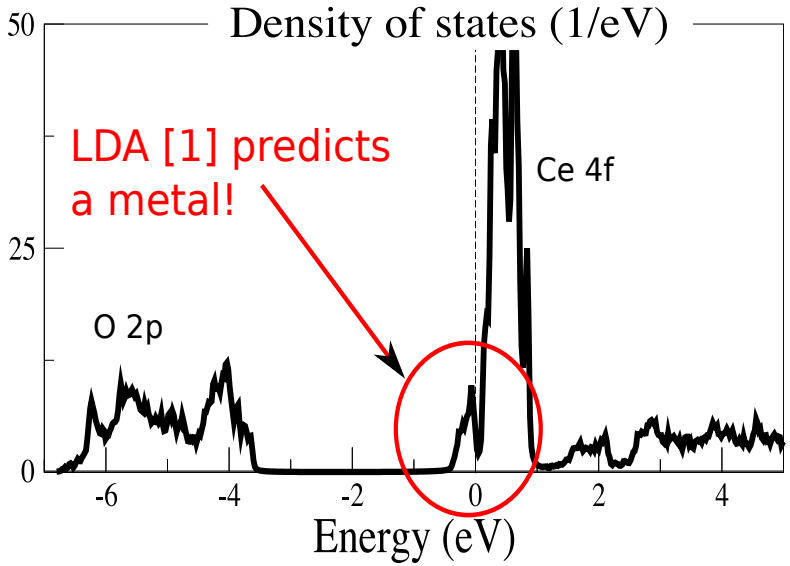
Key concepts of LDA+DMFT and QSGW+DMFT (in the Questaal package)

Lorenzo Sponza

The Hubbard model in a nutshell

Physical problem: Mott insulator

Ce_2O_3 is an antiferromagnet with a **gap of 2.5 eV**.
However, **according to band theory**,
it should be a metal since the *4f* levels are not filled.



[1] L. V. Pourovskii, B. Amadon, S. Biermann and A. Georges, Phys. Rev. B **76**, 235101 (2007)

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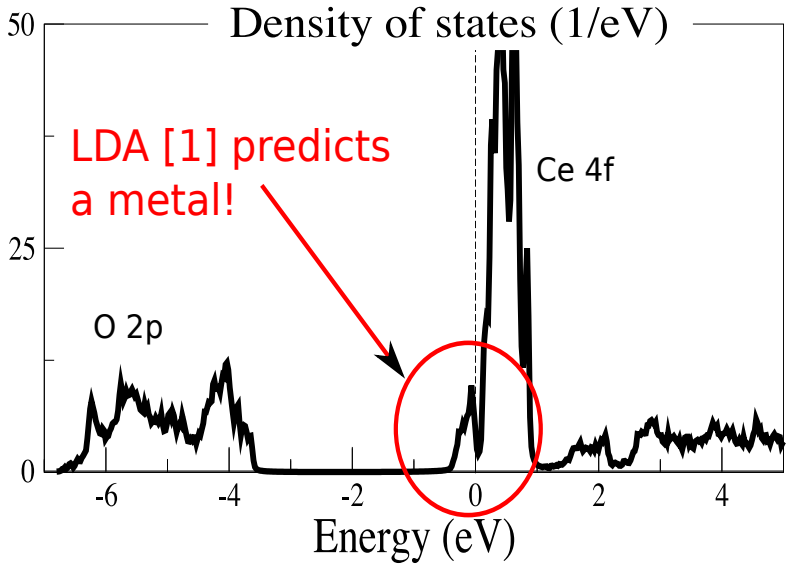

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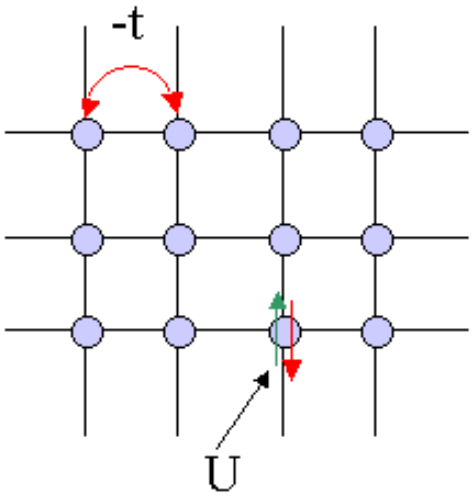
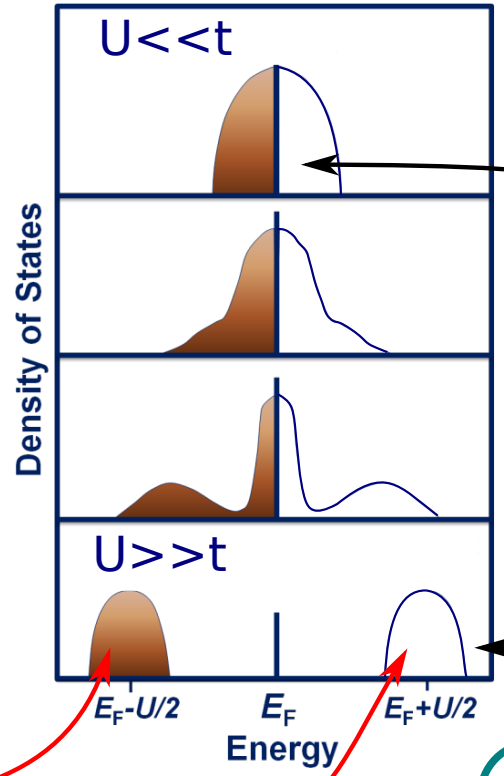


- lattice of **localised sites**
- electrons **can hop** (-t)
- **on-site repulsion U**



metallic density of states
There is energy gain in hopping

Insulating density of states
There is no energy gain in hopping

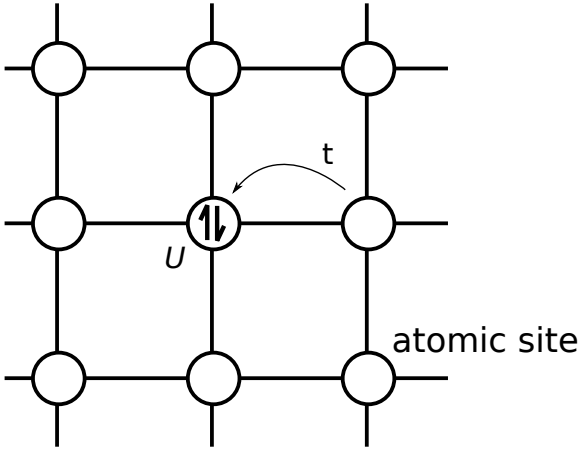
lower and upper
Hubbard bands

LDA does not capture the importance of **local correlations** (on-site repulsion) in *d*- and *f*-electron systems.

[1] L. V. Pourovskii, B. Amadon, S. Biermann and A. Georges, Phys. Rev. B **76**, 235101 (2007)

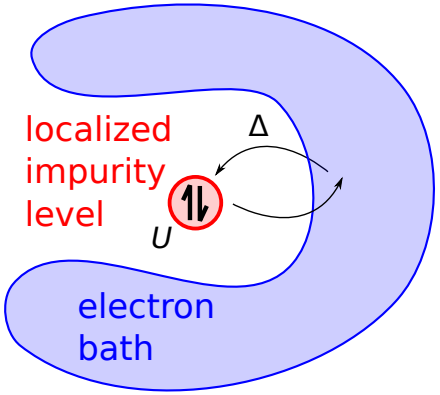
A similar problem: Anderson's impurity model

Hubbard model



a lattice of sites
coupled to each other

Anderson's impurity model



a single site coupled with a bath
(at thermodynamic equilibrium)

From a lattice of sites to a single site, with **identical on-site physics**
Anderson's model can be solved numerically (solver).

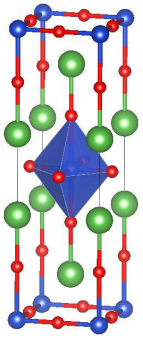
Improving LDA by treating as impurities the localised electrons

Anderson's model is an ideal framework to be coupled with LDA
impurity = *d*- or *f*-electron atomic-like levels
electron bath = independent particle electrons from LDA

LDA+DMFT (general scheme)

Lattice model

- physical system
- **local**, **static**
- atomistic simulation
- interacrtions = **functional**



The LDA Loop

$$\rho(\mathbf{r})$$

$$H(i\omega_n) = H^{LDA} + \bar{\Sigma}(i\omega_n)$$

$$G(i\omega_n) = (i\omega_n + \mu - H(i\omega_n))^{-1}$$

[2] K. Haule, C.-H. Yee and K. Kim, Phys Rev B **81**, 195107 (2010)

[3] P. Werner *et al.*, Phys. Rev. Lett. **97**, 076405 (2006) and K. Haule, Phys. Rev. B **75**, 155113 (2007)

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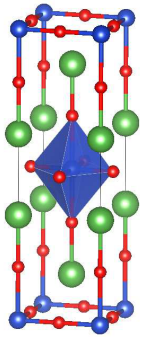

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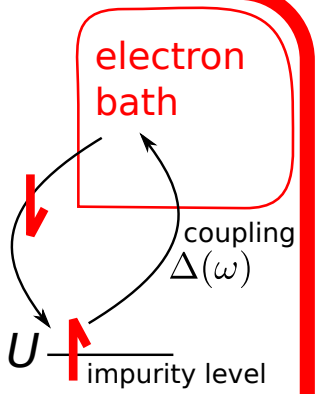
Impurity model



- Anderson impurity model
- **local**, **dynamic**
- CTQMC (solver) [2]
- **diagrammatic** expansion

The DMFT Loop

$$\bar{\Sigma}(i\omega_n) = \Sigma^{imp}(i\omega_n) - E^{DC}$$

$$G^{imp}(i\omega_n)$$

$$\Delta(i\omega_n)\ ;\ U$$

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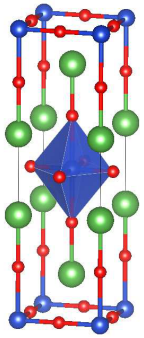

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embedding $\hat{E}$

$\hat{P}$ **projection**

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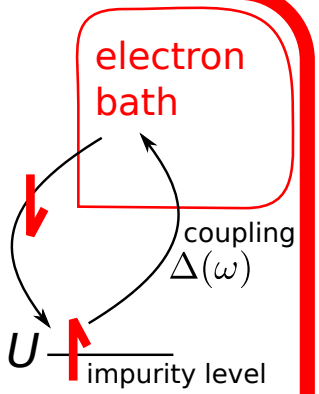

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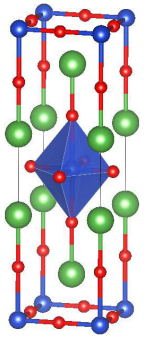

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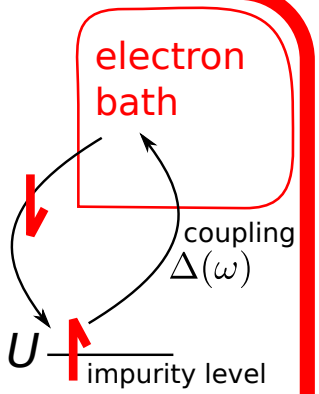

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$$G^{imp}(i\omega_n)$$

$$\Delta(i\omega_n)\ ;\ U$$

projection $\hat{P}$

- In the LDA Loop $\bar{\Sigma}$ is kept constant, while in the DMFT loop G (lattice) is kept constant.
- The two loops are alternated until the **self-consistent condition** $\hat{P}G = G^{imp}$ is met.
- E^{DC} is the **double counting** term, accounting for exchange-correlation terms already included during the LDA loop (via the XC-potential).
- There are **several projection and embedding schemes**. We adopted [2].
- Fermionic Matsubara's frequencies $\omega_n = (2n-1)\pi/\beta$ are adopted because of the solver used (CTQMC = Continuous time quantum Monte Carlo) [3].

[2] K. Haule, C.-H. Yee and K. Kim, Phys Rev B **81**, 195107 (2010)

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More details about the LDA loop

Hamiltonian (corrected by DMFT self-energy)

$$H_{ij\mathbf{k}}(i\omega_n) = H^{LDA}_{ij\mathbf{k}} + \bar{\Sigma}_{ij\mathbf{k}}(i\omega_n)$$

in the first loop $\bar{\Sigma} = 0$ i.e., the loop is a standard LDA calculation

Diagonalization

$$\varepsilon_{i\mathbf{k}}(i\omega_n)\ ;\ \psi^{R/L}_{ij\mathbf{k}}(i\omega_n)$$

LDA loop

Charge neutrality

Adjust the chemical potential μ using $\varepsilon_{i\mathbf{k}}(i\omega_n)$

Embedding the DMFT self-energy

$$\bar{\Sigma}_{ij\mathbf{k}}(i\omega_n) = \sum_{LL'} \hat{E}^{LL'}_{ij\mathbf{k}} \bar{\Sigma}_{LL'}(i\omega_n)$$

$$\rho(\mathbf{r}) = \sum_{\mathbf{k},l}\left[\frac{2}{\beta}\sum_n \mathrm{Re}\left(\psi^R_{l\mathbf{k}}(\mathbf{r},i\omega_n)\frac{1}{i\omega_n+\mu-\varepsilon_{l\mathbf{k}}(i\omega_n)}\psi^{L*}_{l\mathbf{k}}(\mathbf{r},i\omega_n)\right)\right]$$

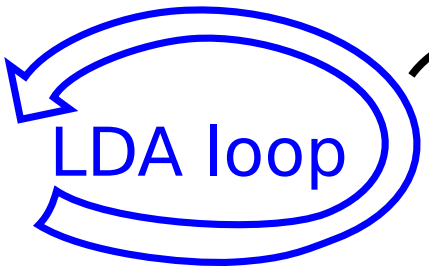
New density with dynamical contributions from DMFT

Lattice Green's function

$$G_{ij\mathbf{k}}(i\omega_n) = \left[i\omega_n + \mu - H_{\mathbf{k}}(i\omega_n)\right]^{-1}_{ij}$$

DMFT loop

More details about the DMFT loop



Project the lattice Green's function

$$G_{LL'}(i\omega_n) = \sum_{ij\mathbf{k}} \hat{P}^{ij\mathbf{k}}_{LL'} G_{ij\mathbf{k}}(i\omega_n)$$

effective in-site interactions

U ; J

U and J can be computed *ab-initio* from local quantities (cLDA or cRPA). But they are some times treated as tunable parameters.

The hybridization function

$$\Delta_{LL'}(i\omega_n) = i\omega_n - G^{-1}_{LL'}(i\omega_n) - E^{imp}_{LL'} - \Sigma^{imp}_{LL'}(i\omega_n)$$

DMFT loop

CTQMC

Solver

The impurity self-energy

$$\Sigma^{imp}_{LL'}(i\omega_n)$$

Double counting term

$$E^{DC} = U\left(n - \frac{1}{2}\right) - J\left(\frac{n}{2} - \frac{1}{2}\right)$$

n=nominal occupation of the impurity level

Remove the double counting

$$\bar{\Sigma}_{LL'}(i\omega_n) = \Sigma^{imp}_{LL'}(i\omega_n) - E^{DC}\delta_{LL'}$$

Ensure charge neutrality

In order to adjust the chemical potential μ , one has to make **a step back to the lattice problem** and repeat part of the LDA loop:

i) embed $\bar{\Sigma}_{LL'}(i\omega_n)$

ii) diagonalise $H_{ij\mathbf{k}}(i\omega_n)$

iii) adjust μ using $\varepsilon_{i\mathbf{k}}(i\omega_n)$

iv) construct and project $G_{ij\mathbf{k}}(i\omega_n)$

Summary of some key aspects of LDA+DMFT

- The LDA+DMFT scheme is composed by **TWO loops**.

- The **LDA loop leads to a converged lattice Green's function** with fixed local corrections, the **DMFT loop leads to converged local self-energy** (self-consistent solution of the Anderson's impurity model) at fixed lattice Green's function.

- Part of the local correlations are already included in the LDA scheme, so they have to be removed from the solution of the local problem. This is done by subtracting the **DOUBLE COUNTING** term from the impurity self-energy. Because of the **lack of diagrammatic expansions** in LDA, this is probably the most delicate aspect of the approach and a lot of effort is done to solve this issue (in the context of LDA + DMFT see for instance [4]).

- The passage from a loop to the other is done through suitably defined **PROJECTION** operations (from lattice G to local G) and **EMBEDDING** operations (from impurity self-energy to local corrections) Intrinsiquely related to the choice of these operators are the **effective on-site interactions U and J**.

- The two loops are performed alternately with the output of one loop being the input for the other until the self-consistent condition is met: **the impurity Green's function is equal to the projected lattice Green's function**.

[4] K. Haule, Phys. Rev. Lett. **115**, 196403 (2015)

Few words about Projection and Embedding

How to go from the lattice Green's function of LDA to the impurity Green's function?
How to map the electronic band structure into the Anderson's electron bath?

two possibilities

implemented in Questaal

PROJECTION/EMBEDDING [2]
- Projection on atomic spheres
- large energy window
- cut out from full problem
- locality assured by construction

DOWNFOLDING [5]
- Wannier functions
- small energy window
- effective local Hamiltonian (simple)
- locality some times questionable

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[5] F. Lechermann *et al*, Phys. Rev. B **74**, 125120 (2006)

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The Questaal implementation: Projection/Embedding

$\hat{P}^{ij\mathbf{k}}_{LL'} := \sum_{ij\mathbf{k}} U^{i\mathbf{k}\dagger}_{L} : U^{j\mathbf{k}}_{L'}$ and $\hat{E}^{LL'}_{ij\mathbf{k}} := \sum_{LL'} U^{i\mathbf{k}}_{L} : U^{j\mathbf{k}\dagger}_{L'}$ where $U^{i\mathbf{k}}_{L} \propto \sum_{\kappa} \mathcal{A}^{\kappa}_{iL}(\mathbf{k})\Phi^{\kappa}$

coefficient inside the atomic sphere

wave function and derivatives

They are defined to satisfy four requirements:
1) **No leak of spectral weight** after projection (the same as LDA one),
2) Ensure **causality of the DMFT equations**,
3) **Accuracy of the hybridization function** (close to LDA estimate),
4) Accurate **kinetic energy and electronic density** (do not deteriorate LDA results)

IMPORTANT: P and E are **not inverse of each other** since $\hat{P} \circ \hat{E} = \mathbf{1}$, but $\hat{E} \circ \hat{P} \neq \mathbf{1}$

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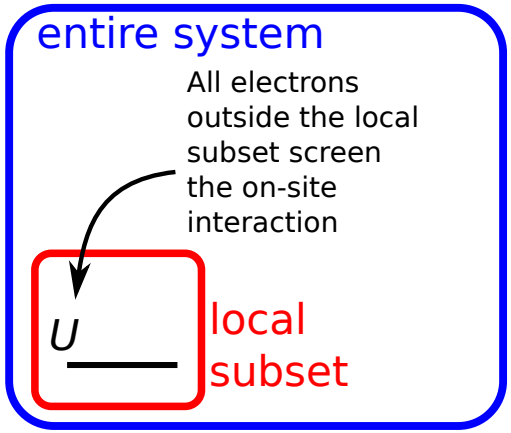
Few words about the effective interactions

U and J are effective local interactions:
they depends on HOW you project/downfold.
In particular, **the more screening processes are excluded from the local subset, the more they screen the repulsive on-site interaction.**



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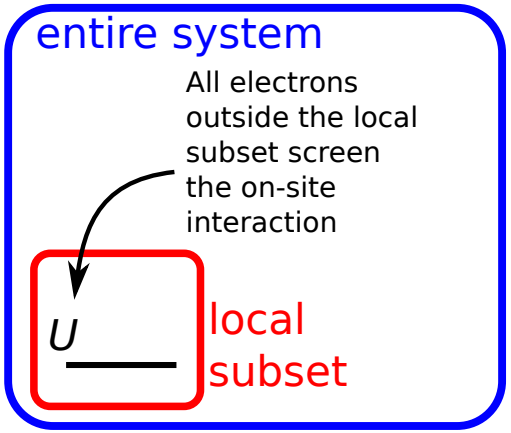
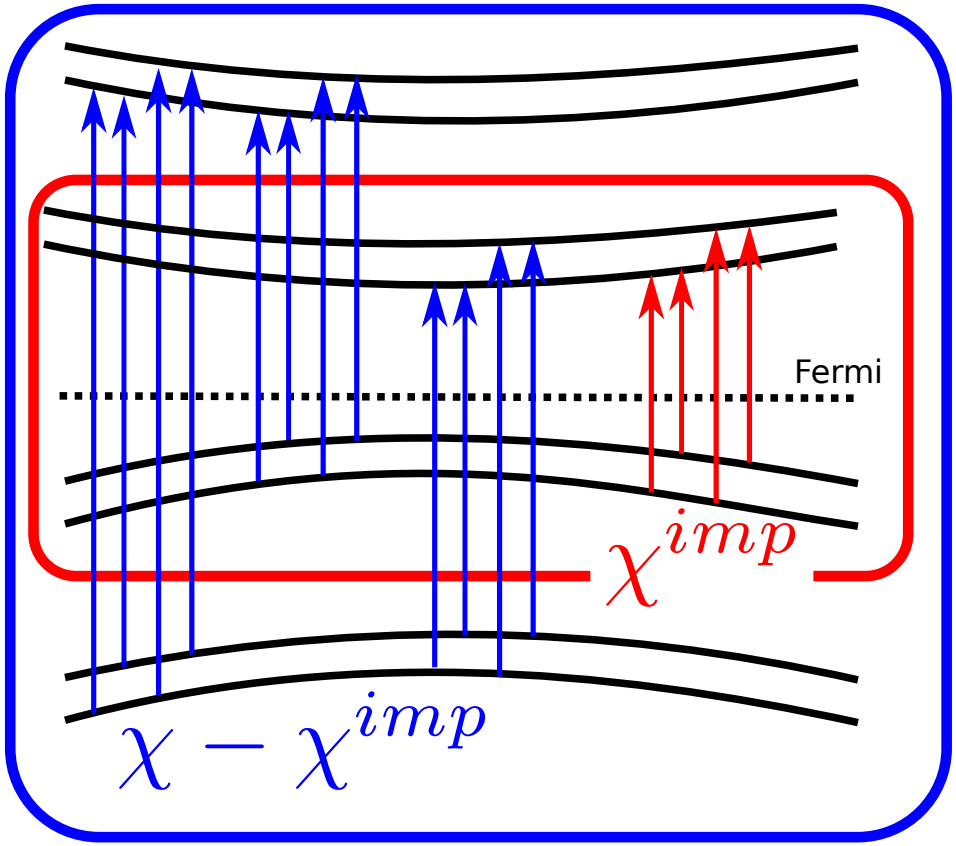


A sketch of the **constrained RPA** (cRPA) scheme [6]



$\chi = -iGG$ Total polarizability: all screening channels

$\chi^{imp} = -iG^{imp}G^{imp}$ Local polarizability: screening inside the local subset

The effective local interaction *U* as the **static limit** of the **Coulomb interaction screened by transitions** involving at least one state **outside the local subset**.

$$W^r = [1 - v(\chi - \chi^{imp})]^{-1} v$$ ← bare Coulomb

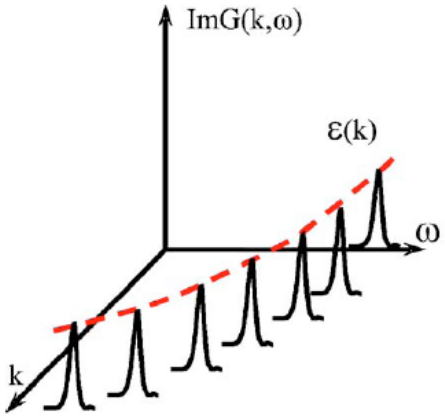
$$U = W^r(\omega = 0)$$ cRPA on-site interaction

[6] F. Aryasetjawan *et al*, Phys. Rev. B **70**, 195104 (2004)

What about spectral functions?

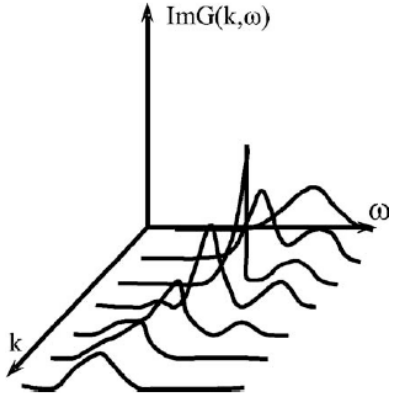
Static Hamiltonian (LDA and QSGW)

The Green's function has
delta-like poles at ω-H.
Band structure



Dynamic Hamiltonian (X+DMFT)

The Green's function has
structures for poles at ω-$H(\omega)$.
Spectral function



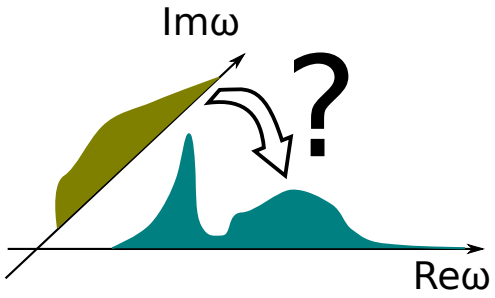
Problem:
$H(i\omega_n)$ is defined on the imaginary frequency axis.
To compare with measured specral functions (e.g. photoemission)
we have to analytically continue the solution
from Matsubara's frequencies to real frequencies.

A few words on analytic continuation

The problem:

Get the spectral function $A(\omega)$ from the Green's function $G(i\omega_n)$.
Formally the problem is known, and solved in the imaginary time:

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



However, *G* has poles on the real axis, whereas it is very smooth on the imaginary axis, hence the inversion problem is **prone to severe numerical problems**.

Possible methods:

All methods rely on **creating a trial spectral function** $\tilde{A}(\omega)$ on the real axis, getting **the corresponding** $\tilde{G}(i\omega_n)$ on imaginary frequencies and judge on the quality of the agreement.
Possible methods are, e.g. the **Maximum Entropy** [7] or the Stochastic optimization [8].

[7] J. E. Gubernatis *et al*, Phys. Rev. B **44**, 6011 (1991)
[8] A. S. Mishchenko: www.cond-mat.de/events/correl12/manuscripts/mishchenko.pdf

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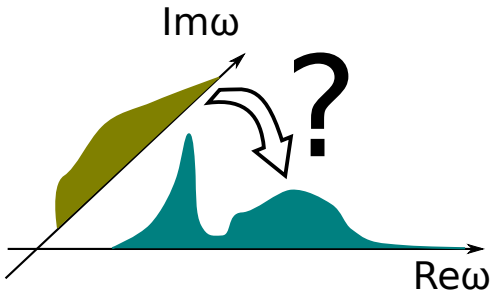

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The Maximum Entropy Method:

The solution is the spectral function $\tilde{A}(\omega)$ that **minimizes**
i) the distance between $\tilde{A}(\omega)$ and a **model function** $m(\omega)$ that embodies the information known a-priori on the solution (positivity for instance) and
ii) the distance between its corresponding $\tilde{G}(i\omega_n)$ and **the given** $G(i\omega_n)$.
The space of possible spectral functions is **sampled randomly** with Metropolis algorithms.

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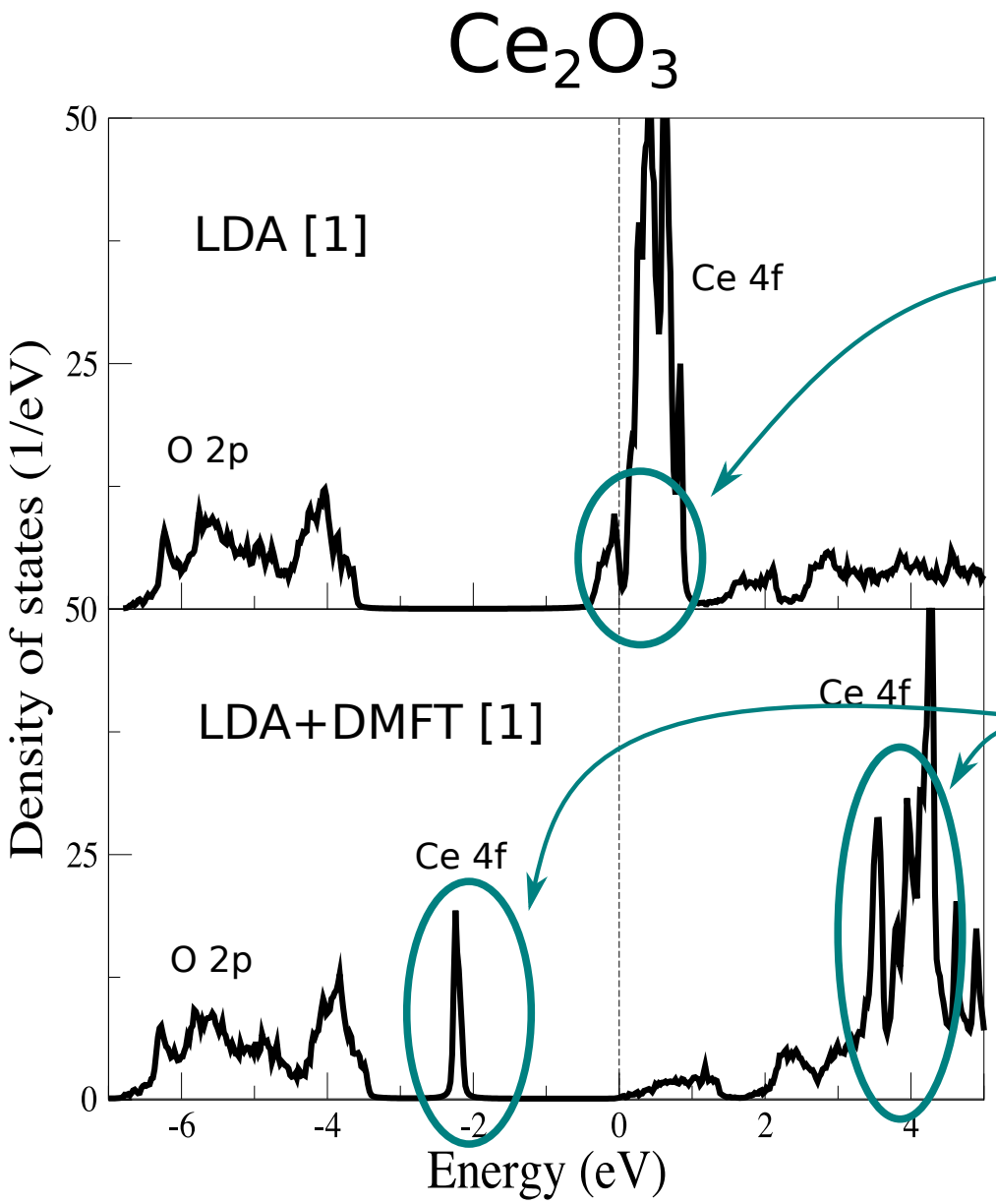
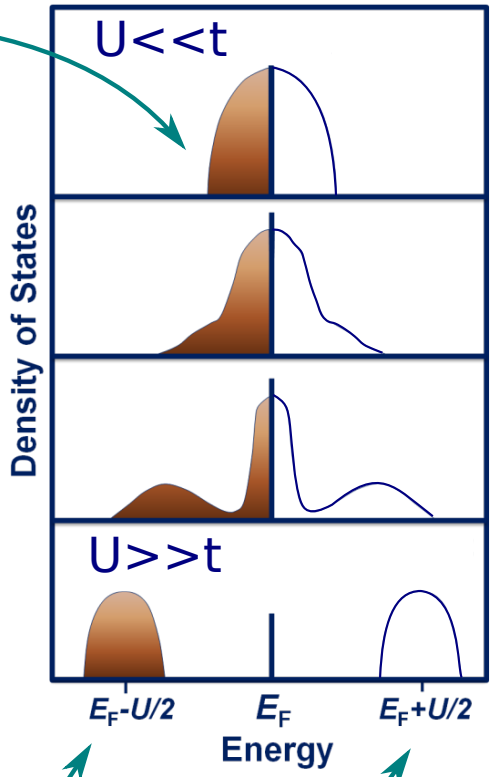
Back to the original problem: Mott insulator



LDA
predicts a **metal**

LDA+DMFT
opens a gap
correctly separates
the 4f-states
(Hubbard bands)

Hubbard physics



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Motivations of the QSGW+DMFT approach

Problems of the LDA + DMFT approach

- Ambiguities in the definition of the **double cunting terms**.
  The difficulty stems from the lack of diagrammatic expansion of the XC-potential of LDA
  Despite recent advances [4], the subject is still under debate.
  A **Many body scheme** to solve the lattice problem constitutes a more rigorous framework.

- Both LDA and DMFT are local theories, so **non-local physics is absent** (or very crudely represented).
  There are aspects (for instance band alignment) that require a proper treatment of non-local correlations.
  In particular **low-order diagrams appear to be highly non-local with weak dynamical dependance, while high-order diagrams appear to be highly local with strong dynamical dependance** [9].
  This makes QSGW (static and non-local) an ideal theory for the lattice problem.

- LDA provides often a **poor description** of the band structure and wave functions.
  A **better representation** of the electrons in the lattice obtained from QSGW should lead to a **faster convergence** of the entire QSGW+DMFT scheeme.

[4] K. Haule, Phys. Rev. Lett. **115**, 196403 (2015)
[9] J. M. Tomczak, M. van Schilfgaarde, G. Kotliar, Phys. Rev. Lett. **109**, 237010 (2012)

The QSGW+DMFT loops in Questaal: QSGW loop

For the time being, the implementation of the QSGW+DMFT scheme is very similar to the LDA+DMFT one.
However because of the way QSGW corrections are added to the LDA Hamiltonian, the QSGW loop requires some additional attention.

$\rho(\mathbf{r}) = \sum_{i,\mathbf{k},\omega_n} \ldots$

$\varepsilon_{i\mathbf{k}}(i\omega_n)\ ;\ \psi^{R/L}_{i\mathbf{k}}(i\omega_n)\ ;\ \mu$

$G_{LL'}(i\omega_n) = \hat{P} G_{ij\mathbf{k}}(i\omega_n)$

$H = H^{LDA} + \Delta V^{QSGW}_{xc} + \bar{\Sigma}$

$\Delta_{LL'}(i\omega_n) \propto G^{-1}_{LL'}(i\omega_n) + \Sigma^{imp}_{LL'}(i\omega_n)$

Since QSGW is implemented as a correction to the LDA Hamiltonian, updating the density improves only on the LDA contribution.
Once the density is converged, ΔV^{QSGW}_{xc} has to be evaluated again through **a full QSGW calculation at fixed density**.

μ same procedure as in the LDA + DMFT loop

CTQMC

U From **external source** (literature, models)

$\Sigma^{imp}_{LL'}(i\omega_n)$

$E^{DC} = U\left(n - \frac{1}{2}\right) - J\left(\frac{n}{2} - \frac{1}{2}\right)$

Standard (simple) double counting term.
At the moment we still can not exploit the real advantage of having a complete many-body formalism.
This is a (temporary) limitation.

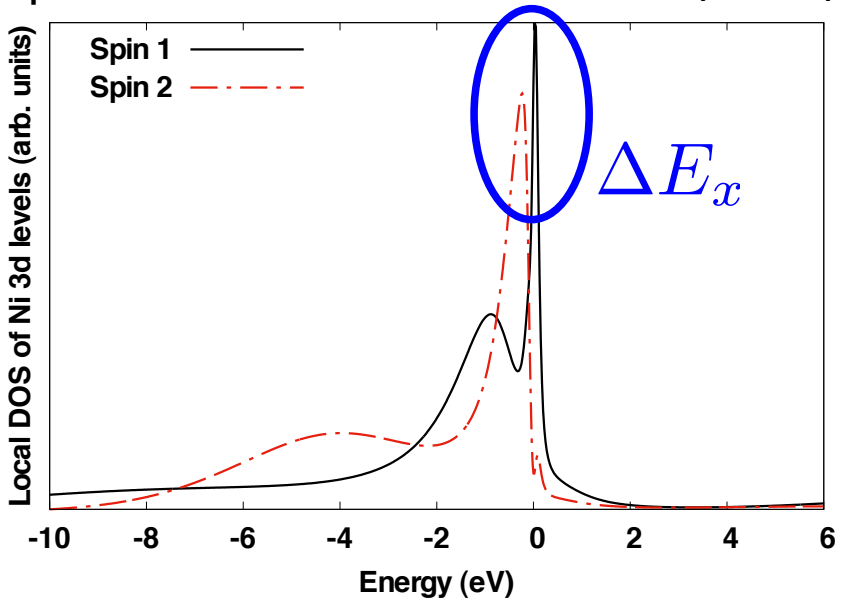
$\bar{\Sigma}_{LL'}(i\omega_n) = \Sigma^{imp}_{LL'}(i\omega_n) - E^{DC}$

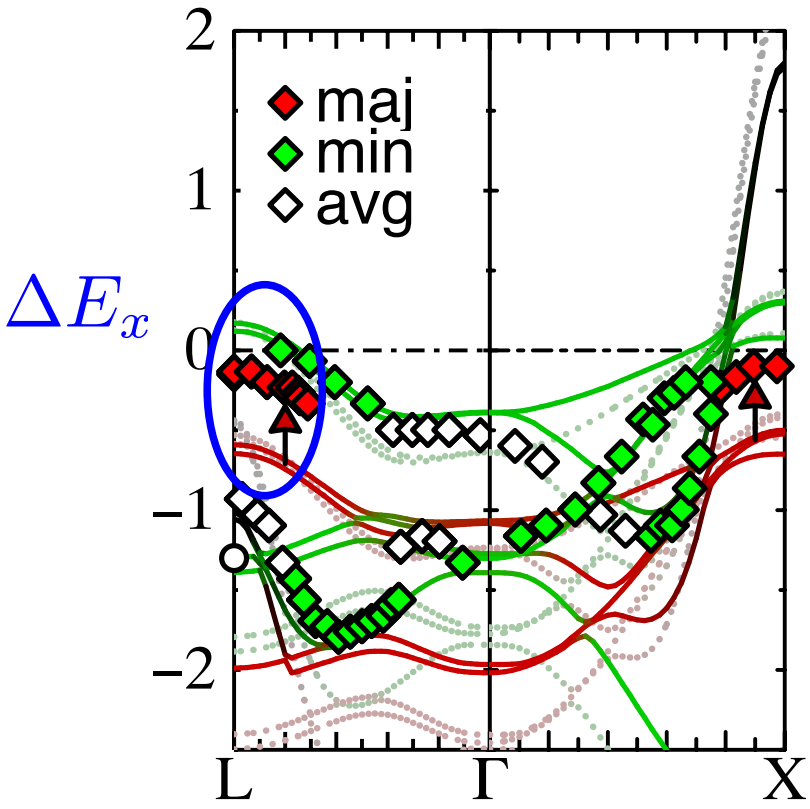
An example of QSGW+DMFT on Ni [10]

ΔE_x: exchange splitting: band shift between the two spin channels
M: magnetic moment: difference between occupation of the two spin channels

	M (Bhor)	ΔE_x @ L (eV)
LSDA	0.62	0.71
QSGW	0.75	0.77
QSGW+DMFT	0.51	0.30
Experiment	0.57	0.31

spectral function of d states (MEM)

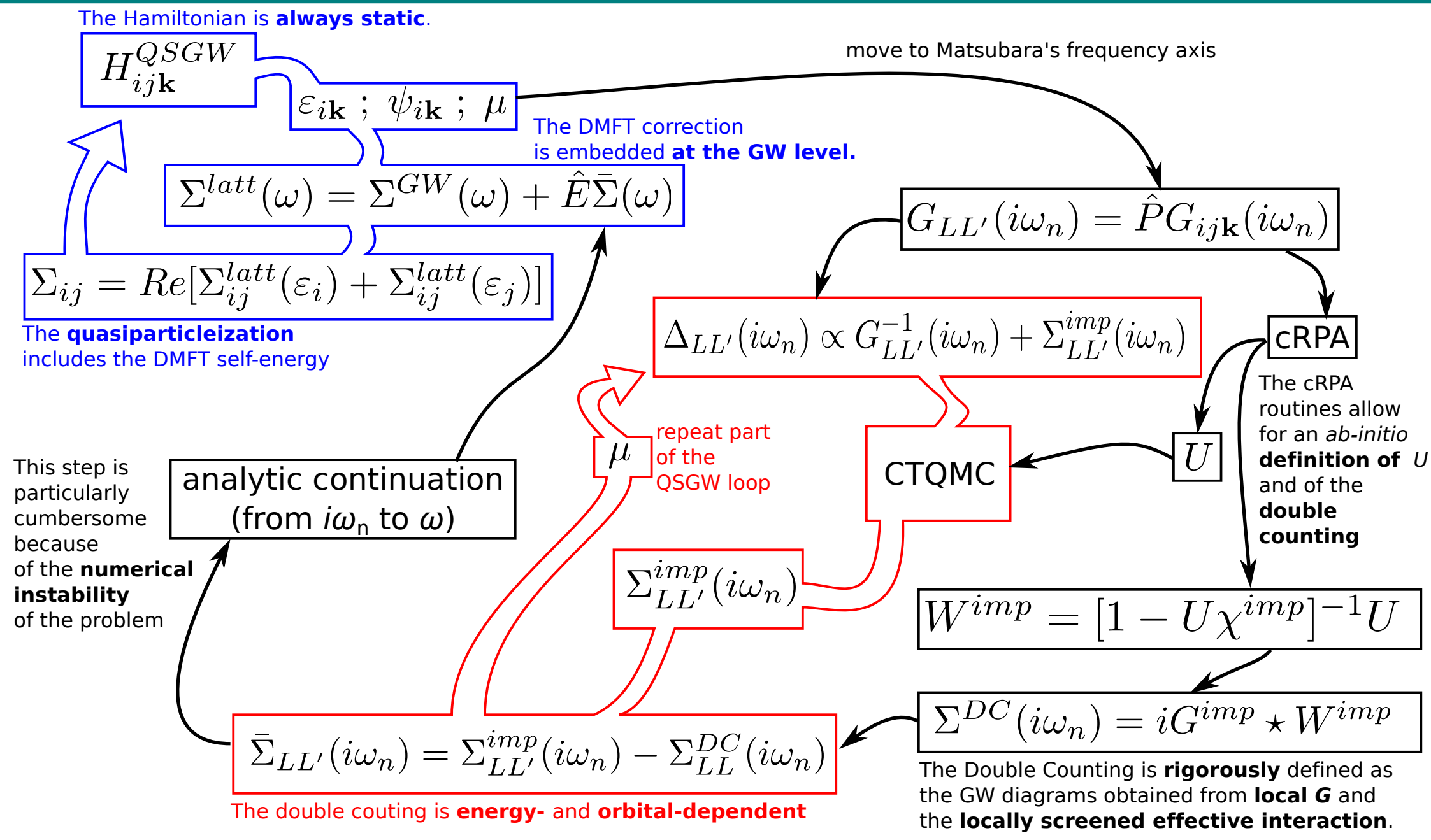




LDA (dotted curves)
QSGW (solid lines
EXP (diamonds and circles)

[10] L.S., P. Pisanti, A. Vishina, D. Pashov, C. Weber, M. van Schilfgaarde, S. Acharya and G. Kotliar, Phys. Rev. B **95**, 041112(R) (2017)

The full QSGW+DMFT scheme (to be done)



Conclusions on the QSGW+DMFT scheme

Advantages of the QSGW+DMFT scheme

- **Non-local correlations** included at the RPA level through GW
- **More accurate** representation of the electron bath
- **Double counting** terms can be defined rigorously

Open questions (related to each other)

- Which **projection scheme** is preferable (and for which scope?)
- **Static or dynamic U?**
- Is **RPA accurate enough** for non-local correlations [9]?

...

The implementation in Questaal

- Better starting point than LDA
- No cRPA for U, but there is some part of it
- So far, standard double counting, but other schemes are possible
- Loop closes on the density instead of the self-energy (more natural in MBPT)

...

[9] J. M. Tomczak, M. van Schilfgaarde, G. Kotliar, Phys. Rev. Lett. **109**, 237010 (2012)