High-precision DMFT algorithms for double perovskite models

Nils Blümer

Outline

Double perovkite models and 1-band Hubbard model

Dynamical mean-field theory (DMFT)

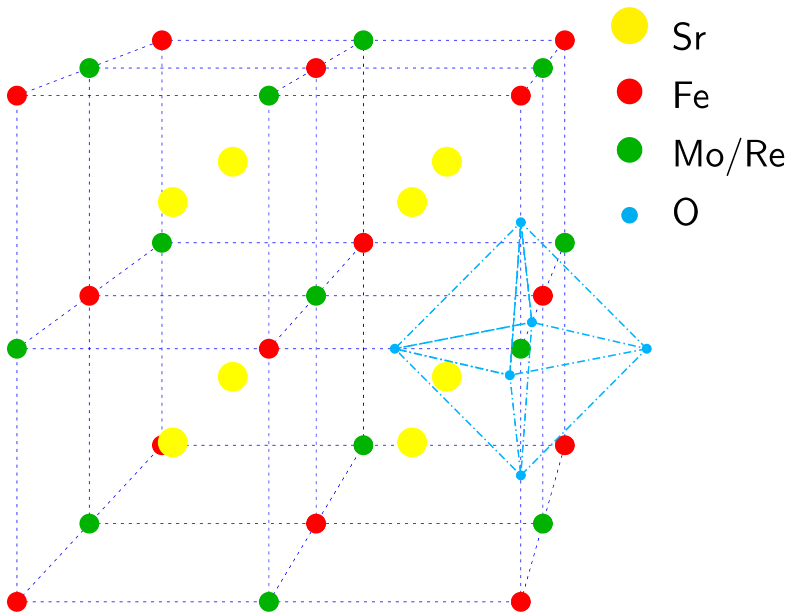
Quantum Monte Carlo (QMC) approach – improved QMC algorithm

Extreme-precision QMC results for V_2O_3 model

Summary and Outlook

Double perovkites models

Sr_2FeMoO_6 and Sr_2FeReO_6



Valences:	Sr^{2+}	[Kr]
	Fe^{3+}	[Ar] $3d^5$
	Mo^{5+}	[Kr] $4d^1$
	O^{2-}	[Ne]

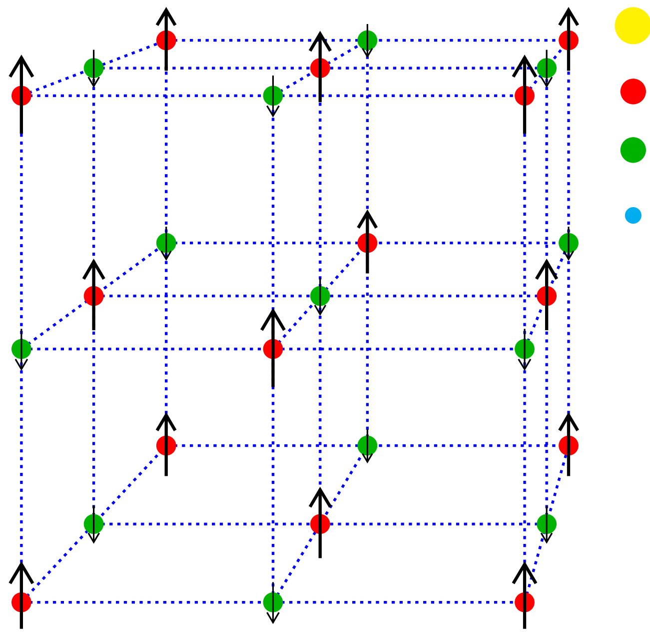
Cubic symmetry: $d \rightsquigarrow$ 3 degenerate t_{2g} bands,
2 degenerate e_g bands

Hybridization Fe - O - Mo/Re

Net ferromagnetic spin polarization, $T_c \approx 400$ K

Double perovkites models

Sr_2FeMoO_6 and Sr_2FeReO_6



Valences:	Sr^{2+}	[Kr]
	Fe^{3+}	[Ar] $3d^5$
	Mo^{5+}	[Kr] $4d^1$
	O^{2-}	[Ne]

Cubic symmetry: $d \rightsquigarrow$ 3 degenerate t_{2g} bands,
2 degenerate e_g bands

Hybridization Fe - O - Mo/Re

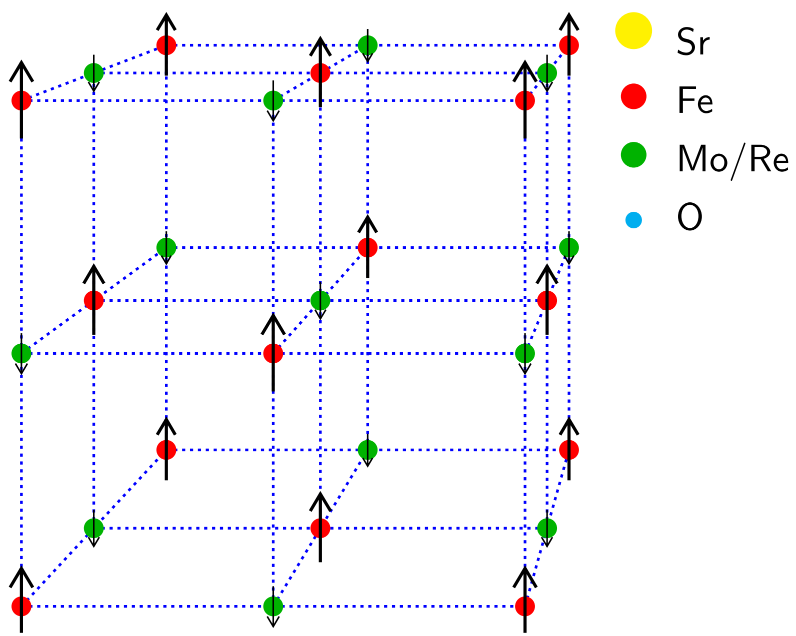
Net ferromagnetic spin polarization, $T_c \approx 400$ K

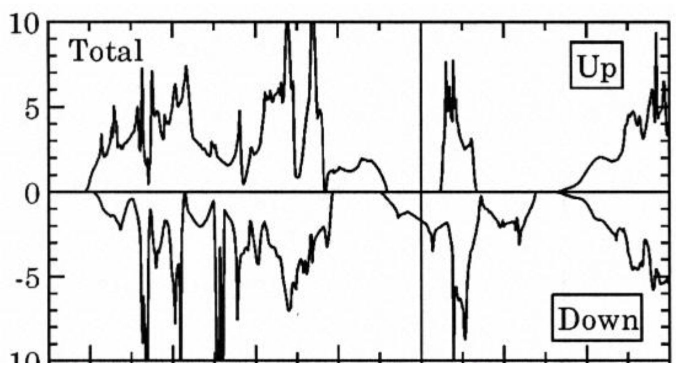
Calculations: total system ferrimagnetic ($\sim$ AF)

Large magnetoresistance for ordered probes

Double perovkites models

Sr_2FeMoO_6 and Sr_2FeReO_6





[LSDA+U for Sr_2FeMoO_6, Saitoh et al. (2002)]

Valences:	Sr^{2+}	[Kr]
	Fe^{3+}	[Ar] $3d^5$
	Mo^{5+}	[Kr] $4d^1$
	O^{2-}	[Ne]

Cubic symmetry: $d \rightsquigarrow$ 3 degenerate t_{2g} bands,
2 degenerate e_g bands

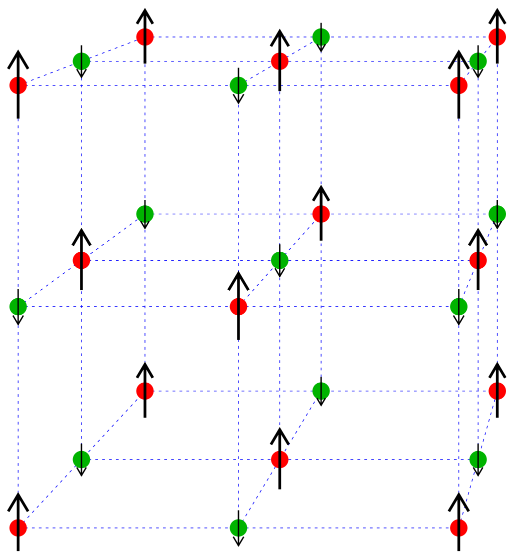
Hybridization Fe - O - Mo/Re

Net ferromagnetic spin polarization, $T_c \approx 400$ K

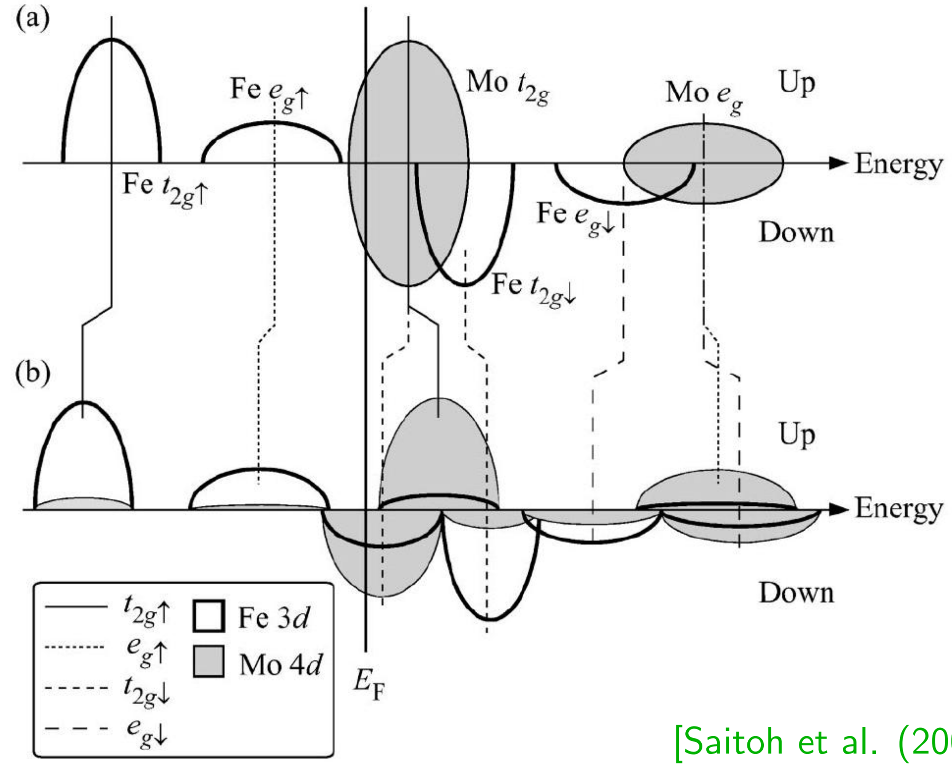
Calculations: total system ferrimagnetic ($\sim$ AF)

Large magnetoresistance for ordered probes

Calculations/heuristics: systems half-metallic
($\sim$ Mott insulator)



Scheme: hybridization of hypothetical ionic states



[Saitoh et al. (2002)]

Microscopic theory (5-band model)

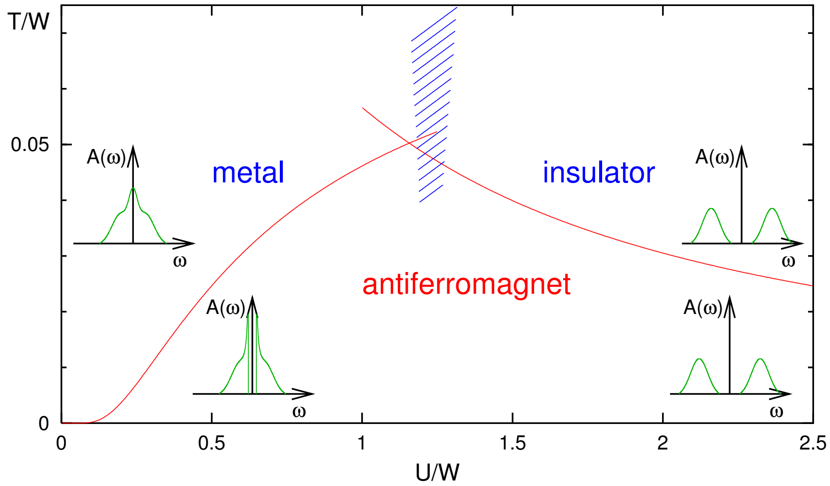
$\alpha \equiv (\nu, \sigma)$

$$H = \epsilon^f \sum_{i\alpha} n^f_{i\alpha} + \epsilon^m \sum_{i\alpha} n^m_{i\alpha} + \sum_{i,\,\alpha\neq\alpha'} U^f_{\alpha\alpha'}\, n^f_{i\alpha} n^f_{i\alpha'} + \sum_{j,\,\alpha\neq\alpha'} U^m_{\alpha\alpha'}\, n^m_{j\alpha} n^m_{j\alpha'}$$
$$+ \sum_{\langle ij\rangle\,\alpha} t^{fm}\,(f^\dagger_{i\alpha} m_{j\alpha} + \text{hc}) + \sum_{\langle jj'\rangle\,\alpha} t^{mm}\, m^\dagger_{j\alpha} m_{j'\alpha} + \sum_{\langle ii'\rangle\,\alpha} t^{ff}\, f^\dagger_{i\alpha} f_{i'\alpha}$$

$t^{fm} \approx 0.25\text{eV} \quad > \quad t^{mm} \approx 0.15\text{eV} \quad \gg \quad t^{ff} \approx 0.03\text{ eV}$ [Phillips et al. (2003)]

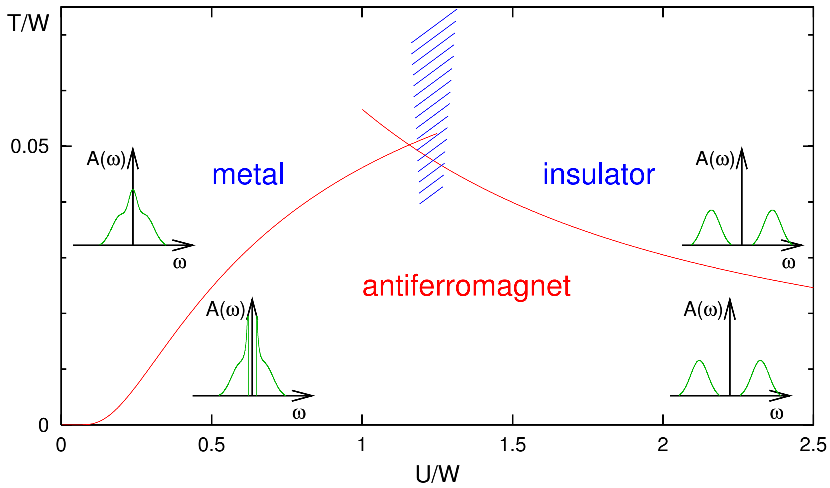
Related physics of 1-band Hubbard models

Half filling $\rightsquigarrow$ AF, MI-crossover

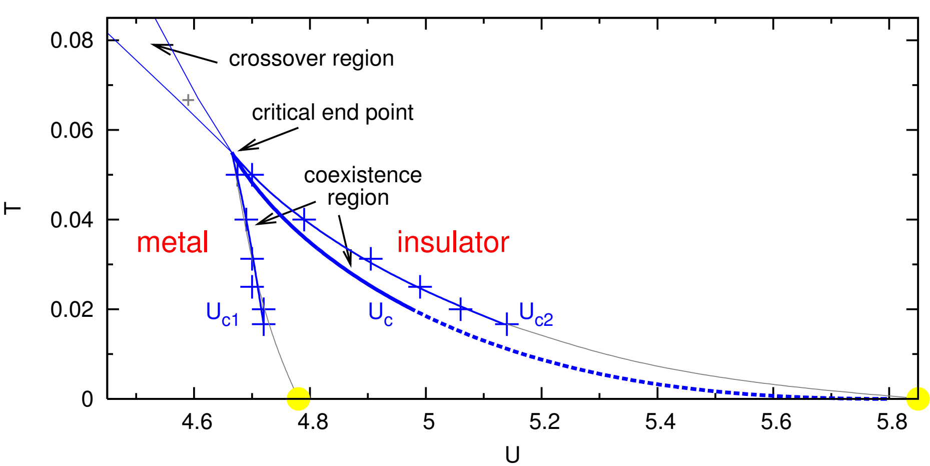


Related physics of 1-band Hubbard models

Half filling ⇝ AF, MI-crossover

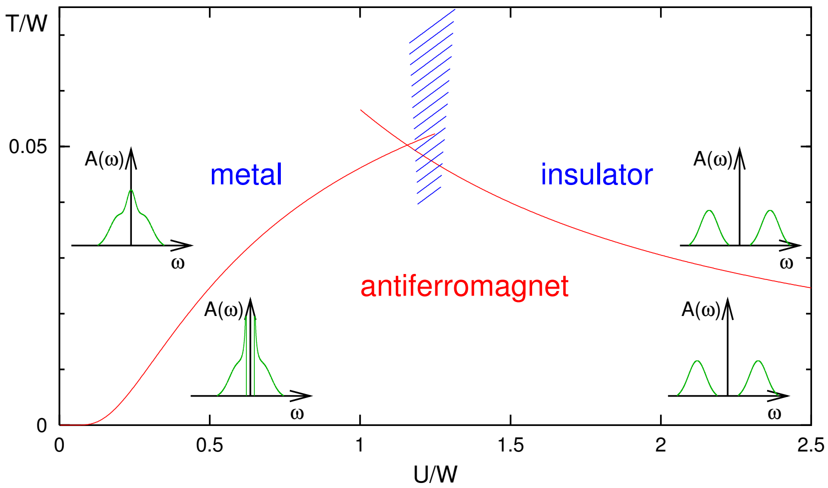


Frustration ⇝ Metal-insulator transition

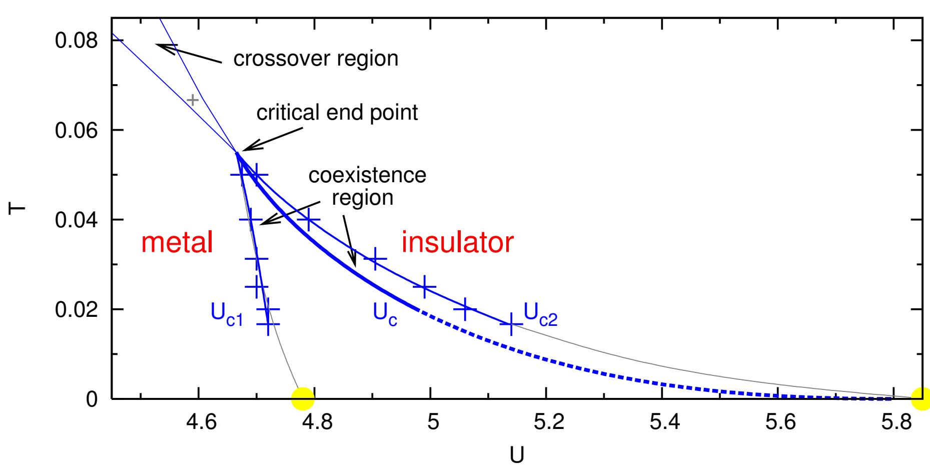


Related physics of 1-band Hubbard models

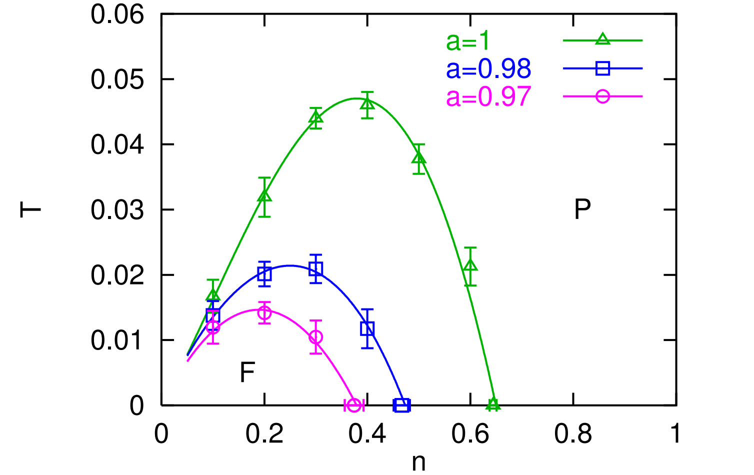
Half filling ⇝ AF, MI-crossover



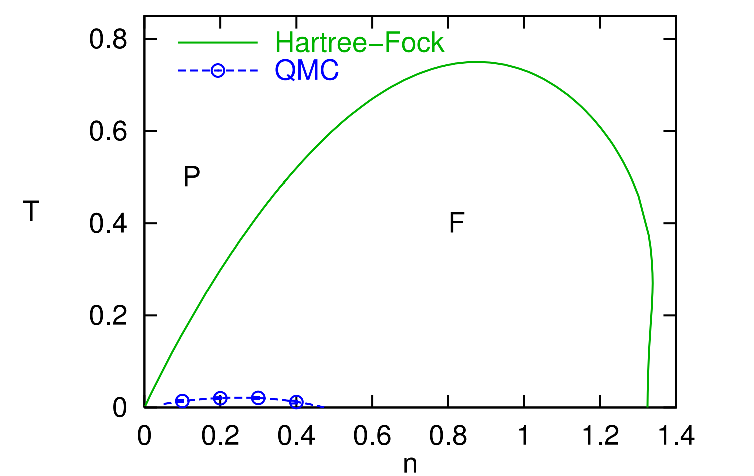
Frustration ⇝ Metal-insulator transition



Asymmetric DOS ⇝ ferromagnet



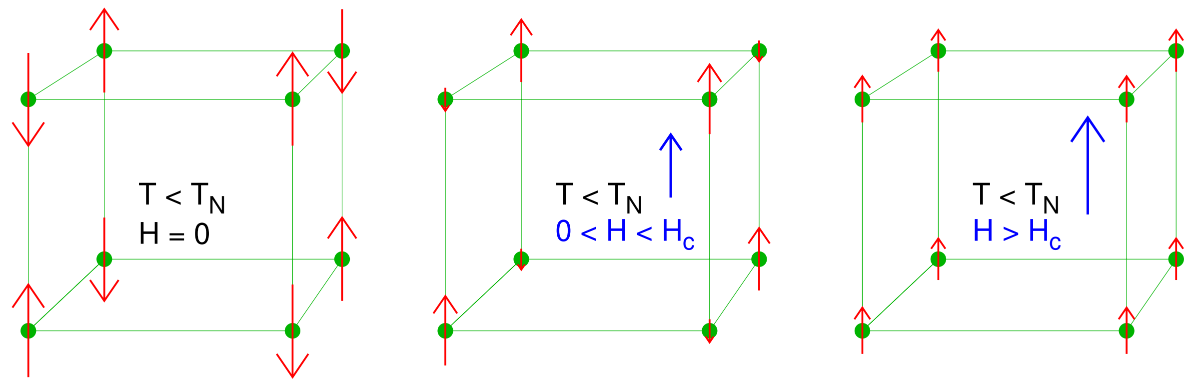
Curie temperature T_C vs. doping n for $U = W$



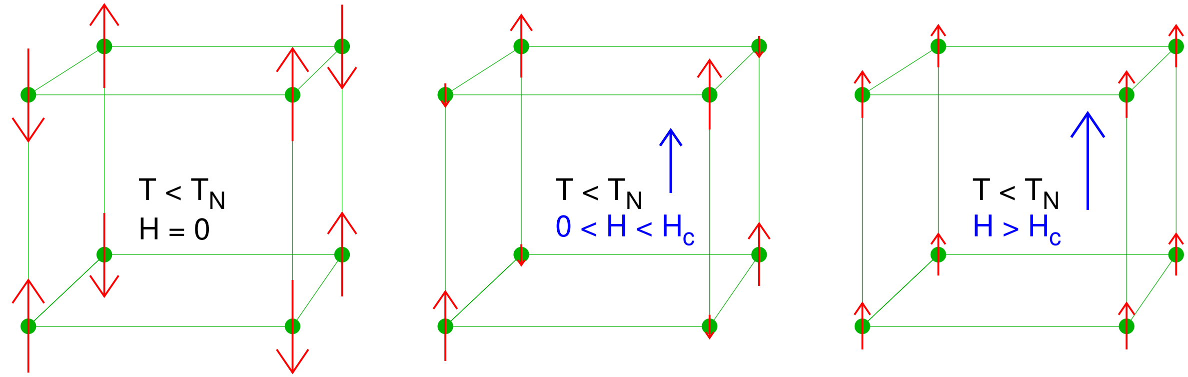
T_C vs n for $U = W = 4$ and $a = 0.98$

[Wahle, NB, Schlipf, Held, Vollhardt (1998)]

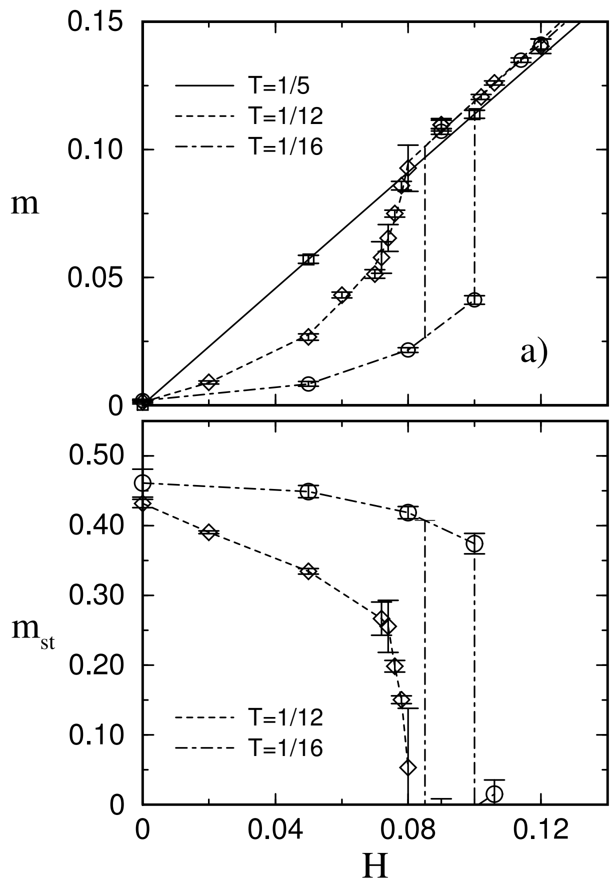
Metamagnetism

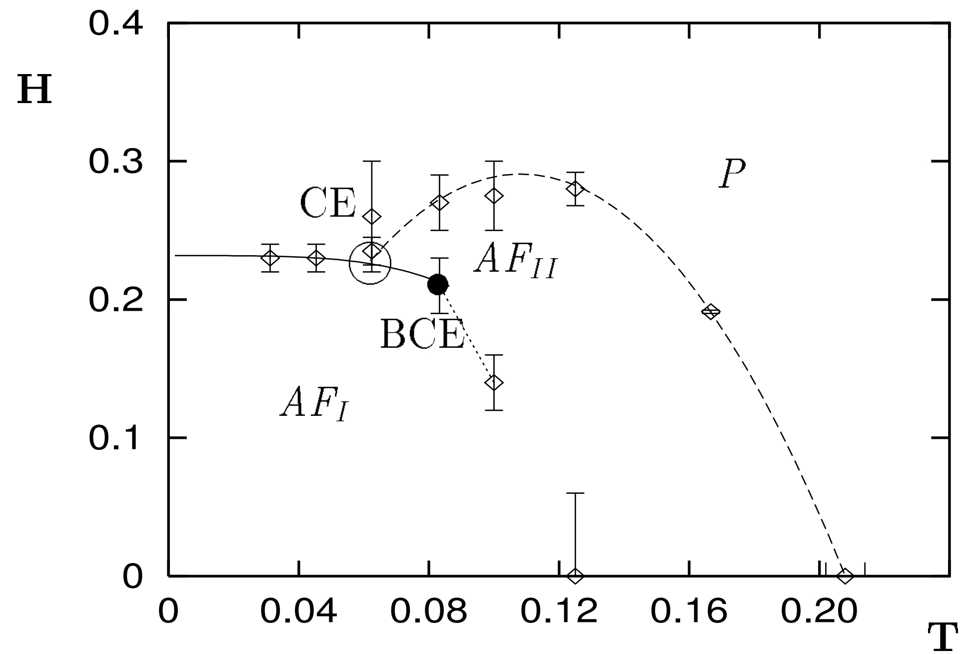


Metamagnetism



Hubbard model with
easy axis, $n=1$, $U=2$





Only DMFT-QMC captures 1^{st} and 2^{nd} order!

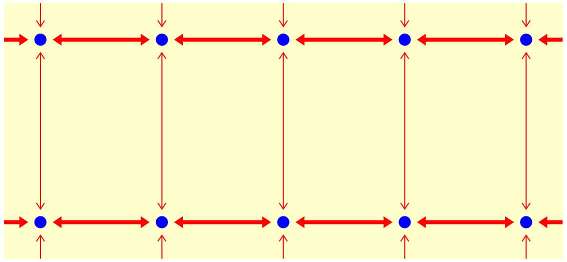
[K. Held, M. Ulmke, NB, D. Vollhardt (1997)]

Dynamical mean-field theory (DMFT)

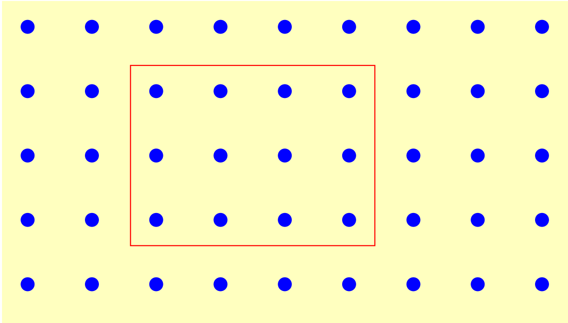
How to solve Hubbard-type models?

$$\hat{H} = \sum_{(i,j),\sigma} t_{ij}(\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + \text{h.c.}) + U\sum_{i}\hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$

$d = 1$: Bethe ansatz, DMRG



finite clusters: ED, QMC



Perturbation theory, e.g.

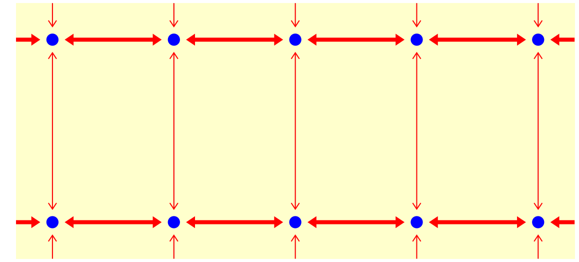
- $U \longrightarrow 0$: Hartree-Fock, 2[nd] order PT, . . .
- $t/U \longrightarrow 0$ at half filling $\rightsquigarrow$ Heisenberg model
- . . .

Dynamical mean-field theory (DMFT)

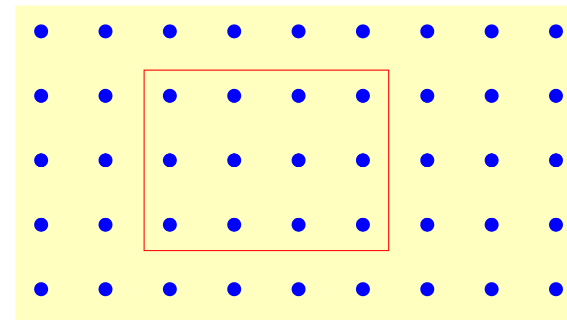
How to solve Hubbard-type models?

$$\hat{H} = \sum_{(i,j),\sigma} t_{ij} (\hat{c}^{\dagger}_{i\sigma} \hat{c}_{j\sigma} + \text{h.c.}) + U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

$d = 1$: Bethe ansatz, DMRG



finite clusters: ED, QMC



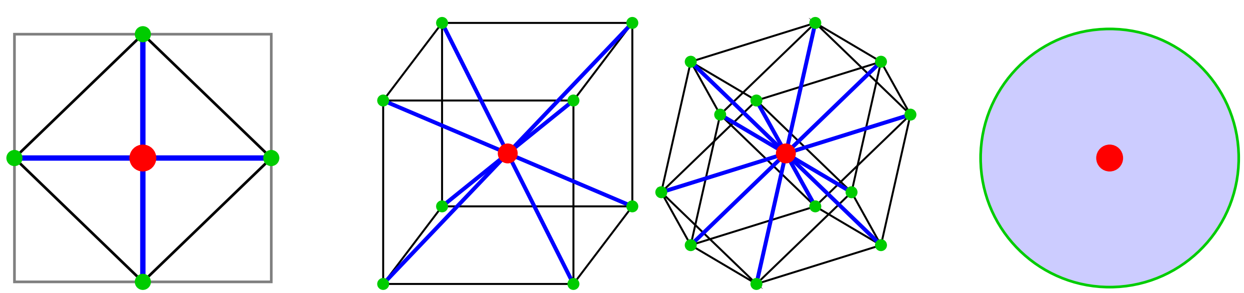
Perturbation theory, e.g.

- $U \longrightarrow 0$: Hartree-Fock, 2^{nd} order PT, ...
- $t/U \longrightarrow 0$ at half filling $\rightsquigarrow$ Heisenberg model
- ...

Dynamical mean-field theory (DMFT): local self-energy $\Sigma(\boldsymbol{k}, \omega) \equiv \Sigma(\omega)$

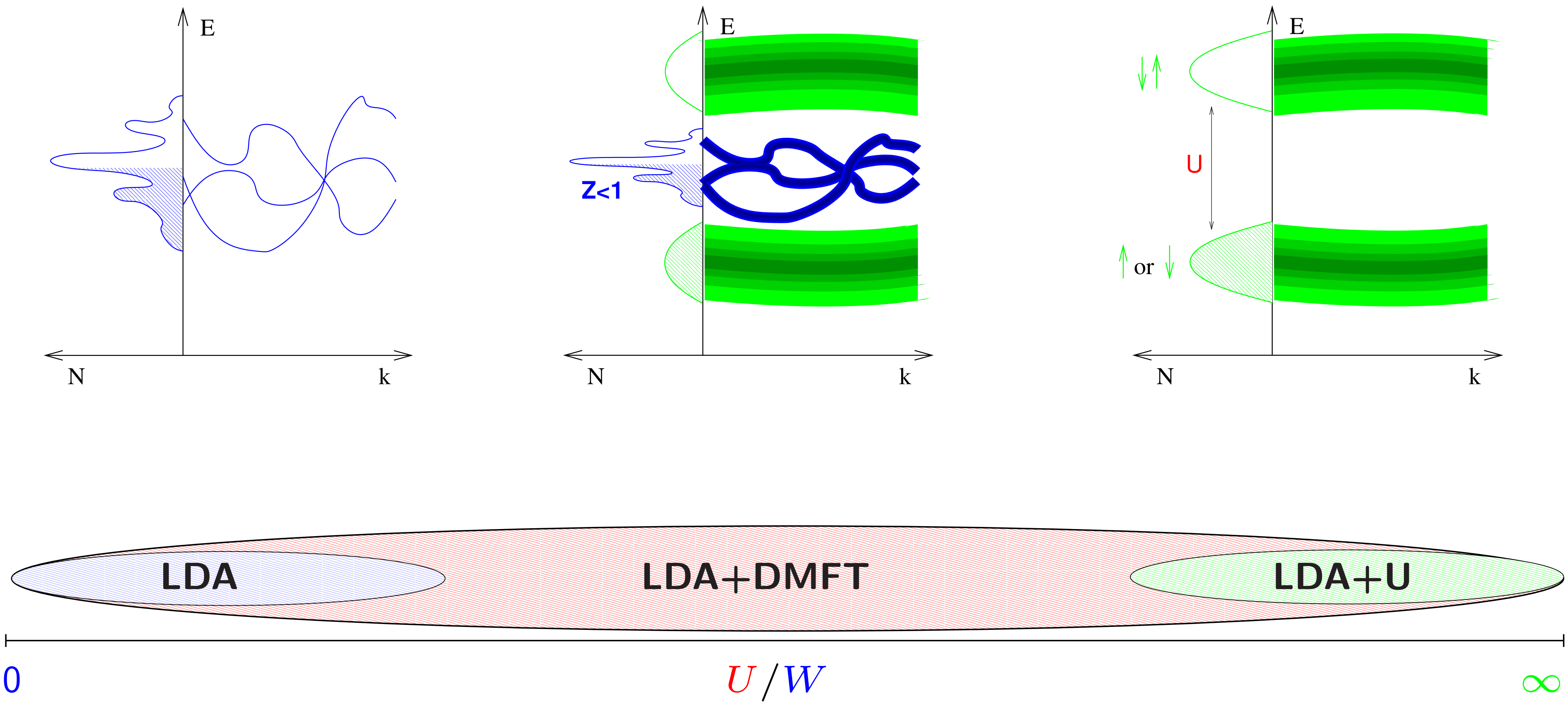
+ non-perturbative
  $\rightsquigarrow$ valid at MIT
+ dynamical on-site correlations preserved
+ in thermodynamic limit

+/− exact for $Z \to \infty$



d=2: $Z = 4$ bcc: $Z = 8$ fcc: $Z = 12$ DMFT: $Z = \infty$

Realistic band structure calculations: LDA+DMFT



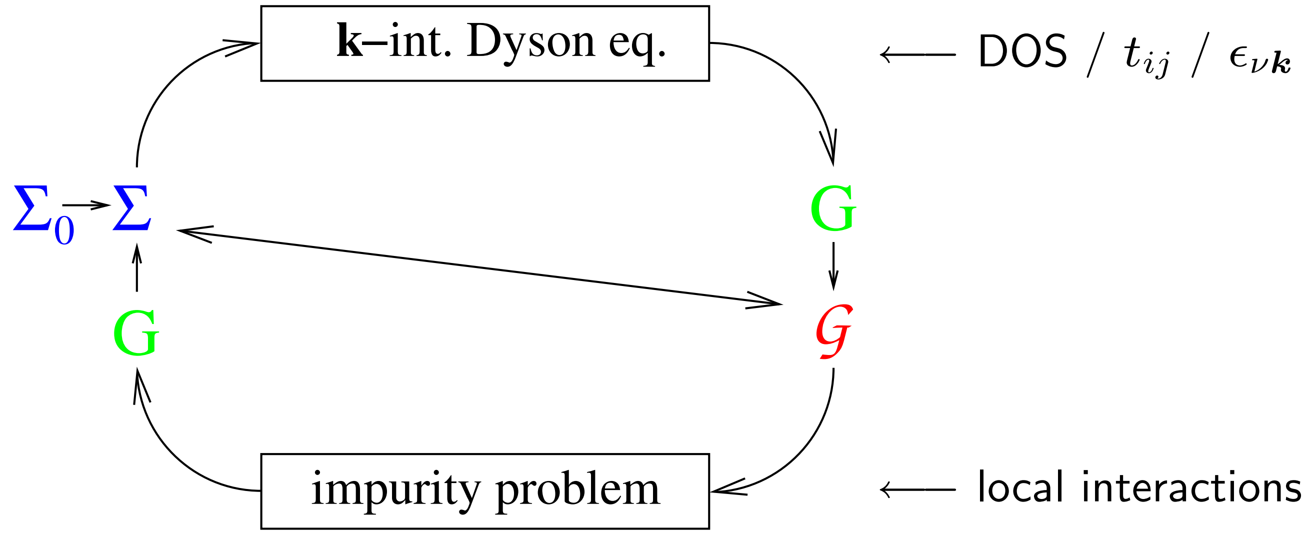
[Held (2004)]

Important: complex, frequency-dependent self-energy $\Sigma(\omega)$

Often less important: momentum-dependence of self-energy (neglected by DMFT)

Iterative solution of

DMFT equations



Impurity solver:

- Quanten-Monte-Carlo (QMC)
- Iterative perturbation theory (IPT; not controlled)
- Non-crossing approximation (NCA; not controlled)
- Exact diagonalization (ED; large finite-size errors)
- Numerical renormalization group (NRG; 1-2 bands)
- Density matrix renormalization group (DMRG)
- Self-energy functional theory (SFT) + ED

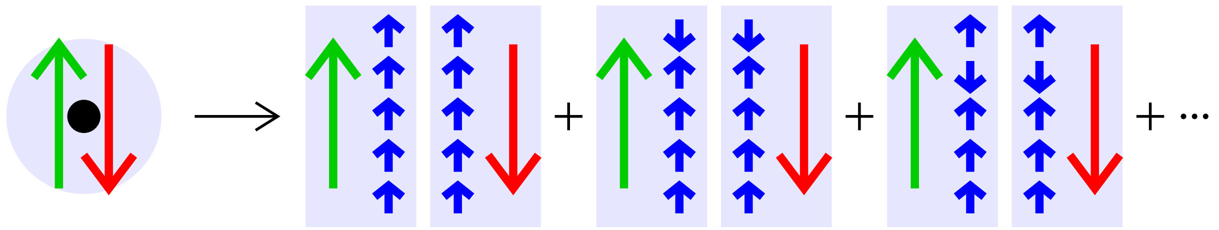
Direct $d = \infty$ solution: PT, ePT

Quantum Monte Carlo (QMC) approach

Wanted: Green-Funktion $G(\omega)$

Treatment in imaginary time using fermionic Grassmann variables ψ, ψ^*,

discretization $\beta = \Lambda \, \Delta\tau$, Trotter decoupling, discrete Hubbard-Stratonovich transformation



Wick theorem:

$$G = \frac{\sum M \det\{M\}}{\sum \det\{M\}}$$

Metropolis MC importance sampling over auxiliary Ising field, 2^Λ configurations, $50 \lesssim \Lambda \lesssim 400$

$+$ nonperturbative, numerically exact

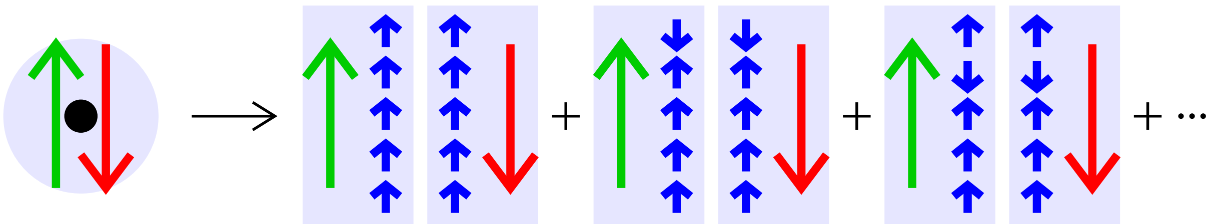
$-$ effort scales as T^{-3}

Quantum Monte Carlo (QMC) approach

Wanted: Green-Funktion $G(\omega)$

Treatment in imaginary time using fermionic Grassmann variables ψ, ψ^*,

discretization $\beta = \Lambda\,\Delta\tau$, Trotter decoupling, discrete Hubbard-Stratonovich transformation



Wick theorem:

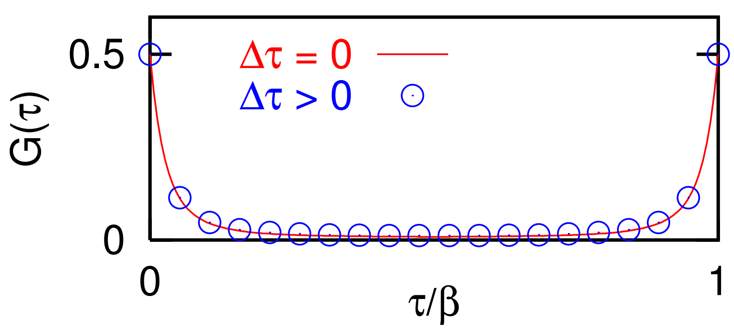
$$G = \frac{\sum M \det\{M\}}{\sum \det\{M\}}$$

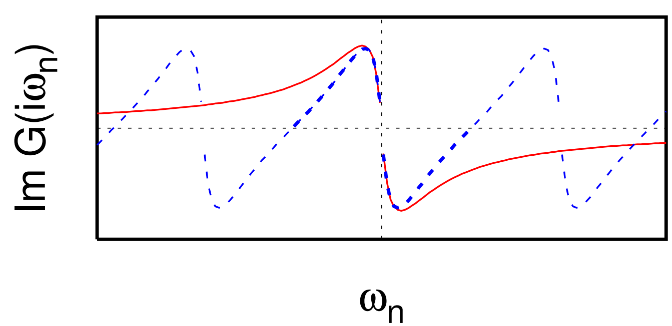
Metropolis MC importance sampling over auxiliary Ising field, 2^Λ configurations, $50 \lesssim \Lambda \lesssim 400$

$+$ nonperturbative, numerically exact

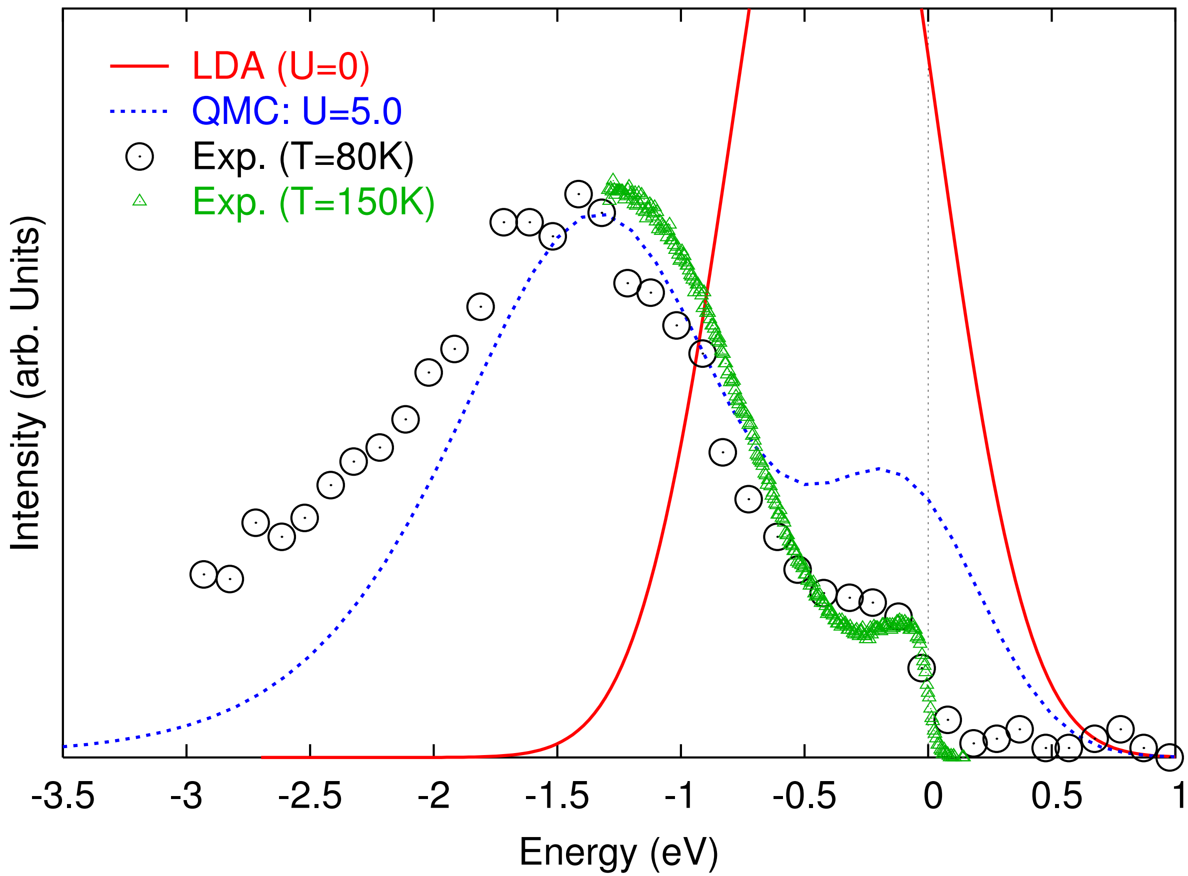
$-$ effort scales as T^{-3}

$-$ no information for $\omega \gtrsim \omega_{\text{Nyquist}}$ $\rightsquigarrow$ problem in Fourier transformation:





Application: LDA+DMFT photoemission spectra for $La_{1-x}Sr_xTiO_3$ (x=0.06)



[Nekrasov, Held, NB, Poteryaev, Anisimov, Vollhardt, (2000)]

Reasonable accuracy, drastic improvement over LDA

But: old code not accurate near MIT $\rightsquigarrow$ possible problems for half-metallic systems

High algorithmic efficiency a must for realistic double perovskite calculations!

Improved QMC algorithm with analytical high-frequency corrections

Really numerically exact: arbitrary precision for

1. large number of spin updates (e.g. 10^6)
2. large number of DMFT iterations (e.g. 40)
3. small minimal discretization (e.g. range $0.1 \leq \Delta\tau \leq 0.3$)
4. high precision even in extrapolation $T \to 0$

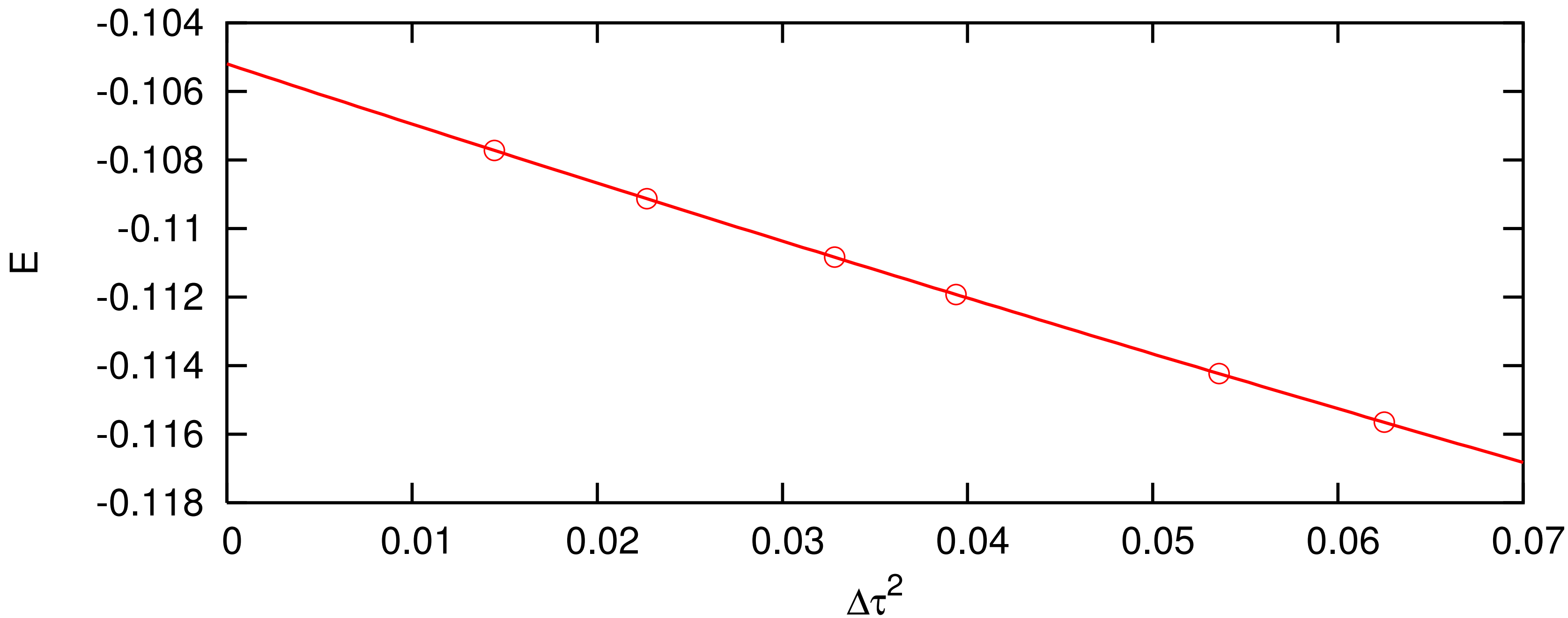
Improved QMC algorithm with analytical high-frequency corrections

Really numerically exact: arbitrary precision for

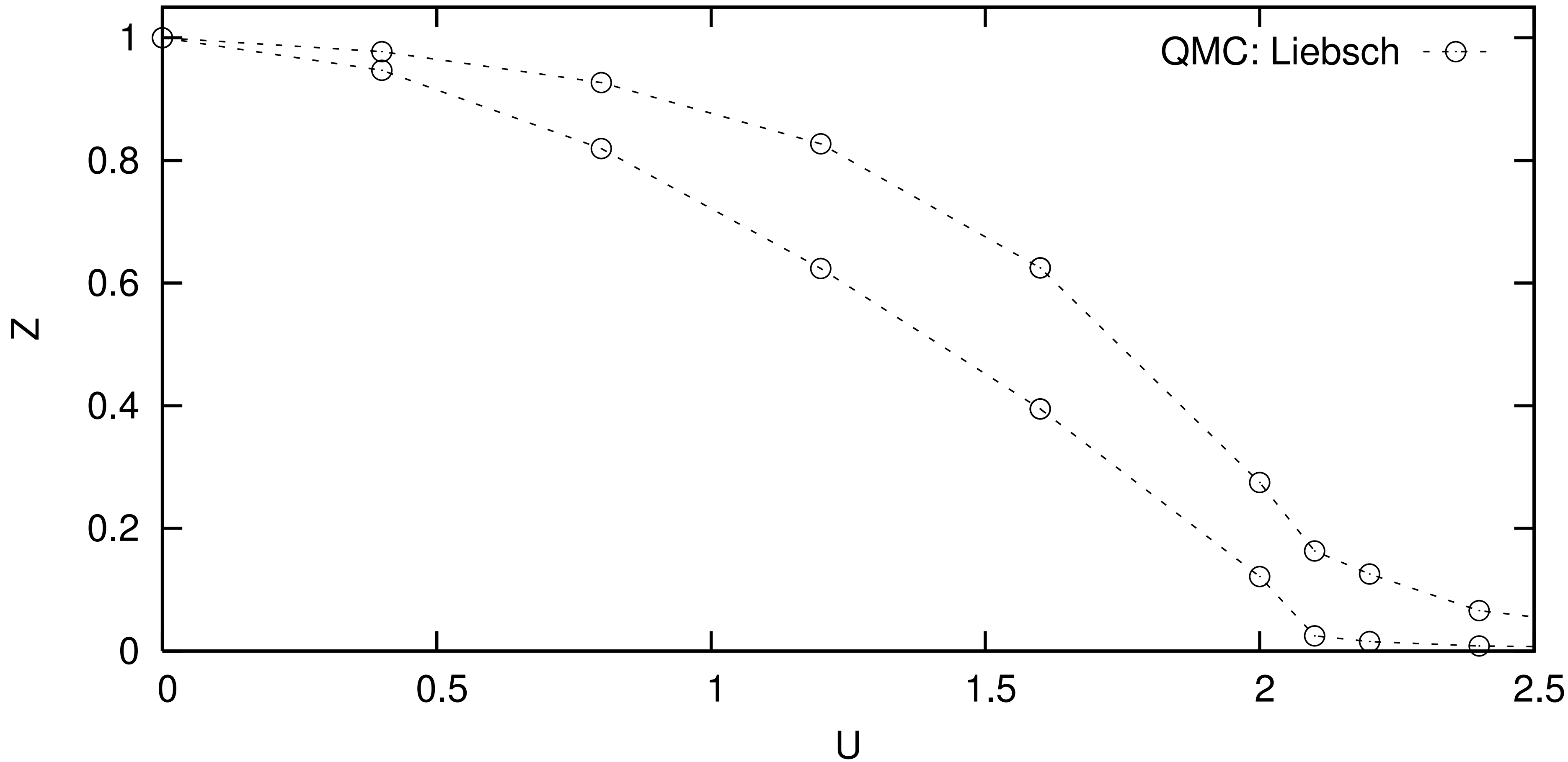
1. large number of spin updates (e.g. 10^6)
2. large number of DMFT iterations (e.g. 40)
3. small minimal discretization (e.g. range $0.1 \leq \Delta\tau \leq 0.3$)

4. high precision even in extrapolation $T \to 0$

Example: $\Delta\tau$ extrapolation of energy for frustrated 1 band model, $U = 5$, $T = 0.04$

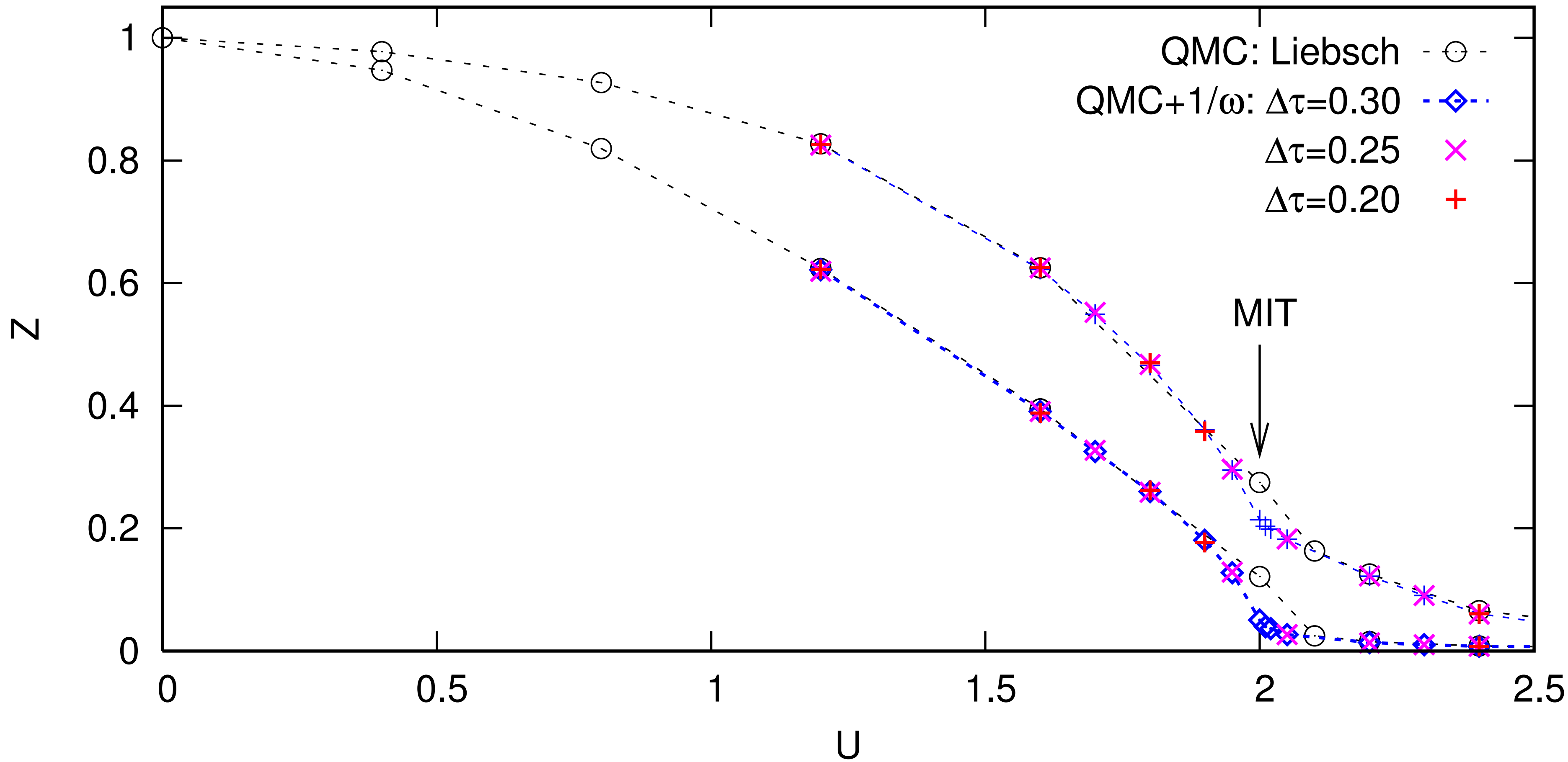


Application/test: quasiparticle weight $Z = m/m*$ of anisotropic 2-band model



[Comparison with Liebsch, PRB **70**, 165103 (2004)]

Application/test: quasiparticle weight $Z = m/m*$ of anisotropic 2-band model



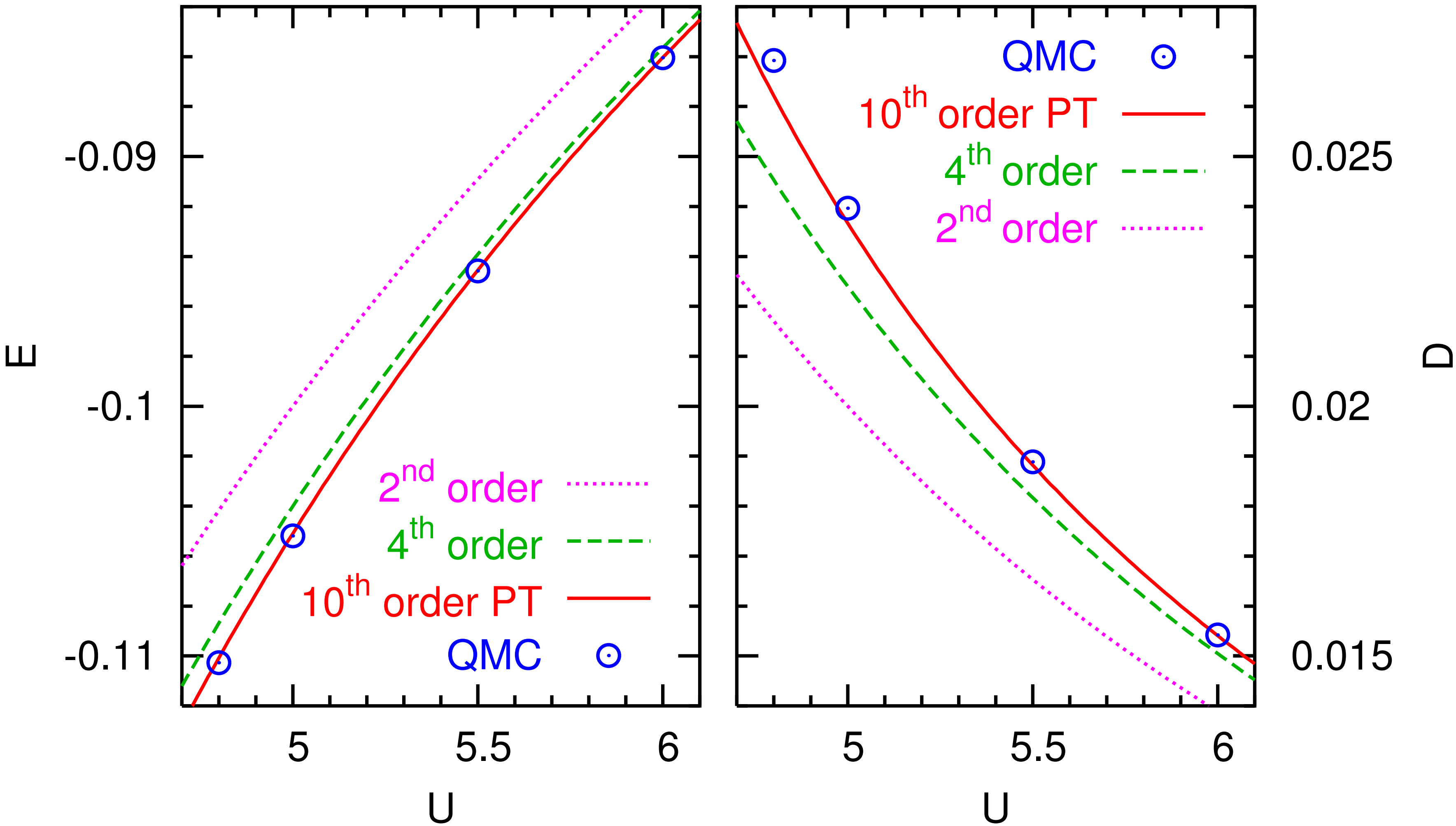
[Comparison with Liebsch, PRB **70**, 165103 (2004)] [C. Knecht, unpublished]

New algorithm clearly exposes (single) metal-insulator transition (MIT)

Very small dependence on discretization $\Delta\tau$.

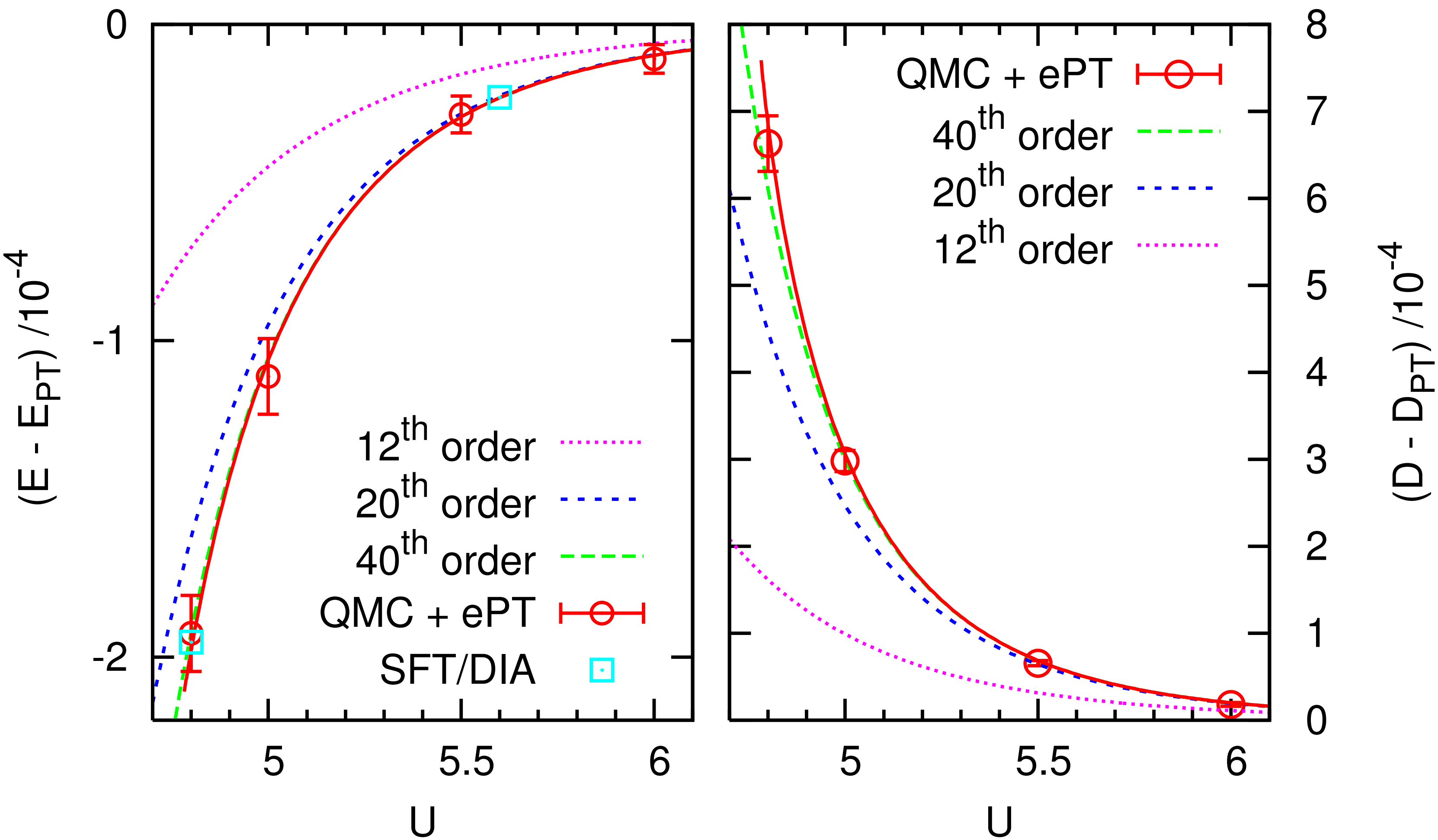
Extreme-precision QMC results for V_2O_3 model

QMC vs. strong-coupling PT for insulating phase



Excellent agreement at $U = 6.0$, deviations below.

Higher resolution plots: differences w.r.t. 10^{th} order PT

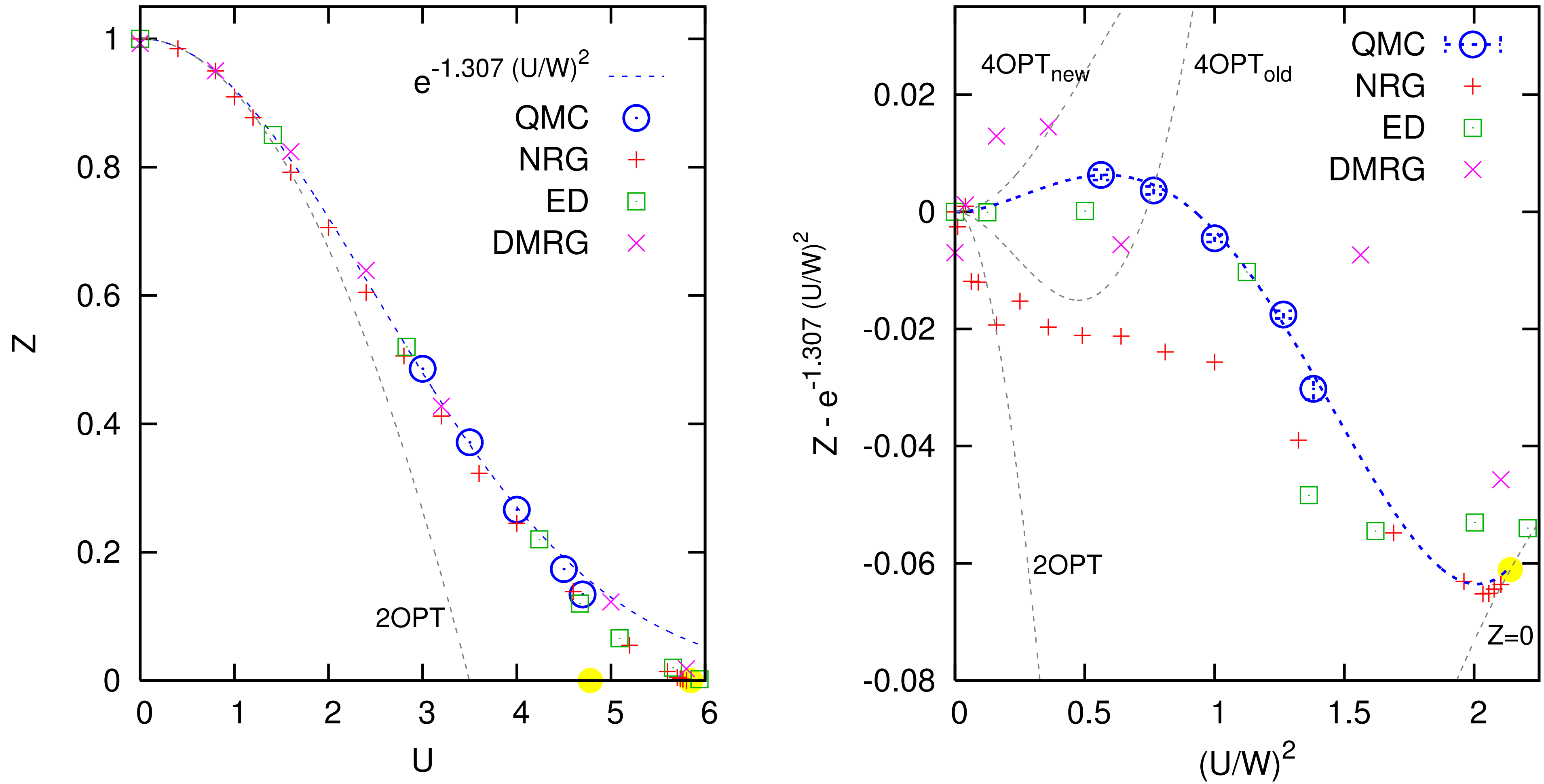


ePT: extrapolation of PT to infinite order [NB, Kalinowski, Phys. Rev B, in press]

$\rightsquigarrow$ critical interaction U_{c1}, critical exponents, benchmark $(\Delta E \lesssim 10^{-6})$

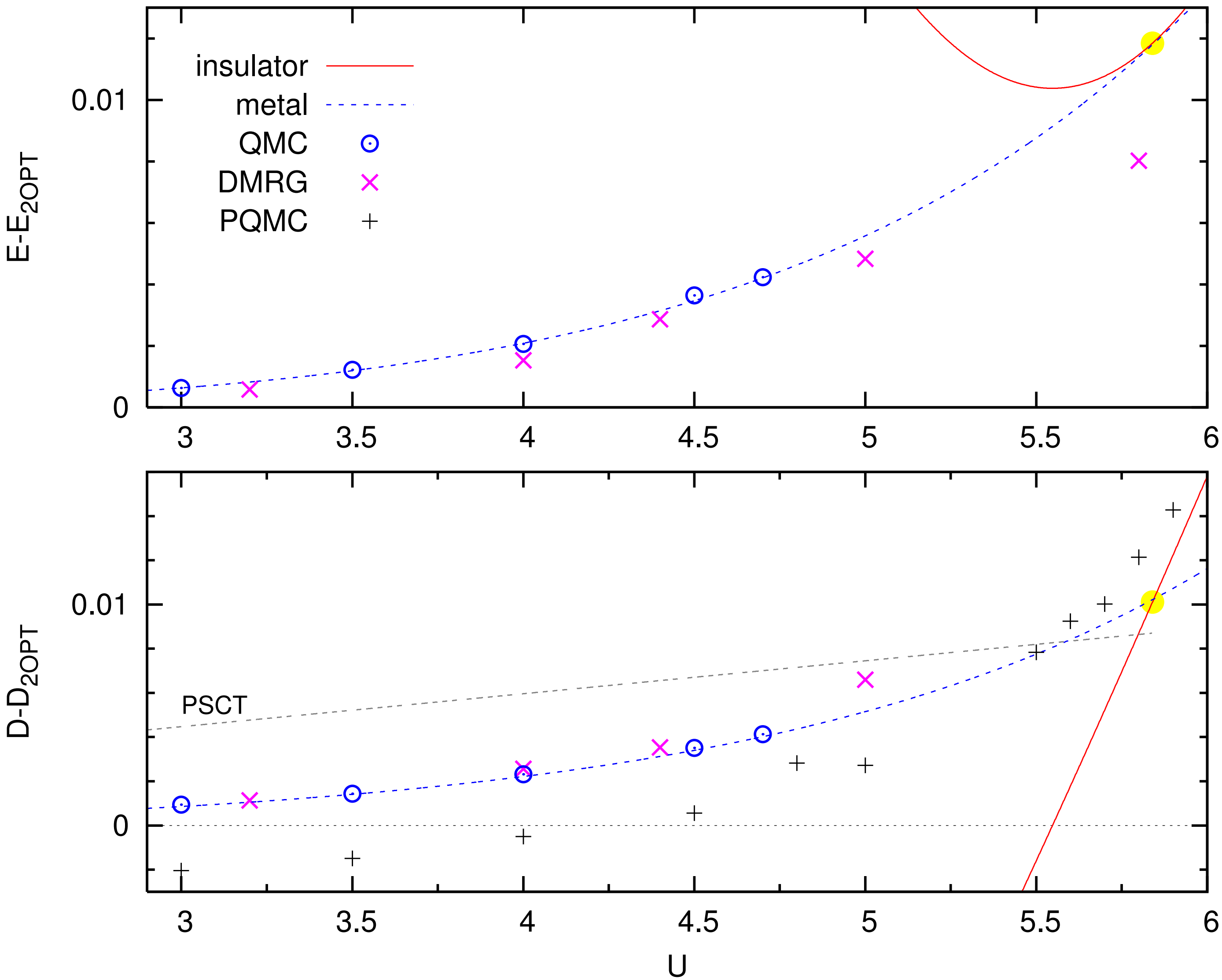
QMC algorithm has passed only available authoritative (1-band) test!

Quasiparticle weight/mass enhancement $Z = m/m^*$ in metal at $T = 0$



T-extrapolated QMC even beats all ground state methods!

Energetics: differences w.r.t. 2^{nd} order weak-coupling PT for E and D



QMC-fit consistent with ePT for insulator; large deviations of PQMC, PSCT.

Summary and Outlook

Modeling of double perovskites

1-band model: similar phenomena and difficulties

Improved quantum Monte Carlo algorithm (QMC$+1/\omega$)

Extreme precision for V_2O_3 model, even at $T = 0$

Important correlated electron model finally well understood (new: exponents)

Only other method that can touch QMC accuracy: self-energy functional theory

Summary and Outlook

Modeling of double perovskites

1-band model: similar phenomena and difficulties

Improved quantum Monte Carlo algorithm (QMC$+1/\omega$)

Extreme precision for V_2O_3 model, even at $T = 0$

Important correlated electron model finally well understood (new: exponents)

Only other method that can touch QMC accuracy: self-energy functional theory

Strategy for DPs: Treat correlations for Fe ($\sim$ multi-band "PAM")

First: calculations for fixed e_g spin ($\rightsquigarrow$ 3 bands $+$ int. field)

Later: Mo/Re correlations, 5-band model