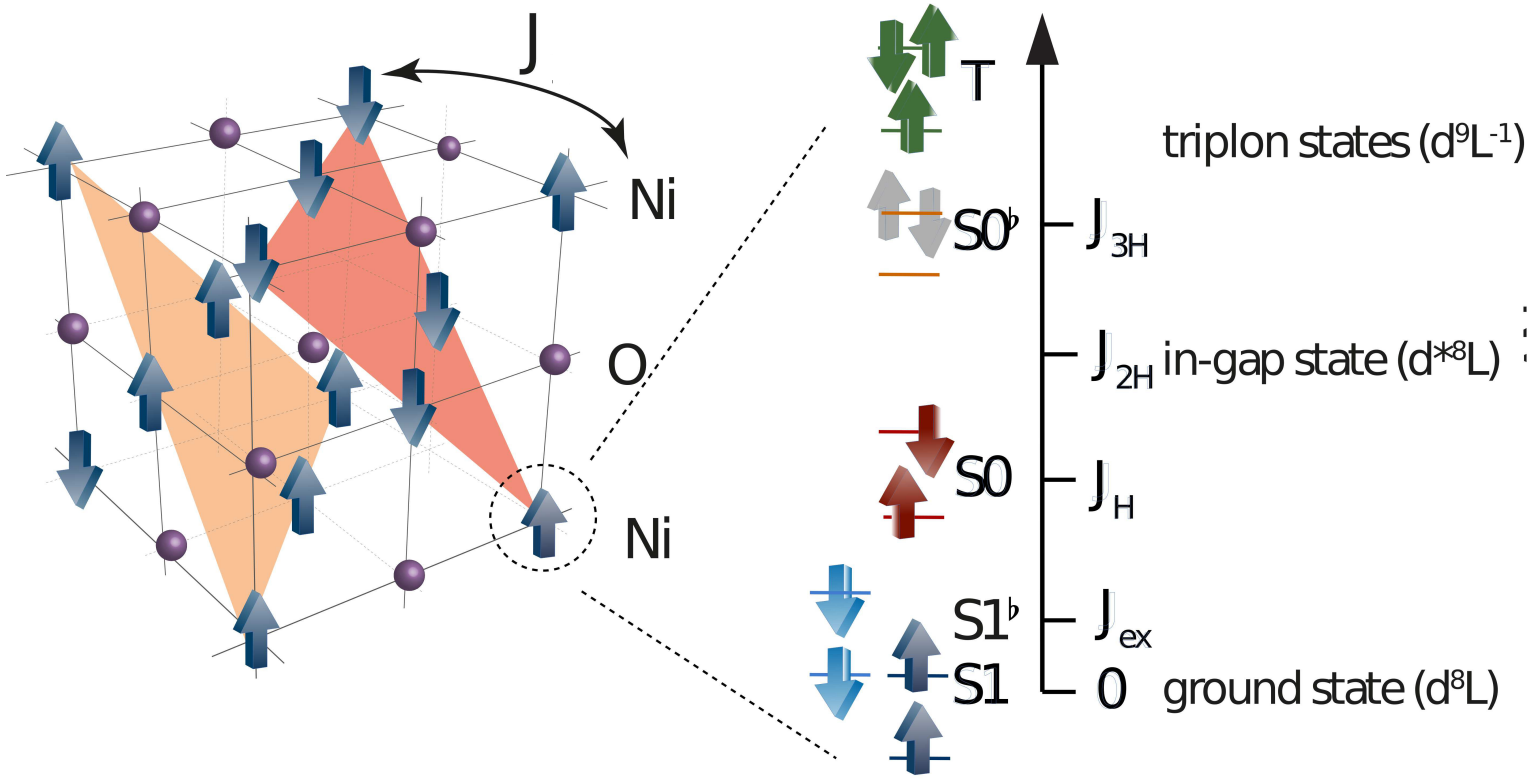
Lattice structure

- Inter-penetrating antiferromagnetic planes
- AFM is dominant



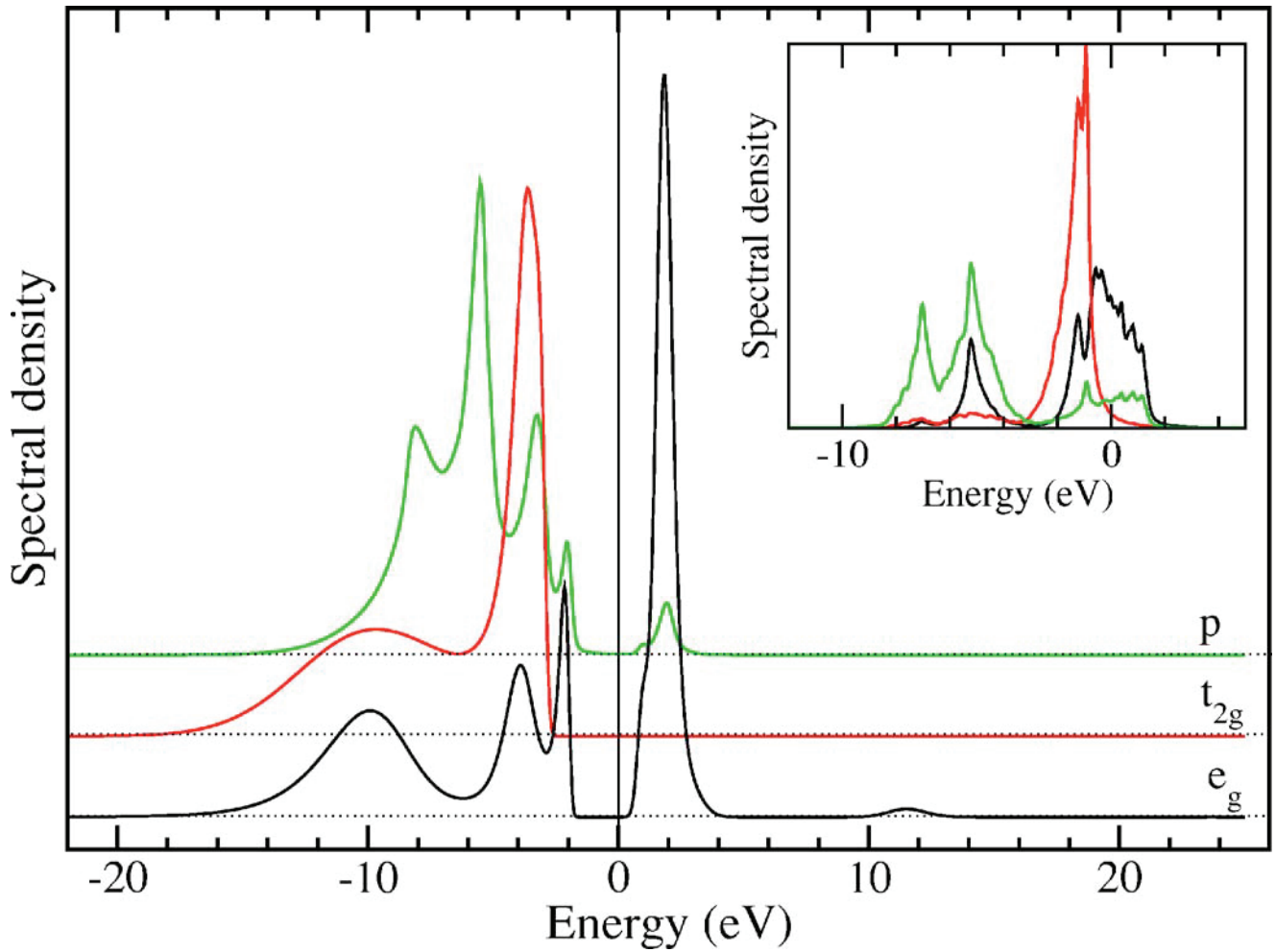
Electronic structure

- Two electrons in two e_g-orbitals
- Excitations: magnons, Hund and CT excitations



Electronic structure

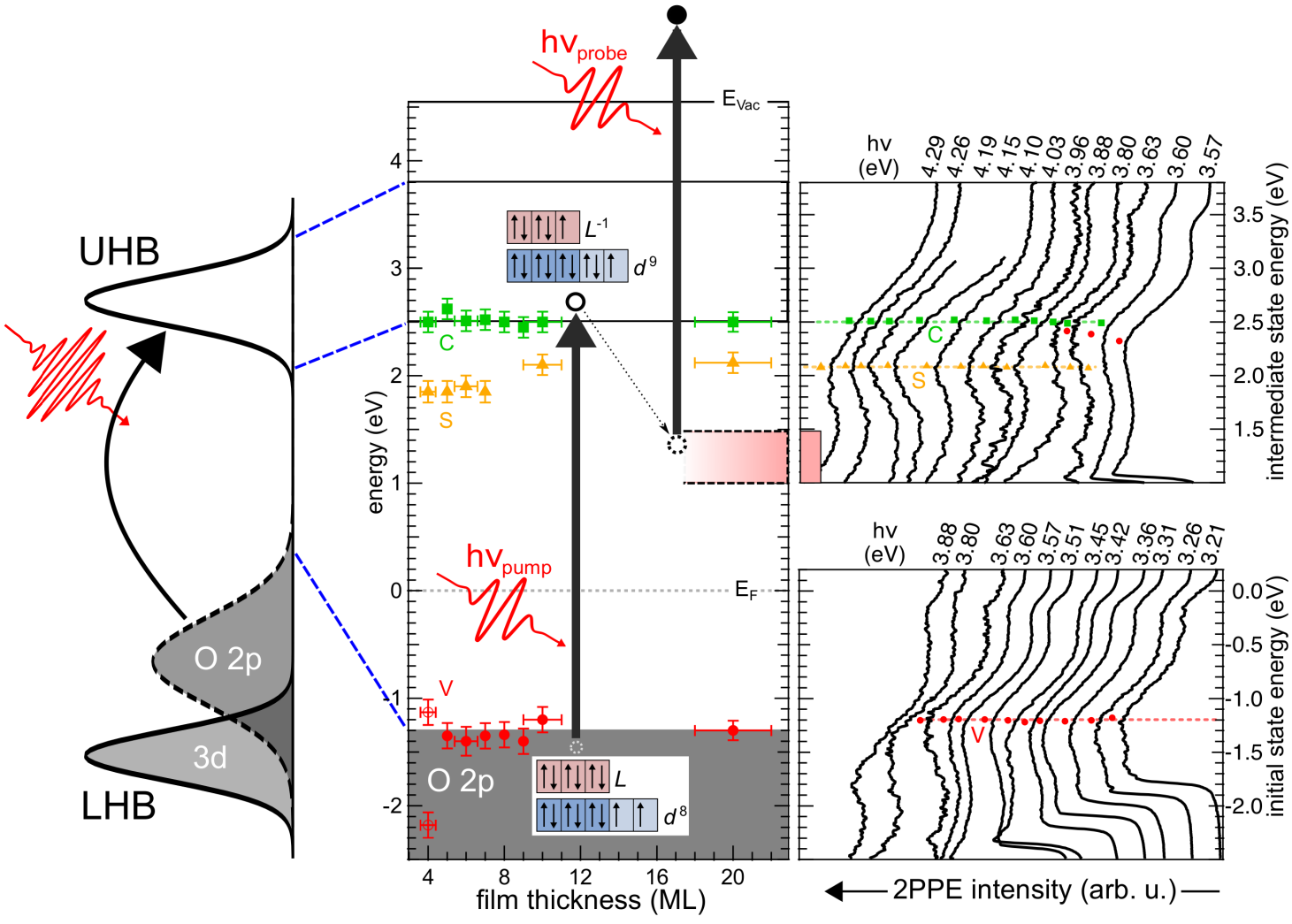
- LDA + DMFT description



J. Kunes, et.al. PRB 75, 165115 (2017)

2PPE - experiment

- Charge transfer excitation
- Surface states



2PPE - experiment

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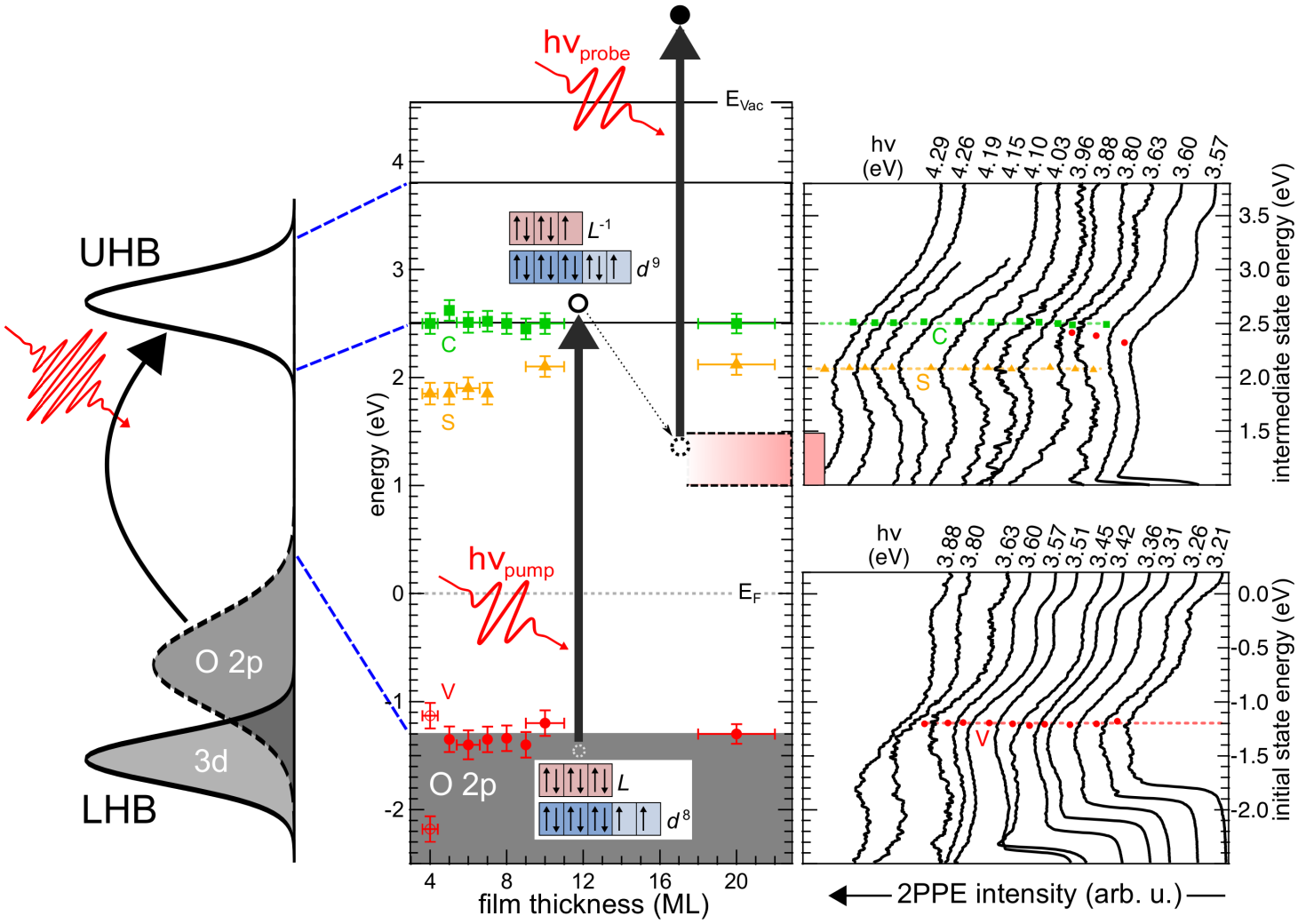
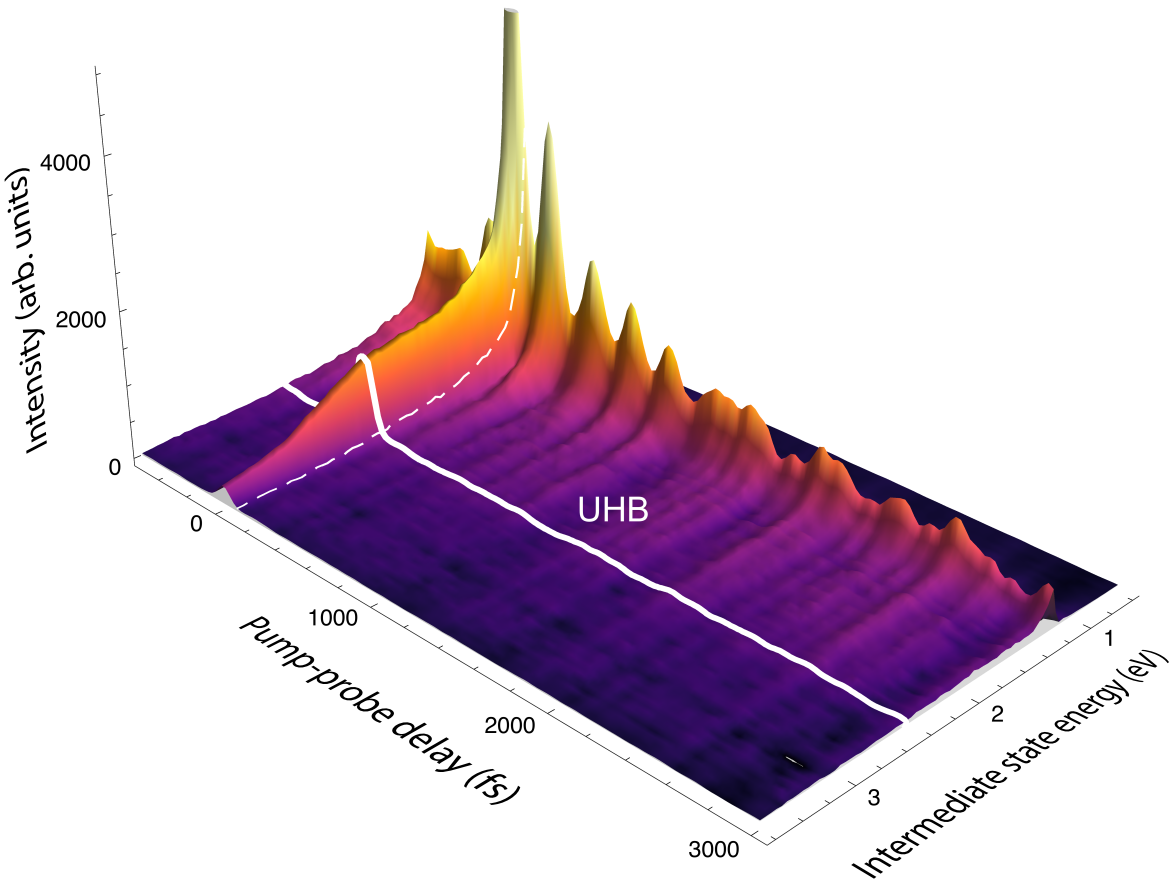


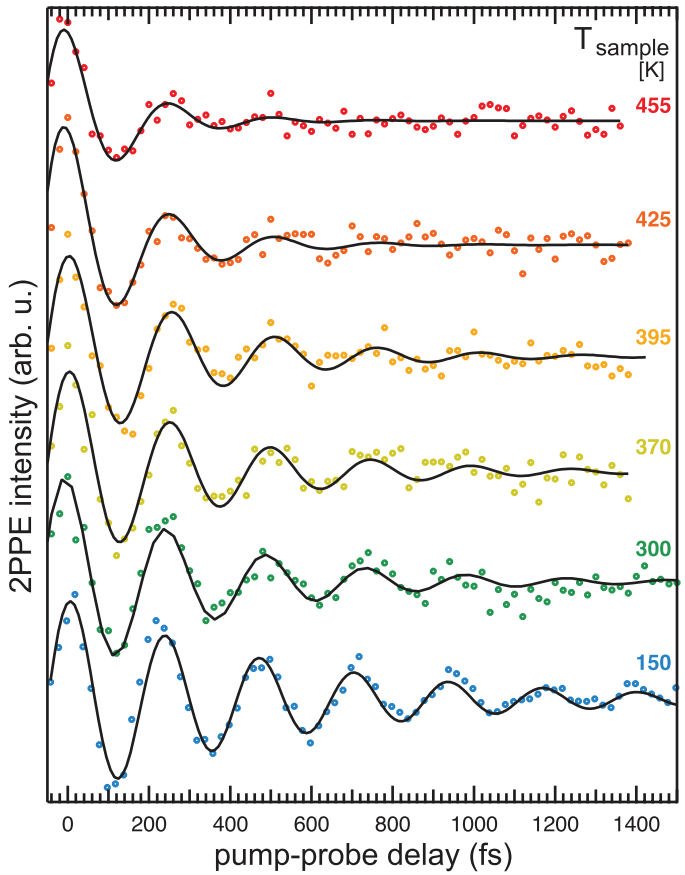
Pump-probe

- Pump $h\nu_P$=4.2 eV
- Ultra-fast relaxation
- Oscillating photo-induced in-gap state



In-gap state

- Long-lived coherent dynamics of in-gap state
- Strongly damped at Neél temperature



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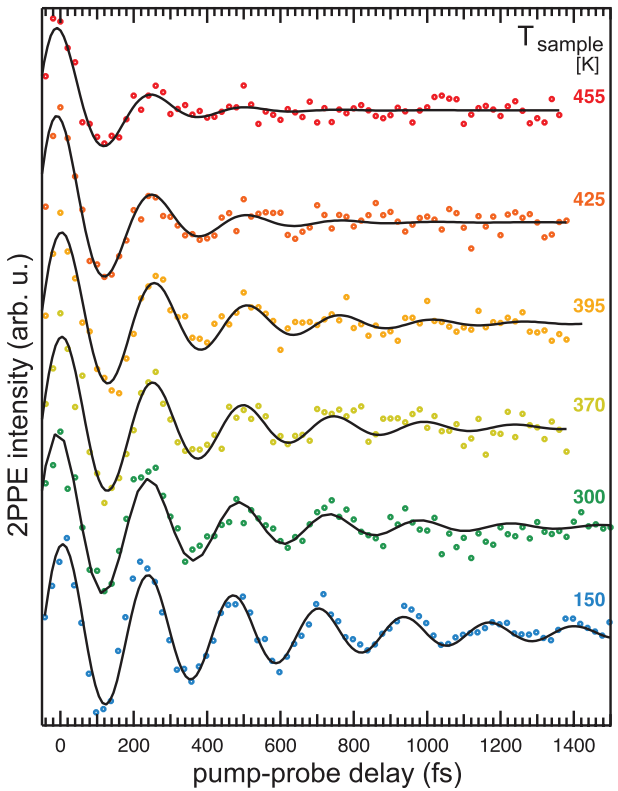
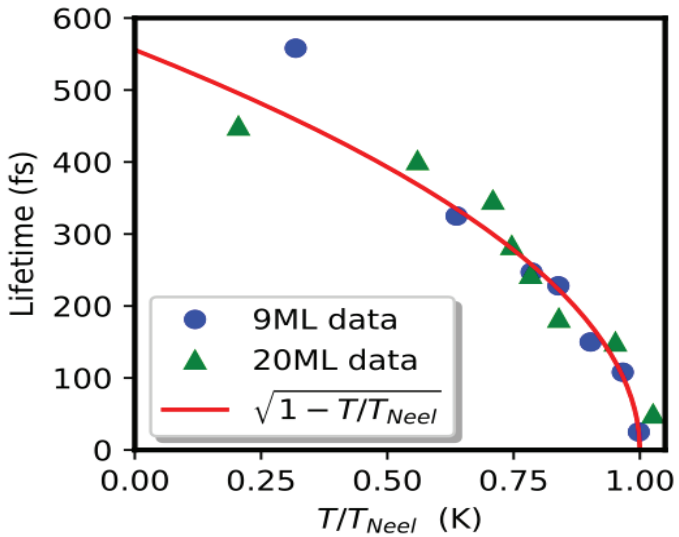
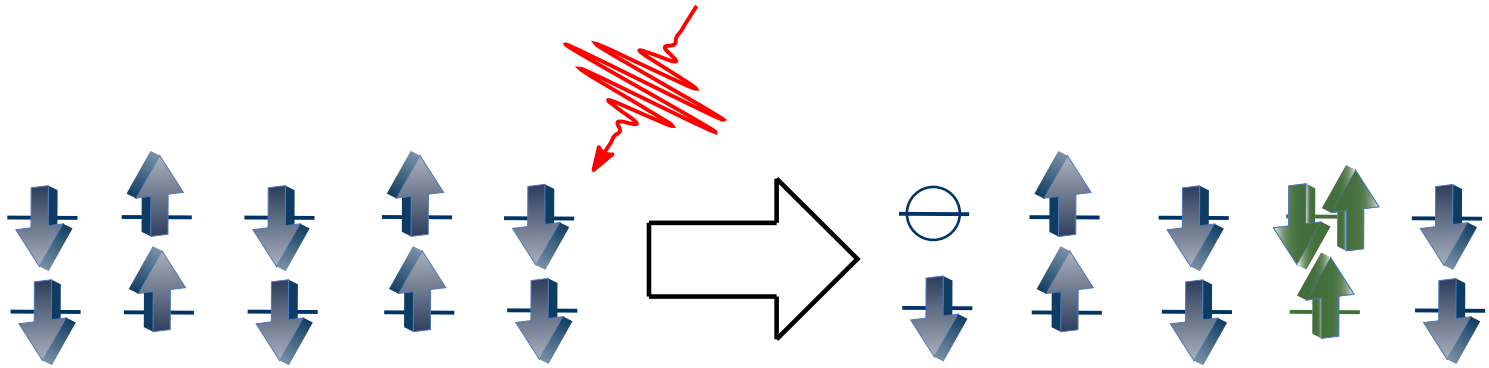




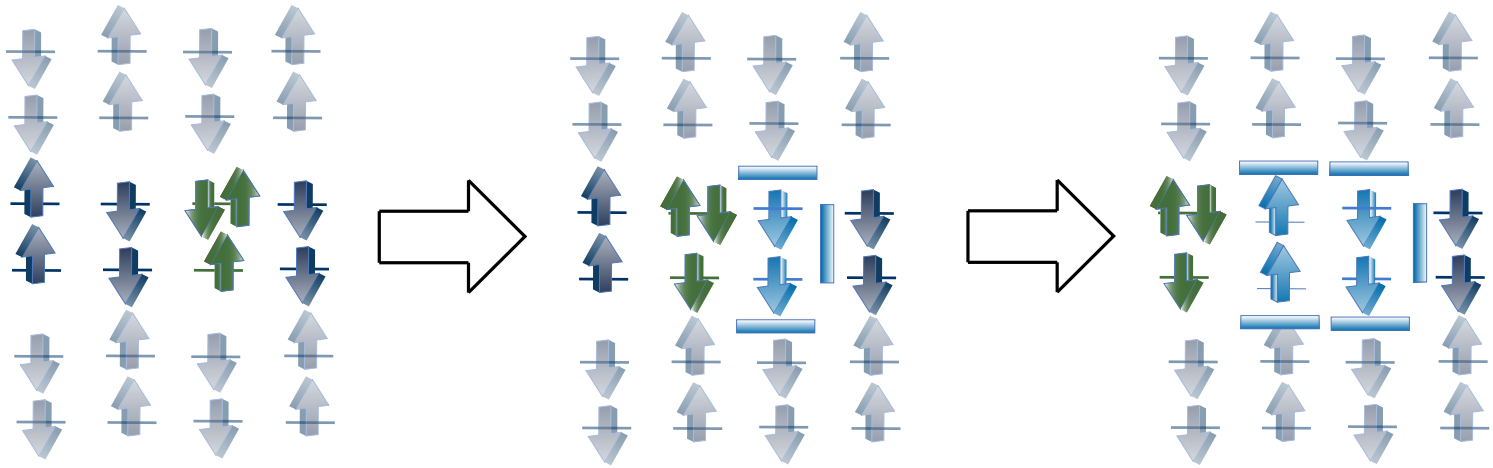
Modeling

- String states



Modeling

- Ground state: High-spin AFM
- Photo-induced triplon and hole
- String states



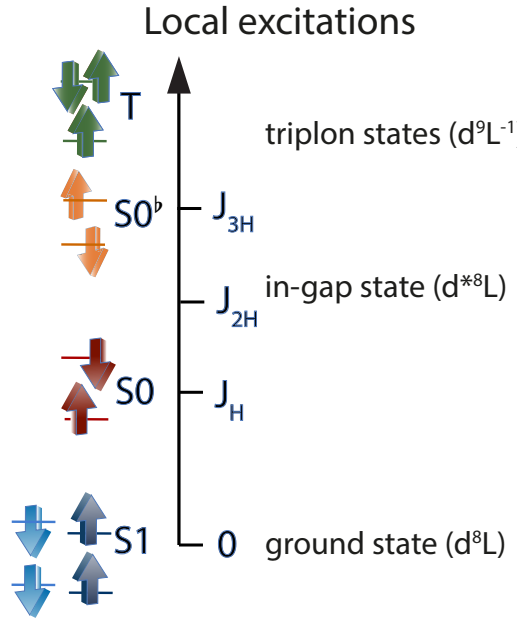
Multiband model

- Neglect excitonic effects
- Mapping to two-band t-J problem (Zhang-Rice construction)
- Atomic, kinetic and AFM contribution

$$H = \hat{H}_{\text{loc}} + H_{\text{kin}} + H_{\text{ex}} \tag{1}$$

Multiband model

- Kanamori interaction for d orbitals
- J_H=1 eV
- Hubbard and Hund physics
- Ground state: high-spin state
- Solve within DMFT

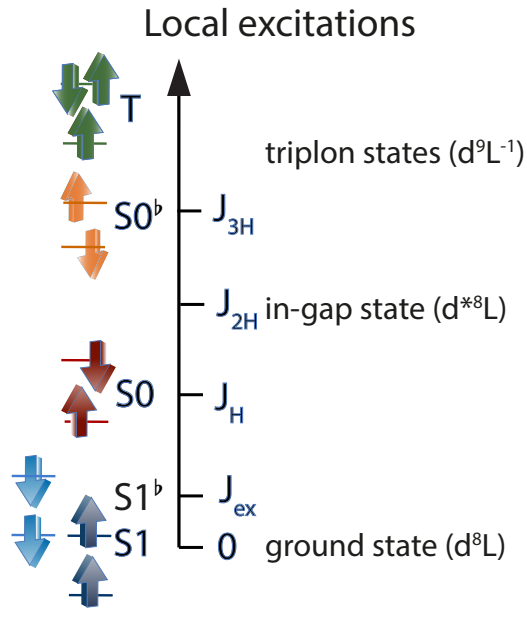


$$\hat{H}_{\text{loc}} = U\sum_{i,\alpha} n_{i,\alpha\uparrow}n_{i\alpha\downarrow} - \mu\sum_{i\alpha\sigma}\tilde{n}_{i\alpha\sigma} + \sum_{i,\alpha<\beta}\sum_{\sigma,\sigma'}(U' - J_H\delta_{\sigma\sigma'})n_{i\alpha\sigma}n_{i\beta\sigma'}$$
$$+ \gamma J_H \sum_{i,\alpha<\beta}\left(\tilde{c}^{\dagger}_{i\alpha\uparrow}\tilde{c}^{\dagger}_{i\alpha\downarrow}\tilde{c}_{i\beta\downarrow}\tilde{c}_{i\beta\uparrow} + \tilde{c}^{\dagger}_{i\alpha\uparrow}\tilde{c}^{\dagger}_{i\beta\downarrow}\tilde{c}_{i\alpha\downarrow}\tilde{c}_{i\beta\uparrow}\right) \tag{2}$$

$$H_{\text{kin}} = -t_0\sum_{\langle i,j\rangle}\sum_{a\sigma}(\tilde{c}^{\dagger}_{ia\sigma}\tilde{c}_{ja\sigma} + \tilde{c}^{\dagger}_{ja\sigma}\tilde{c}_{ia\sigma}) \tag{3}$$

Multiband model

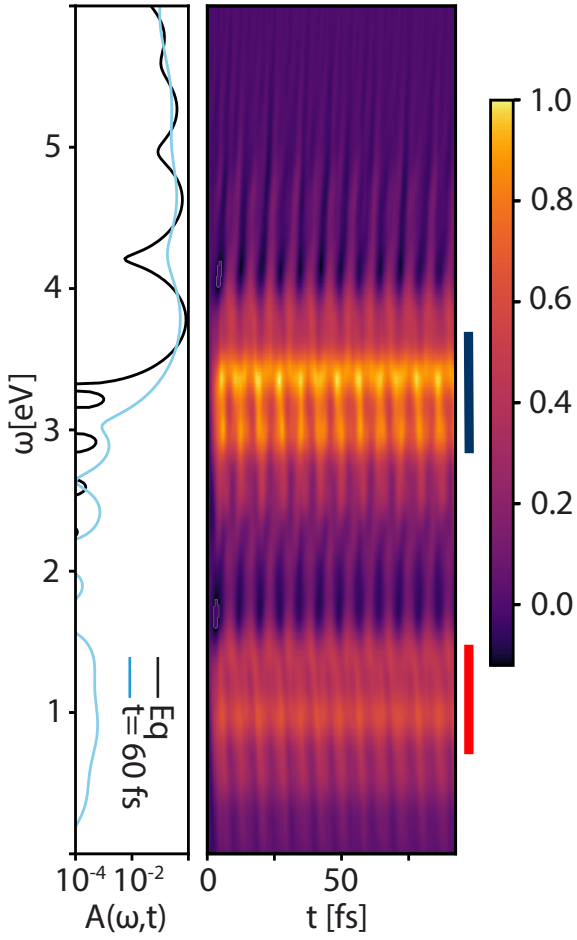
- Superexchange interaction J_{ex}
- Mean-field approximation



$$H_{\text{ex}} = J_{\text{ex}} \sum_{\langle ij \rangle} \mathbf{S}_{ia} \cdot \mathbf{S}_{ja} + \mathbf{S}_{ib} \cdot \mathbf{S}_{jb} \rightarrow J_{\text{ex}} \sum_{\langle ij \rangle} \mathbf{S}_{ia} \cdot \langle \mathbf{S}_{ja} \rangle + \langle \mathbf{S}_{ib} \rangle \cdot \mathbf{S}_{jb} \tag{4}$$

PES - theory

- Photo-induced in-gap state
- Hund excitation



PES - theory

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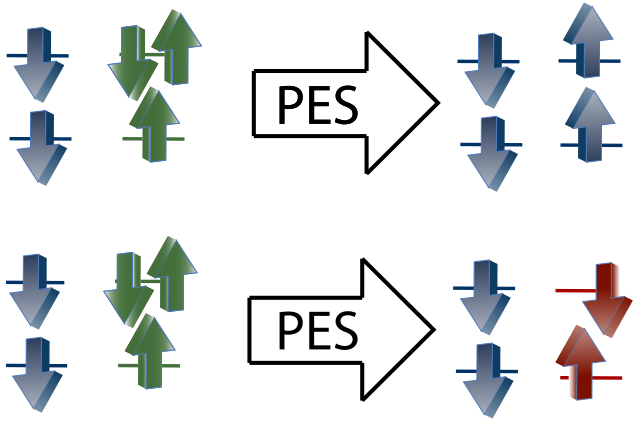
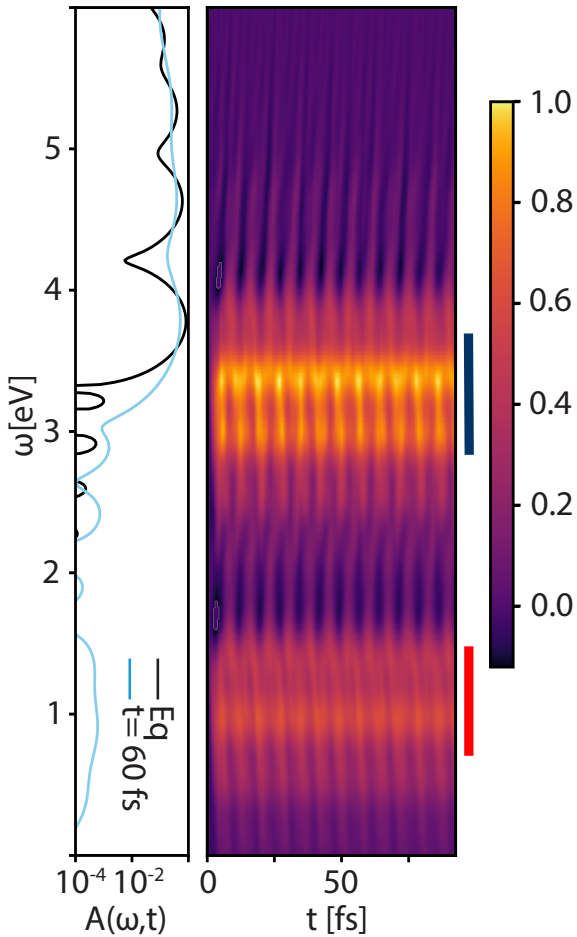
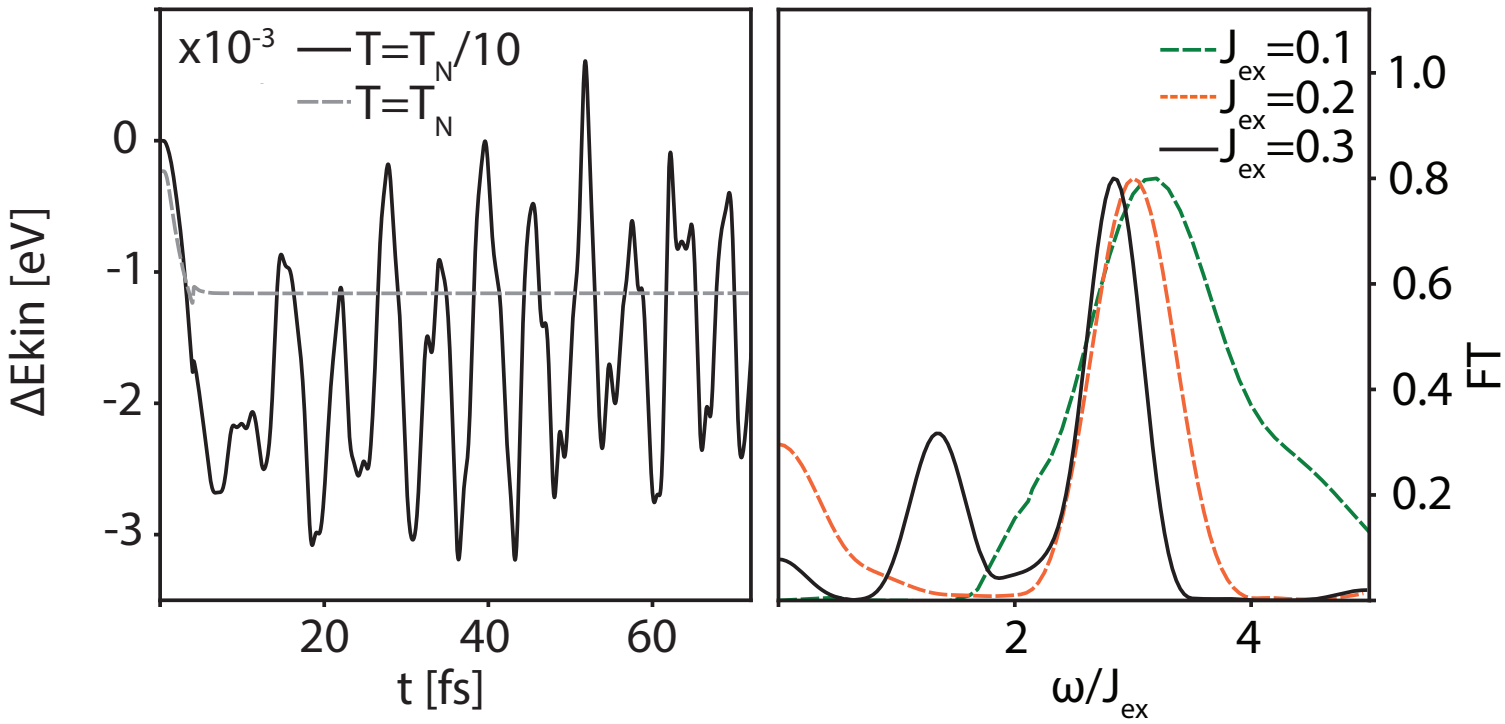




H. Strand, et.al. PRB 96, 165104 (2017)

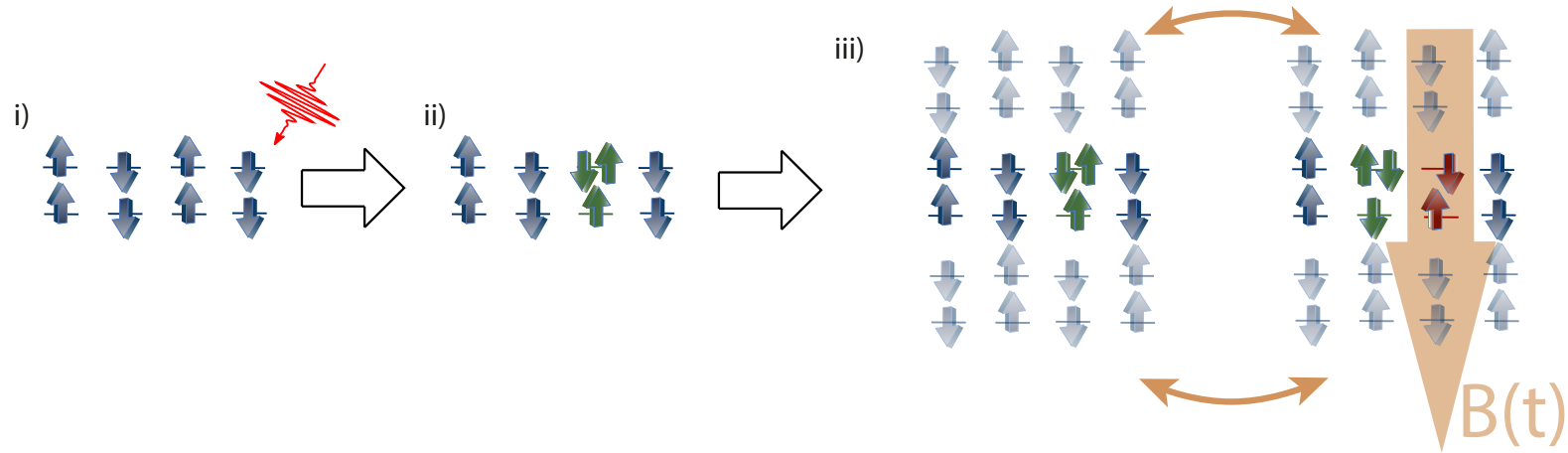
Kinetic energy

- Fast Hund oscillations
- Slow oscillations proportional to J_{ex}
- Coherent dynamics only below T_N



Hund and AFM

- Coupling of AFM and Hund
- String-like excitations
- Zeeman splitting for low-spin state



Hund and AFM

- Coupling of AFM and Hund
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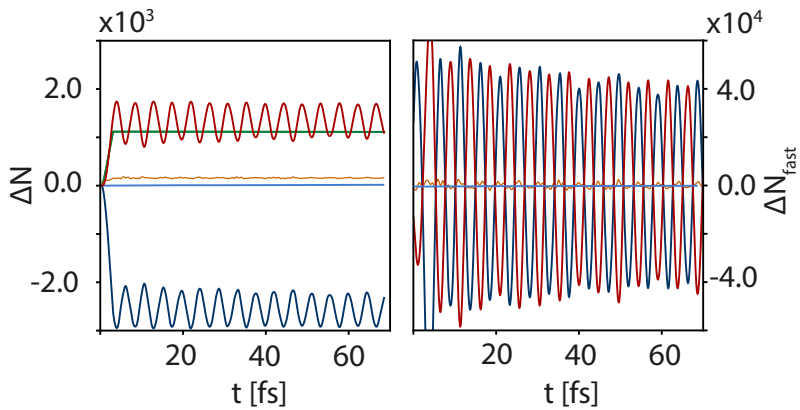
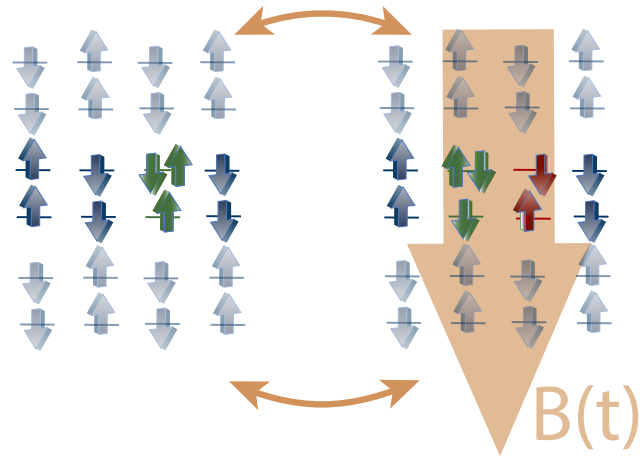
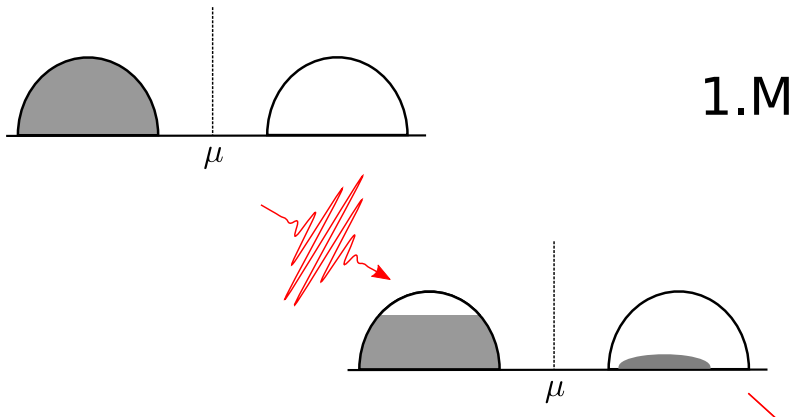




Message II

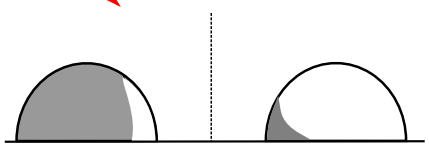
- Photo-induced in-gap state
- Coherent many-body oscillations (2 ps)
- Interplay of Hund and AFM physics

Conclusions



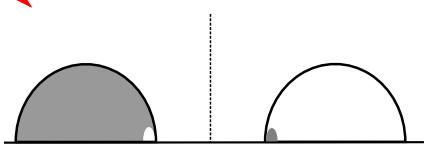
CuO
1.Multiband + dynamical screening
2. Band-gap renormalization
3.PES and optics

cooling



NiO
1. Photo-induced in-gap state
2. Many-body coherence
3. Hund and AFM

thermalization



Collaborators

- Philipp Werner (University of Fribourg)
- Lewin Boehnke
- Nikolaj Bittner
- Martin Eckstein (FAU Erlangen)

NiO - experiment

- Wolf Widdra (Martin-Luther Universität - Halle)
- Konrad Gillmeister
- Cheng-Tien Chiang
- Yaroslav Pavlyukh

Non Equilibrium Systems SImulation (NESSI) library

Numerical library for Greens functions on the Kadanoff-Baym contour

Functionalities:

1. Set up Feynman diagrams and solve EOM
2. High-order propagation scheme
3. MPI parallelization
4. Examples: Hubbard chains, Migdal-Eliashberg, GW, ...

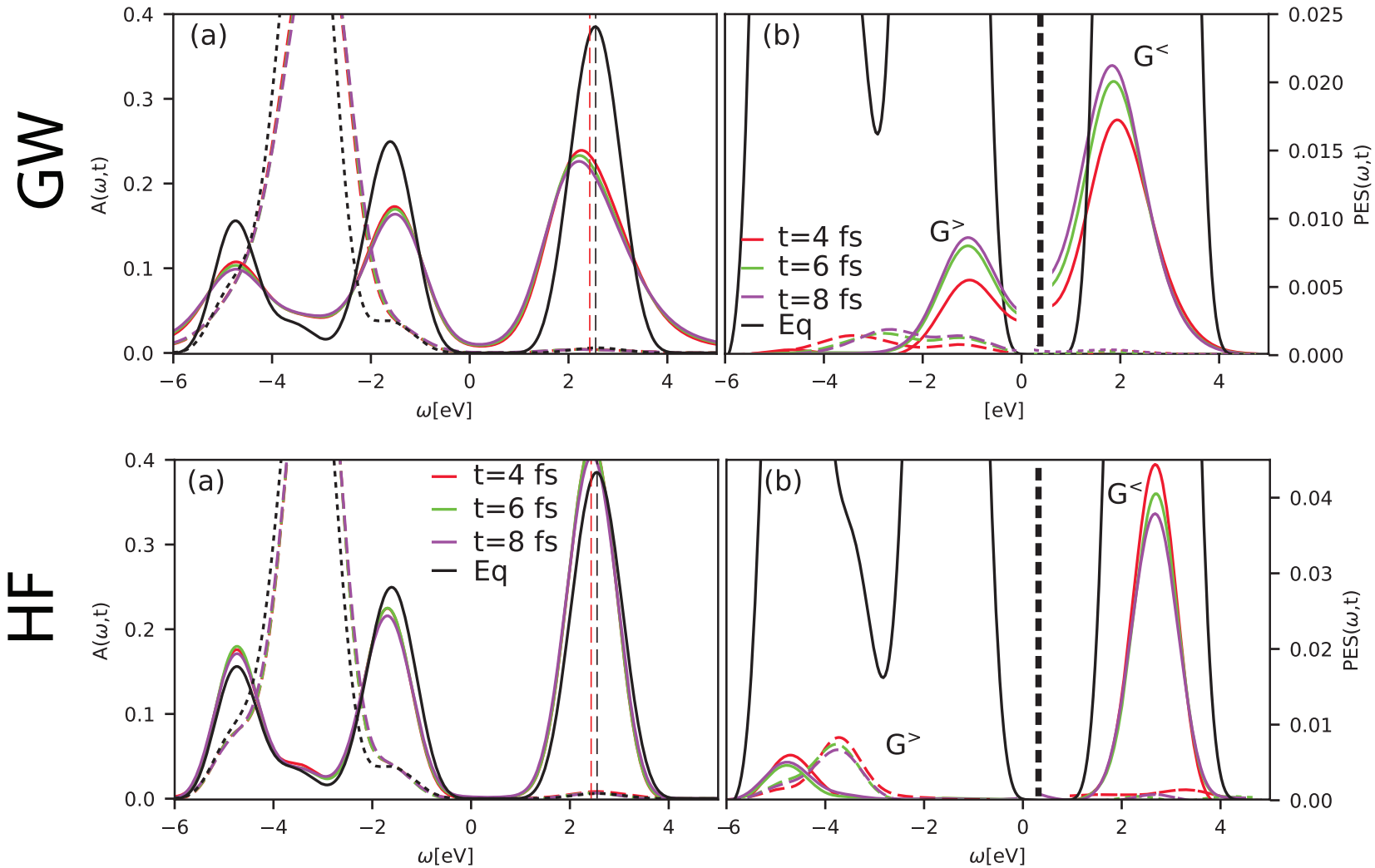
To be released ...

Thank you

Publications:

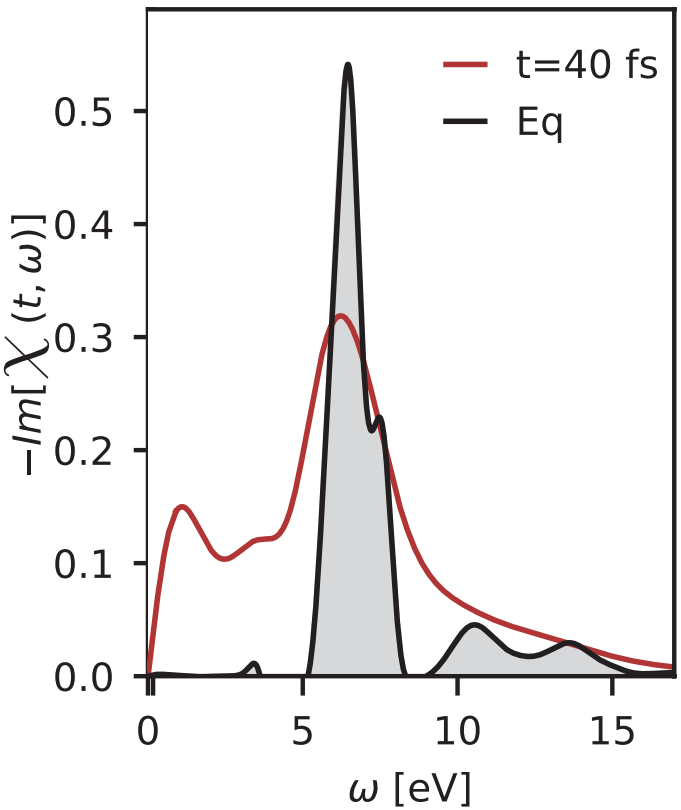
1. DG, L. Boehnke, M. Eckstein, P. Werner, PRB 100 (4), 041111 (2019)
2. DG, M. Eckstein, P. Werner, arXiv:1903.08713 (2019)
3. K. Gilmeister, DG, et.al. (in preparation)

Relaxation



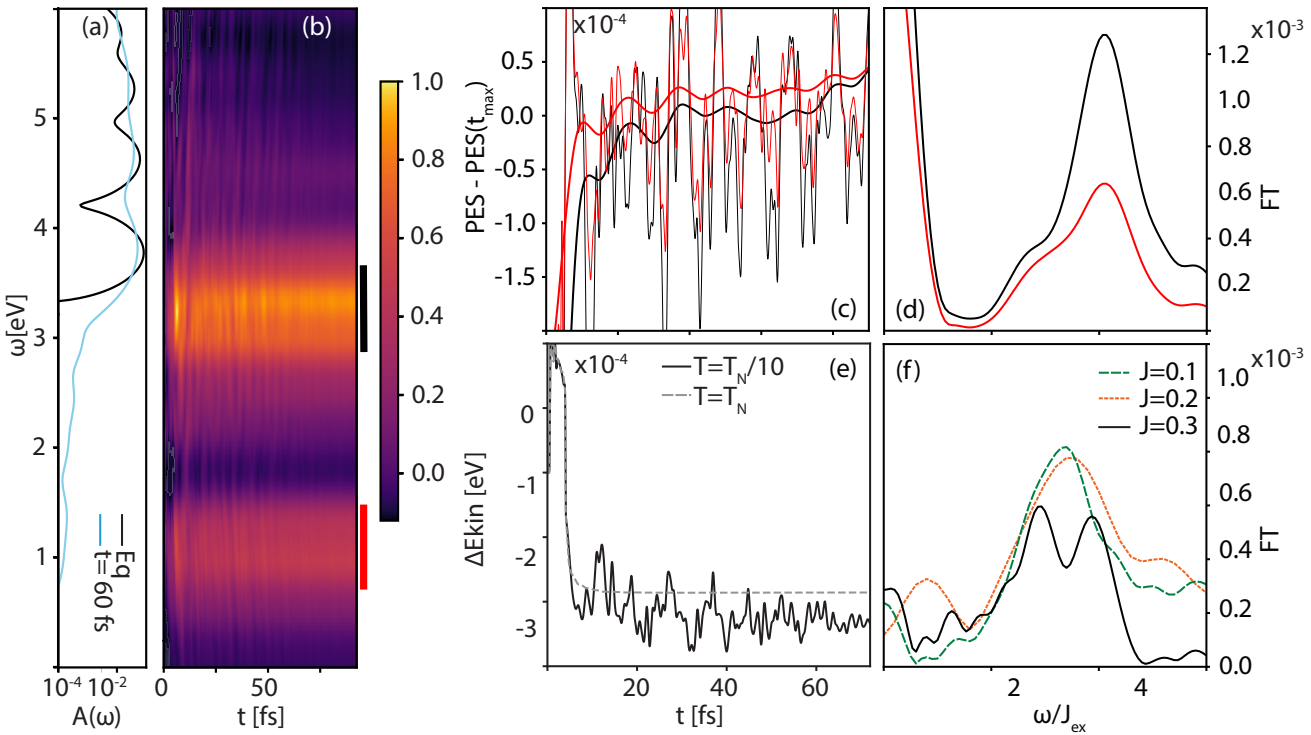
Screening

1. Photo-induced screening channel
2. Strong scattering with plasmons $\rightarrow$ broadening of spectrum



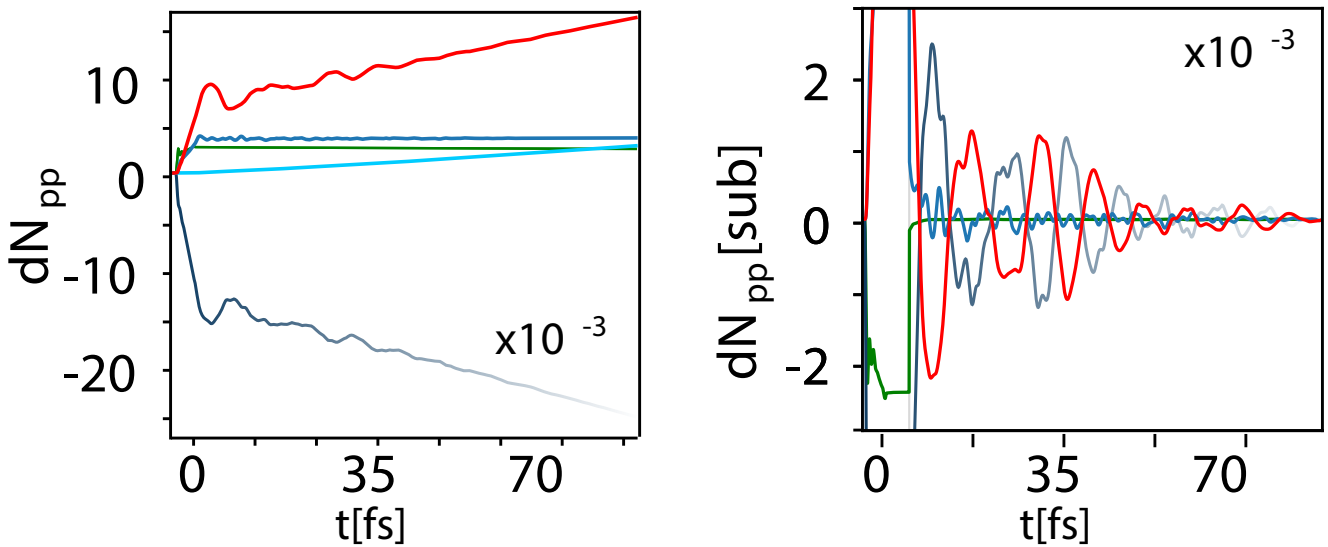
Lifetime of coherent dynamics I

1. High-frequency excitations



Lifetime of coherent dynamics II

1. High-frequency excitations
2. Finite lifetime
3. Increase of AFM defects



Lifetime of coherent dynamics II

1. High-frequency excitations
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