Beyond RPA via Dynamical Mean-Field Theory: Fully self-consistent GW+DMFT

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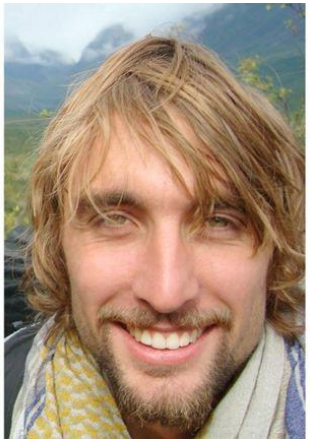


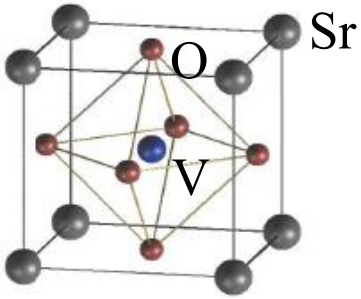
Fredrik Nilsson
(Lund)

Philipp Werner
(Fribourg)

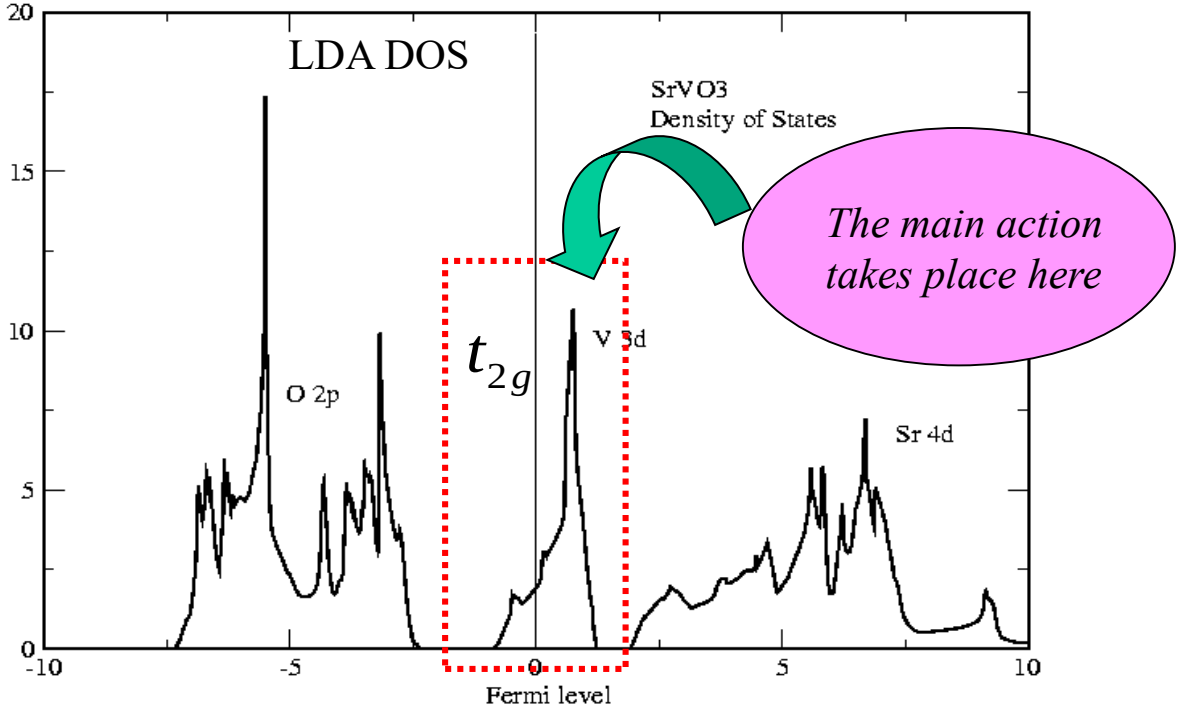
Outline

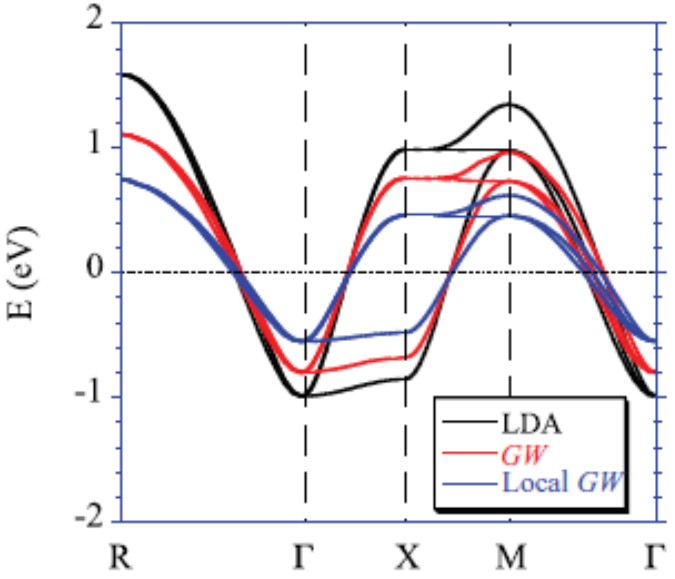
- Problems with *GW* (RPA)
- Vertex corrections via DMFT: *GW*+DMFT
- Fully self-consistent *GW*+DMFT: SrVO3 as a test case
- Results:
  New interpretation of satellite features:
  Plasmons instead of Hubbard bands

Prototype of correlated materials



Cubic perovskite SrVO3





Miyake, PRB 87, 115110 (2013)

Partially filled narrow band originating from 3d or 4f orbitals.
Kinetic energy and onsite Coulomb repulsion become comparable.

Goal:
First-principles description of electronic structure of correlated materials

Experiment vs *GW* Theory (RPA)

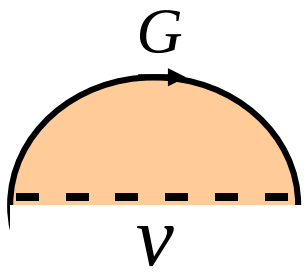
The GW approximation for the self-energy

Lars Hedin, *Phys. Rev.* **139,** A796 (1965)

Hartree-Fock approximation:

$$\Sigma_x(r,r') = iG(r,r';t)\,v(r-r')$$

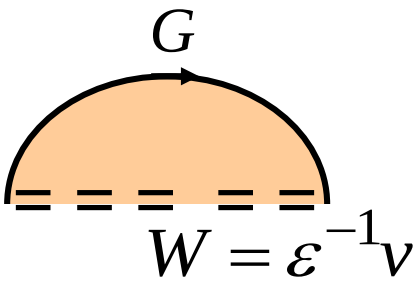
$$= -\sum_k^{occ} \psi_k(r)\psi_k(r')\,v(r-r')$$



bare interaction

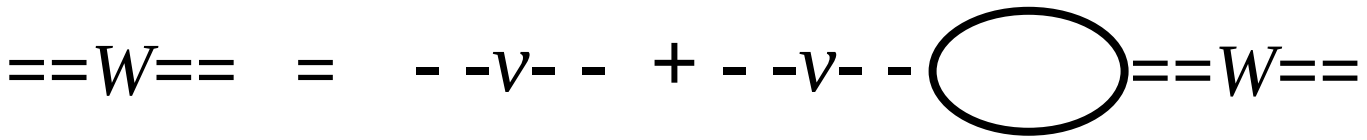
GW approximation:

$$\Sigma_{xc}^{GW}(r,r';t) = iG(r,r';t)W(r,r';t)$$

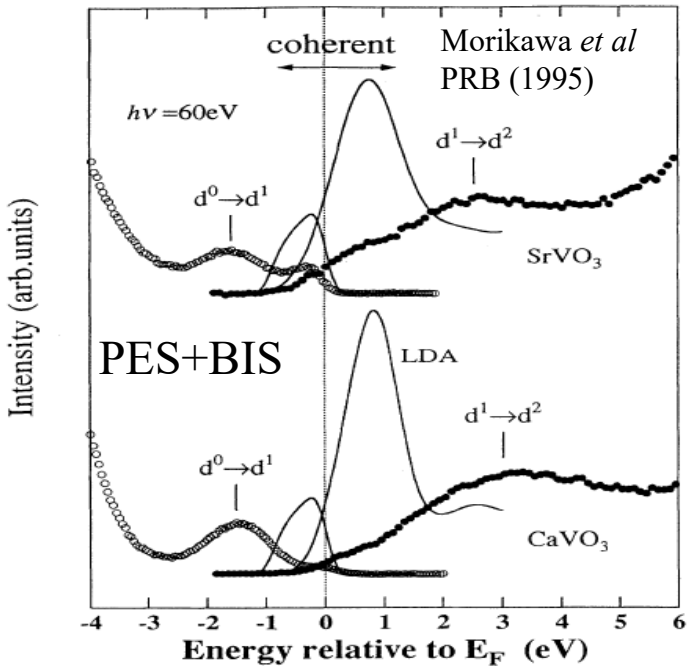


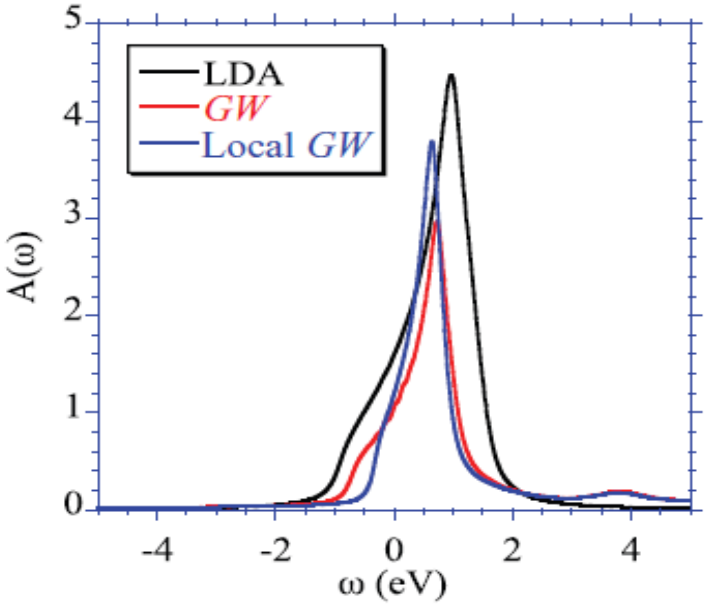
W is a screened interaction

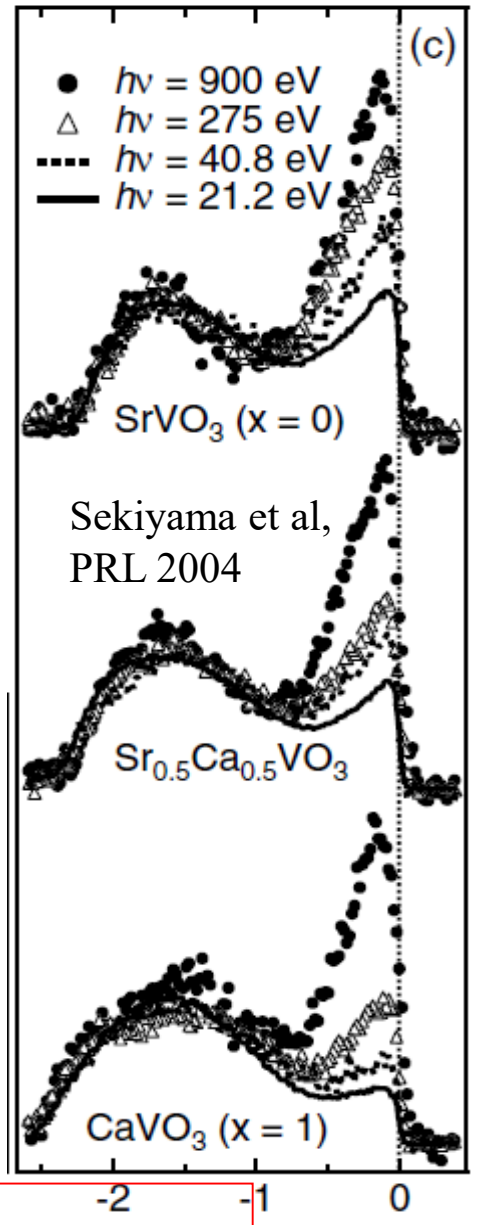
RPA

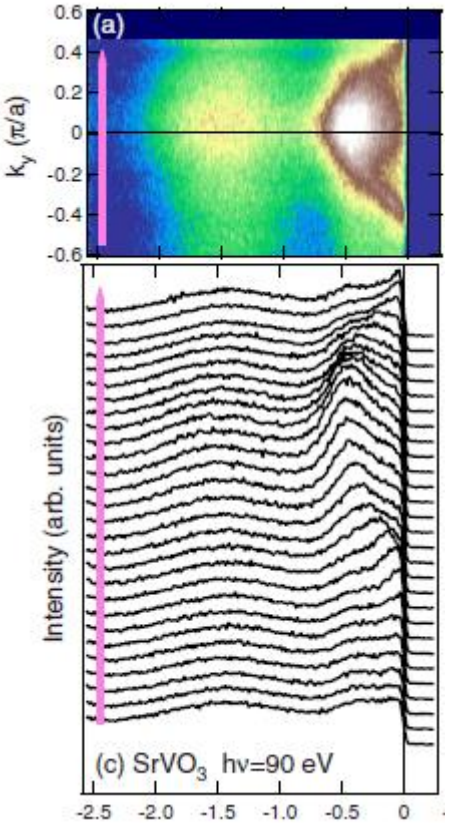


Photoemission spectra of SrVO3





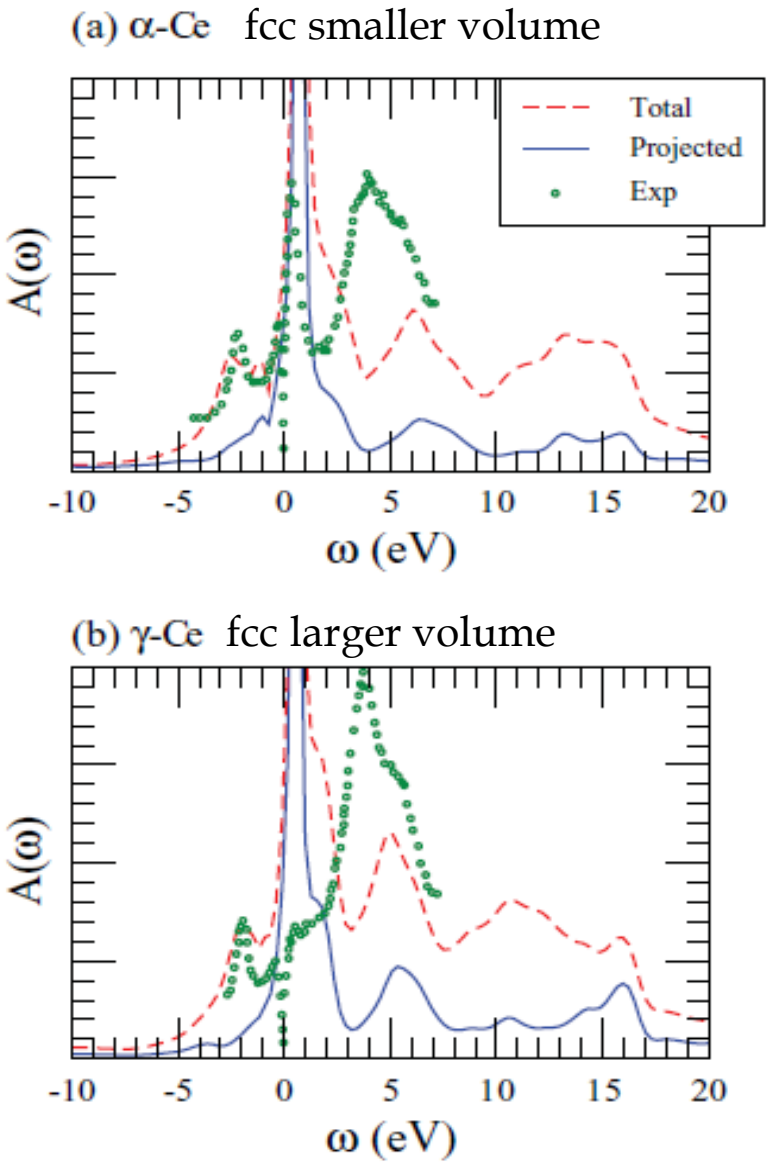




Yoshida,
PRB 82, 085119 (2010)

- Too large LDA band width.
- Absence of satellites in LDA (too high binding energies in GW).

Failure of the *GW* approximation in strongly correlated systems: The case of α–γ transition in Cerium



Sakuma, PRB 86, 245126 (2012)

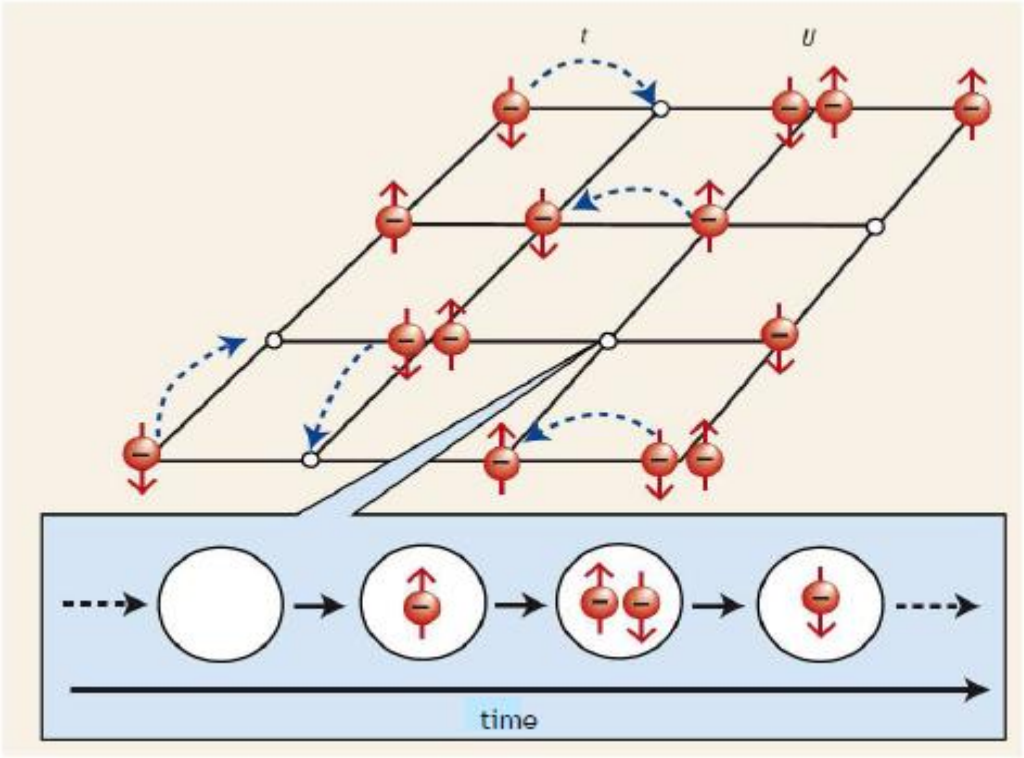
The α–γ transition in Ce is isostructural (fcc). γ phase has a larger volume than α phase.

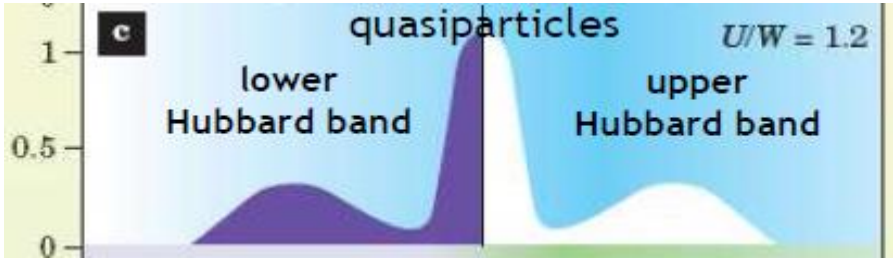
Experimental spectra:
drastic reduction in the QP weight in going from the α–γ phase.

GW spectra: little change.

Dynamical Mean-Field Theory

Georges, *Rev. Mod. Phys.* ***68***, *13 (1996)*





Taken from Vollhardt's talk

Kotliar and Vollhardt,
Physics Today (March 2004)

May be thought of as
the Anderson impurity model
with self-consistent hybridisation

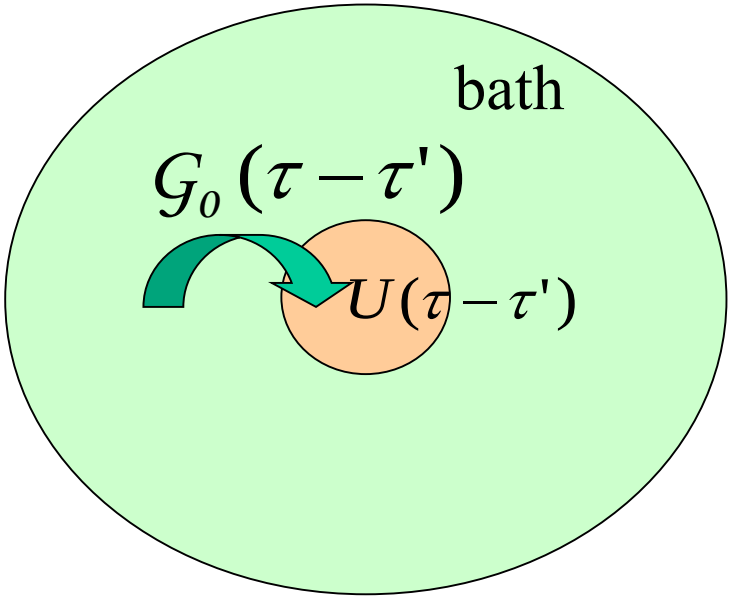
$$H = \sum_k \varepsilon_k c_k^+ c_k + \varepsilon_f c_f^+ c_f + U n_{f\uparrow} n_{f\downarrow} + \sum_k (V_k c_k c_f^+ + c.c.)$$

Effective dynamics for an impurity problem

The Coulomb interaction is fully taken into account in one site (impurity), the rest of the sites (medium) is treated as an effective field

$$S_{eff} = -\iint d\tau d\tau' \sum_{\sigma} c_{\sigma}^{+} \mathcal{G}_0^{-1}(\tau - \tau') c_{\sigma}(\tau') + \int d\tau d\tau' \, n_{\uparrow}(\tau) U(\tau, \tau') n_{\downarrow}(\tau')$$

with the dynamical mean-field $\mathcal{G}_0^{-1}$



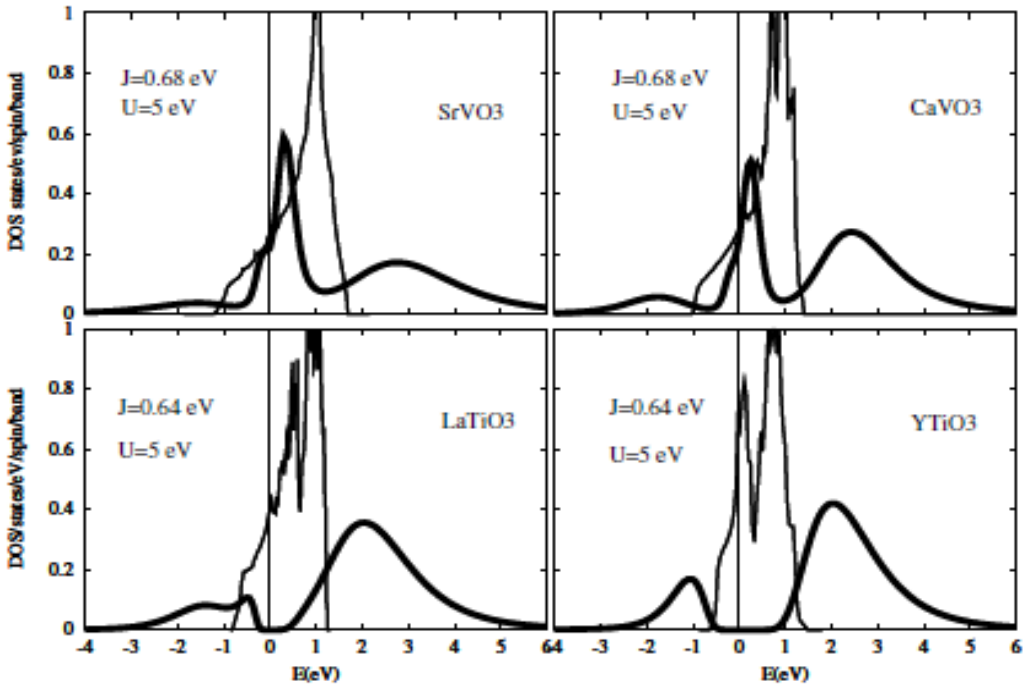
Self-consistency condition:

$$G_{loc} = G_{imp}$$

$$G_{loc} = \sum_k \frac{1}{\omega - \varepsilon_k - \Sigma_{imp}(\omega)}$$

Solve impurity problem using continuous-time QMC
Werner and Millis (PRL 2007, 2010)

SrVO3 spectra from Dynamical Mean-Field Theory (DMFT)



metallic

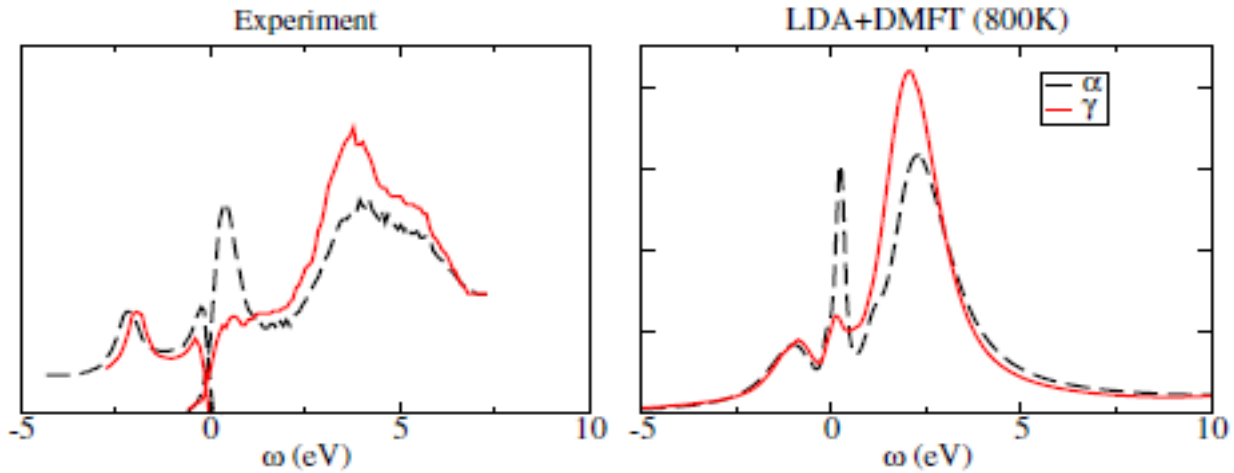
insulating

FIG. 3. DMFT spectral function at $T = 770$ K (thick line) and LDA DOS (thin line). $\mu \equiv 0$.

Pavarini, PRL 92, 176403 (2004)

Qualitatively DMFT gives the correct description of the spectral functions.

Cerium DOS in Dynamical Mean-Field Theory (DMFT)



Amadon , Biermann, Georges, and FA, PRL 96, 066402 (2006)

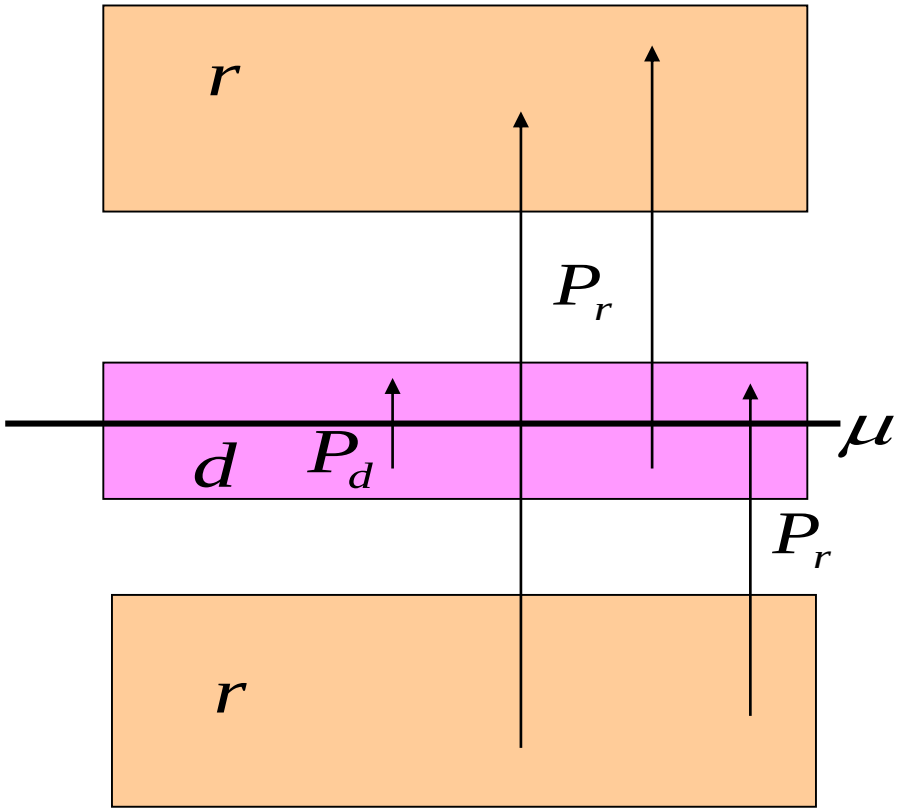
Qualitatively, DMFT gives the correct description of the spectral function in the $\alpha - \gamma$ isostructural transition.

Fundamental problems in DMFT

- Ad hoc U
- The self-energy is local (onsite).
- Double-counting problem

Constrained RPA (cRPA): Determining the Hubbard U from first-principles

Polarisation: $P = P_d + P_r$

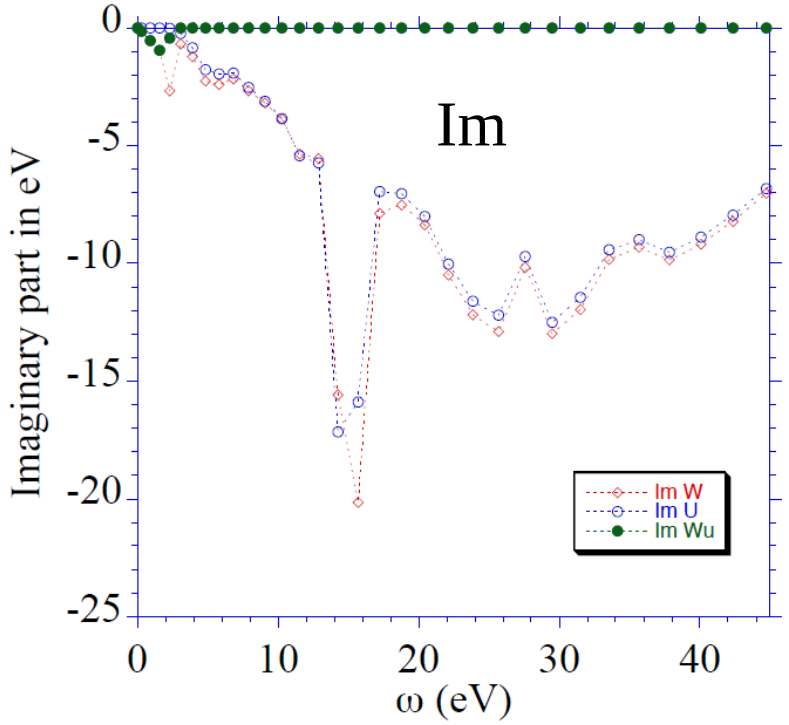


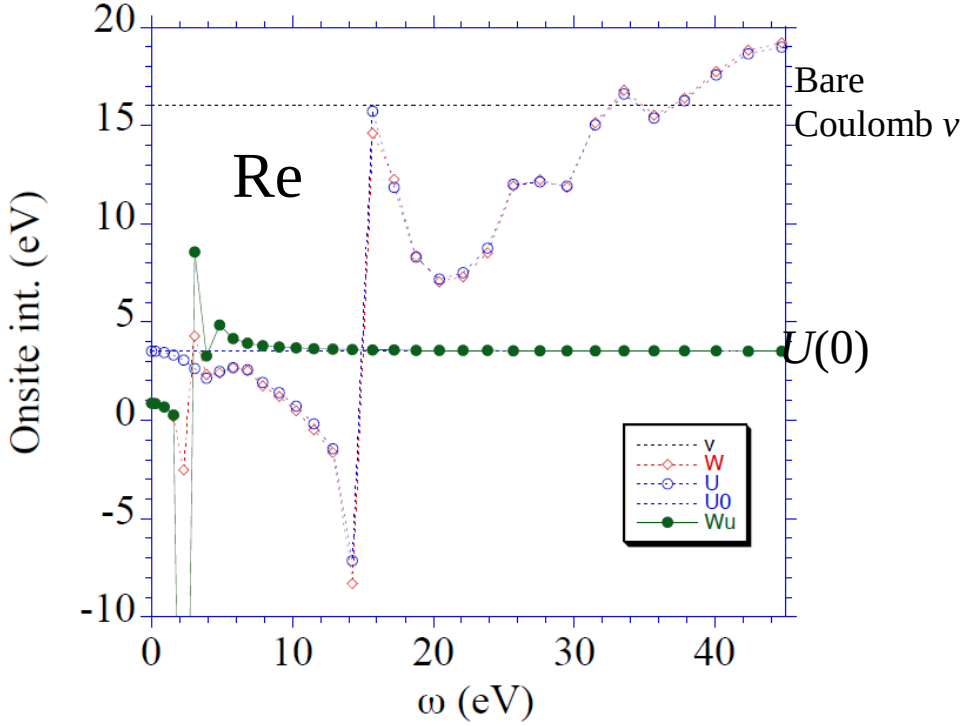
$$W = U + UP_dW$$

$$U = v + vP_rU$$

PRB 70, 195104 (2004)

Dynamic *U* in SrVO3

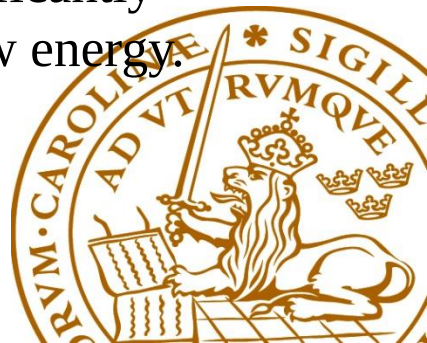




The screened interaction of the static *U* (green circles) differs significantly from the screened interaction of the dynamic *U* (red=*W)* even at low energy.

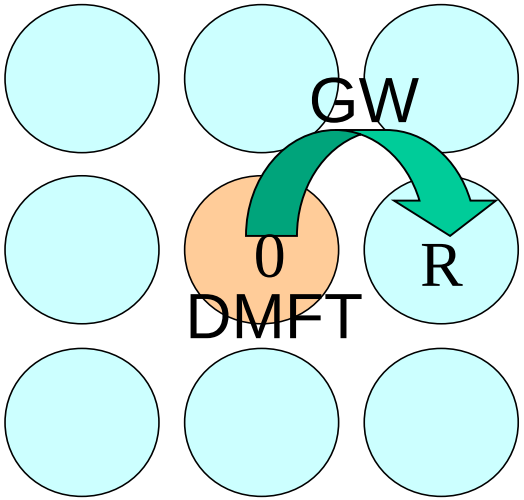
$$W(\omega) = \frac{U(\omega)}{1 - U(\omega)P_d(\omega)}$$

$$W_U(\omega) = \frac{U(0)}{1 - U(0)P_d(\omega)}$$



Combine long-range and short-range self-energies: Ab-initio *GW* + Dynamical Mean-Field Theory (DMFT)

Biermann, FA, and Georges, PRL **90**, 86402 (2003)
Sun and Kotliar, PRB **66**, 085120 (2002).



$$\Sigma_{00}(\omega) = \Sigma_{00}^{DMFT}(\omega)$$

$$\Sigma_{0R}(\omega) = \Sigma_{0R}^{GW}(\omega)$$

DMFT describes short-range local correlations.
GW describes long-range correlations (electron-gas)

Can be rigorously formulated by treating G and W as variables in the Luttinger-Ward functional

Almbladh, von Barth and van Leeuwen, *Int. J. Mod. Phys. B* **13**, 535 (1999)
R. Chitra and G.Kotliar, PRB 63, 115110 (2001)

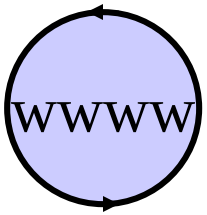
$$\Gamma[G,W] = tr\ln G - tr[G/G_0 - 1] - \frac{1}{2}tr\ln W + \frac{1}{2}tr[W/V - 1] + \Psi[G,W]$$

General Framework

Generalized Luttinger-Ward free-energy functional

$$\Gamma[G,W] = tr\ln G - tr[G/G_0 - 1] - \frac{1}{2} tr\ln W + \frac{1}{2} tr[W/V - 1] + \Psi[G,W]$$

$$\Psi_{GW}[G,W] = -\frac{1}{2} trGWG = -\frac{1}{2}$$



$$\frac{\delta\Gamma}{\delta G} = 0 \rightarrow G^{-1} = G_0^{-1} - \Sigma, \qquad \Sigma = \frac{\delta\Psi}{\delta G}$$

$$\frac{\delta\Gamma}{\delta W} = 0 \rightarrow W^{-1} = V^{-1} - P, \qquad P = 2\frac{\delta\Psi}{\delta W}$$

Almbladh, von Barth and van Leeuwen, *Int. J. Mod. Phys. B* **13**, 535 (1999)

Chitra and Kotliar, *Phys. Rev. B* **63,** 115110 (2001)

Approximation for Ψ

$$\Psi = \Psi_{GW}^{off-site}[G^{RR'}, W^{RR'}] + \Psi_{imp}^{on-site}[G^{RR}, W^{RR}]$$

$$\frac{\delta\Psi}{\delta G} = \Sigma, \qquad \frac{\delta\Psi}{\delta W} = 2P$$

$$\Sigma(k, i\nu) = \Sigma_{GW}(k, i\nu) - \sum_k \Sigma_{GW}(k, i\nu) + \Sigma_{imp}(k, i\nu)$$

$$P(k, i\omega) = P_{GW}(k, i\omega) - \sum_k P_{GW}(k, i\omega) + P_{imp}(k, i\omega)$$

At self-consistency

$$\sum_k \Sigma_{GW}(k, \tau) = -G_{imp}(\tau) W_{imp}(\tau)$$

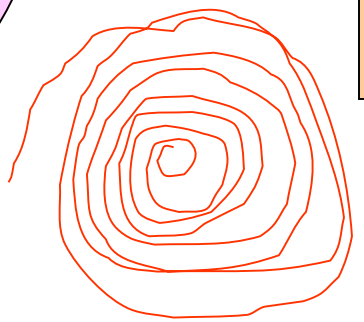
GW+DMFT self-consistency loop

Impurity: given Weiss field $\mathcal{G}$ and $U(\omega)$

$$\Sigma_{imp} = \mathcal{G}^{-1} - G_{imp}^{-1}$$
$$W_{imp} = U - U\chi_{imp}U$$
$$P_{imp} = U^{-1} - W_{imp}^{-1}$$

New Weiss field $\mathcal{G}$ and $U(\omega)$

$$\mathcal{G}^{-1} = G_{loc}^{-1} + \Sigma_{imp}$$
$$U^{-1} = W_{loc}^{-1} + P_{imp}$$

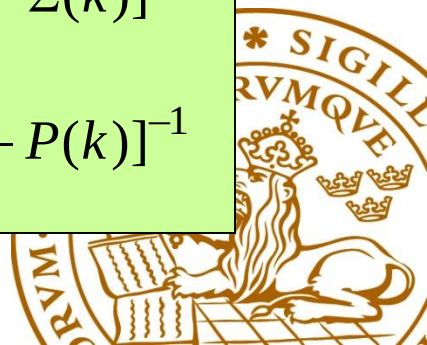


Combine Σ_{GW} and Σ_{imp}

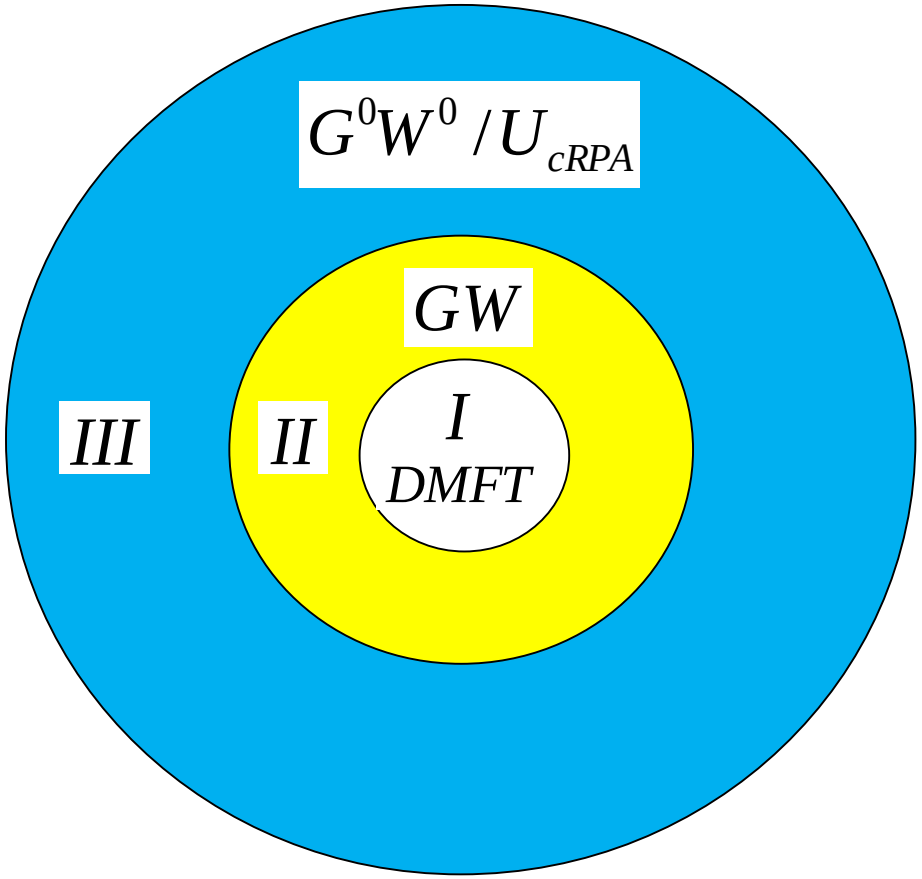
$$\Sigma(k) = \Sigma_{GW}(k) - \sum_q \Sigma_{GW}(q) + \Sigma_{imp}$$
$$P(k) = P_{GW}(k) - \sum_q P_{GW}(q) + P_{imp}$$

Check self-consistency:
G_{loc}=G_{imp}? W_{loc}=W_{imp}?

$$G_{loc} = \sum_k [G_0^{-1}(k) - \Sigma(k)]^{-1}$$
$$W_{loc} = \sum_k [V^{-1}(k) - P(k)]^{-1}$$



Multi-tier self-consistent *GW*+DMFT



$$G^{-1} = G_0^{-1} - [\Sigma_{G^0W^0}^{I+II+III} - \Sigma_{G^0W^0}^{I+II}] \quad \text{Tier } III$$

self-consistent

$$-[\Sigma_{GW}^{I+II} - \Sigma_{GW}^{I}] \quad \text{Tier } II$$

$$-\Sigma_{DMFT}^{I} \quad \text{Tier } I$$

$$G_0^{-1} = G_{LDA}^{-1} - v_{xc}$$

At self-consistency

$$\sum_k \Sigma_{GW}(k,\tau) = -G_{imp}(\tau)W_{imp}(\tau)$$

Previous works on SrVO3 within GW+DMFT

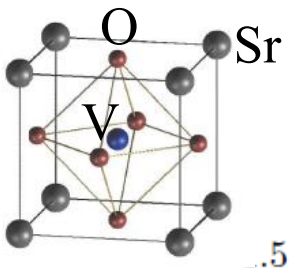
Tomczak et al, Europhys. Lett. 100, 67001 (2012)

Taranto et al, PRB 88, 165119 (2013)

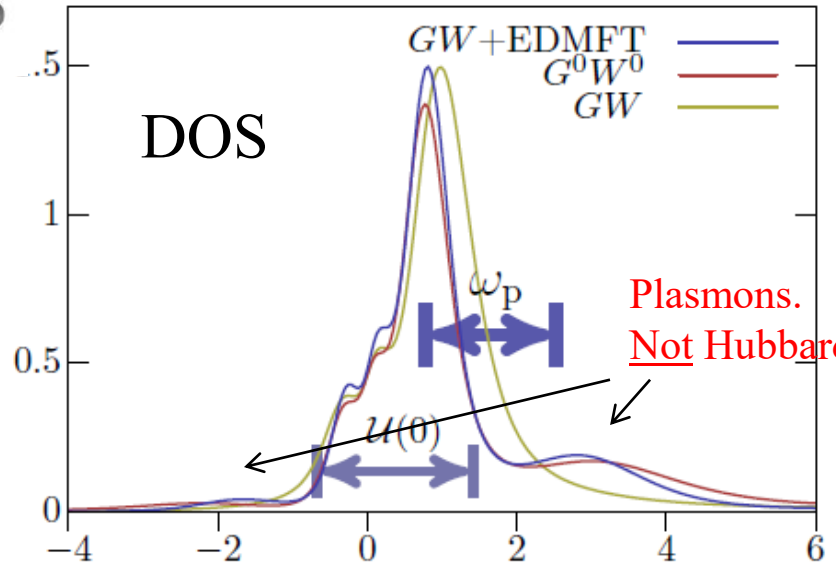
Sakuma et al, PRB 88, 235110 (2013)

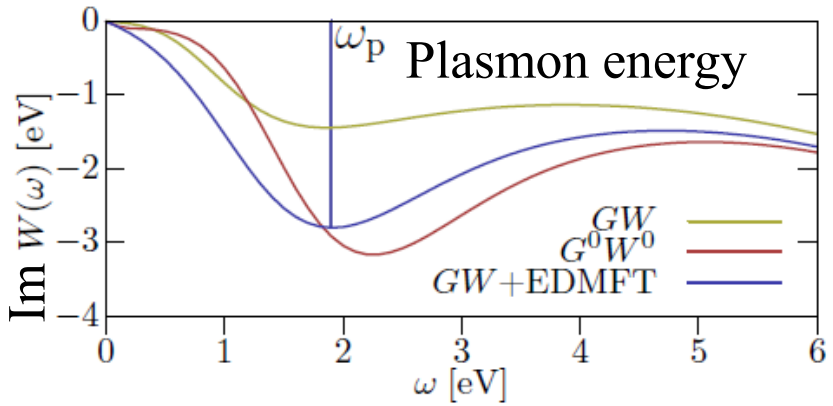
Tomczak et al, PRB 90, 165138 (2014)

Not self-consistent

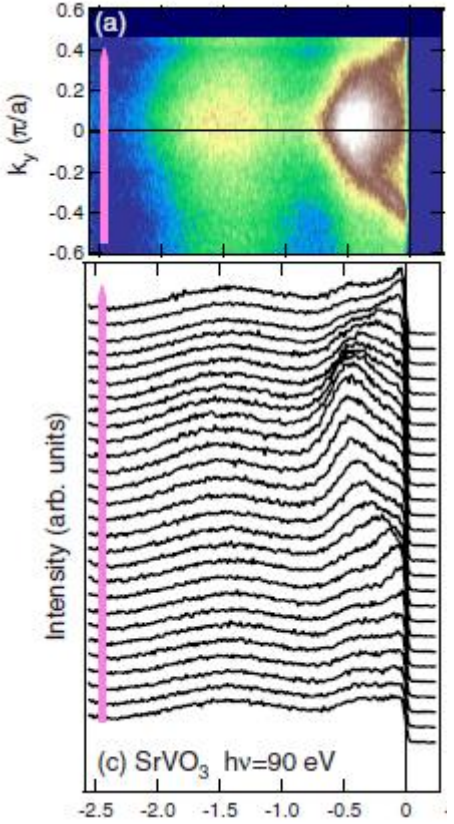


Self-consistent *GW*+DMFT: SrVO3 photoemission spectrum





ARPES

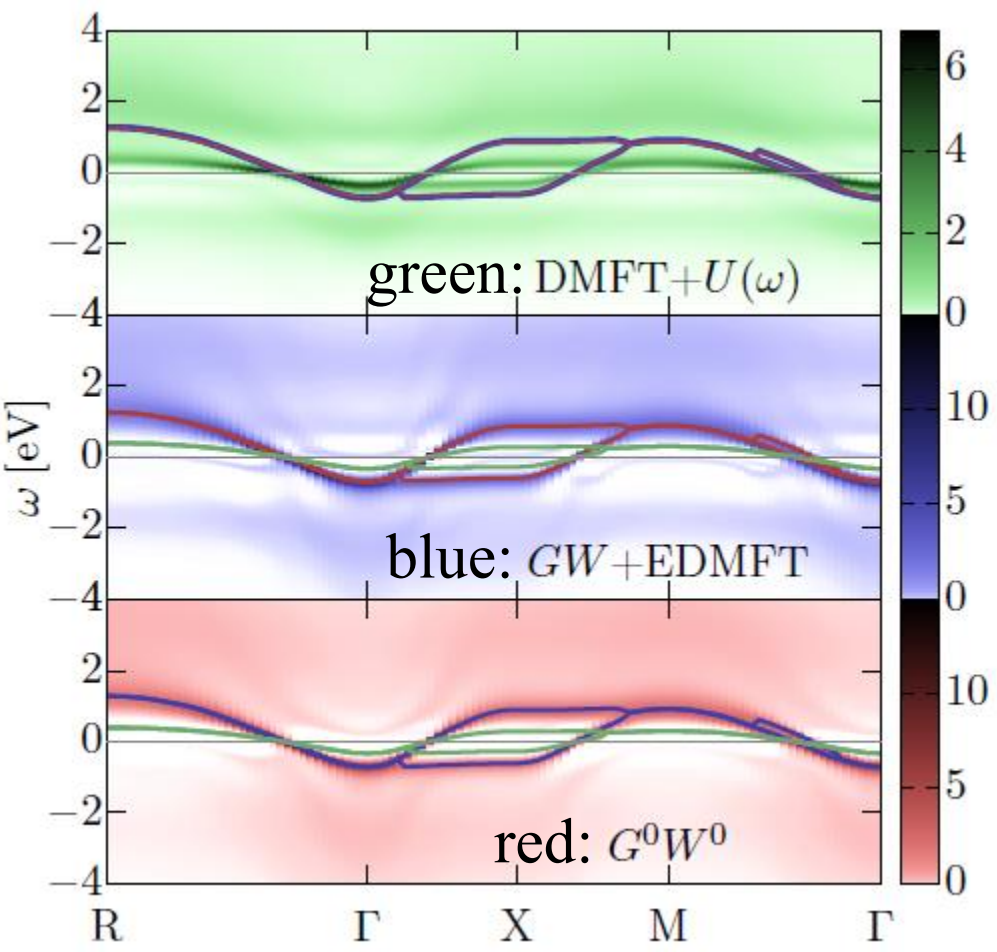


Yoshida et al, PRB 82, 085119 (2010)

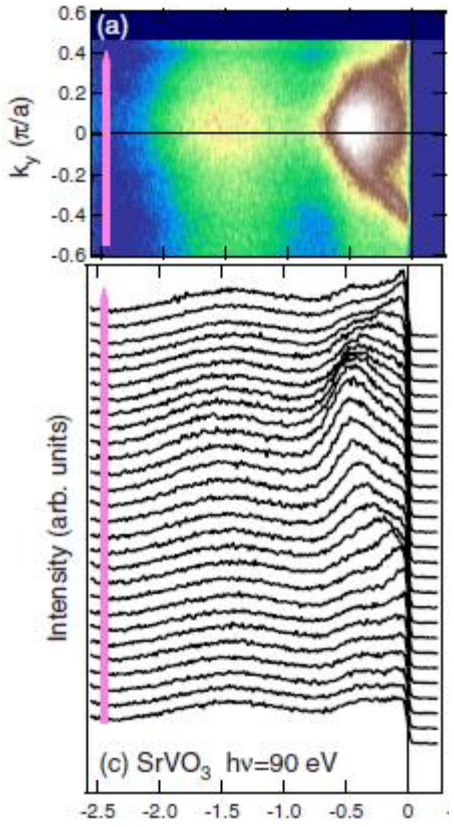
U(0) is *too small* to produce Hubbard bands.
Self-consistency reduces U from 3.5 to 2.2 eV.

Boehnke, Nilsson, FA, and Werner, PRB 94, 201106(R) (2016)

Quasiparticle dispersion of SrVO3



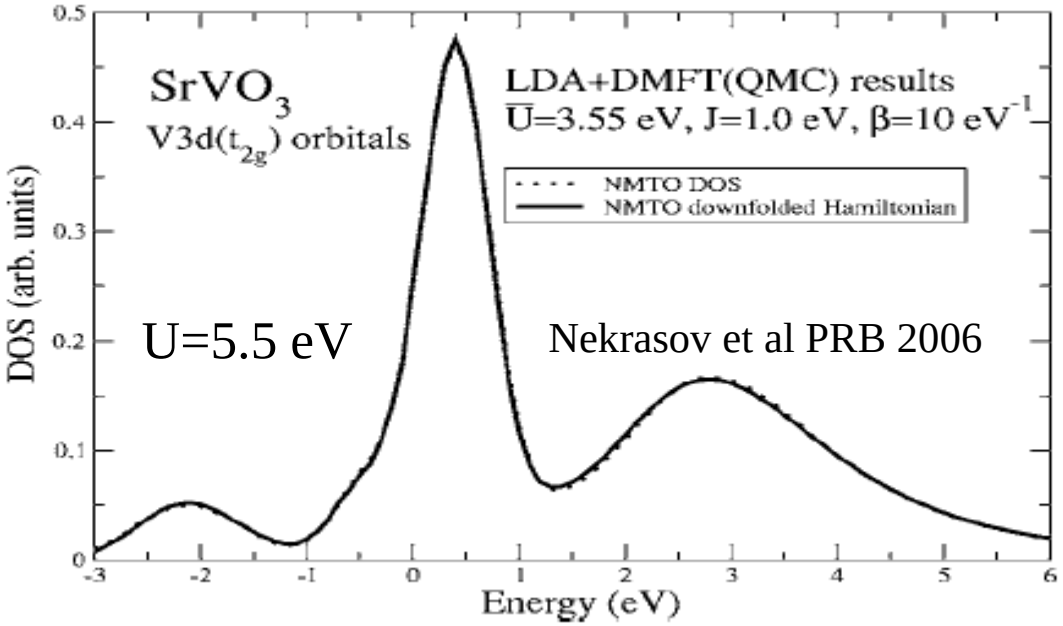
ARPES



Yoshida et al, PRB 82, 085119 (2010)

Importance of non-local self-energy
in widening the band dispersion.

Comparison with LDA+DMFT with static U

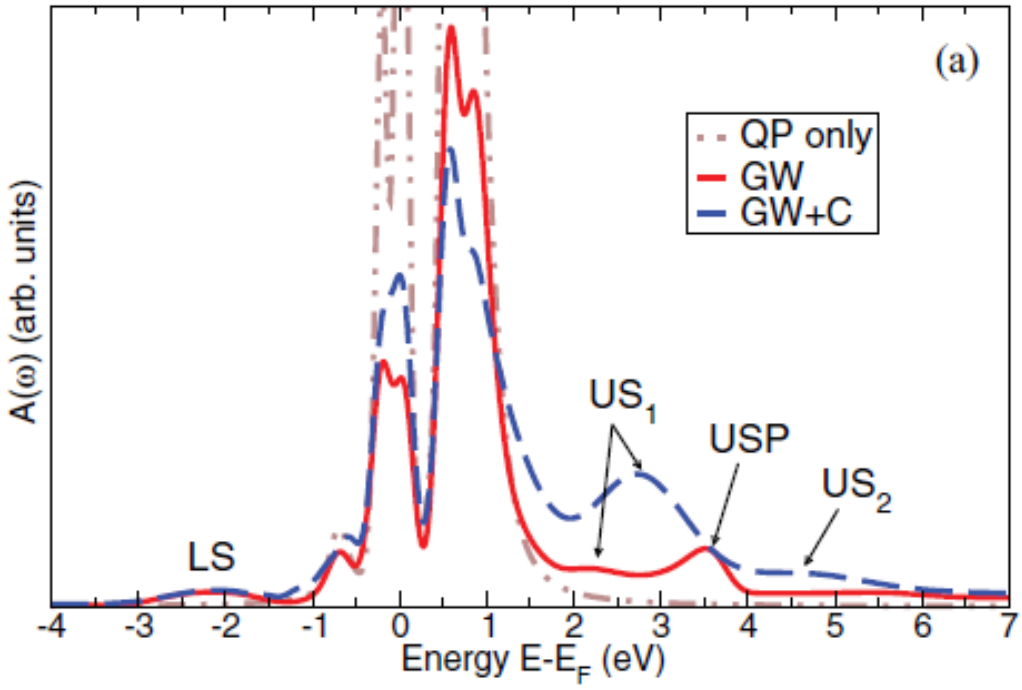


Static U:

Need a large U to get the right band narrowing but the separation of the Hubbard bands becomes too large.

Comparison with the cumulant expansion

Gatti and Guzzo, PHYSICAL REVIEW B **87**, 155147 (2013)



Summary

- Self-consistency reduces impurity U (from 3.5 eV to 2.2 eV in SrVO3)
- Re-interpretation satellite features in SrVO3 as plasmons.
- Vertex corrections via DMFT reduce RPA plasmon energy.
- Importance of non-local vertex corrections in reducing band width.
- GW+DMFT offers a promising method for calculating the electronic structure and spectral functions of strongly correlated materials from first-principles.