Spectroscopy of strongly correlated materials: beyond DMFT

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European XFEL

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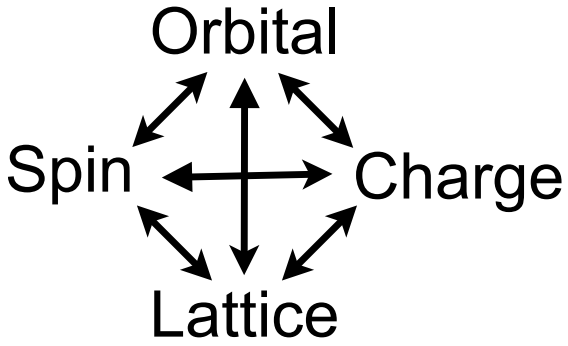
Hiroshima, 05.07.2017

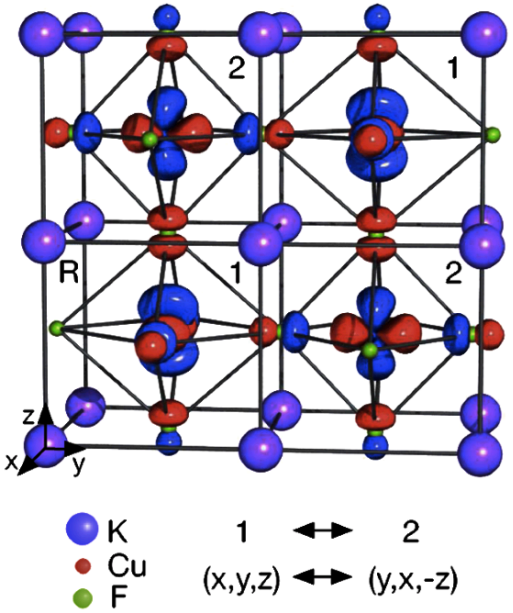
Outline

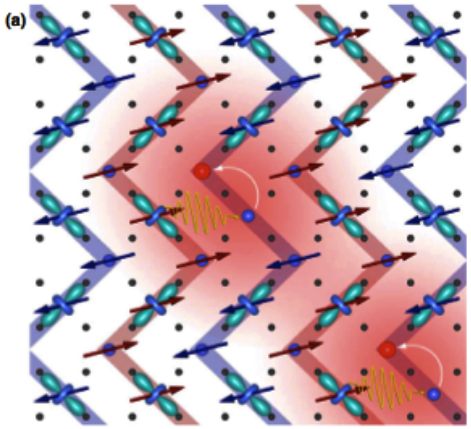
- Spectroscopy of strongly correlated materials: realistic DMFT
- Non-equilibrium dynamics of correlated materials: XFEL-ultra-strong pulses
- Non-local correlations: cluster and dual scheme

Strongly correlated materials

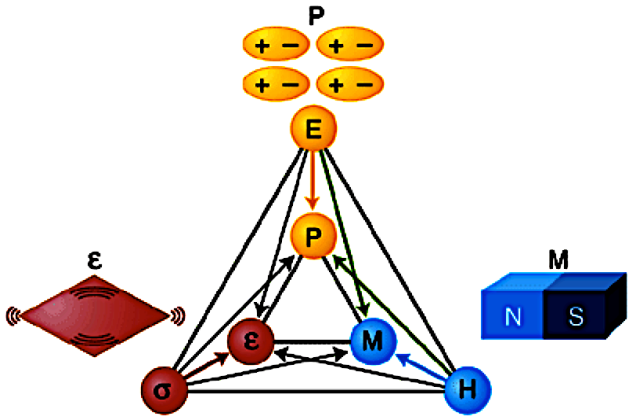
- Interplay of degrees of freedom
- Coupling of control parameters
- Complicated ordering phenomena







H. Ehrke et al.,
PRL 106, 217401 (2011)



Spaldin & Fiebig, Science 309:391 (2005)

E. Pavarini, E. Koch, and A. L. PRL **101**, 266405 (2008)

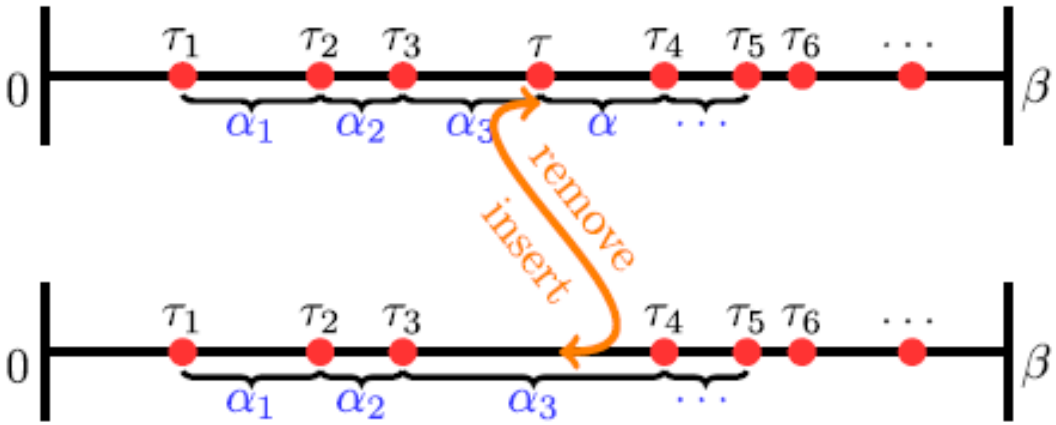
Continuous Time Quantum Monte Carlo

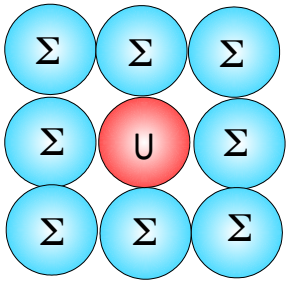
Partition function: $H = H_0 + V$

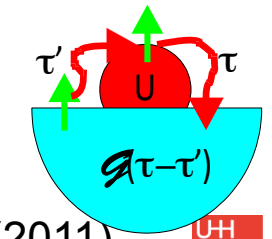
$$Z = \mathbf{Tr}\left[e^{-\beta H_0}\mathbf{T}_\tau e^{-\int_0^\beta d\tau V(\tau)}\right]$$

Stochastic expansion – “Diagrammatic Monte Carlo” (N. Prokof’ev)

$$Z = \sum_{k=0}^{\infty}\int_0^\beta d\tau_1 \int_{\tau_1}^\beta d\tau_2 \ldots \int_{\tau_{k-1}}^\beta d\tau_k \mathbf{Tr}\left[e^{-\beta H_0} e^{-\tau_k H_0}(-V)\ldots e^{-(\tau_2-\tau_1)H_0}(-V)e^{-\tau_1 H_0}\right]$$

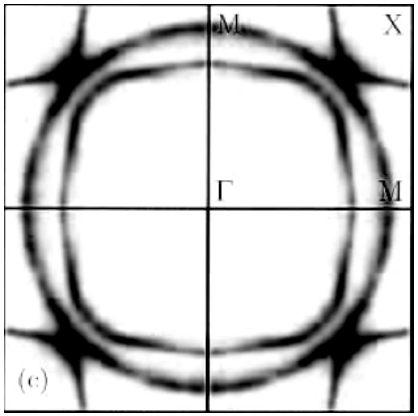
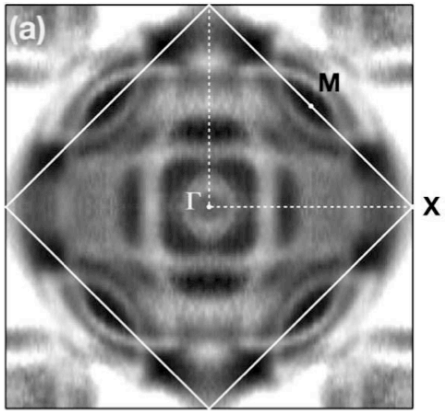
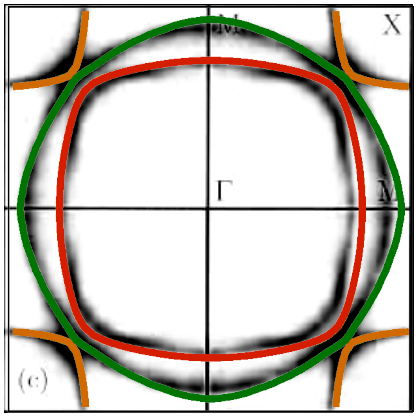
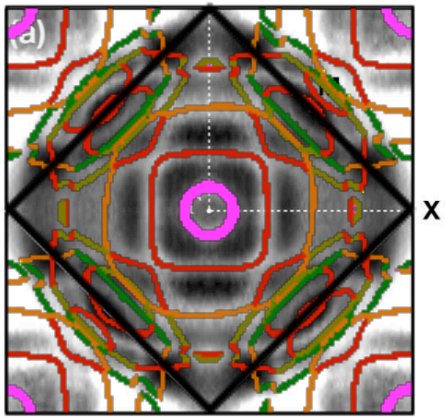


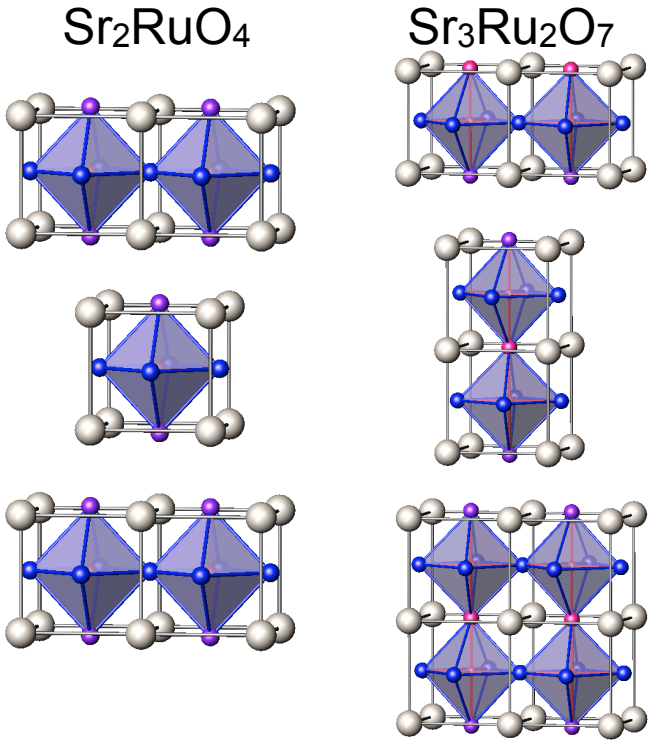




E. Gull, A. Millis, A.L., A. Rubtsov, M. Troyer, Ph. Werner, Rev. Mod. Phys. **83**, 349 (2011)

Fermi surface for Ruthenites: LDA+DMFT

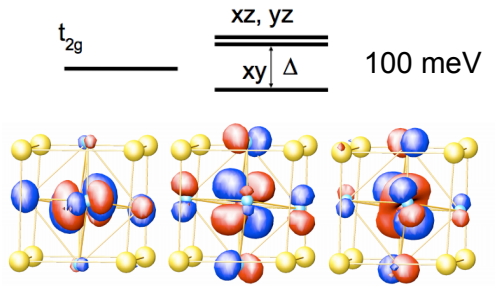
	Sr_2RuO_4	$Sr_3Ru_2O_7$
ARPES		
	A. Damascelli et al., PRL 85, 5194 (2000)	A. Tamaiet et al., PRL 101, 026407 (2008)
Theory		
	G. Zhang, E. Gorelov, E. Sarvestani, and E. Pavarini, accepted to PRL (2016)	E. Gorelov, G. Zhang, and E. Pavarini (unpublished)



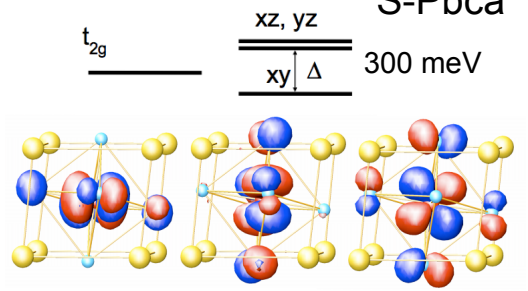
Orbital ordering in Ca_2RuO_4

Crystal-field orbitals of Ru 4d t_{2g} manifold

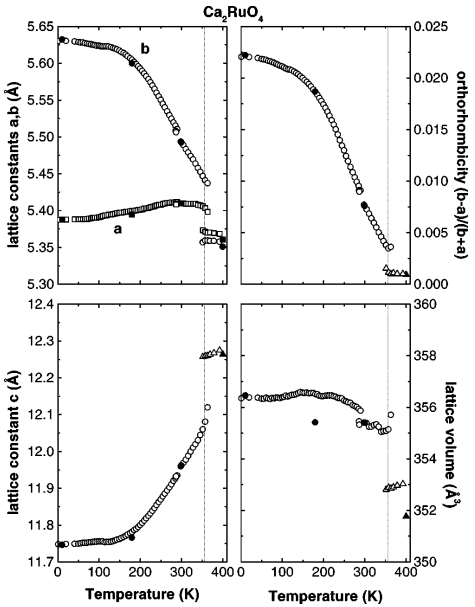
High-temperature phase: L-Pbca



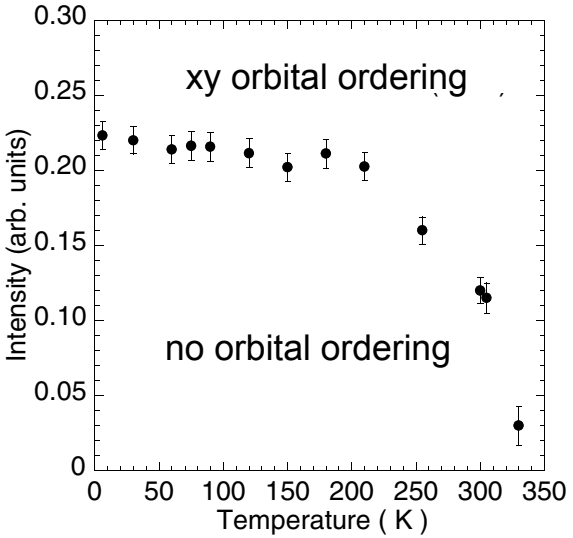
Low-temperature phase: S-Pbca

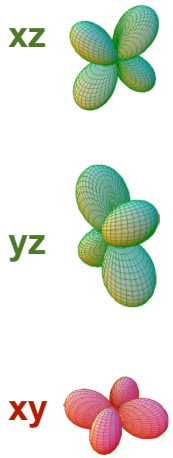


Structural transition



RXS signal for the d_{xy} ordering



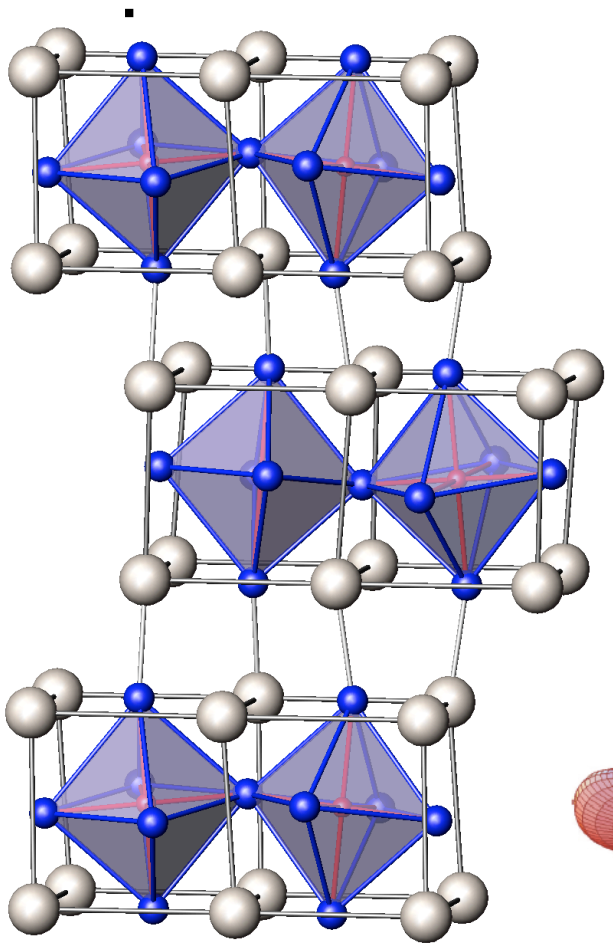


E. Gorelov, M. Karolak, T. Wehling, F. Lechermann, A. L, E. Pavarini, PRL 104, 226401 (2010)

O. Friedt et al, PRB 63, 174432 (2001)

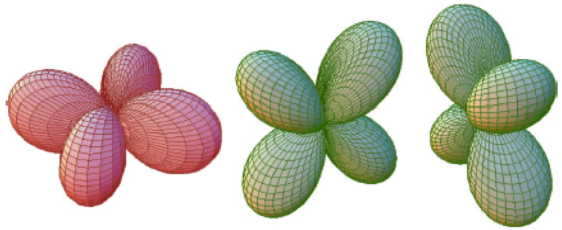
M. Kubota et al., PRL 95, 026401 (2005)

Metal-Insulator transition in Ca_2RuO_4

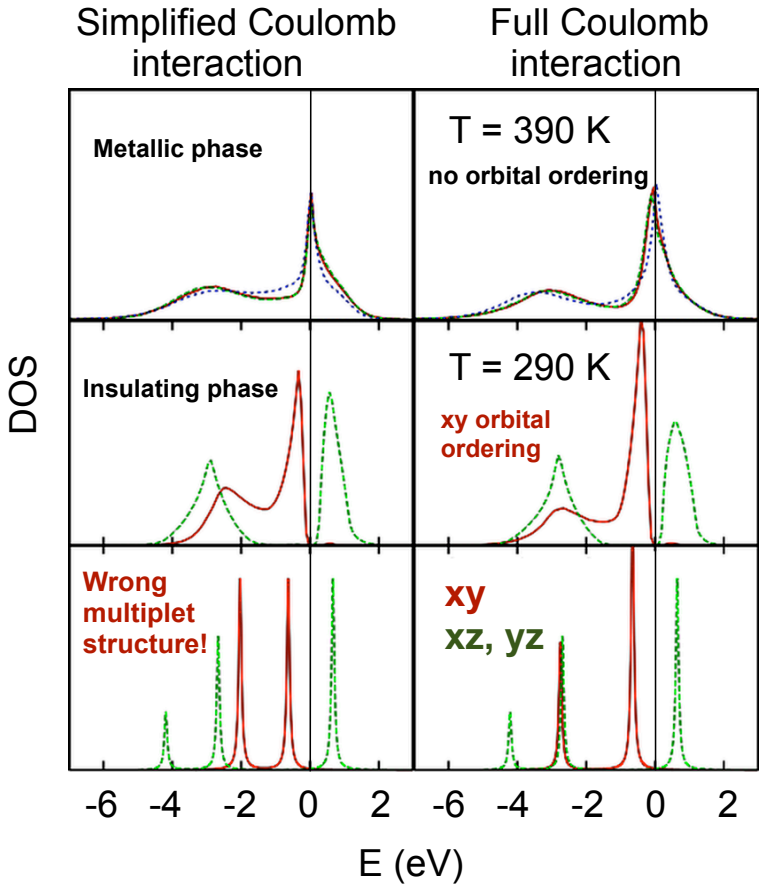


Ru ● O ●
Ca ●

Ru 4d t_{2g} orbitals



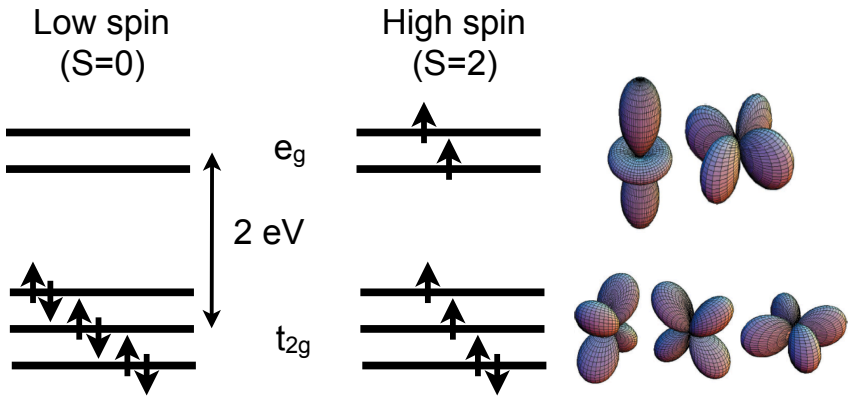
xy xz yz

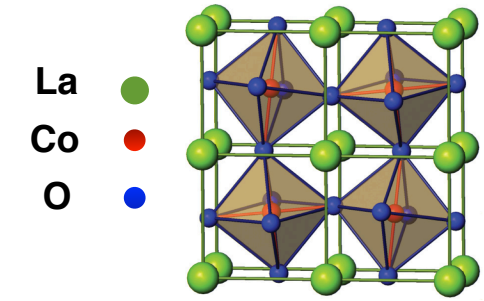


E. Gorelov, M. Karolak, T. O. Wehling, F. Lechermann, A. L. , and E. Pavarini, PRL **104**, 226401 (2010)

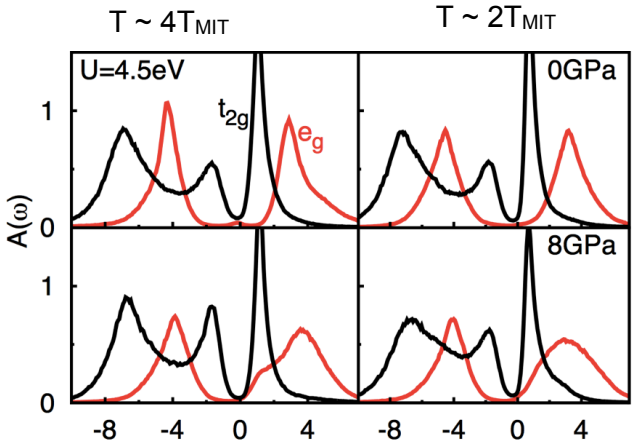
Spin state transition in $LaCoO_3$

Co 3d states



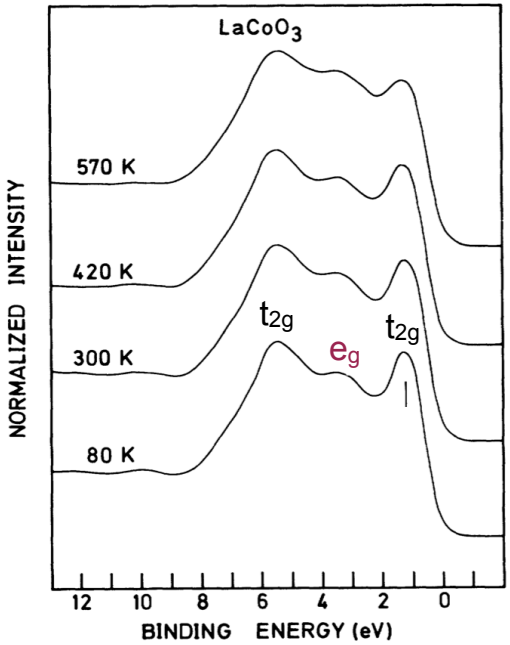


High spin state, density of states for Co 3d orbitals



G. Zhang, E. Gorelov, E. Koch, and E. Pavarini, PRB 86, 184413 (2012)

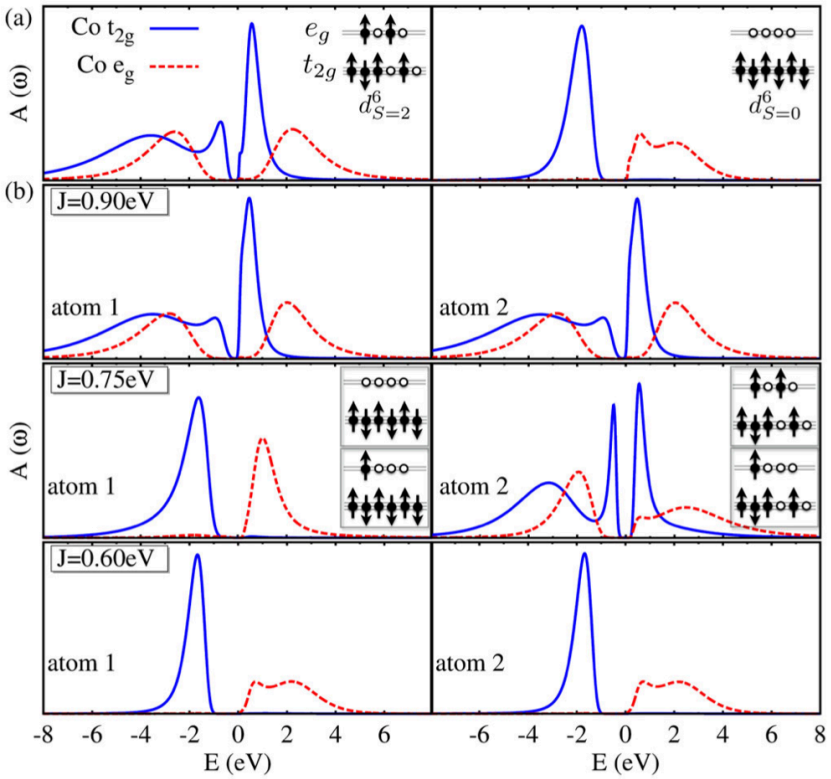
Valence-band XPS



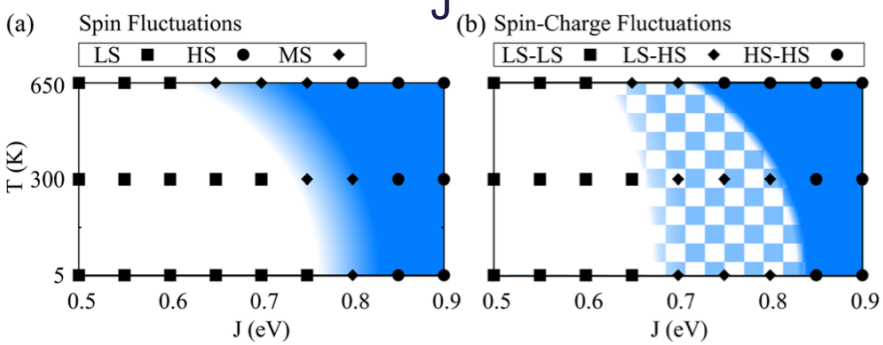
M. Abbate et al., PRB 47, 16124 (1993)

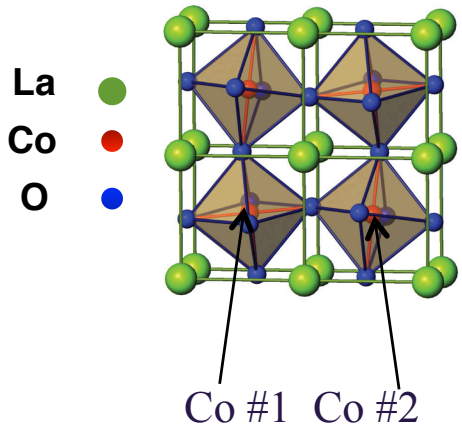
Spin and Charge modulations in $LaCoO_3$

Density of states for Co 3d orbitals



Phase diagram as a function of the lattice temperature and the Hund's rule coupling J



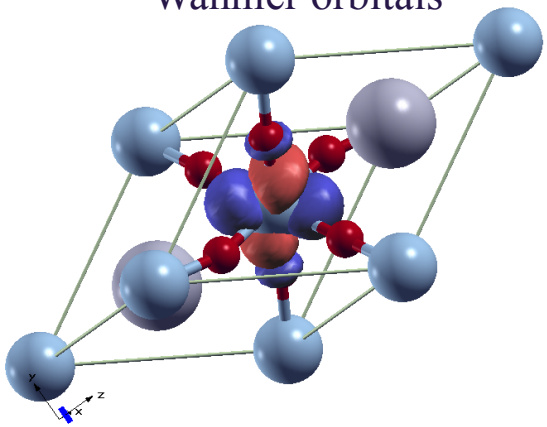


M. Karolak, M. Izquierdo, S. L. Molodtsov, and A. L., PRL **115**, 046401 (2015)

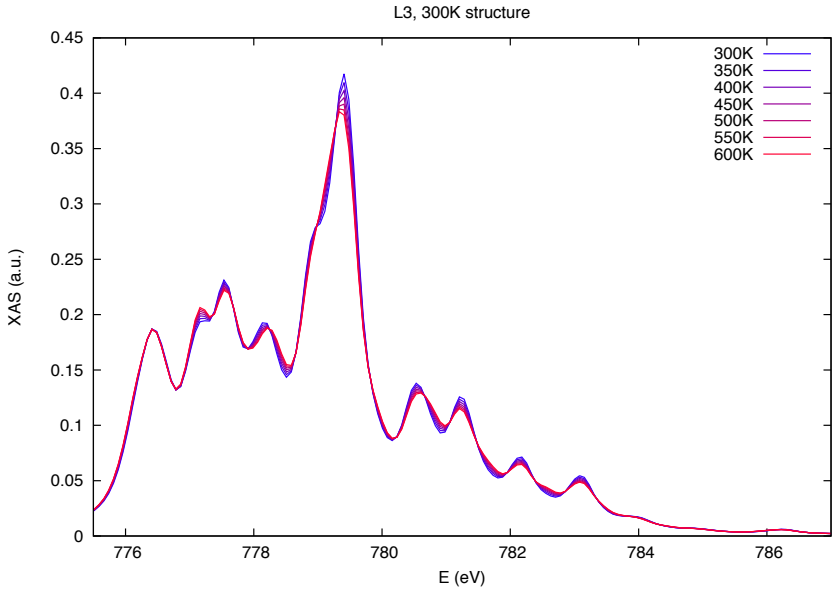
Cluster model for calculation of XAS spectra in $LaCoO_3$

- Includes Co-d and O-p orbitals
- Electron Hopping between Co-d and Ligands is allowed
- Full Coulomb vertex at Co-d orbitals is included

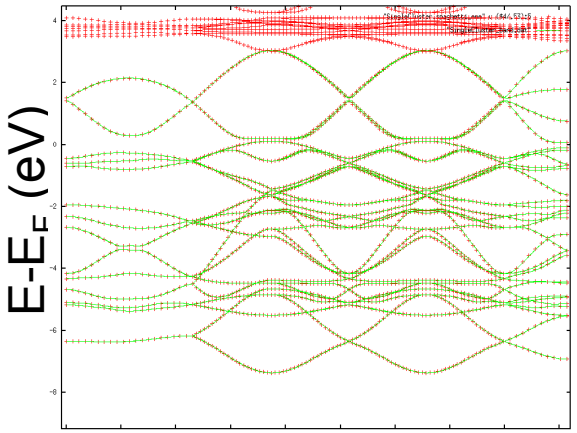
Wannier orbitals



Co $L_{2,3}$ XAS spectra with fixed structure and different electronic temperatures



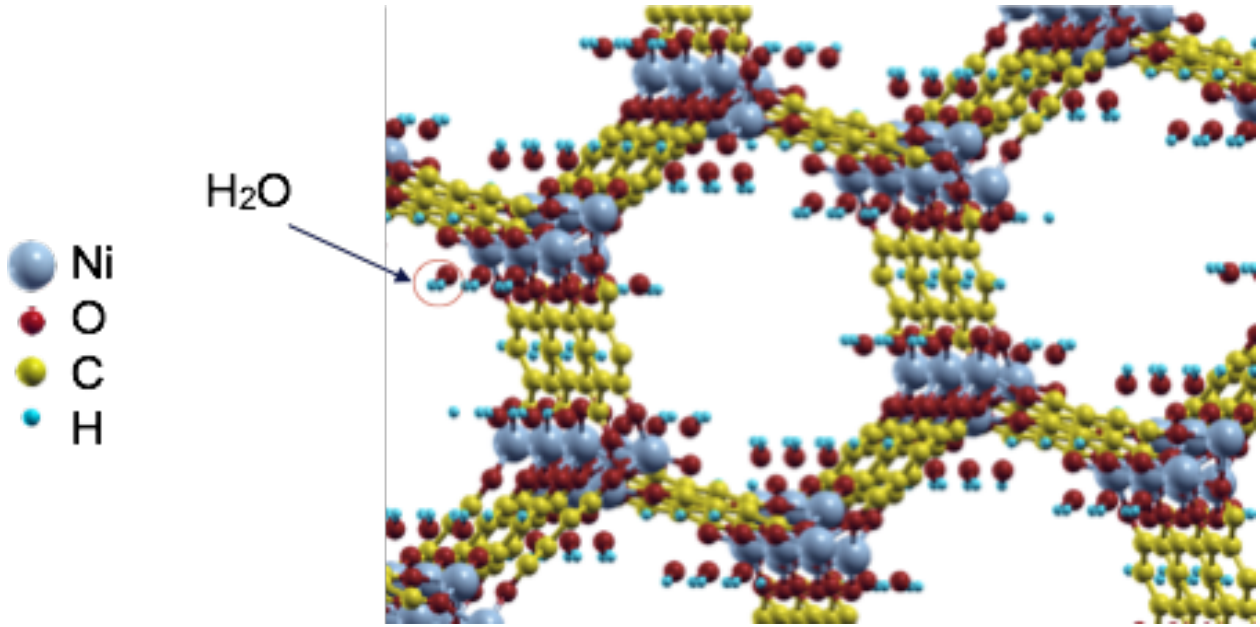
Co and O bands (green)

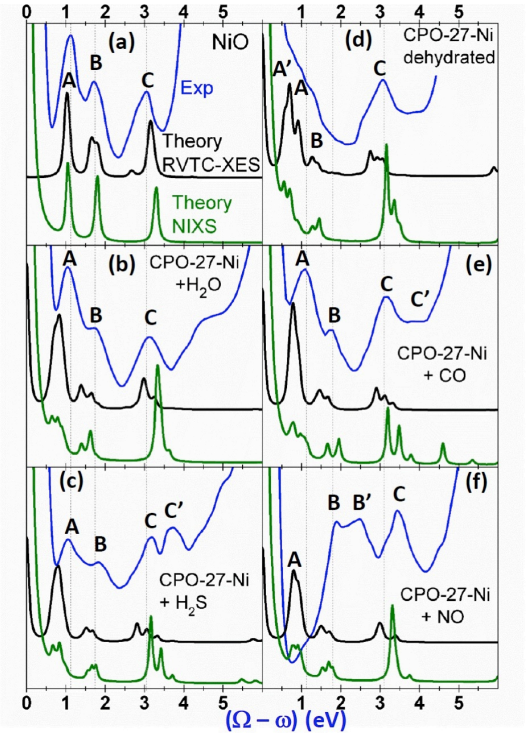


- E. Gorelov, to be published

Spectral properties of correlated materials: CPO-27-Ni XES

- Catalytic metal-organic framework CPO-27-Ni
  - Ni valence to core K-edge XES
  - Ni d-d excitations reflects Ni environment, where catalytic activity takes place
  - Study of catalytic activity in-situ is possible



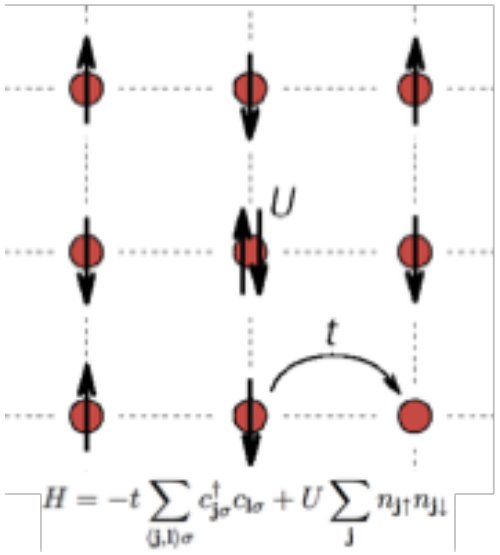


- E. Gallo, E. Gorelov, A. A. Guda, F. Bonino, E. Borfecchia, D. Gianolio, S. Chavan, and C. Lamberti, submitted to Inorg. Chem.
- Full Coulomb interaction for Co-d orbitals
- Cluster spectrum codes: XTLS by Arata Tanaka (Hiroshima); Quanty by Maurits W. Haverkort (Heidelberg)

Correlated materials in strong time-dependent electric fields

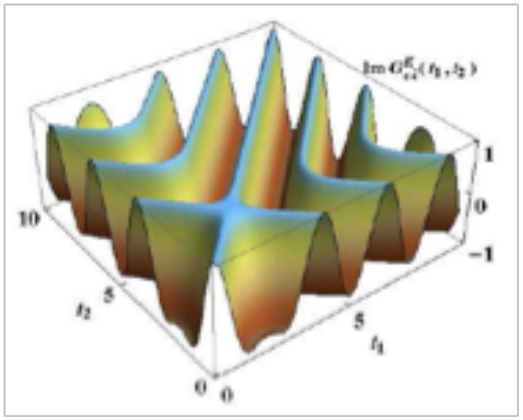
- Investigation of behaviour of correlated materials in presence of strong electric fields
  - Hubbard model is used to model correlated materials
  - IPT on Keldysh contour with non-equilibrium dynamical mean-field theory
  - Electric field is included as Pierls substitution for hoppings

Hubbard model in Electric fields



Peierls substitution: $t_{ij} \to t_{ij} \exp\left(ie\mathbf{E}(\mathbf{R}_j - \mathbf{R}_i)t\right)$

$\mathbf{E}(t) = -\partial_t \mathbf{A}(t)$



E-Puls $E(t) = E\theta(t)\theta(t_f - t)$

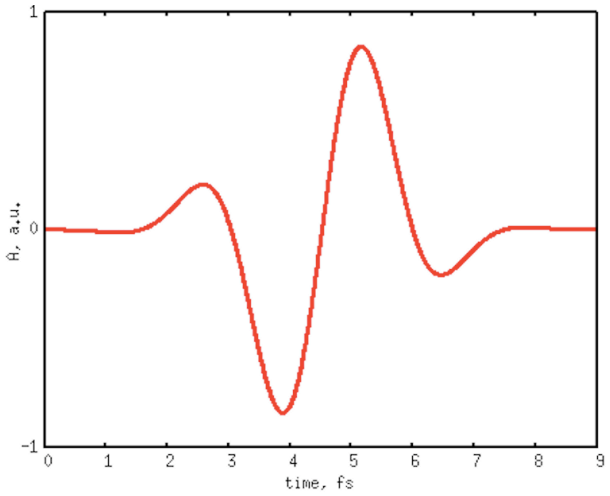
E-switch $A(t) = E\theta(t)$

A. Joura, J. K. Freericks, A.L., Phys. Rev. B **91**, 245153 (2015)

Correlated materials in strong time-dependent electric fields

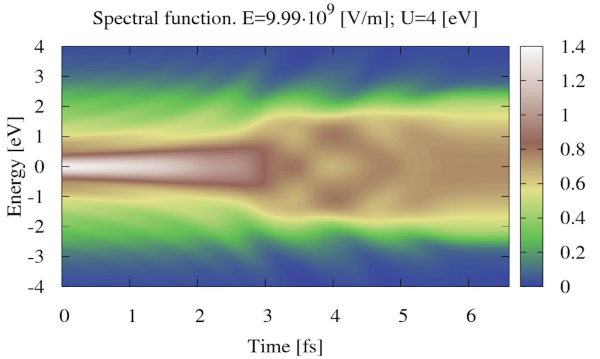
- Investigation of behaviour of correlated materials in presence of strong electric fields
  - We study single-band Hubbard model with anisotropic hoppings
  - Arbitrary field shape and strength can be considered
  - Multi-orbital extension allowing realistic calculations is feasible

Vector potential

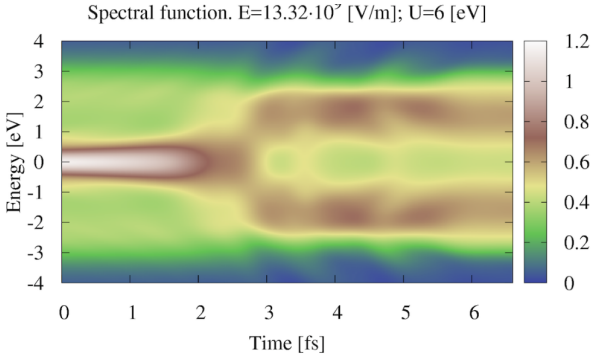


Spectral function

U=4 eV



U=6 eV



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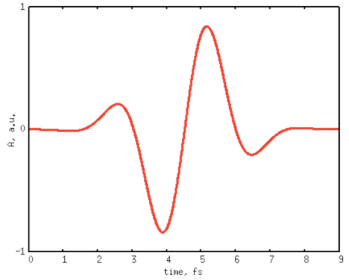
European XFEL

Correlated materials in strong time-dependent electric fields

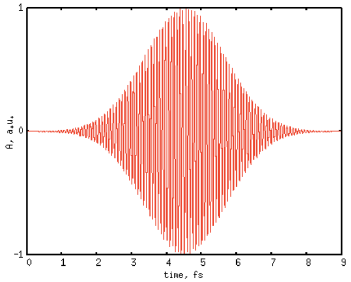
- Infrared vs X-ray

Vector potential

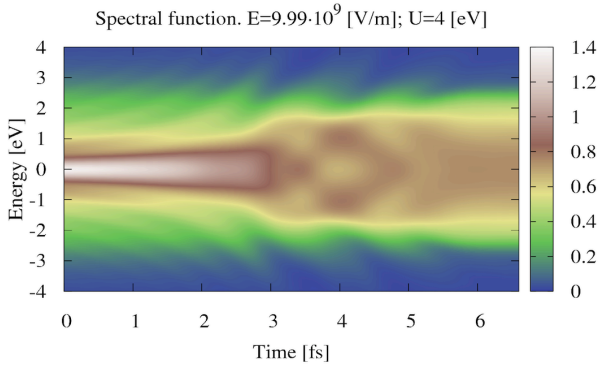
IR

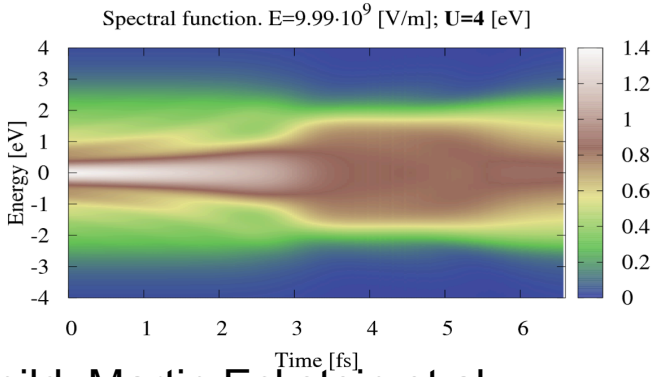


Xray

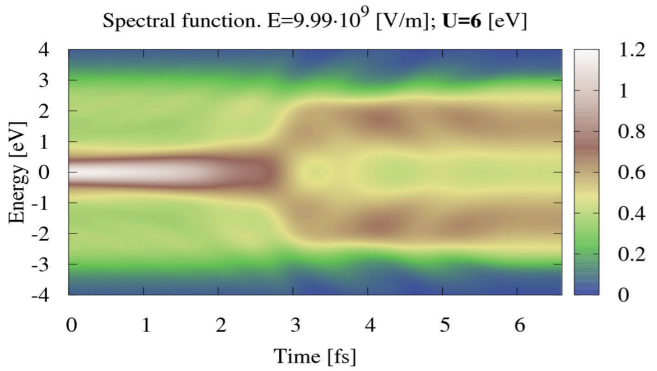


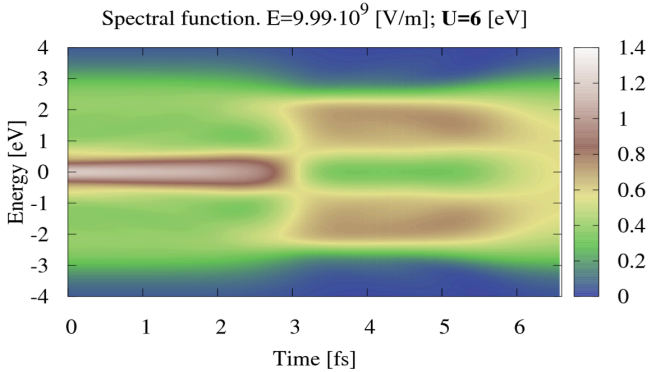
U=4 eV





U=6 eV

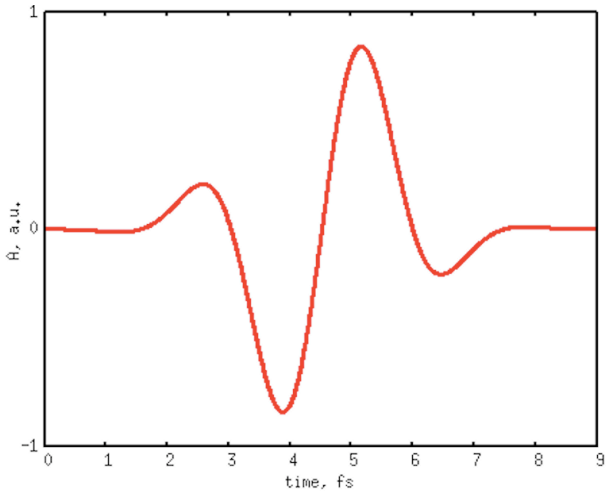


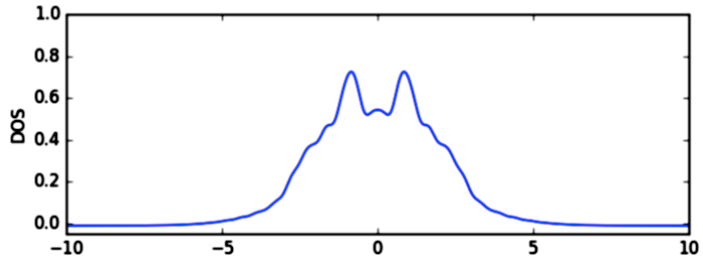


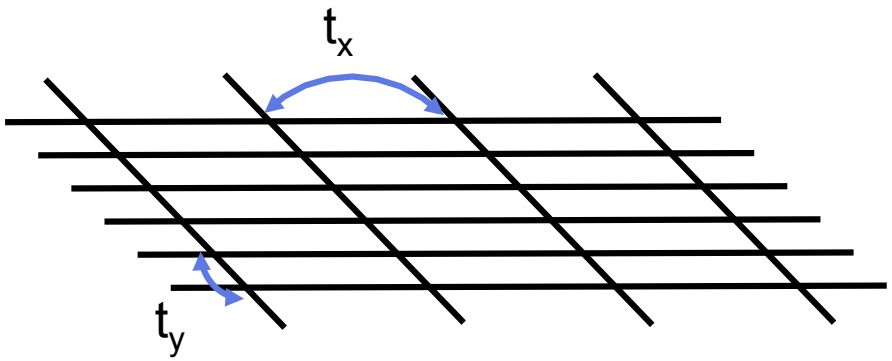
- Viktor N. Valmispild, Martin Eckstein et al.

Correlated materials in strong time-dependent electric fields

- Polarization dependence on rectangular lattice
  - We study anisotropic Hubbard model, $t_y = 2t_x$
  - We use infrared pulse, $\omega = 1.4$ eV
  - Linear polarization along X or Y axis is used





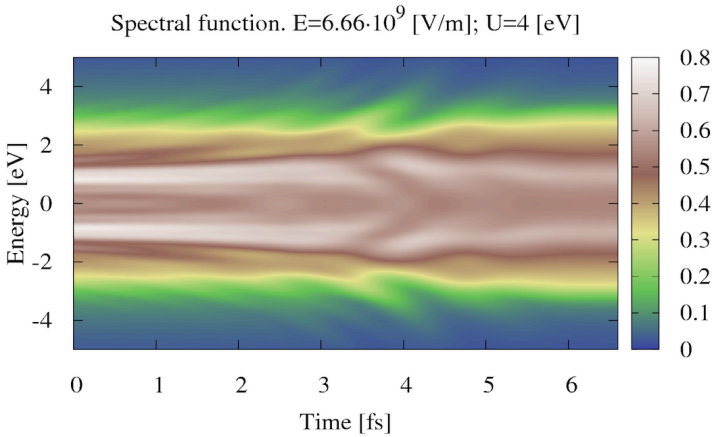


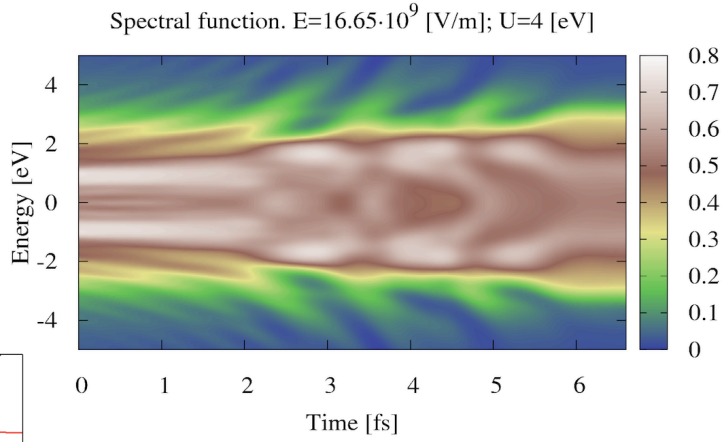
Lattice distortion and time-dependent electric fields

Small Field

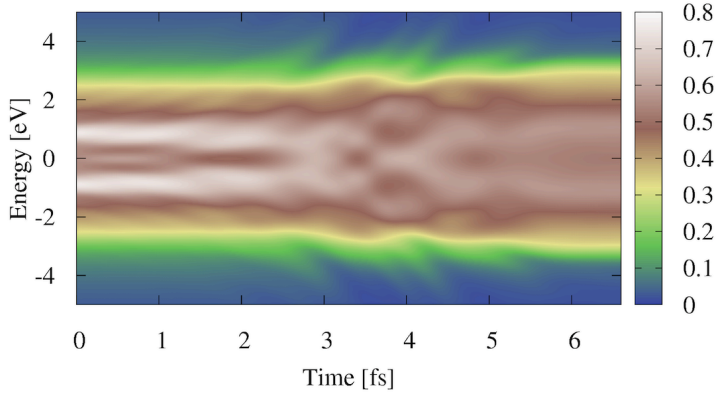
Large Field

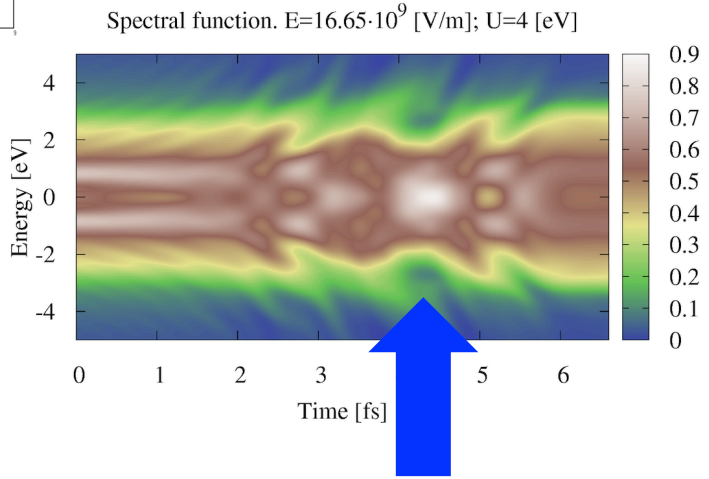
E || x

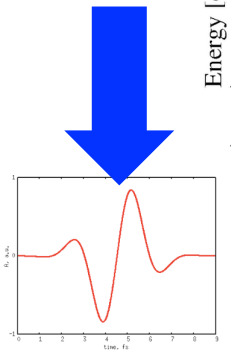




E || y



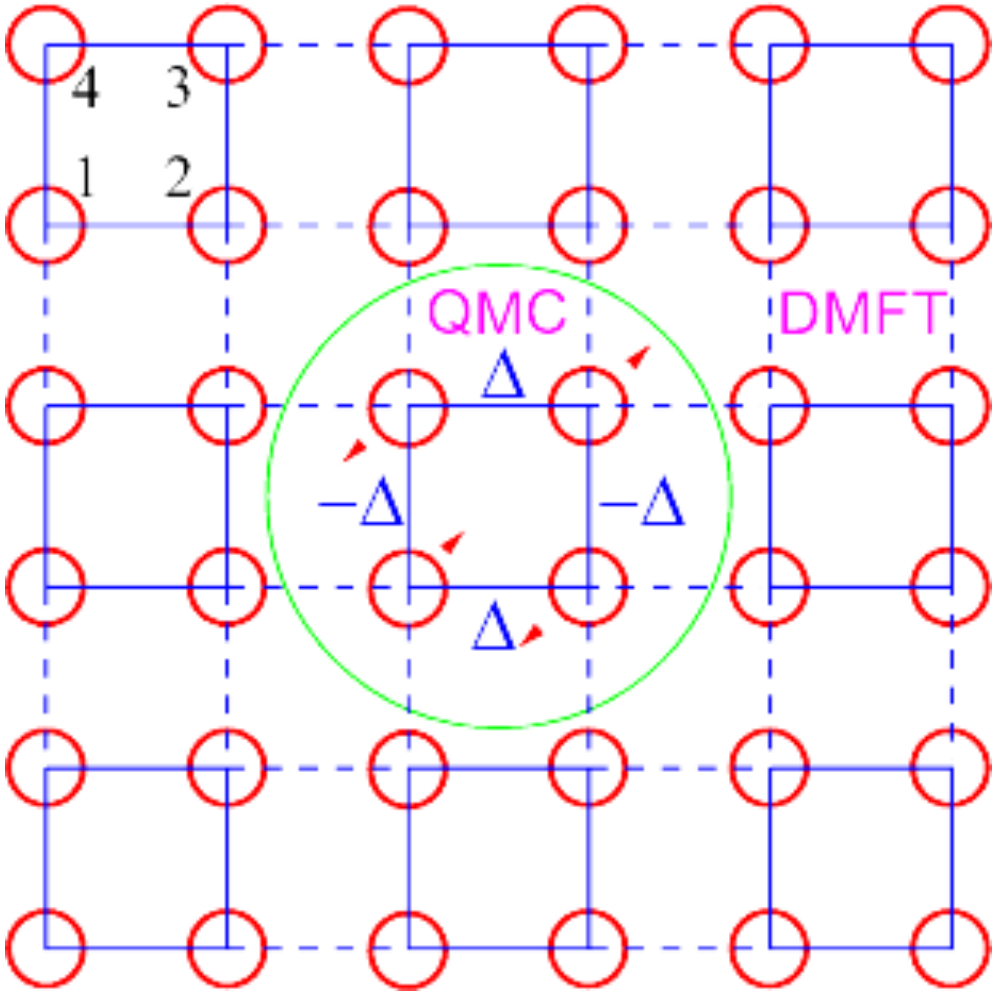


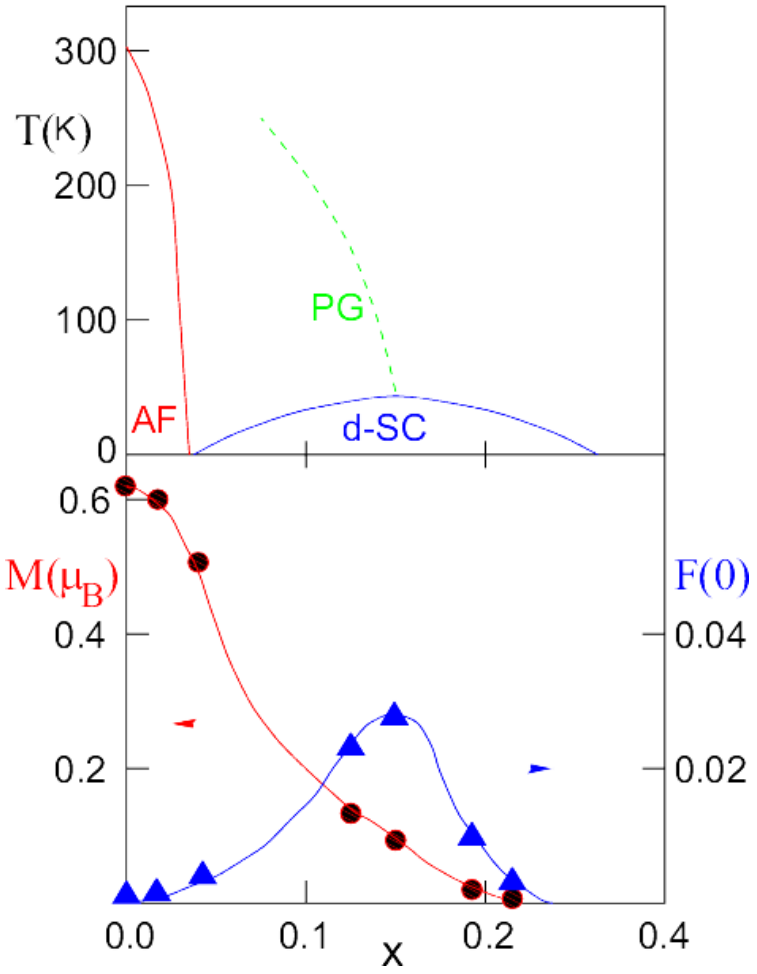


Viktor N. Valmispild, Martin Eckstein et al.

European XFEL

Non-Local correlations: AFM and d-wave in HTSC



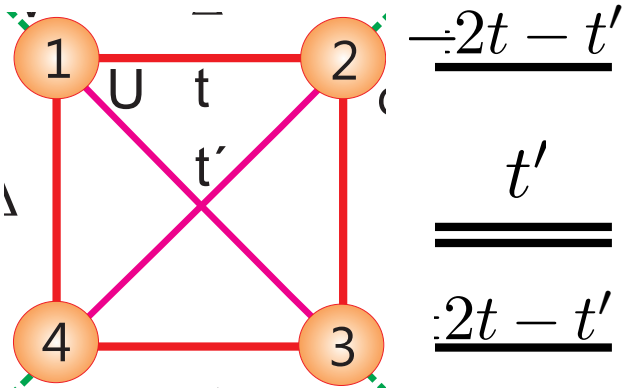


A.L. and M.Katsnelson, PRB **62**, R9283 (2000)

Plaquette Valence Bond Theory of HTSC

$$H_p = \sum_{(i,j)=1..4} h^0_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \sum_{i=1..4} U n_{i\uparrow} n_{i\downarrow}$$

$$-\hat{h}_0 = \begin{pmatrix} \mu & t & t' & t \\ t & \mu & t & t' \\ t' & t & \mu & t \\ t & t' & t & \mu \end{pmatrix}$$



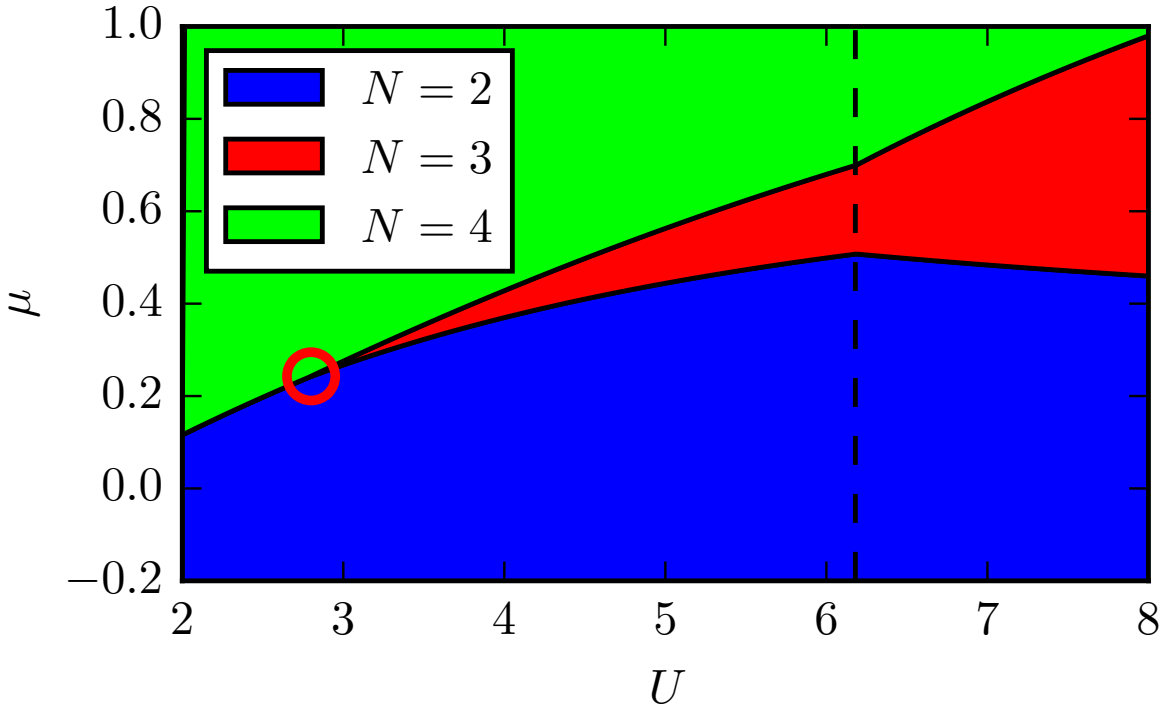
$-2t - t'$

t'

$-2t - t'$

t=-1 t‘=0.3

„Plaquette Quantum Critical Point“



Periodic BC: t->2t and U->2U

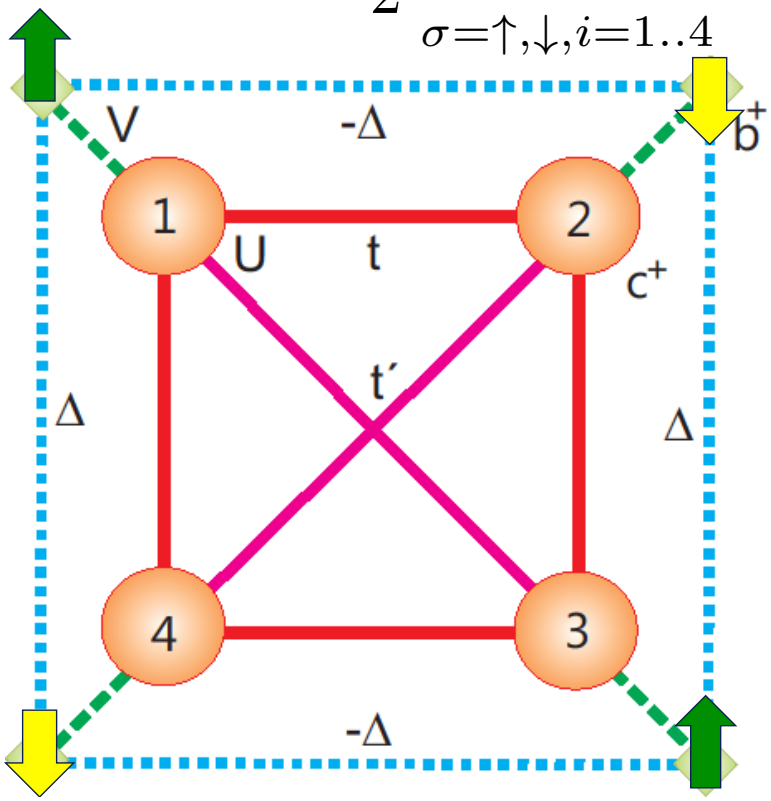
M. Harland, M. Katsnelson, A.L. PRB 94, 125133 (2016)

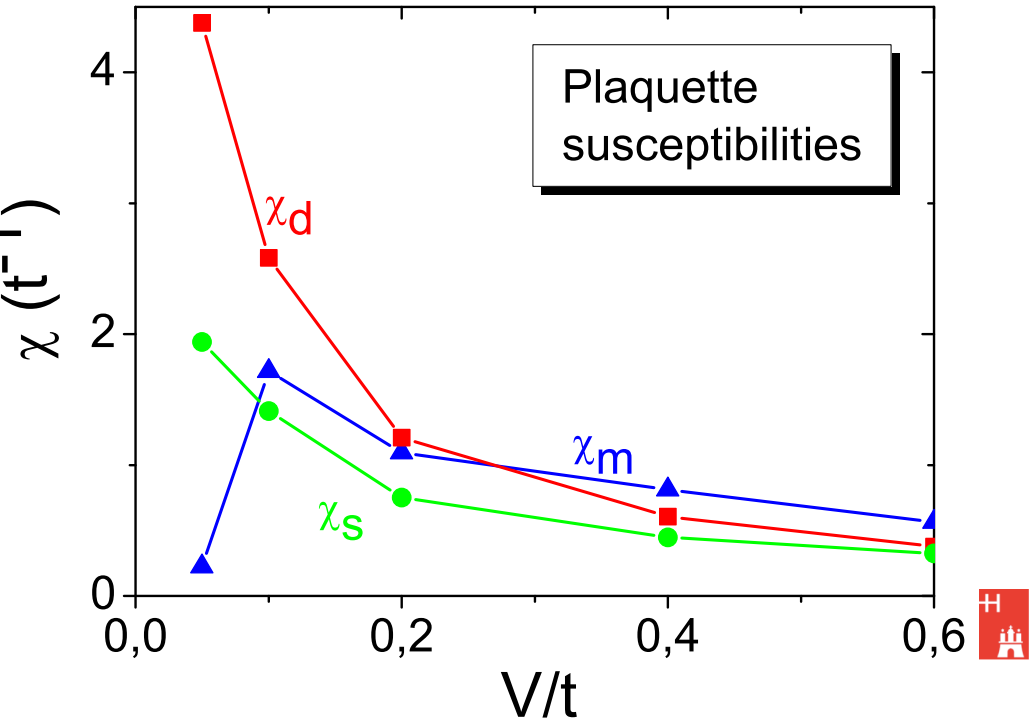
Plaquette susceptibility: one level bath

$$h_d = \sum_{\sigma=\uparrow,\downarrow, i=1..4} (-1)^i \sigma \Delta_d (b_{i,\sigma} b_{i+1,-\sigma} + h.c.),$$

$$h_s = \sum_{\sigma=\uparrow,\downarrow, i=1..4} (-1)^i \sigma \Delta_s (b^\dagger_{i,\sigma} b_{i+1,-\sigma} + h.c.),$$

$$h_m = \frac{1}{2} \sum_{\sigma=\uparrow,\downarrow, i=1..4} (-1)^i \sigma \Delta_m b^\dagger_{i,\sigma} b_{i,\sigma}.$$

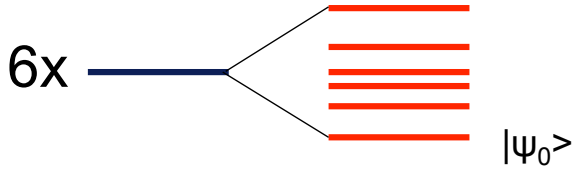


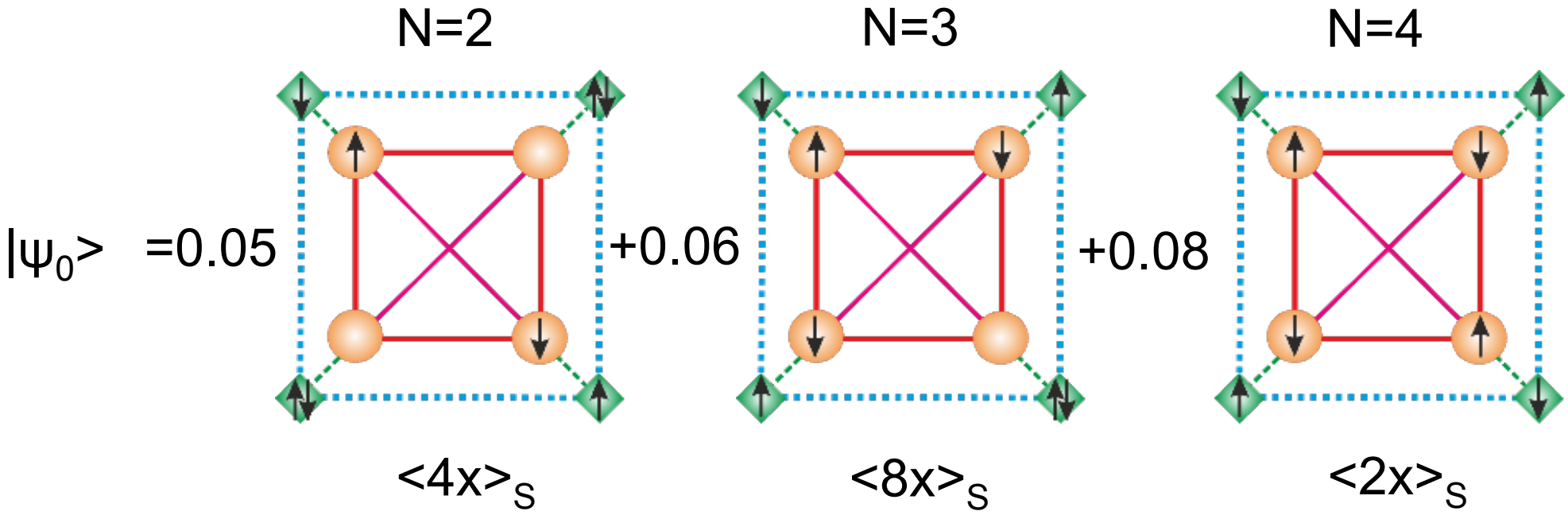


Singlet State of SC-plaquette with bath

Isolated Plaquete:

Plaquete in SC-Bath: $V=0.2$, $\Delta_d=0.05$

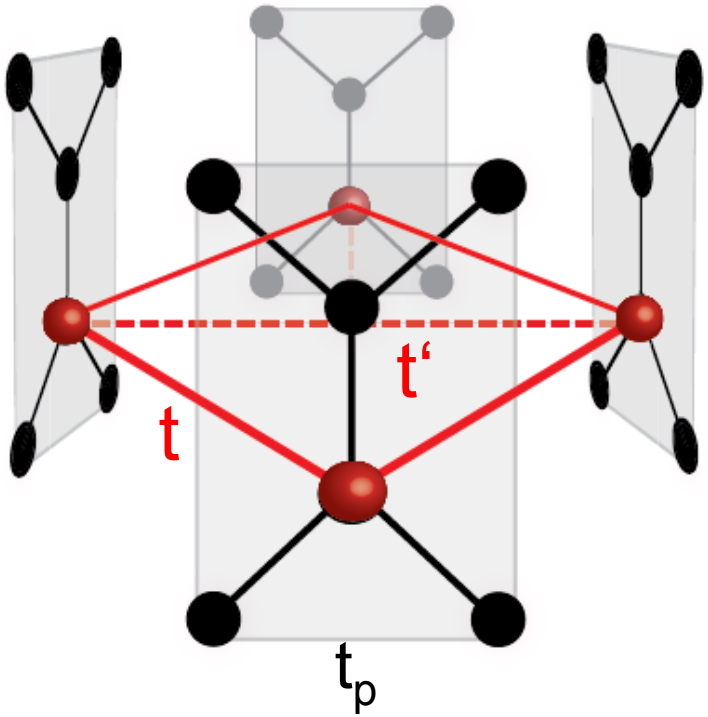




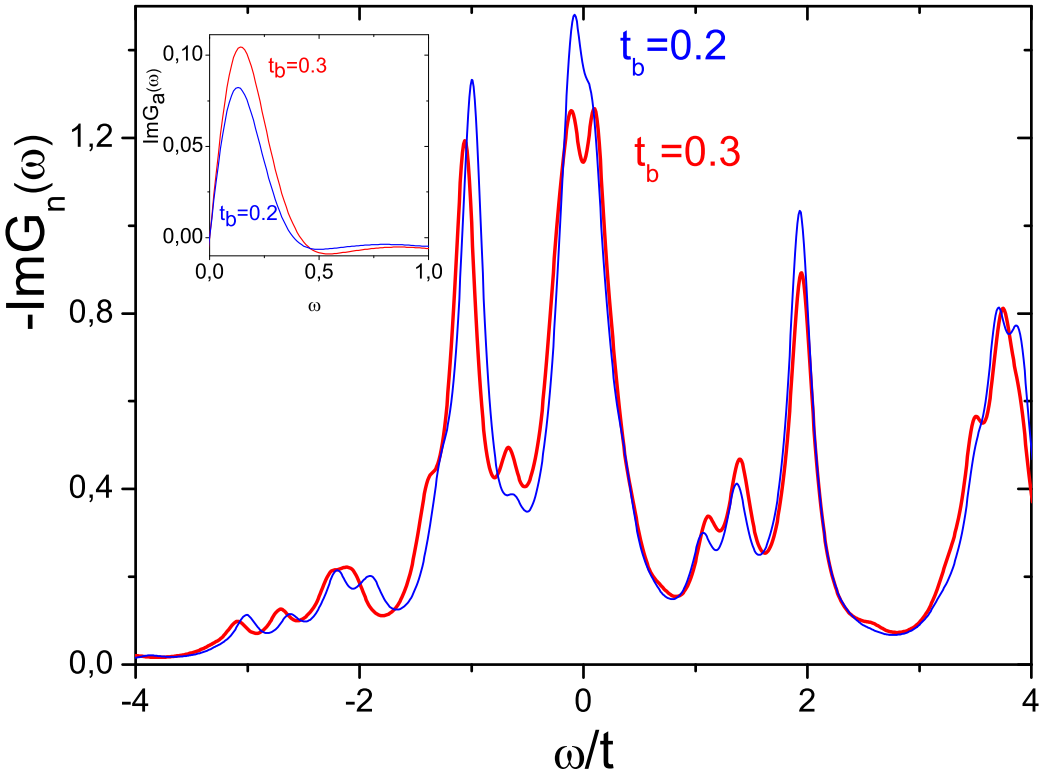
Plaquette-Bethe lattice

$$\hat{\mathcal{G}}^{-1}(i\omega) = \left(i\omega + \mu - \hat{h}_0\right)^{-1} - t_b \sigma_z \hat{G}(i\omega)\, \sigma_z t_b$$

G. Moeller, V. Dobrosavljevic, and A. E. Ruckenstein,
Phys. Rev. B **59**, 6846 (1999).

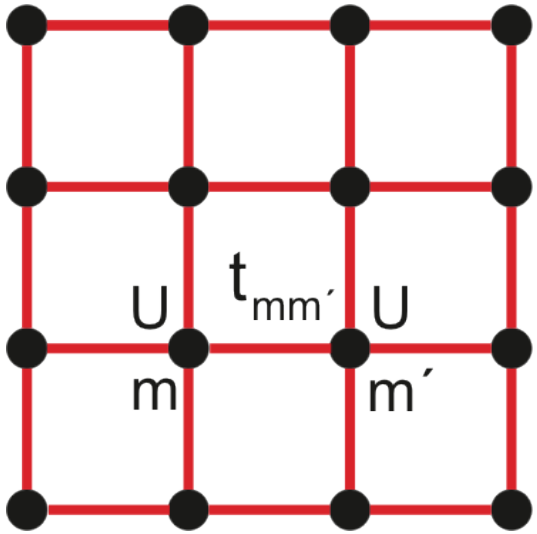


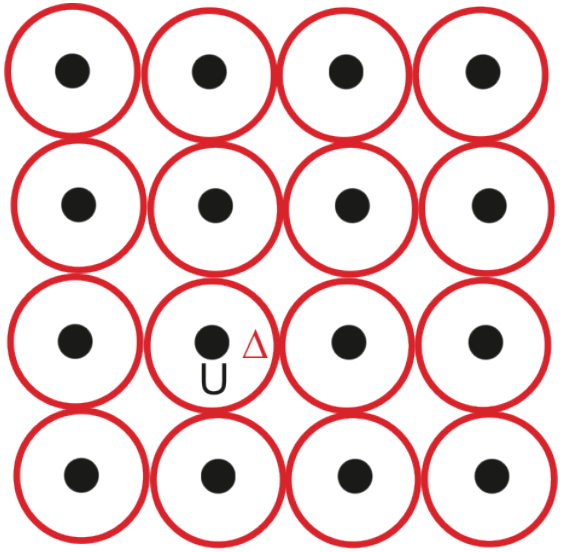
Optimal Bethe-hopping: t_p =0.25

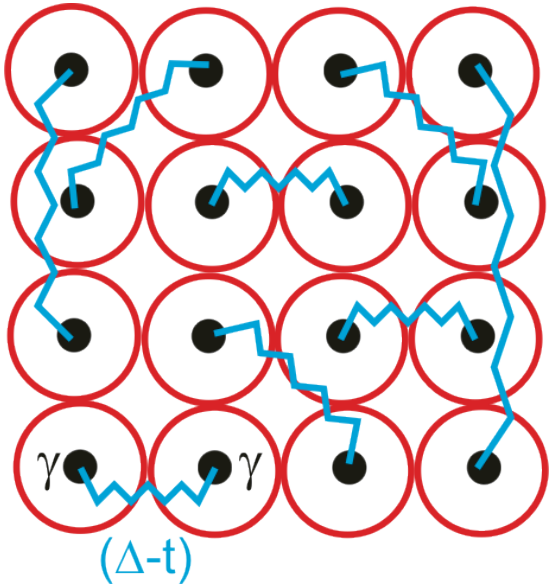


M. Harland, M. Katsnelson, A.L. PRB 94, 125133 (2016)

DMFT-functional





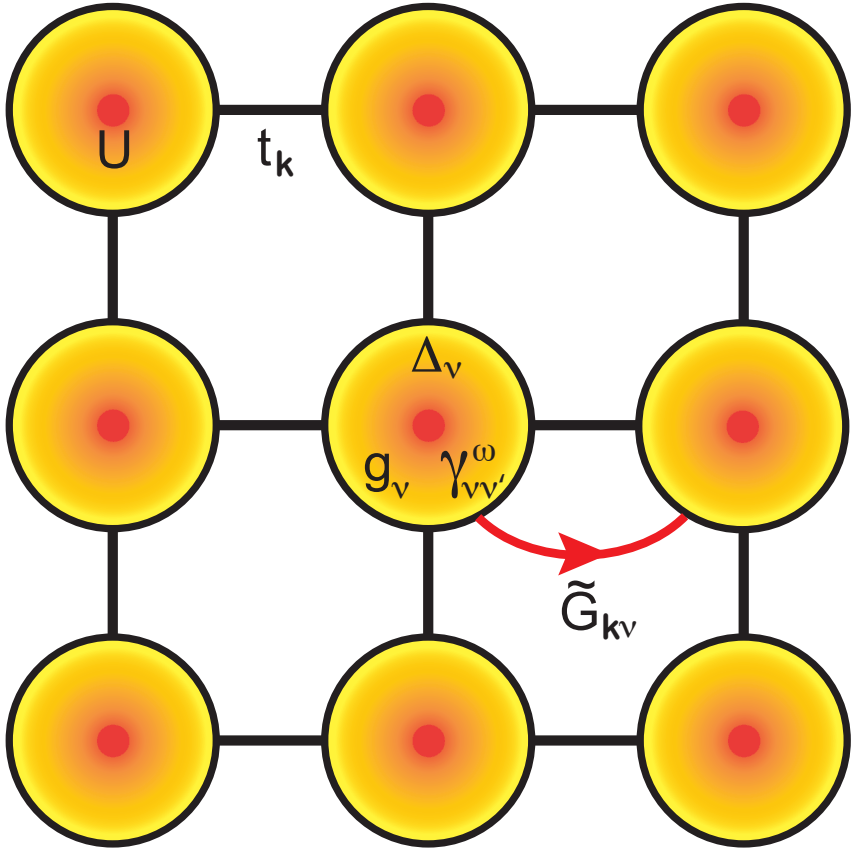


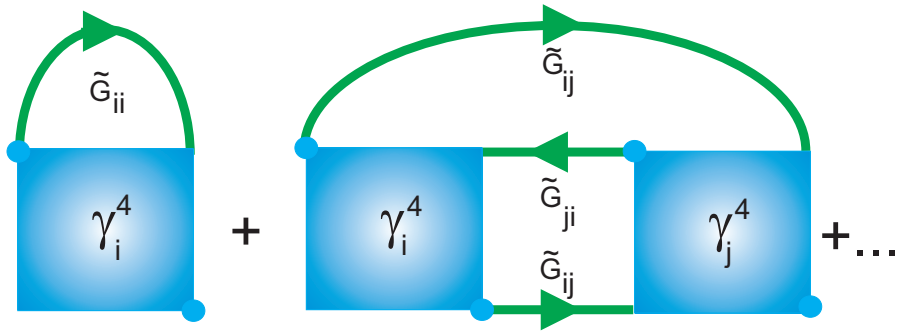
Start from Correlated Lattice

Find the optimal Reference System Bath hybridization

Expand around DMFT solution

Dual Fermion: Beyond DMFT

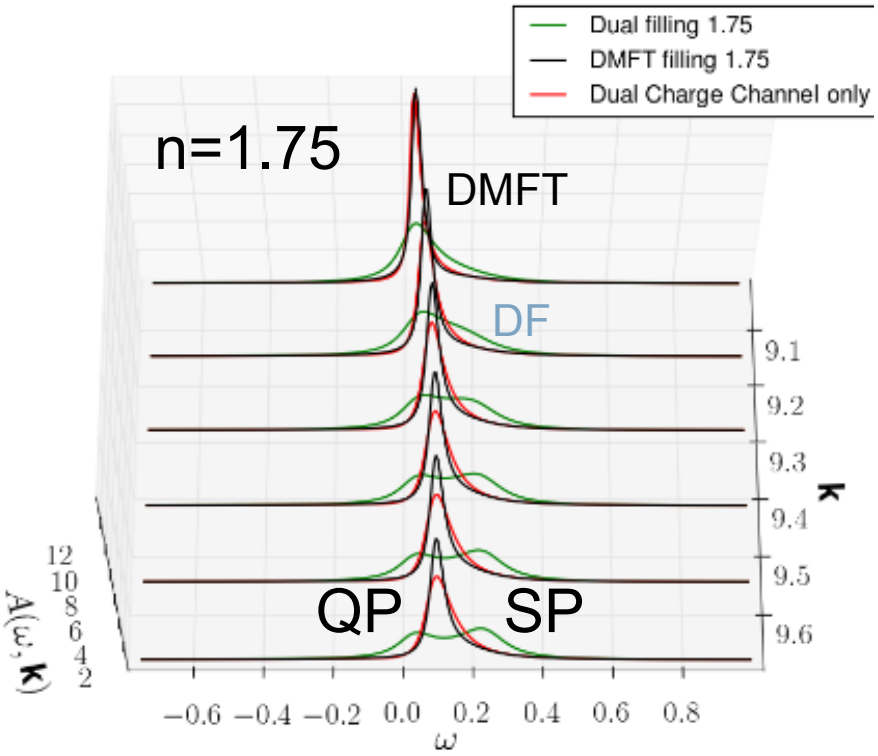


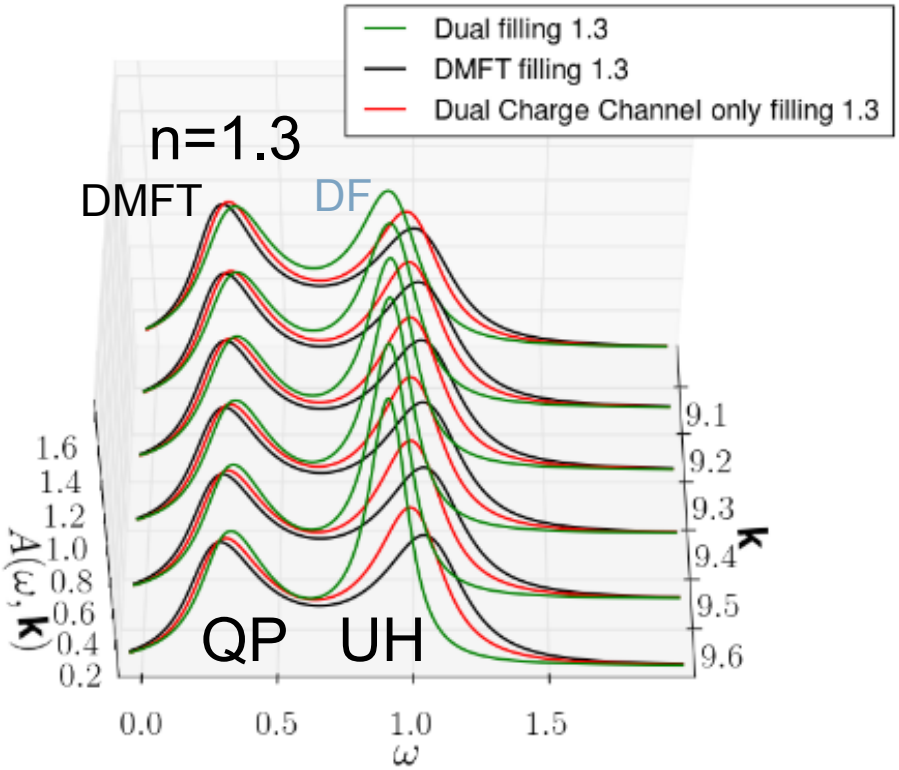


A. Rubtsov, et al, PRB **77**, 033101 (2008)

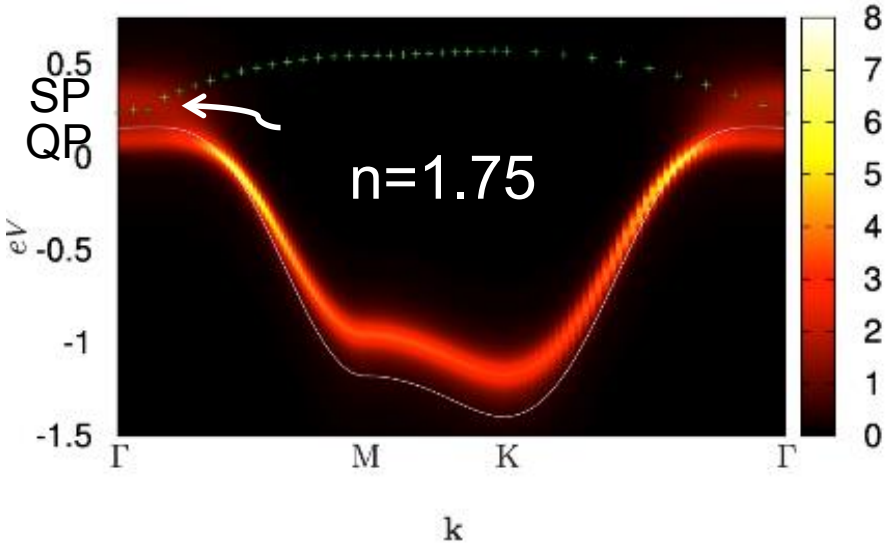
$$G_{k\omega} = [(g_\omega + g_\omega \tilde{\Sigma}_{k\omega} g_\omega)^{-1} + \Delta_\omega - t_k]^{-1}$$

Spin-Polaron near van Hove singularity in real Material: Na_xCoO_2





DF-spectral function Na_xCoO_2



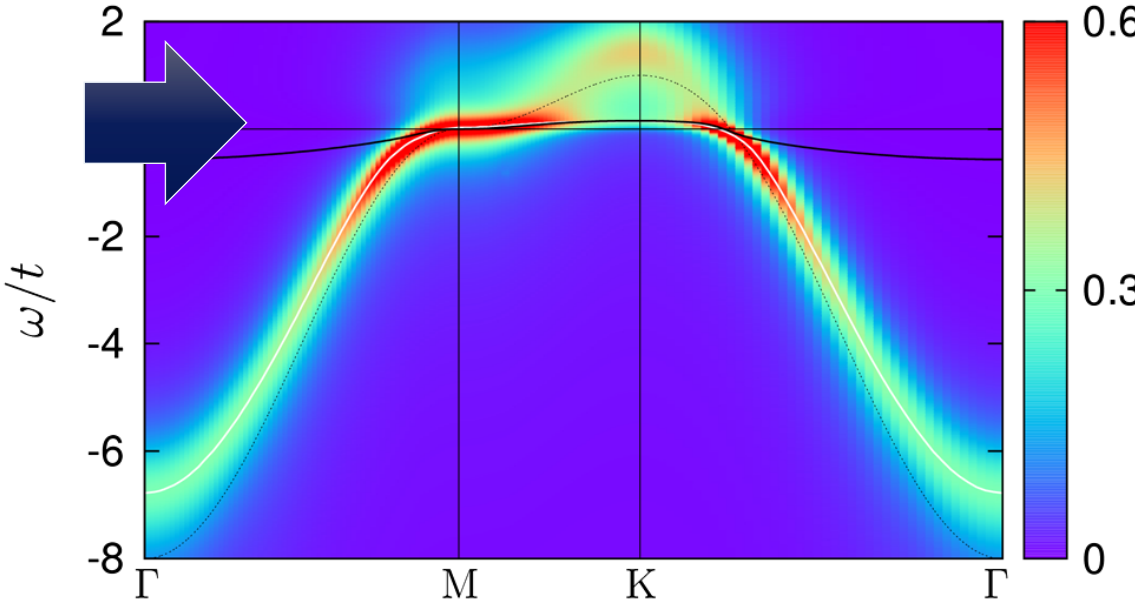
A. Wilhelm, F. Lechermann, H. Hafermann, M. Katsnelson, A. L. Phys. Rev. B 91, 155114 (2015)

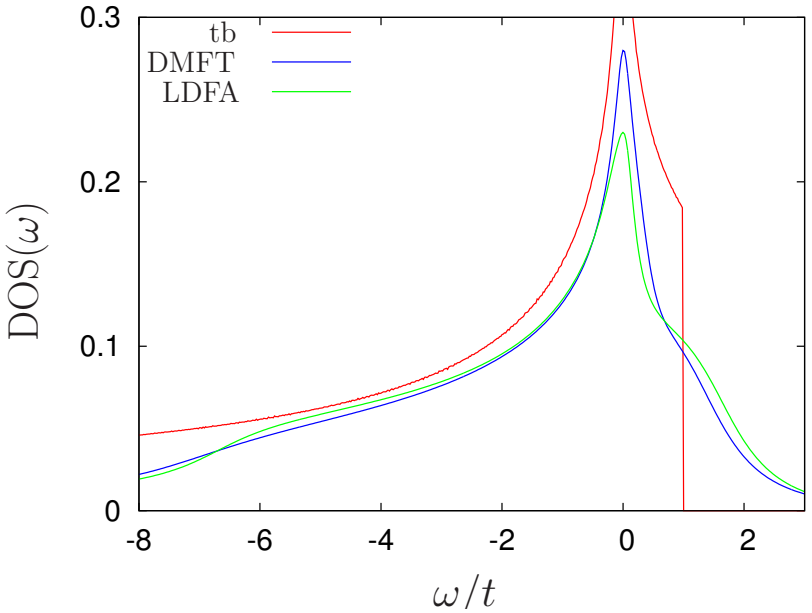
Spin-Polaron physics for n=1.75

$$E(k) = -\frac{2t\left(t - J\cos k\right)}{|t| + J}$$

1d t-J model, M. Katsnelson (1982)

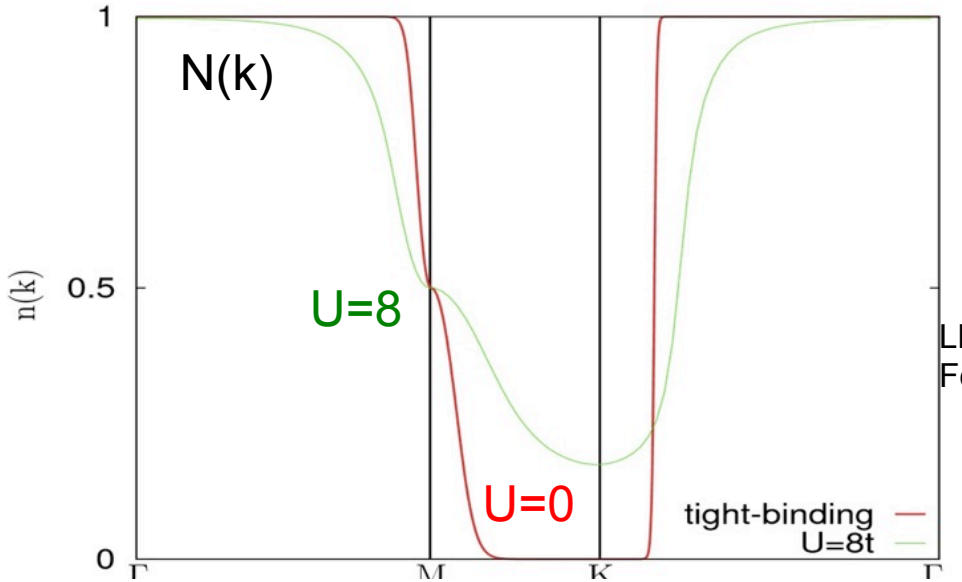
Extended van Hove singularity: triangular lattice



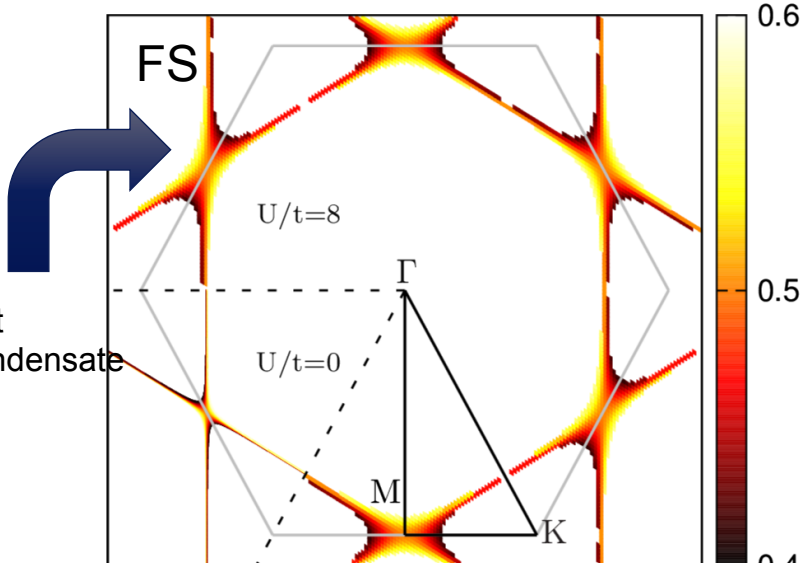


Spectral Function

D. Yudin, D. Hirschmeier, et al., PRL **112**, 070403 (2014)

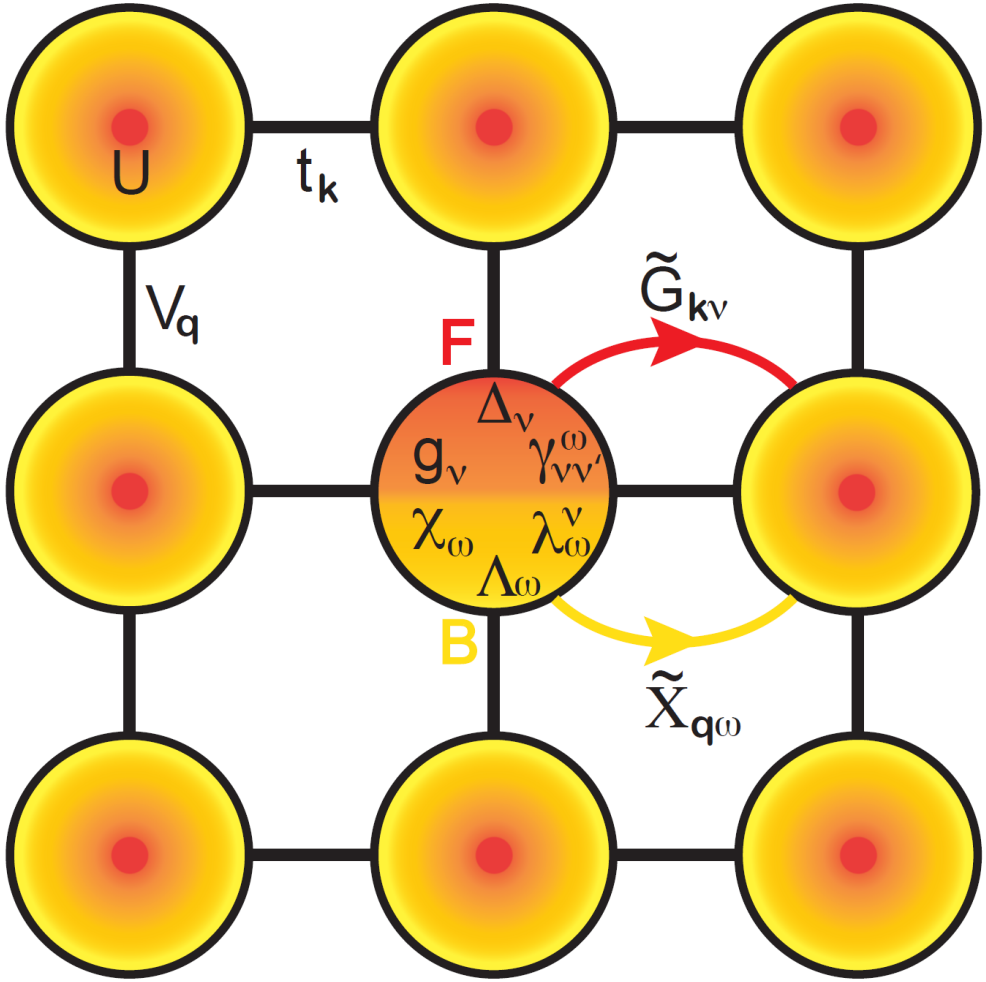


LDFA U=8t
Fermion Condensate



Dual Boson: non-local interaction + screening

$$S_{imp} = \int\int_0^\beta d\tau d\tau' \left[-\sum_\sigma \mathcal{G}^{-1}_{\tau\tau'} c^*_{\tau\sigma} c_{\tau'\sigma} + \frac{1}{2}\left(U + \Lambda^c_{\tau\tau'}\right) n_\tau n_{\tau'} + \frac{1}{2}\Lambda^s_{\tau\tau'}\, \vec{s}_\tau \cdot \vec{s}_{\tau'} \right]$$

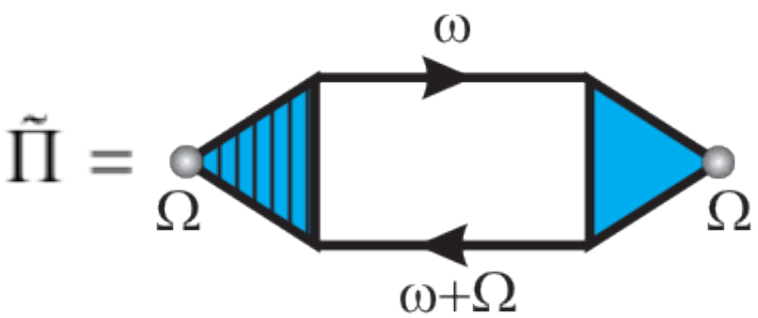


HTSC

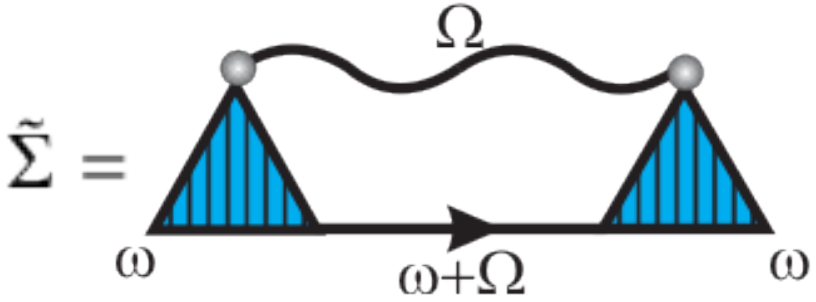
$$\Lambda_\omega \sim J_{\tau\tau'} \vec{S}_\tau \cdot \vec{S}_{\tau'}$$

DB-diagrammatic scheme

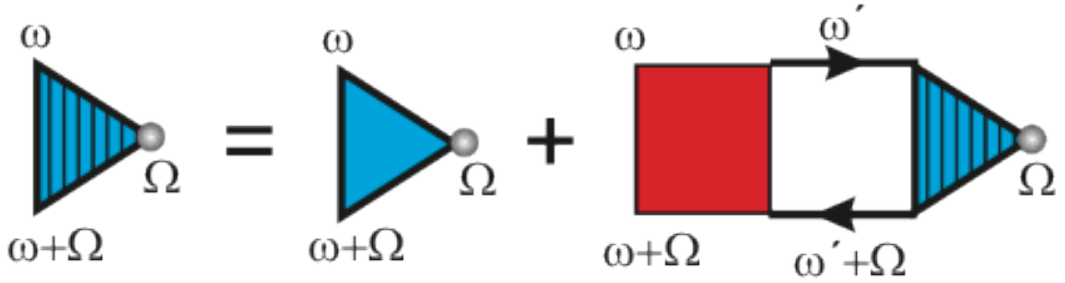
Bosonic Selfenergy



Fermionic Selfenergy



Renormalized vertex:



Fermionic and Bosonic Green Functions

$$G_{k\nu} = [(g_\nu + g_\nu \tilde{\Sigma}_{k\nu} g_\nu)^{-1} + \Delta_\nu - t_k]^{-1}$$

$$X_{q\omega} = [(\chi_\omega + \chi_\omega \tilde{\Pi}_{q\omega} \chi_\omega)^{-1} + \Lambda_\omega - V_k]^{-1}$$

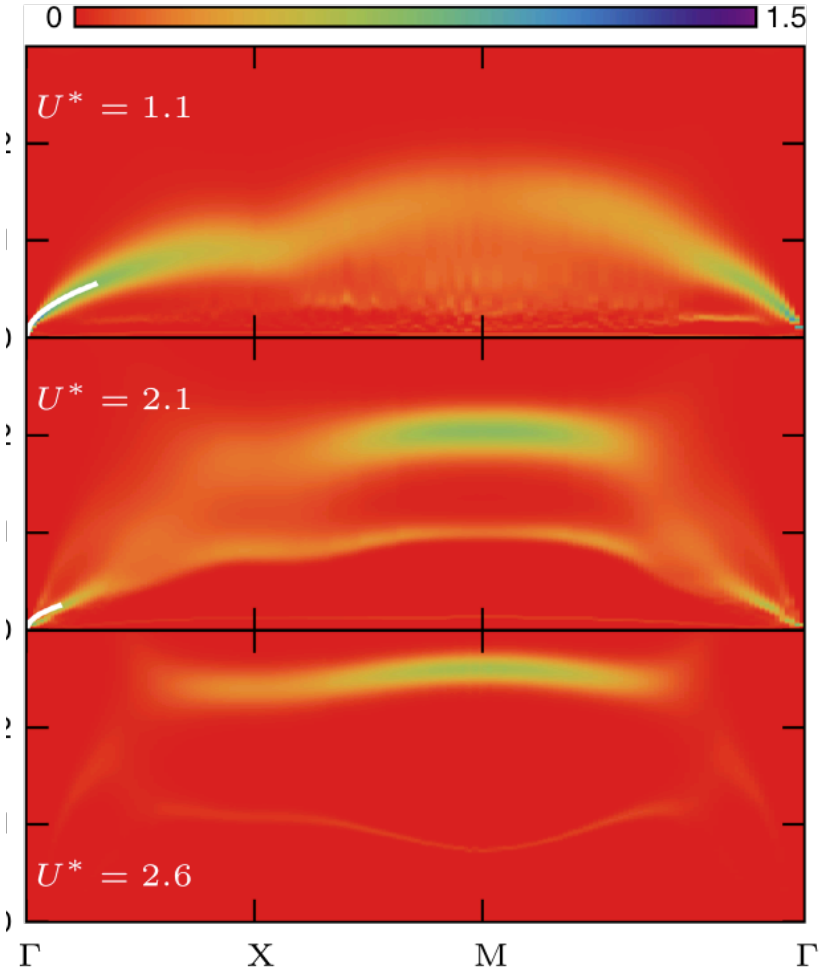
SCF-condition

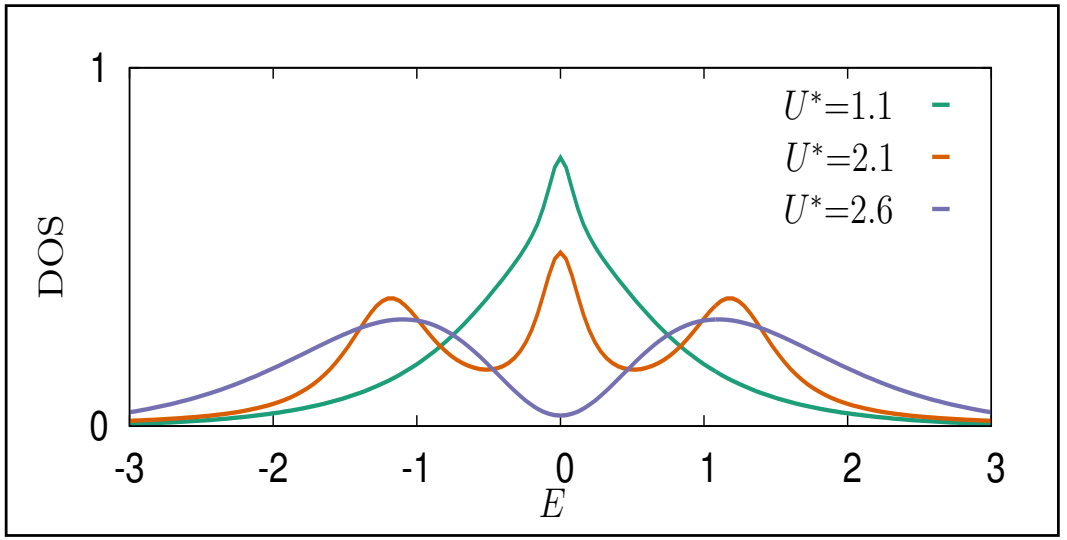
$$\sum_k G_{k\nu} = g_\nu \quad \Rightarrow \quad \Delta_\nu$$

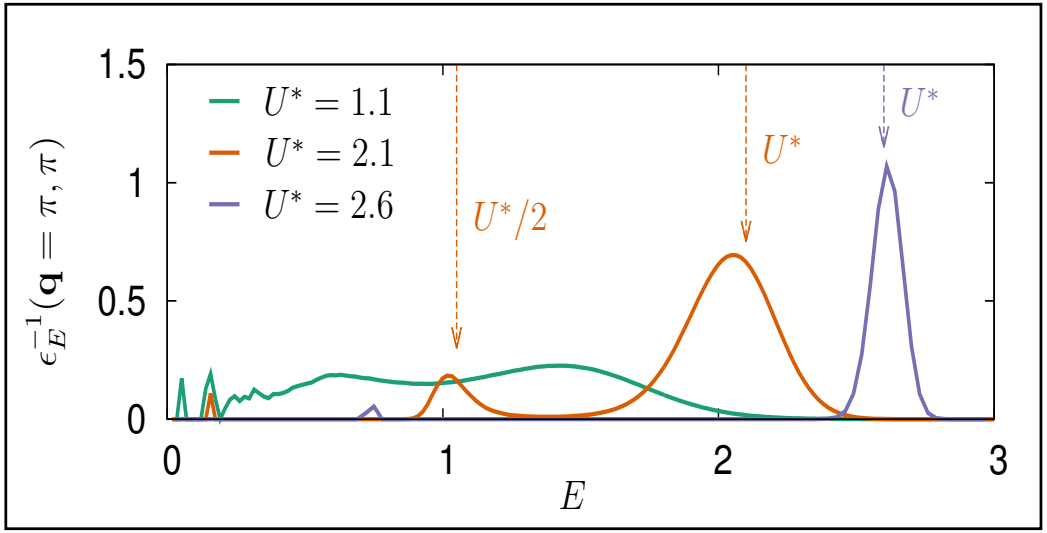
$$\sum_q X_{q\omega} = \chi_\omega \quad \Rightarrow \quad \Lambda_\omega$$

A. Rubtsov, M. Katsnelson, A. L., Annals of Phys. **327**, 1320 (2012)

Plasmon in strongly correlated materials







Single plasmon mode for q->0

Erik van Loon, et all, PRL **113**, 246407 (2014)

Summary

- We developed general methods for spectroscopic calculations of strongly correlated materials
- Accurate calculations of Fermi-surface, metal-to-insulator transition details, and spectral functions for strongly correlated materials have been done in framework of DFT+DMFT.
- XAS spectra of real materials are calculated in order to assist in interpretation of experimental data.
- Non-equilibrium DMFT approach have been used in order to study behavior of correlated materials in presence of strong electric fields.
- Non-local correlations beyond DMFT can be well described in dual fermion/boson scheme