

From LDA+DMFT to GW+DMFT

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From LDA+DMFT to GW+DMFT

or

Open questions in dynamical mean field
approaches to realistic materials

Silke Biermann

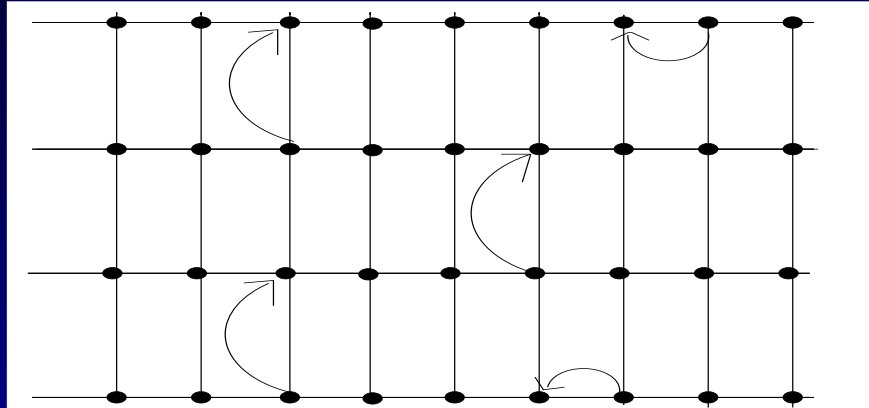
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Outline

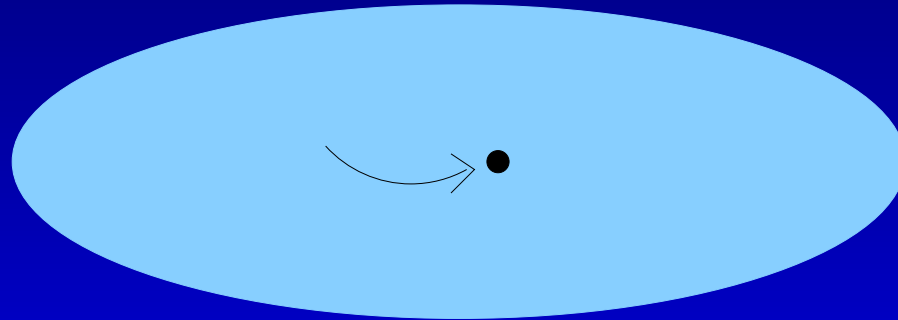
- Reminder: impurity physics = the heart of DMFT
- Discussion of some practical issues in the context of “LDA+DMFT”
(impurity solvers, analytical continuation ...)
- Which conceptual points to be aware of?
(correlated vs. uncorrelated orbitals, Hubbard-U, double counting ...)
- Some perspectives to go beyond “LDA+DMFT”
 (“GW+DMFT” ...)
*S.B., F. Aryasetiawan, A. Georges PRL 90 086402 (2003) +
cond-mat/0401653*
*F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar, S.B.,
A. I. Lichtenstein PRB 70 195104 (2004)*

Dynamical mean field theory ...

... maps a lattice problem



onto a single-site (Anderson impurity) problem



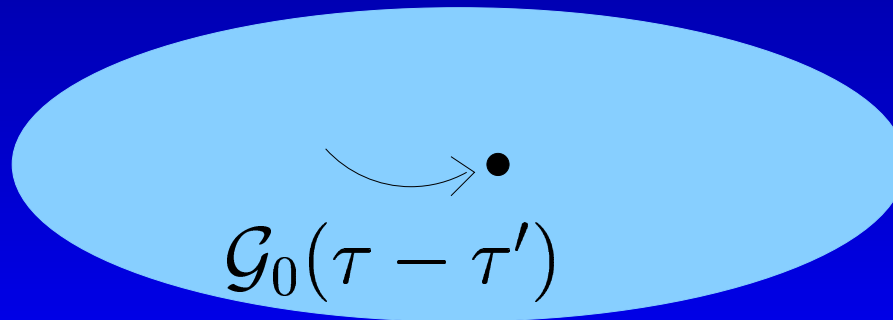
with a self-consistency condition

Effective dynamics ...

... for *single-site* problem

$$S_{eff} = - \int_0^\beta d\tau \int_0^\beta d\tau' \sum_{\sigma} c_{\sigma}^{\dagger}(\tau) \mathcal{G}_0^{-1}(\tau - \tau') c_{\sigma}(\tau') \\ + U \int_0^\beta d\tau n_{\uparrow} n_{\downarrow}$$

with the dynamical mean field $\mathcal{G}_0^{-1}(\tau - \tau')$



DMFT (contd.)

Green's function:

$$G(\tau) = -\langle \hat{T} c(\tau) c^\dagger(0) \rangle$$

Self-energy (k-independent):

$$\Sigma(i\omega) = \mathcal{G}_0^{-1}(i\omega) - G^{-1}(i\omega)$$

DMFT assumption :

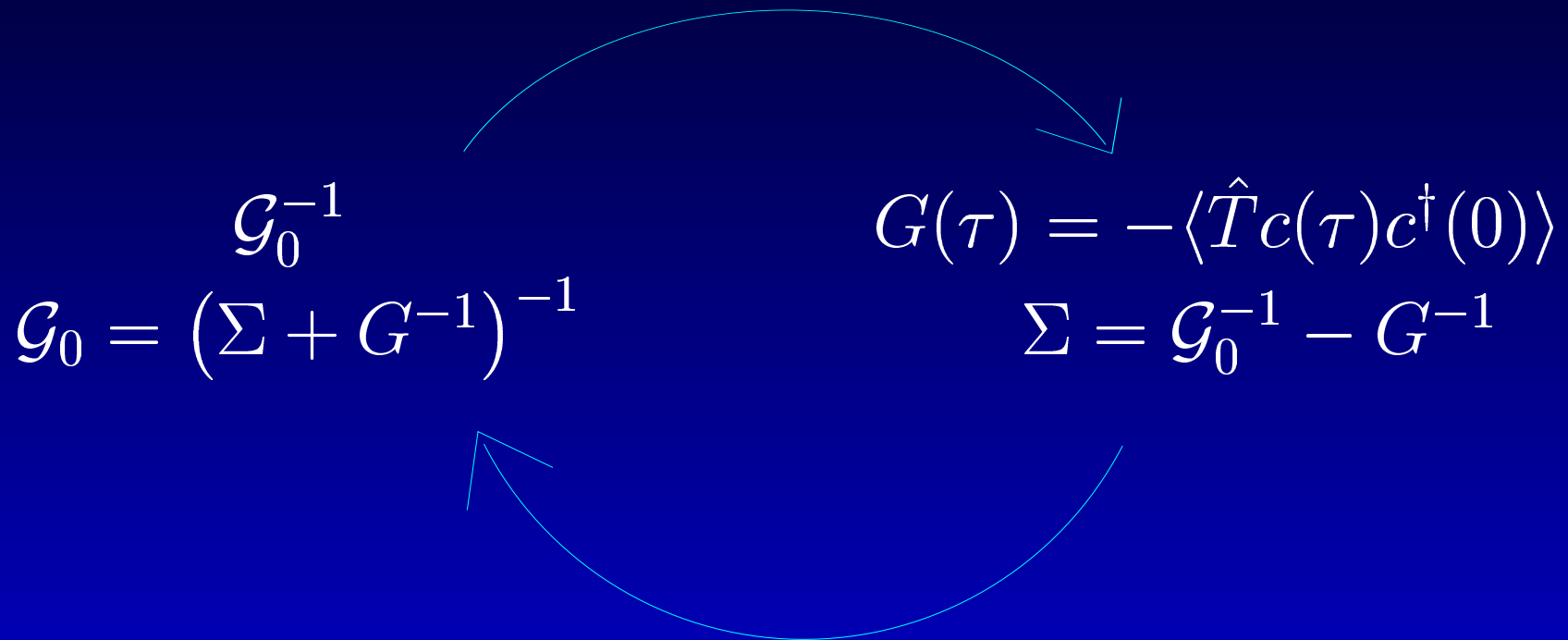
$$\Sigma_{impurity} = \Sigma^{lattice}$$

$$G_{impurity} = G_{local}^{lattice}$$

→ Self-consistency condition for \mathcal{G}_0^{-1}

The DMFT self-consistency cycle

Anderson impurity model solver



Self-consistency condition:

$$G(\omega) = \sum_k \frac{1}{\omega + \mu - \epsilon_k - \Sigma(\omega)}$$

Realistic Approach to Correlations

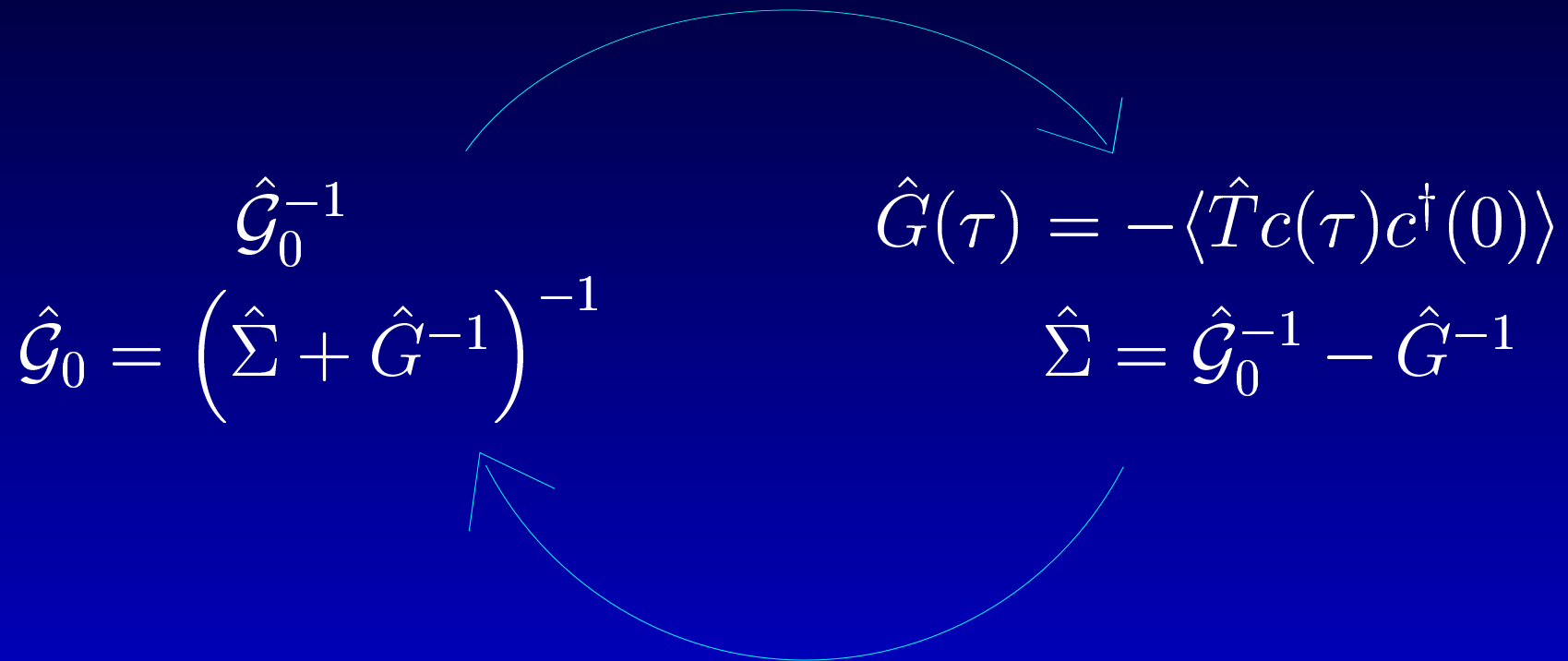
→ combine DMFT with band structure input (LDA)

$$\begin{aligned} H &= \sum_{\{im\sigma\}} (H_{im,i'm'}^{LDA} - H_{im,i'm'}^{double\ counting}) a_{im\sigma}^+ a_{i'm'\sigma} \\ &+ \frac{1}{2} \sum_{imm'\sigma \text{ (correl. orb.)}} U_{mm'}^i n_{im\sigma} n_{im'-\sigma} \\ &+ \frac{1}{2} \sum_{im \neq m' \sigma \text{ (correl. orb.)}} (U_{mm'}^i - J_{mm'}^i) n_{im\sigma} n_{im'\sigma} \end{aligned}$$

- treat within DMFT

The DMFT self-consistency cycle

Anderson impurity model solver

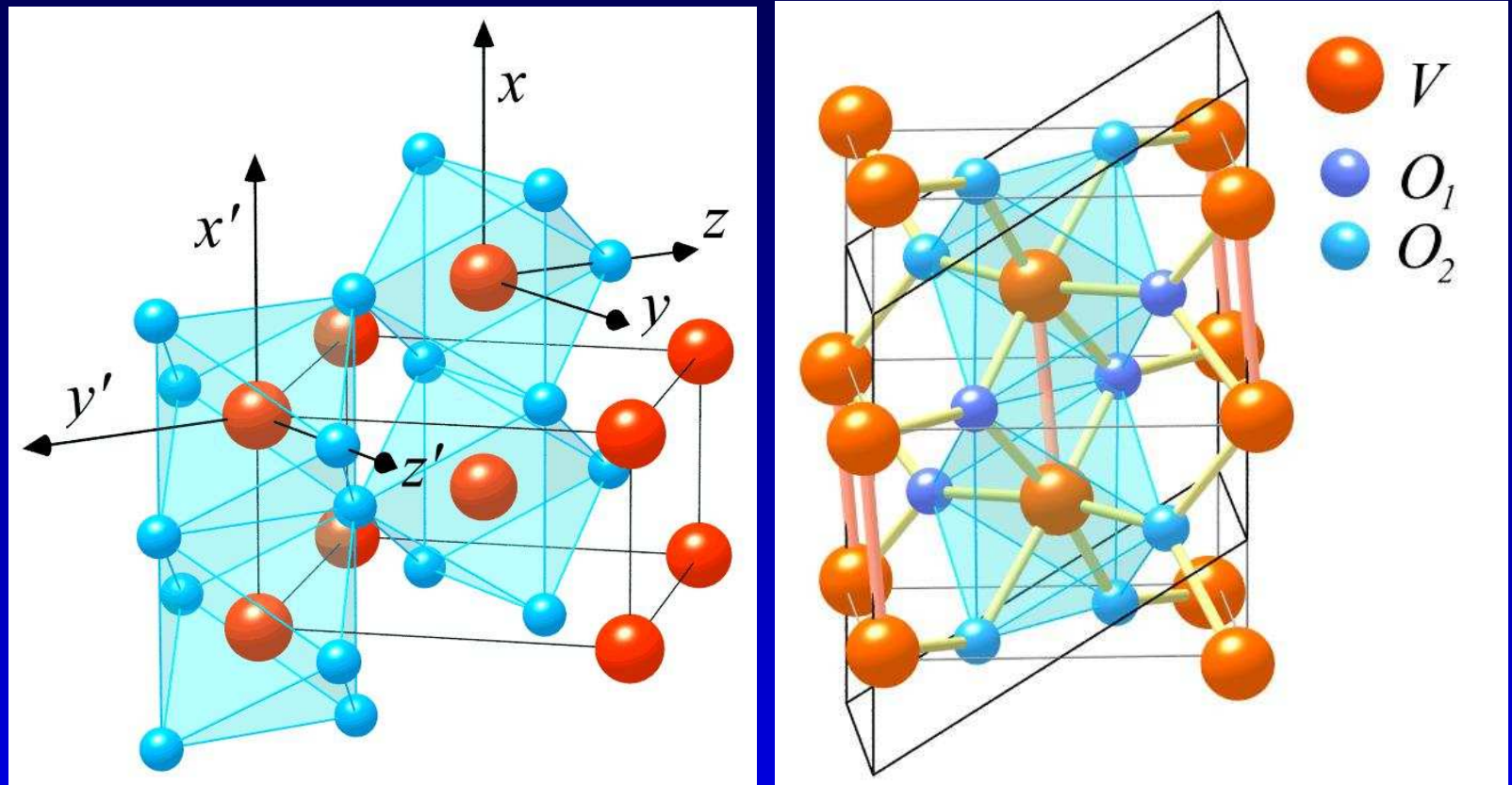


Self-consistency condition:

$$\hat{G}(\omega) = \sum_k \left(\omega + \mu - \hat{H}_o(k) - \hat{\Sigma}(\omega) \right)^{-1}$$

The example of VO_2

- High temperature rutile phase: metallic
- Low temperature monoclinic phase: insulating



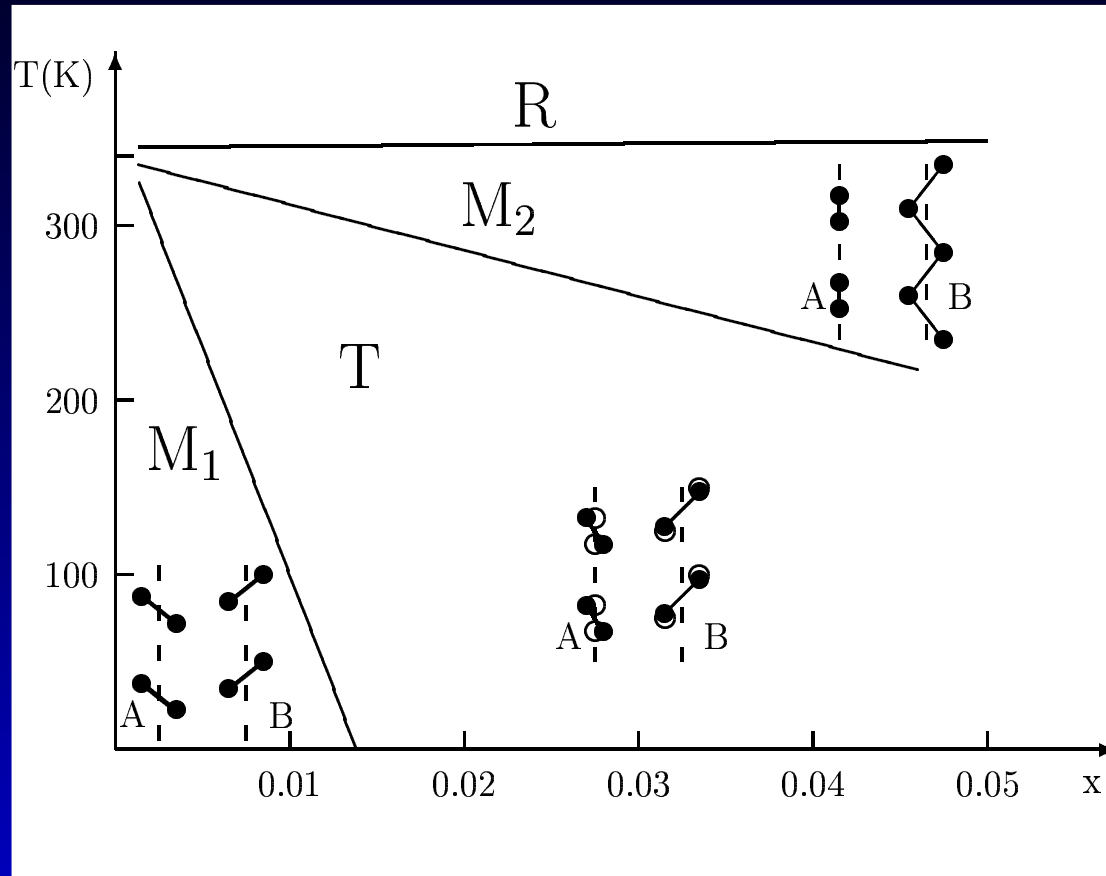
From V. Eyert

Peierls or Mott ?

What is the nature of the insulating phase ?

- LDA describes structural properties well (Wentzcovitch et al.)
→ “more band-like than correlated”
- LDA yields a gap if distortions are exaggerated (Eyert)
→ Peierls insulator ?
- Existence of “mixed” phases (M2, T) proves importance of correlations (Pouget et al.)
→ Mott-Hubbard insulator?

VO_2 mixed phases

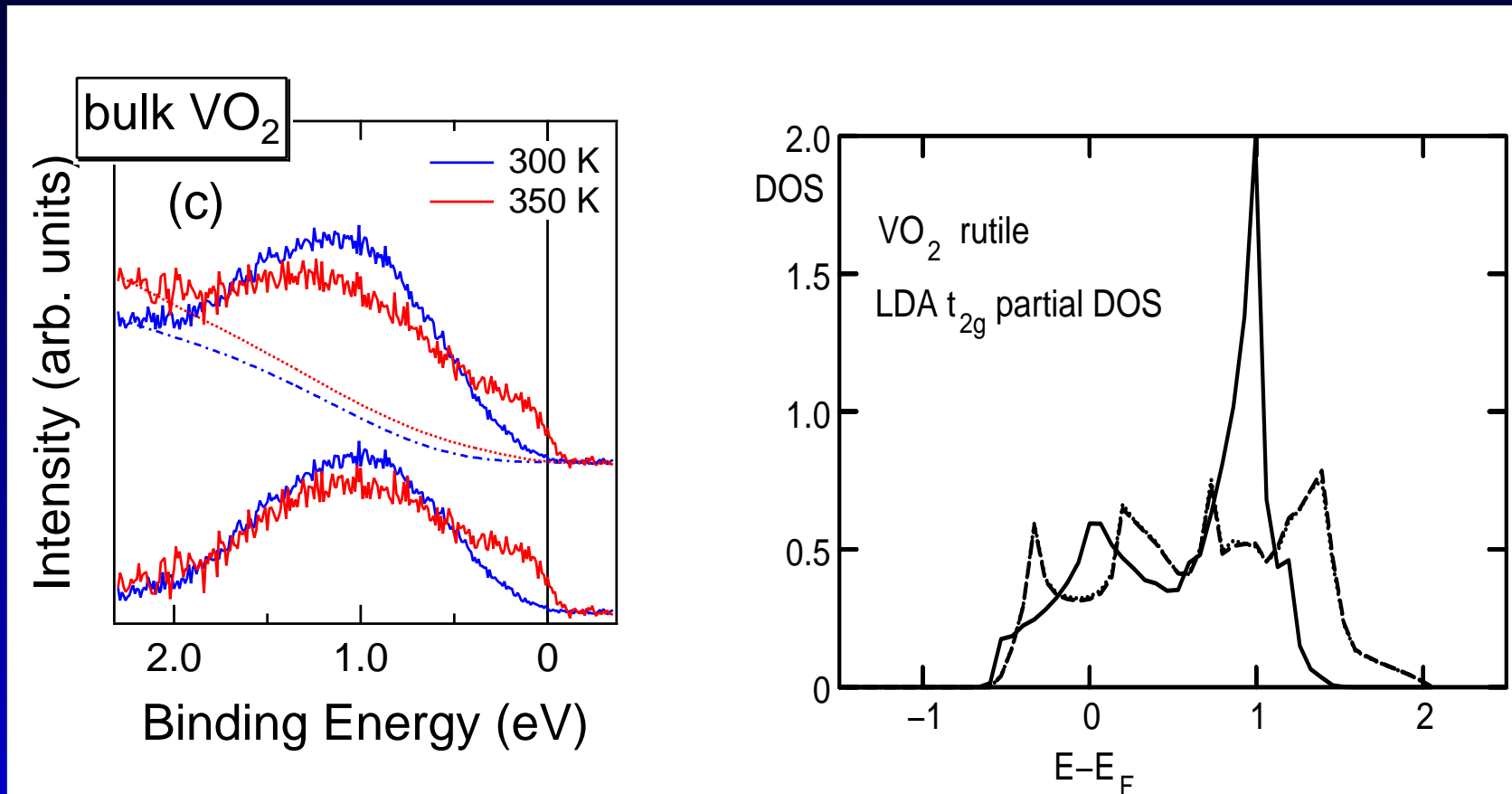


Half of the V atoms dimerize, but do not tilt
Half of the V atoms tilt, but do not dimerize

(Pouget et al.)

VO₂ rutile

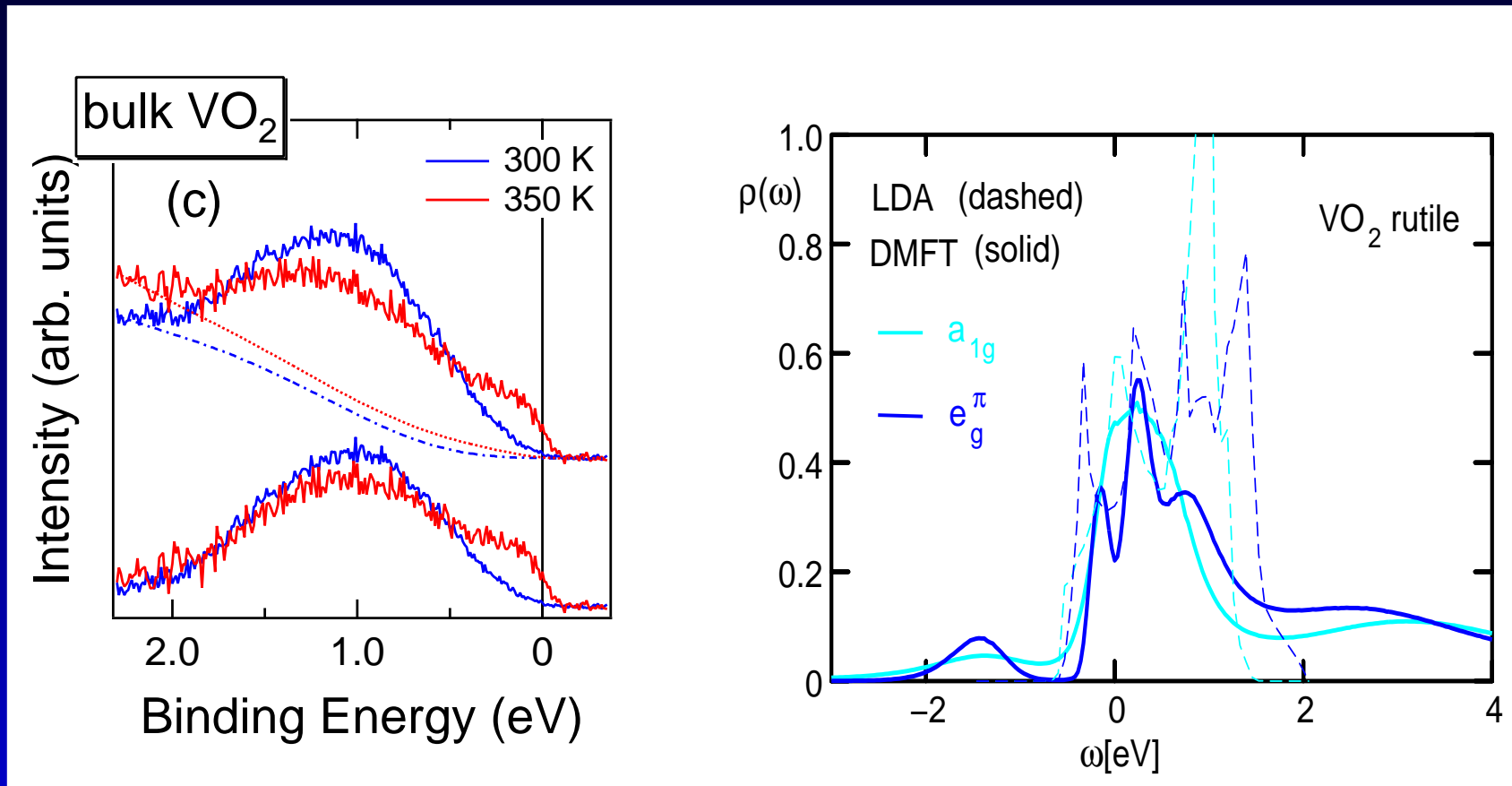
Photoemission versus LDA



(Photoemission data from Okazaki et al., 2004)

VO₂ rutile

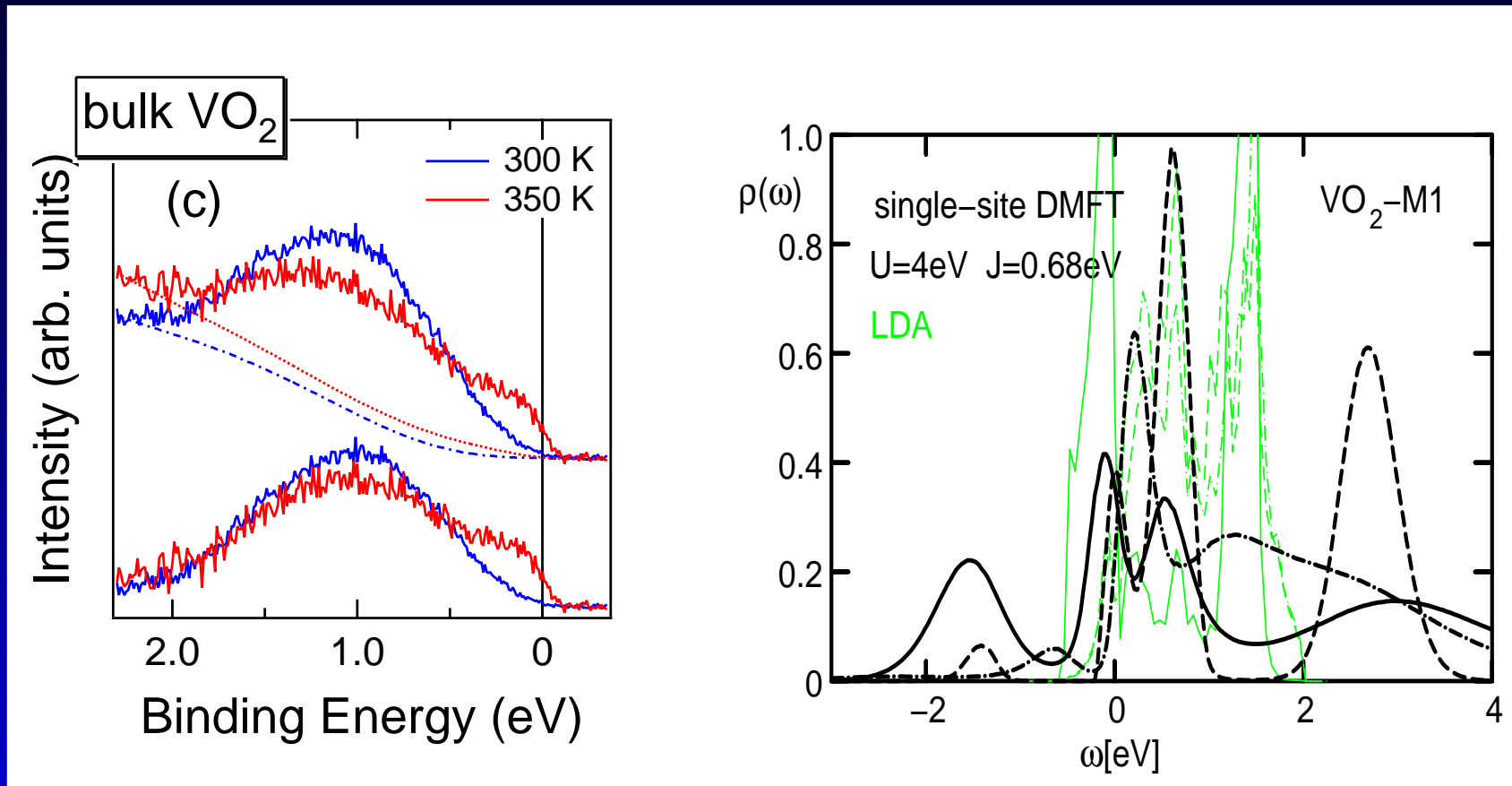
Photoemission versus LDA+DMFT



(Photoemission data : from Okazaki et al., 2004)
(cf also Liebsch et al. PRB 2005)

VO₂ monoclinic phase

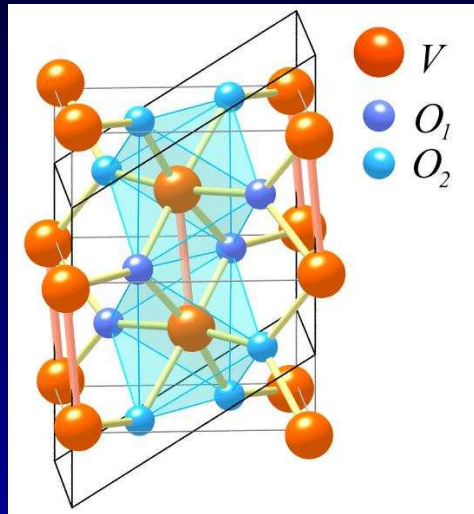
Photoemission versus LDA+DMFT



Standard LDA+DMFT not sufficient !

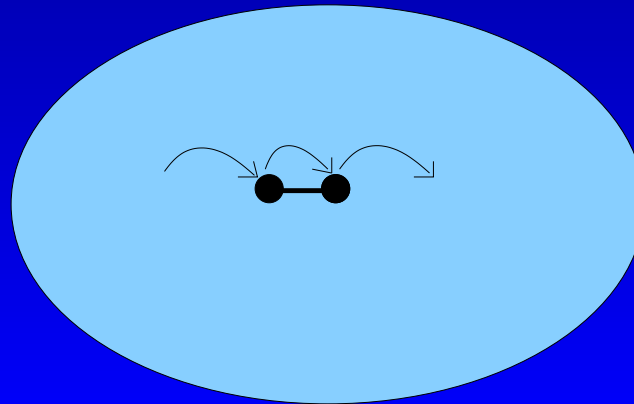
LDA+DMFT simulations ...

for the monoclinic phase have to be done with care:



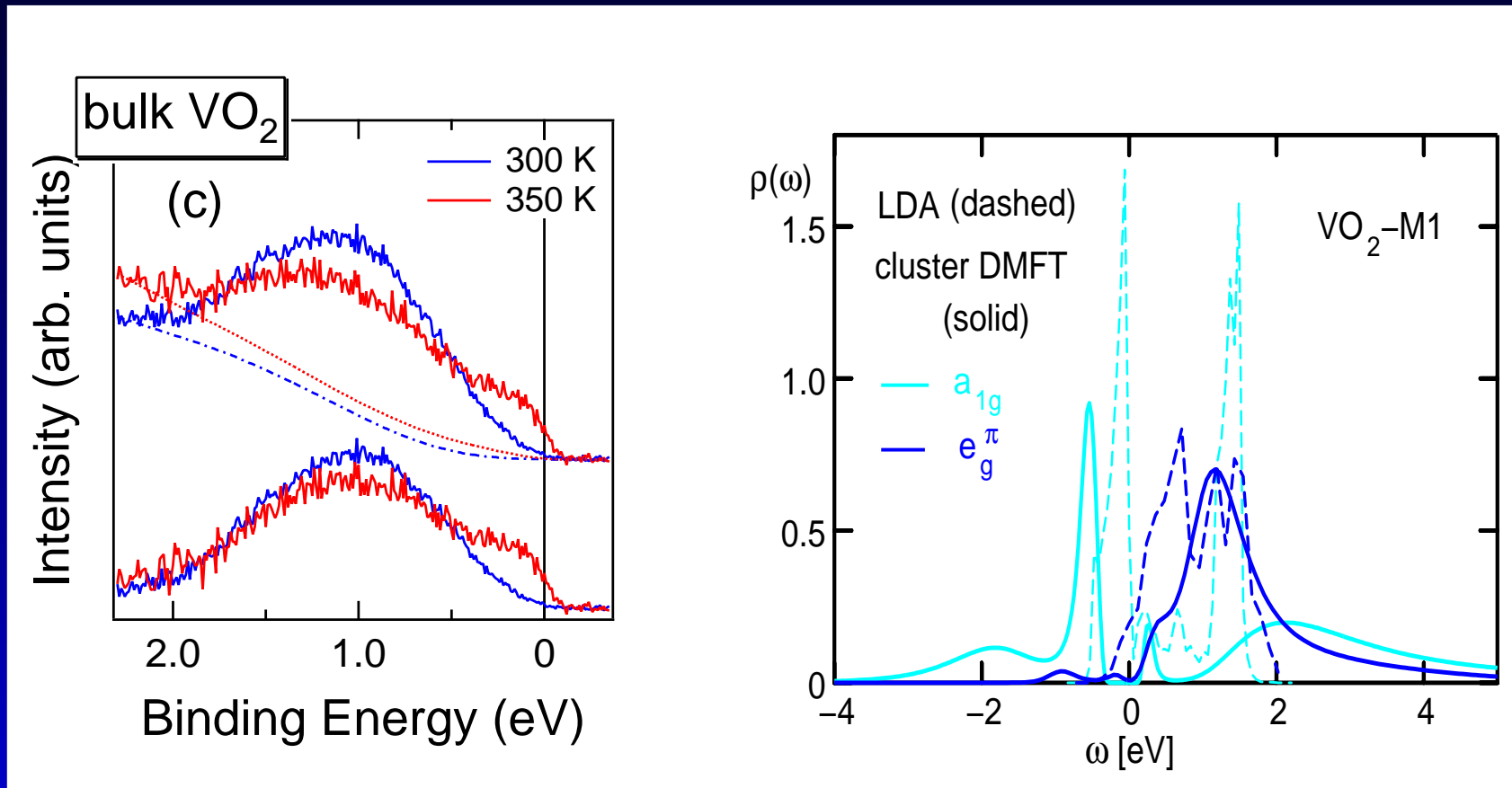
Inter-site (intra-pair)
fluctuations ?

"Cluster-DMFT" :
embed V pair in bath \rightarrow non-local self-energy !



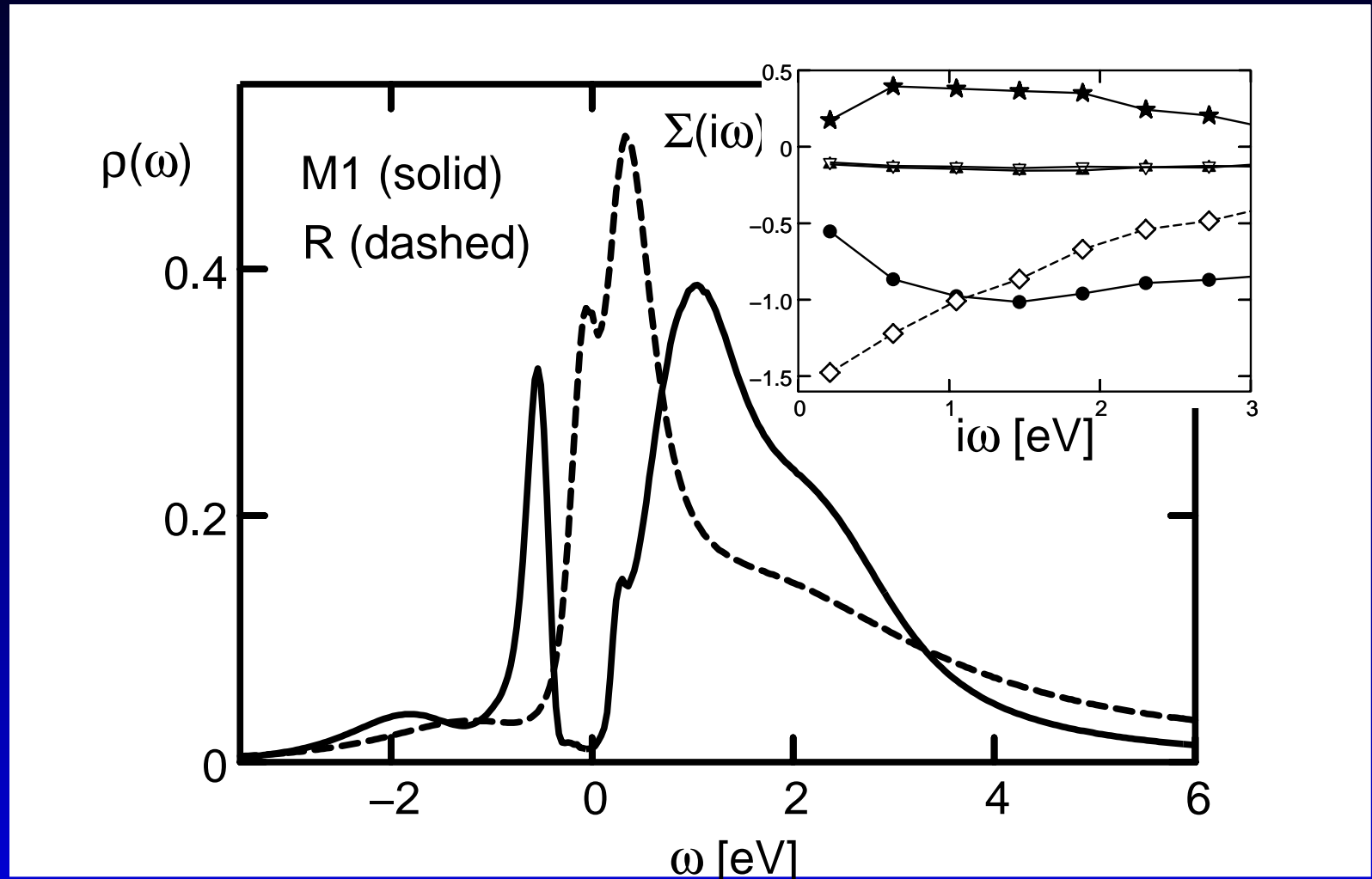
VO₂ monoclinic phase

Photoemission versus LDA+"cluster-DMFT"



Opening of a gap !

Total spectral function



rutile phase vs. monoclinic phase
self-energy does not diverge!

VO₂ monoclinic phase

- opening of the gap is not accompanied by a diverging self-energy
- "Peierls gap" ...
- ... coexists with Hubbard bands
- large intra-pair self-energy
- considerable charge transfer into a_{1g} band

is well described by LDA+"cluster-DMFT" !

Technical issues

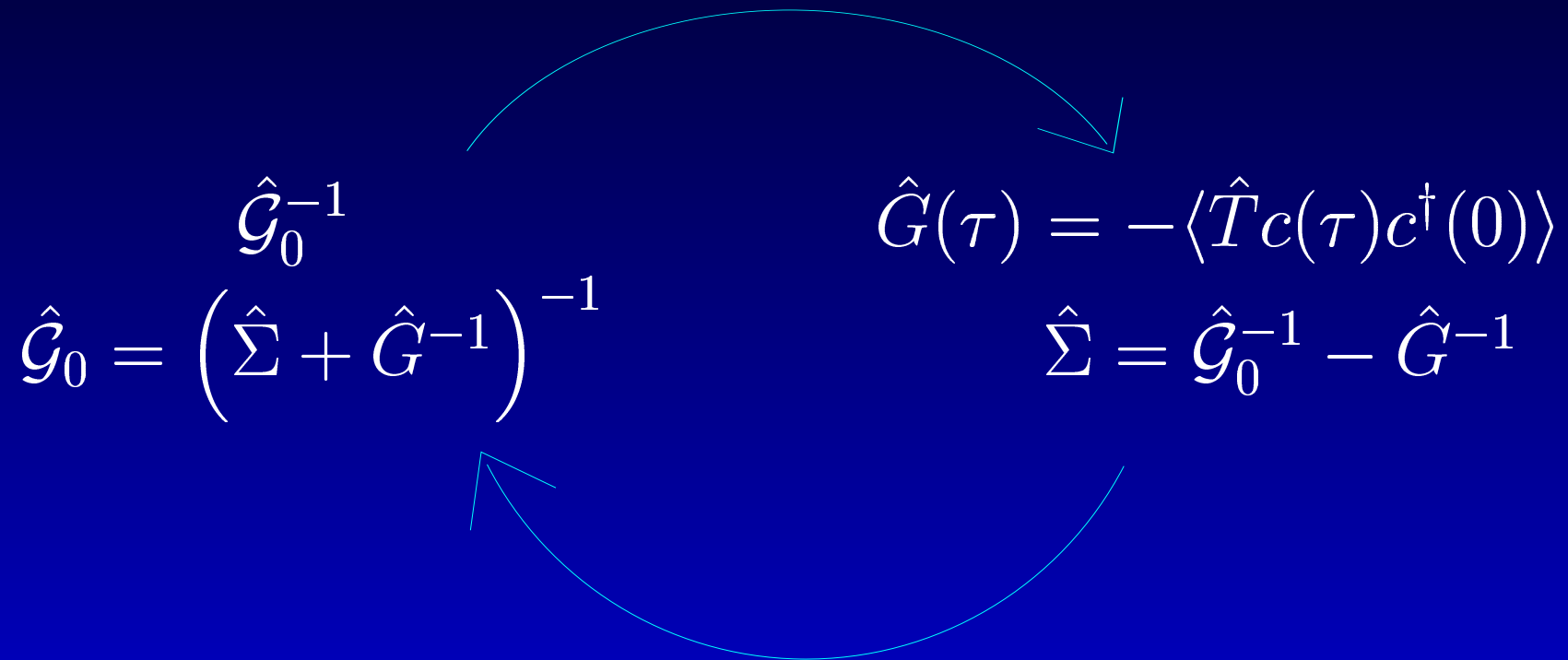
- How to solve the impurity problem?
(QMC, Hubbard approximations, NCA, IPT, DMRG, NRG, Gutzwiller)
If by QMC, then:
- Analytical continuation of imaginary frequency data?
→ Maximum Entropy method

$$G(\tau) = \int_{-\infty}^{\infty} d\omega \rho(\omega) \frac{e^{-\tau\omega}}{1 + e^{-\beta\omega}}$$

Want to obtain $\rho(\omega)$ from $G(\tau)$

The DMFT self-consistency cycle

Anderson impurity model solver



Self-consistency condition:

$$\hat{G}(i\omega) = \sum_k \left(i\omega + \mu - \hat{H}_o(k) - \hat{\Sigma}(i\omega) \right)^{-1}$$

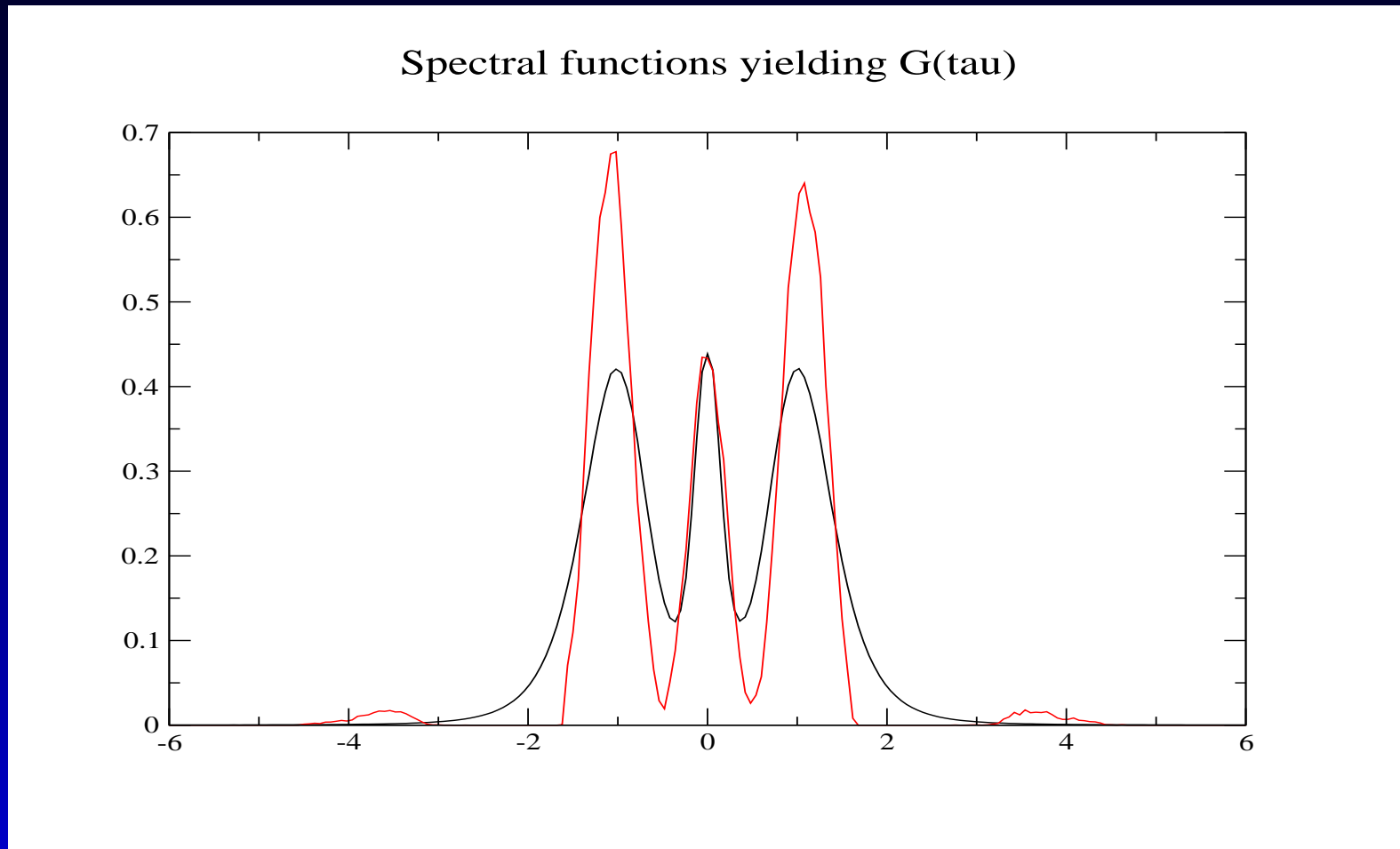
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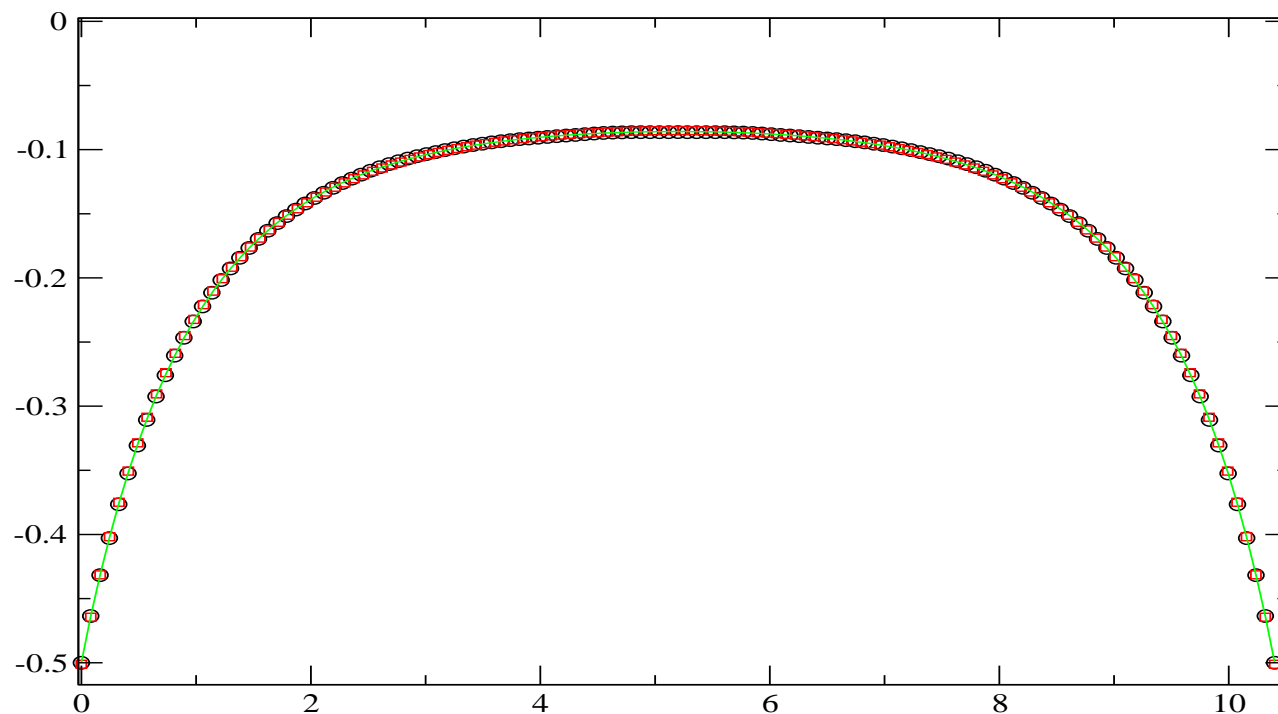
A warning ...



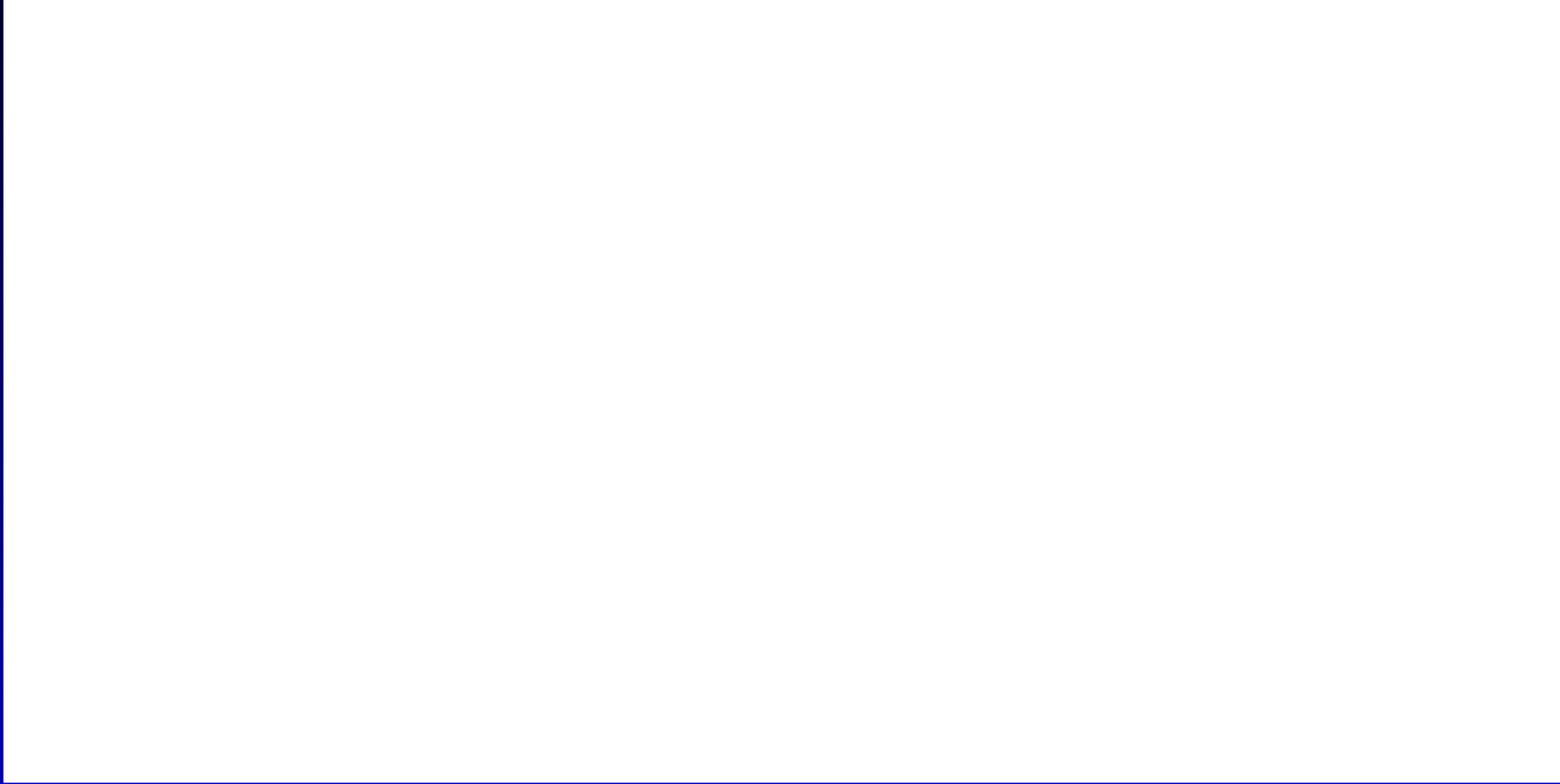
Two different spectral functions yield ...

... the “same” Green’s function!

$G(\tau)$ -- QMC data and two reconstructions ...



Real frequency self-energies



(Jan Tomczak, prelimin. results)

Real frequency self-energies

Obtain $\hat{G}(\omega)$ from the Maximum Entropy method and the Kramers-Kronig relations and invert

$$\hat{G}(\omega) = \sum_k \left(\omega + \mu - \hat{H}_o(k) - \hat{\Sigma}(\omega) \right)^{-1}$$

to obtain $\hat{\Sigma}(\omega)$

(cf also Jarrell et al., PRB 1999)

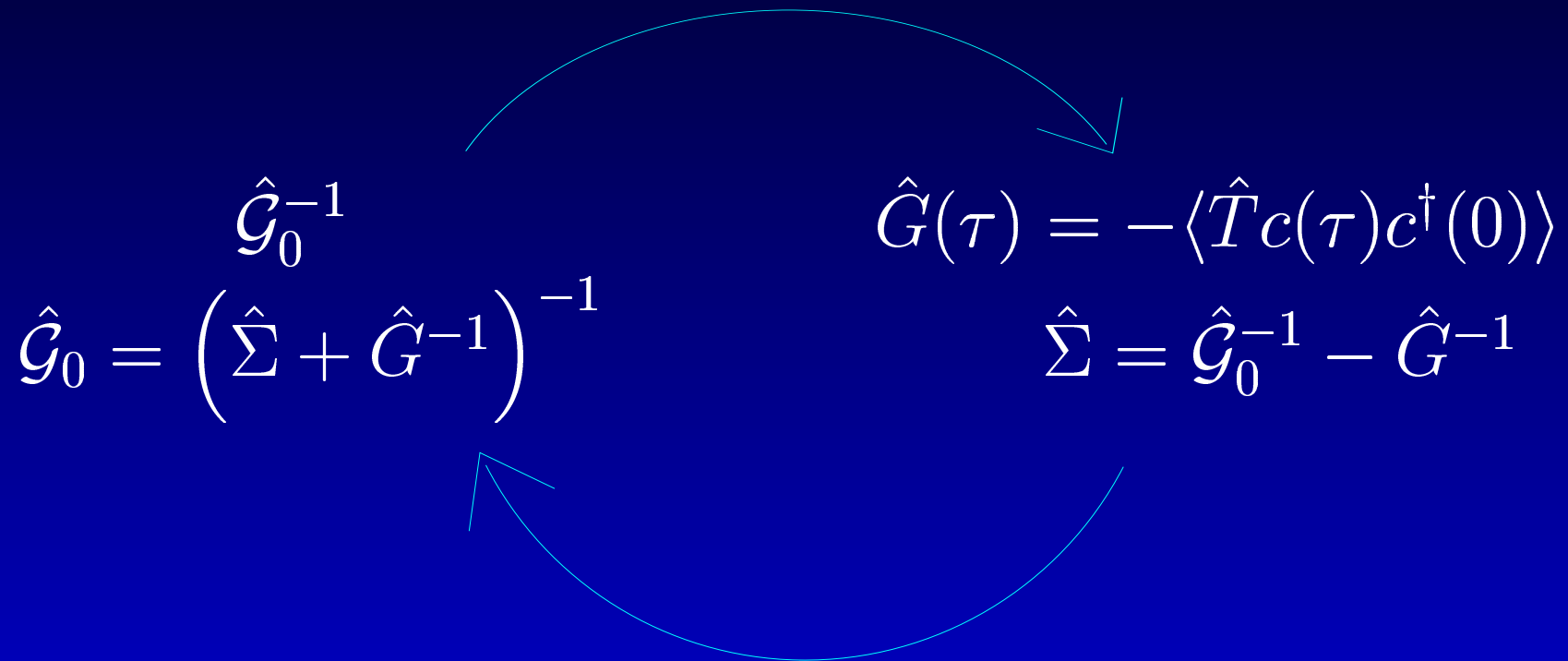
Conceptual remarks

- Global self-consistency
- Notion of locality is basis-set dependent
- Correlated vs. uncorrelated orbitals
- What's U ?
- Double counting?

NB. If only correlated states in low-energy Hamiltonian (present case): assume double counting to be orbital-independent and absorb in global shift

The DMFT self-consistency cycle

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GW+DMFT ...

... a recent attempt to go beyond LDA+DMFT

Why ?

How ?

LDA+DMFT

$$\begin{aligned} H &= \sum_{\{im\sigma\}} (H_{im,i'm'}^{LDA} - H_{im,i'm'}^{double\ counting}) a_{im\sigma}^+ a_{i'm'\sigma} \\ &+ \frac{1}{2} \sum_{imm'\sigma \text{ (correl. orb.)}} U_{mm'}^i n_{im\sigma} n_{im'-\sigma} \\ &+ \frac{1}{2} \sum_{im \neq m'\sigma \text{ (correl. orb.)}} (U_{mm'}^i - J_{mm'}^i) n_{im\sigma} n_{im'\sigma} \end{aligned}$$

What's U in a solid?

What's W in a solid? (Springer and Aryasetiawan, '98)

Screened Coulomb interaction from RPA

$\langle dd|W|dd\rangle$ for Nickel:

Can we do better than using a *static* Hubbard *parameter* U ?

What's U in a solid?

... an answer from RPA:

Divide $P = P_d + P_r$ where P_d = polarization of the correlated orbitals (e.g. 3d orbitals)

Then:

$$\begin{aligned} W &= [1 - vP]^{-1}v \\ &= [1 - W_r P_d]^{-1}W_r \end{aligned}$$

where W_r that does not include 3d-3d screening:

$$W_r(\omega) = [1 - vP_r(\omega)]^{-1}v$$

Identify $U = W_r$!

F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar, S.B., A. I. Lichtenstein PRB 70 195104 (2004)

What's U in a solid?

... what about an answer beyond RPA ??

Can we calculate W_{local} ...

... from a (dynamical) impurity model?

→ Question of representability !

- DMFT: G_{local} calculated from impurity model
- What about W_{local} ?

Self-consistency requirement:

- $G_{impurity} = G_{local}$ of the solid
- $W_{impurity} = W_{local}$ of the solid

→ “GW+DMFT”

The GW approximation

(Hedin, 1965)

$$\Sigma(r, r', \omega) = \frac{i}{2\pi} \int d\omega' G(r, r', \omega + \omega') W(r, r', \omega')$$

$W = \epsilon^{-1}v$ dynamically screened Coulomb interaction

GW successful for sp-metals, semiconductors ...

(Reviews:

Onida et al., Rev. Mod. Phys. 2002;

Aryasetiawan et al., Rep. Prog. Phys. 1998)

A functional point of view

[Chitra & Kotliar, 2001, Almbladh et al. 1999]

$$\Gamma[G, W] = \text{Tr} \ln G - \text{Tr}[(G_H^{-1} - G^{-1})G] \\ - \frac{1}{2} \text{Tr} \ln W + \frac{1}{2} \text{Tr}[(V^{-1} - W^{-1})W] + \Psi[G, W]$$

Free energy Γ is a functional of

- one-electron Green's function

$$G(\mathbf{r}, \mathbf{r}'; \tau - \tau') \equiv -\langle T_\tau \psi(\mathbf{r}, \tau) \psi^\dagger(\mathbf{r}', \tau') \rangle$$

- the screened Coulomb interaction

$$W = V - V\chi V$$

G_H = bare (Hartree) Green's function

Approximations to $\Psi[G, W]$?

$$\text{GW: } \Psi_{GW}[G, W] = -\frac{1}{2} \text{Tr}GWG$$

Extended DMFT (“E-DMFT”):

$$\Psi_{DMFT}[G, W] = \Psi_{impurity}[G_{impurity}, W_{impurity}]$$

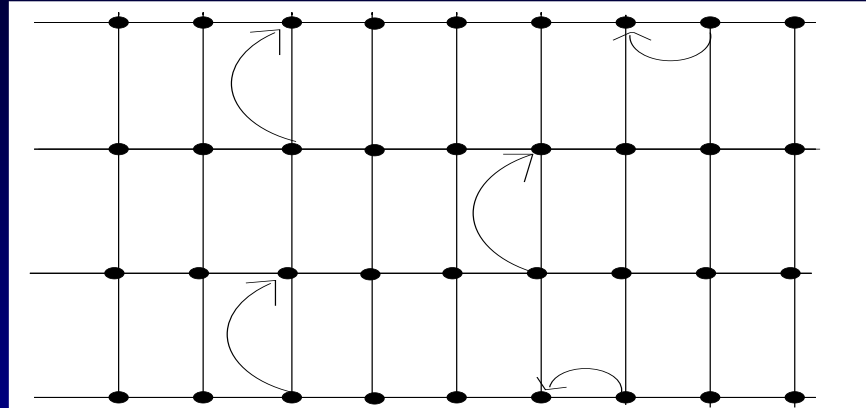
E-DMFT for local part + GW for nonlocal part:

$$\Psi_{GW+DMFT}[G, W] = \Psi_{DMFT} + \Psi_{GW}^{nonlocal}$$

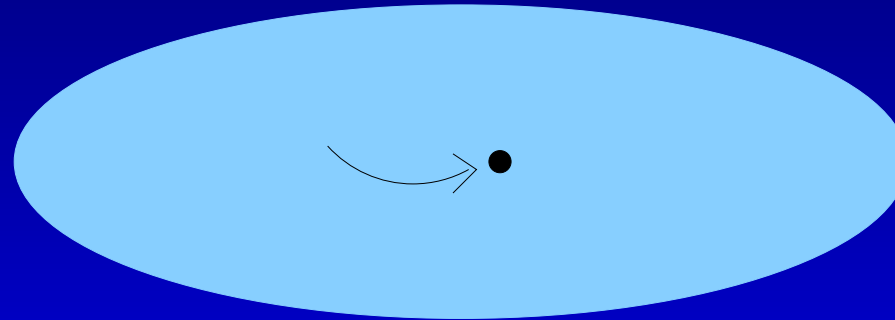
NB: “local” = “onsite” is a basis-set dependent notion!

Extended DMFT ...

... maps a lattice problem



onto a single-site (Anderson impurity) problem



with a *dynamical* interaction $\mathcal{U}(\tau - \tau')$

GW+DMFT: local part

$\Psi_{imp}[G_{imp}, W_{imp}]$ calculated from local impurity model:

$$S_{imp} = \int d\tau d\tau' \left[- \sum c_L^\dagger(\tau) \mathcal{G}_{LL'}^{-1}(\tau - \tau') c_{L'}(\tau') \right. \\ \left. + \sum : c_{L_1}^\dagger(\tau) c_{L_2}(\tau) : \mathcal{U}_{L_1 \dots L_4}(\tau - \tau') : c_{L_3}^\dagger(\tau') c_{L_4}(\tau') \right]$$

$$G_{imp}^{LL'} = - \langle T_\tau c_L(\tau) c_{L'}^\dagger(\tau') \rangle_S$$

$$\chi_{L_1 L_2 L_3 L_4} = \langle : c_{L_1}^\dagger(\tau) c_{L_2}(\tau) :: c_{L_3}^\dagger(\tau') c_{L_4}(\tau') : \rangle_S$$

$$W_{imp} = \mathcal{U} - \mathcal{U} \chi \mathcal{U}$$

GW+DMFT (contd)

Combine local self-energy and polarization

$$\Sigma_{imp}^{xc} \equiv \delta\Psi_{imp}/\delta G_{imp} = \mathcal{G}^{-1} - G_{imp}^{-1}$$

$$P_{imp} \equiv -2\delta\Psi_{imp}/\delta W_{imp} = \mathcal{U}^{-1} - W_{imp}^{-1}$$

with non-local self-energy and polarization:

$$\begin{aligned}\Sigma^{xc}(\mathbf{k}, i\omega_n)_{LL'} &= \Sigma_{GW}^{xc}(\mathbf{k}, i\omega_n)_{LL'} \\ &- \sum_{\mathbf{k}} \Sigma_{GW}^{xc}(\mathbf{k}, i\omega_n)_{LL'} + [\Sigma_{imp}^{xc}(i\omega_n)]\end{aligned}$$

$$\begin{aligned}P(\mathbf{q}, i\nu_n)_{\alpha\beta} &= P^{GW}(\mathbf{q}, i\nu_n)_{\alpha\beta} \\ &- \sum_{\mathbf{q}} P^{GW}(\mathbf{q}, i\nu_n)_{\alpha\beta} + P^{imp}(i\nu_n)_{\alpha\beta}\end{aligned}$$

Self-consistency condition

$$G_{loc}(i\omega_n) = \sum_{\mathbf{k}} [G_H^{-1}(\mathbf{k}, i\omega_n) - \Sigma^{xc}(\mathbf{k}, i\omega_n)]^{-1}$$

$$W_{loc}(i\nu_n) = \sum_{\mathbf{q}} [V_{\mathbf{q}}^{-1} - P(\mathbf{q}, i\nu_n)]^{-1}$$

Update Weiss field and impurity interaction:

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

Iterate until self-consistency ...

Self-consistency loop

Impurity model :

$$\mathcal{G}(\tau), \mathcal{U}(\tau)$$

$$G_{imp} \equiv -\langle T_\tau cc^\dagger \rangle_S \rightarrow \Sigma_{imp}^{xc} = \mathcal{G}^{-1} - G_{imp}^{-1}$$

$$W_{imp} = \mathcal{U} - \mathcal{U}\chi\mathcal{U} \quad P_{imp} = \mathcal{U}^{-1} - W_{imp}^{-1}$$

↑

Update

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

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

↑

Self – consistency

$$G_{loc} = \sum_{\mathbf{k}} [G_H^{-1} - \Sigma^{xc}]^{-1}$$

$$W_{loc} = \sum_{\mathbf{q}} [V_{\mathbf{q}}^{-1} - P]^{-1}$$

↓

Combine :

$$\Sigma = \Sigma_{imp} + \Sigma_{GW}^{nonlocal}$$

$$P = P_{imp} + P_{GW}^{nonlocal}$$

↓

Challenges and questions

- Global self-consistency?
- Choice of orbitals? Hamiltonian?
- Treat all orbitals – localized and delocalized – on equal footing? Downfolding?
- How to solve the dynamical impurity model?
 - P.Sun & G.Kotliar
 - A. Rubtsov
 - S. Florenshere: static approximation

A simplified implementation

Non-selfconsistent GW + local Σ from static impurity model

$$G_{loc}^{\sigma}(i\omega_n) = \sum_{\mathbf{k}} [G_H^{-1}(\mathbf{k}, i\omega_n) - (\Sigma_{GW}^{xc})_{non-loc} - (\Sigma_{imp,\sigma} - \frac{1}{2} \text{Tr}_{\sigma} \Sigma_{imp,\sigma}(0) + V_{xc}^{loc})]^{-1}$$

Nonlocal part: correct Hartree by GW

Local part: correct LDA by DMFT

Nickel

Evidence for many-body effects:

	Exp.	LDA	GW ⁽¹⁾	LDA+DMFT ⁽²⁾
bandwidth	3.3 eV	4.5 eV	3.5 eV	OK!
x-splitting	0.3 eV	0.6 eV	0.6 eV	OK!
satellite	at 6eV	NO!	NO!	YES!

large quasi-particle widths

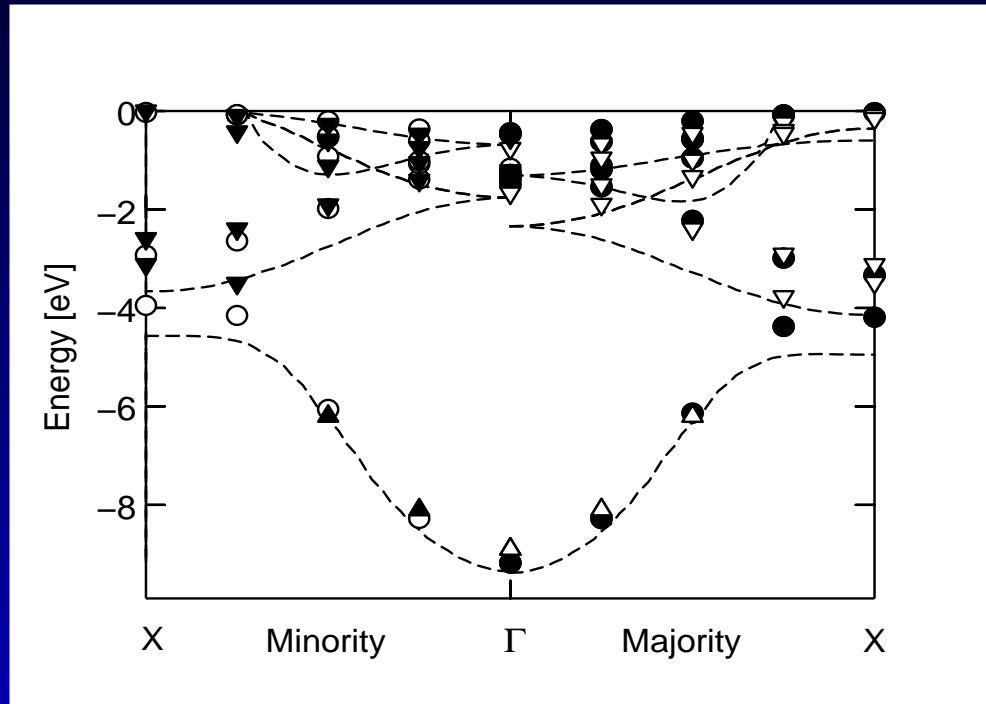
Open question: Fermi surface pocket ?

(1) Aryasetiawan, 1992

(2) Lichtenstein et al., 2001

Simplified GW+DMFT

Ni band structure



Circles: GW+DMFT

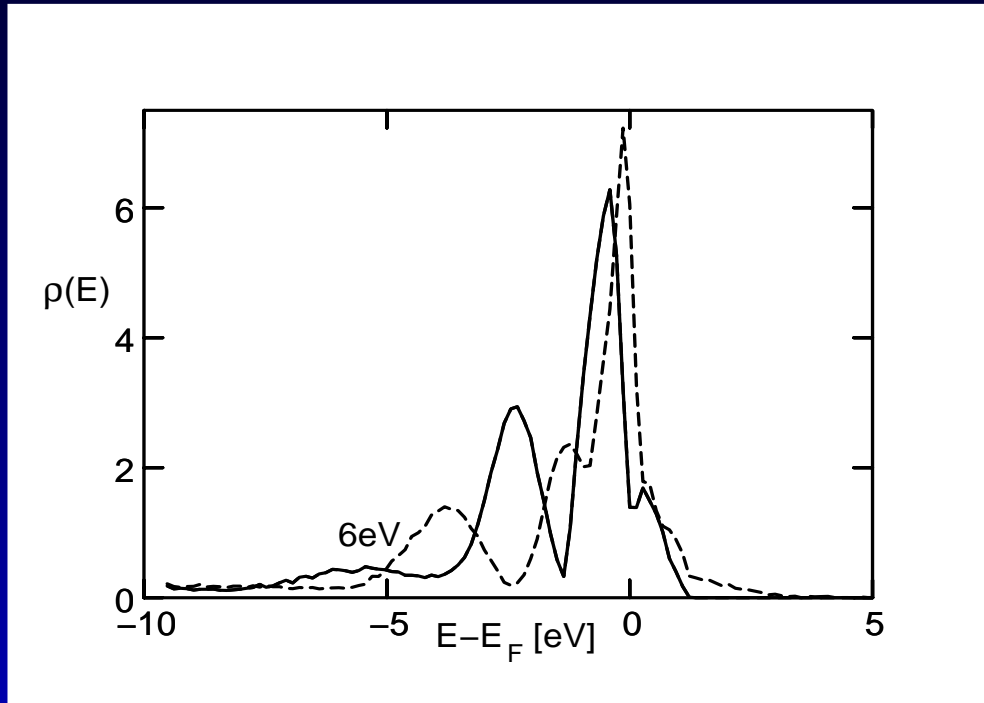
Dashed: LDA

Triangles: photoemission data

(Bünemann et al. 2002, Mårtensson et al., 1984)

Simplified GW+DMFT:

Spectral function of Ni



Majority and minority spins

Satellite at 6eV correct!

Conclusion and perspectives

Combination of LDA and DMFT ...

- a useful tool for electronic structure calculations of strongly correlated materials

Combination of GW and DMFT ...

- describes long-range interactions in an ab initio way
- includes non-local self-energy effects

Questions and perspectives:

- Dynamical impurity models in realistic context?
- Effects of self-consistency?

An exciting field with many open problems!

Collaborations and references

- VO_2 (PRL 2005):
A. Poteryaev, A. Georges, A. Lichtenstein
- GW+DMFT (PRL 2003 + condmat-0401653):
F. Aryasetiawan, A. Georges
- $\mathcal{U}(\omega)$ within RPA (PRB 2004) :
F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar,
A. I. Lichtenstein