LDA+DMFT study for LiV_2O_4 at T→0

Phys. Rev. Lett. 98 166402 (2007)

Ryotaro ARITA (RIKEN)

Collaborators

K. Held
(Max Planck Institute, Stuttgart)

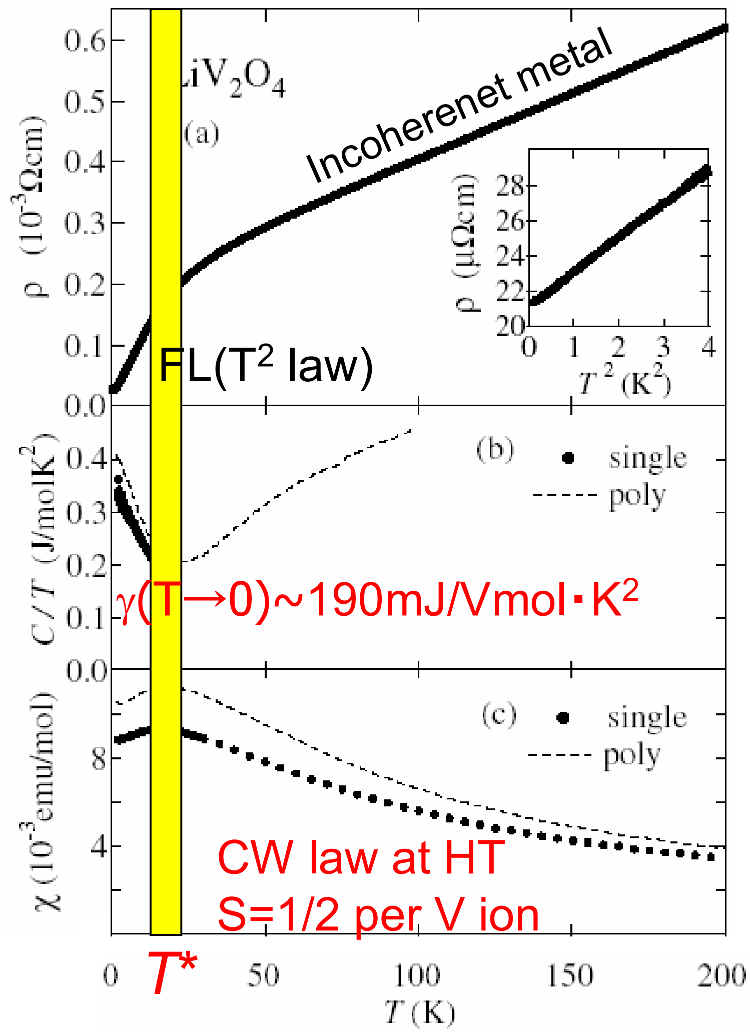
A.V. Lukoyanov
(Ural State Technical Univ.)

V.I. Anisimov
(Institute of Metal Physics)

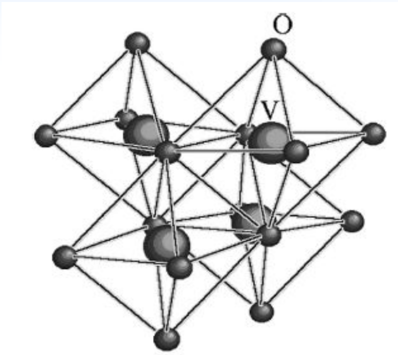
Outline

- Introduction
  - LiV_2O_4: 3d heavy Fermion system
    heavy (sharp) quasi-peak in $A(\omega)$ for low T
- Development of PQMC
  - QMC for $T \rightarrow 0$
  - Application to DMFT calculation
- Results
  - LDA+DMFT(PQMC) for LiV_2O_4
- Discussion
  - Origin of HF behaviors

LiV_2O_4: 3d heavy Fermion system



(Urano et al. PRL85, 1052(2000))



Crossover at T^*~20K

- resistivity: $\rho = \rho_0 + AT^2$ with an enhanced A
- specific heat coefficient: anomalously large
  $\gamma(T \to 0)$~190mJ/V mol･K^2
  cf) $CeRu_2Si_2$ ~350mJ/Ce mol･K^2
  UPt_3 ~420mJ/U mol･K^2
  (Kadowaki-Woods relation satisfied)
- χ: broad maximum (Wilson ratio～1.8)

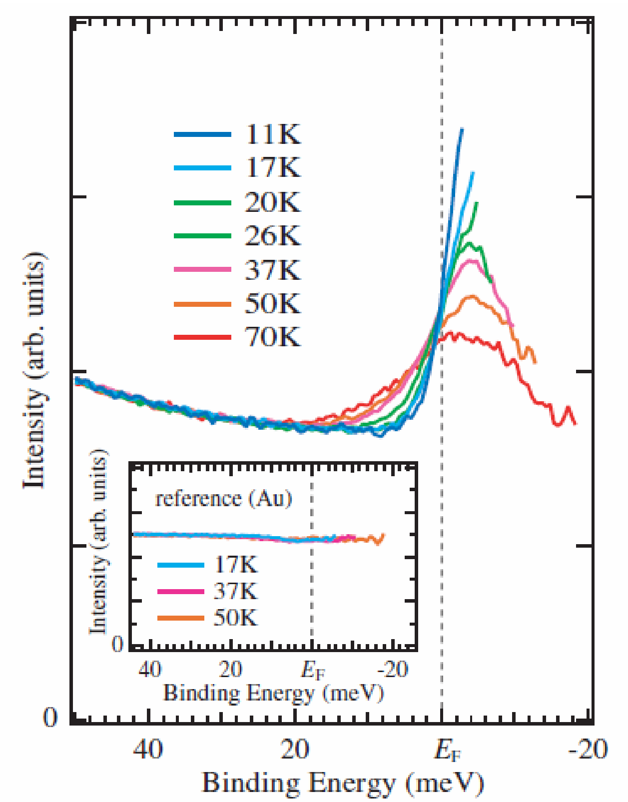
T^*･･ onset of the formation of the heavy mass quasiparticles (m^*～$25m_{LDA}$)

R. Arita

LiV_2O_4: 3d heavy Fermion system

PhotoEmission Spectroscopy

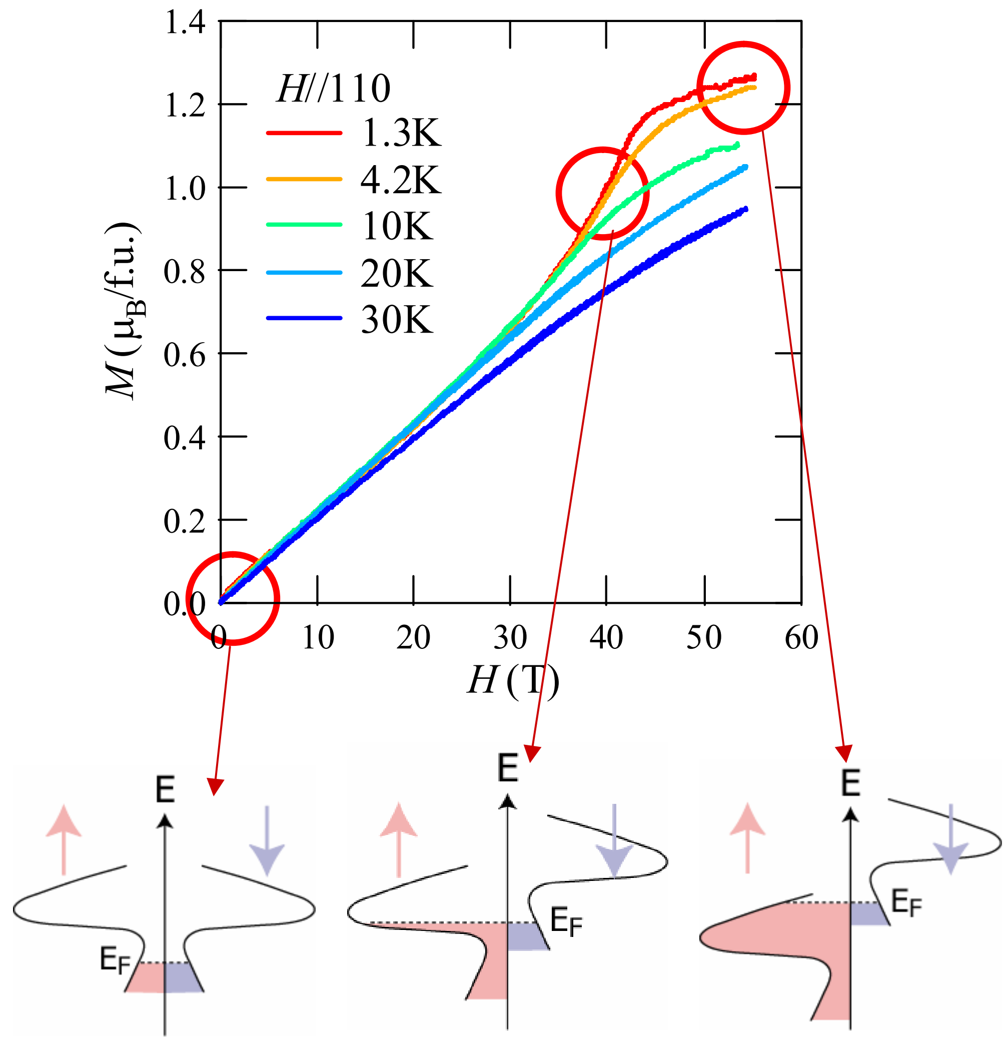
(Shimoyamada et al. PRL 96 026403(2006))



A sharp peak appears
for T<26K
ω=4meV, Δ~10meV

Magnetization curves

(Niitaka et al. '06)

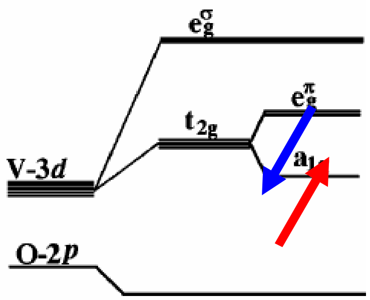


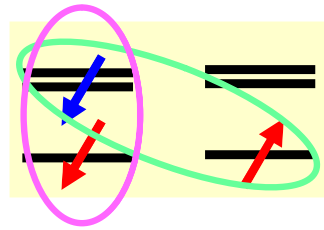
Theoretical studies so far

- Anisimov et al, 99
- Eyert et al, 99
- Matsuno et al, 99
- Kusunose et al, 00
- Lacroix, 01
- Shannon, 01
- Fulde et al, 01
- Burdin et al, 02
- Hopkinson et al, 02
- Fujimoto, 02
- Tsunetsugu, 02
- Yamashita et al, 03
- Laad et al, 03
- Nekrasov et al, 03
- Yushankhai et al, 07

- Kondo scenario

Anisimov et al, PRL 83 364(1999)



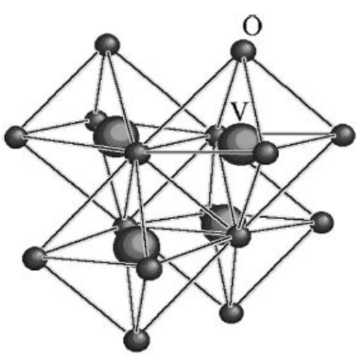


e_g (itinerant)
a_{1g} (localized)

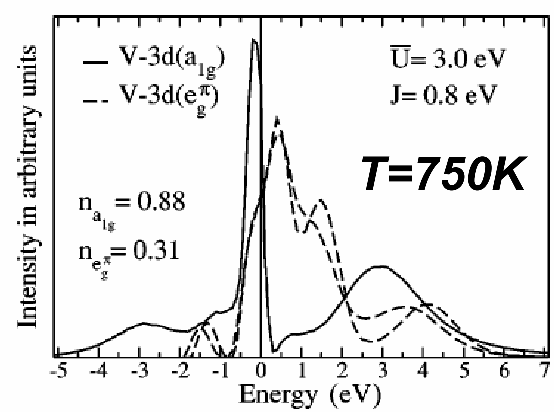
Hund

hybridization
→Kondo?

- Geometrical Frustration



Short range (local) correlations become dominant
⇒ **DMFT** expected to be a good approx.



LDA+DMFT by Nekrasov et al, PRB 67 085111 (2003)

Calculation for T→0

LDA+DMFT(QMC)

- LDA+DMFT
  - Successful beyond-LDA method
    - Applications to various strongly correlated materials

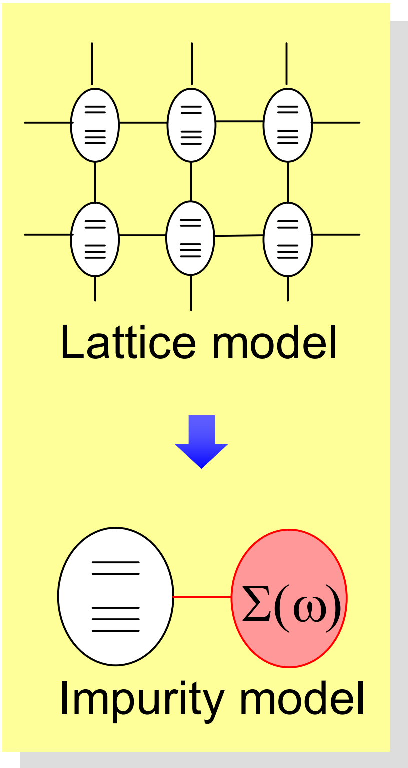
- Solver for the effective impurity model
  - QMC (Hirsch-Fye)
    - Numerically exact
    - Restricted to high *T*

      (Numerically expensive for Low *T*

      effort ~ $1/T^3$)

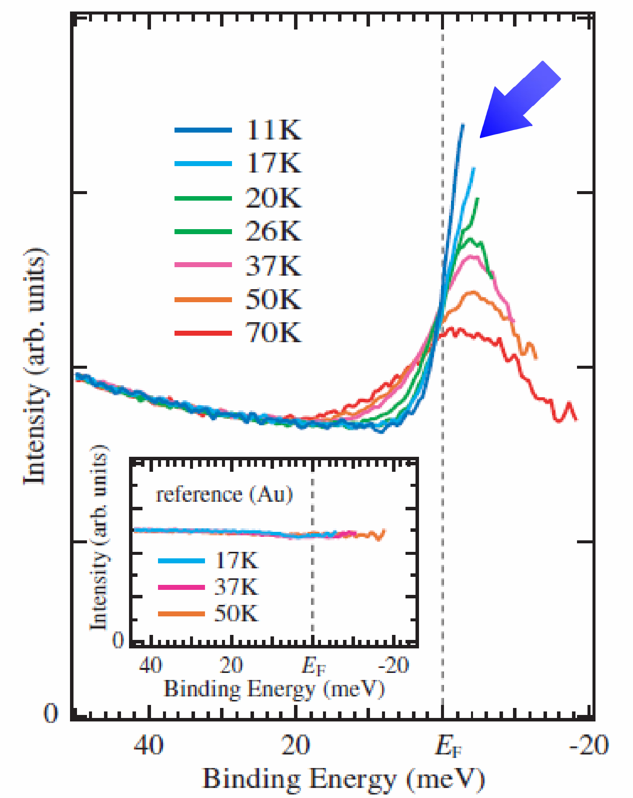
  - **Development of Projective QMC for *T*→*0* and its application to LDA+DMFT calculations for LiV_2O_4**

DMFT



Purpose

- Reproduce the sharp (heavy) quasi-particle peak in A(ω) at T→0 by LDA+DMFT(PQMC)
- Clarify the origin of HF behaviors



(Shimoyamada et al. PRL 96 026403(2006))

Auxiliary-field QMC

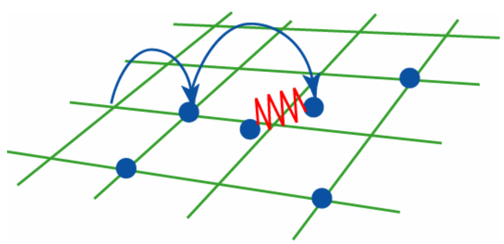
- Suzuki-Trotter decomposition

$$Z = \mathrm{Tr}\, e^{-\beta H} = \mathrm{Tr} \prod_{l=1}^{L} e^{-\Delta\tau H_0} e^{-\Delta\tau H_{\mathrm{int}}} \qquad (\beta = 1/T = L\Delta\tau)$$

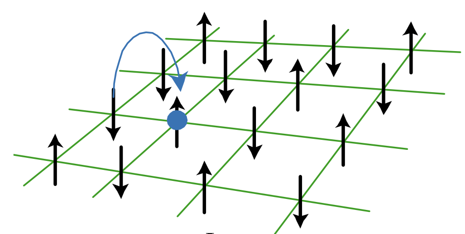
- Hubbard-Stratonovich transformation for H_{int}

$$e^{-\Delta\tau U[n_\uparrow n_\downarrow - \frac{1}{2}(n_\uparrow + n_\downarrow)]} = \frac{1}{2}\sum_{s=\pm 1} e^{\lambda s (n_\uparrow - n_\downarrow)} \qquad (\cosh(\lambda) = \exp[\Delta\tau U/2])$$

- Many-particle system
  $= \sum$ (free one-particle system + auxiliary field)



$$= \sum$$



$$Z = \sum_{s_1 s_2 \ldots s_L} Z_{s_1 s_2 \ldots s_L} \qquad Z_{s_1 s_2 \ldots s_L} \equiv \frac{1}{2^L} \prod_{\sigma} \mathrm{Tr} \prod_{l=1}^{L} [e^{-\Delta\tau H_0^\sigma} e^{\lambda\sigma s_l n_\sigma}]$$

$$\langle A \rangle = \sum_{s_1 s_2 \ldots s_L} \frac{Z_{s_1 s_2 \ldots s_L}}{Z} \langle A \rangle_{s_1 s_2 \ldots s_L}$$, A : arbitrary operator

Monte Carlo sampling

Projective QMC

- Finite-*T* QMC for the Anderson impurity model (Hirsch-Fye 86)

  Integrate out the conduction bands and calculate *G* of the impurity

  Calculate $G_{\{s\}}(\tau_1,\tau_2)$ from $G_0(\tau_1,\tau_2)$

$$G(\tau_1,\tau_2) = -\left\langle T_\tau c_{p\sigma}(\tau_1) c^\dagger_{p\sigma}(\tau_2) \right\rangle = \sum_{\{s\}} w_{\{s\}} G_{\{s\}}(\tau_1,\tau_2)$$

$$0<\tau_1,\tau_2<\beta=1/T,\ \beta=L\Delta\tau$$

Size of *G* $=L^2$
Effort $\sim L^3$
(calculation for low *T* is numerically expensive)

- Projective QMC for T=0

Feldbacher, KH, Assaad, PRL 93 136405(2004)

$$\langle \mathcal{O} \rangle_{T=0} = \frac{\langle \Psi_{\rm GS} | \mathcal{O} | \Psi_{\rm GS} \rangle}{\langle \Psi_{\rm GS} | \Psi_{\rm GS} \rangle}$$

Projective QMC

- Finite-*T* QMC for the Anderson impurity model (Hirsch-Fye 86)

Integrate out the conduction bands and calculate *G* of the impurity

Calculate $G_{\{s\}}(\tau_1,\tau_2)$ from $G_0(\tau_1,\tau_2)$

$$G(\tau_1,\tau_2) = -\left\langle T_\tau c_{p\sigma}(\tau_1) c^\dagger_{p\sigma}(\tau_2) \right\rangle$$
$$= \sum_{\{s\}} w_{\{s\}} G_{\{s\}}(\tau_1,\tau_2)$$

$$0<\tau_1,\tau_2<\beta=1/T,\ \beta=L\Delta\tau$$

Size of $G = L^2$
Effort $\sim L^3$
(calculation for low *T* is numerically expensive)

- Projective QMC for T=0

Feldbacher, KH, Assaad, PRL 93 136405(2004)

$$\langle \mathcal{O} \rangle_{T=0} = \frac{\langle \Psi_T | e^{-\theta/2\,H} \mathcal{O} e^{-\theta/2\,H} | \Psi_T \rangle}{\langle \Psi_T | e^{-\theta H} | \Psi_T \rangle}$$

Projective QMC

- Finite-*T* QMC for the Anderson impurity model (Hirsch-Fye 86)

Integrate out the conduction bands and calculate *G* of the impurity

Calculate $G_{\{s\}}(\tau_1,\tau_2)$ from $G_0(\tau_1,\tau_2)$

$$G(\tau_1,\tau_2) = -\left\langle T_\tau c_{p\sigma}(\tau_1) c^\dagger_{p\sigma}(\tau_2) \right\rangle = \sum_{\{s\}} w_{\{s\}} G_{\{s\}}(\tau_1,\tau_2)$$

$0<\tau_1,\tau_2<\beta=1/T,\ \beta=L\Delta\tau$

Size of *G* $=L^2$
Effort $\sim L^3$
(calculation for low *T* is numerically expensive)

- Projective QMC for T=0

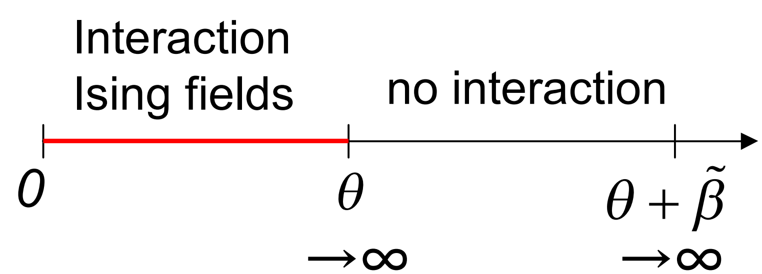
Feldbacher, KH, Assaad, PRL 93 136405(2004)

$$\langle \mathcal{O} \rangle_{T=0} = \frac{\langle \Psi_T | e^{-\theta/2\, H} \mathcal{O} e^{-\theta/2\, H} | \Psi_T \rangle}{\langle \Psi_T | e^{-\theta H} | \Psi_T \rangle}$$

$$\langle \mathcal{O} \rangle_{T=0} = \lim_{\tilde{\beta}\to\infty} \frac{\mathrm{Tr} e^{-\tilde{\beta} H_0} e^{-\theta/2\, H} \mathcal{O} e^{-\theta/2\, H}}{\mathrm{Tr} e^{-\tilde{\beta} H_0} e^{-\theta H}}$$

with $|\Psi_T\rangle$ groundstate of H_0

$\Rightarrow$ same algorithm as Hirsch-Fye QMC but with $G_{T=0}(\tau)$ instead of $G_T(\tau)$

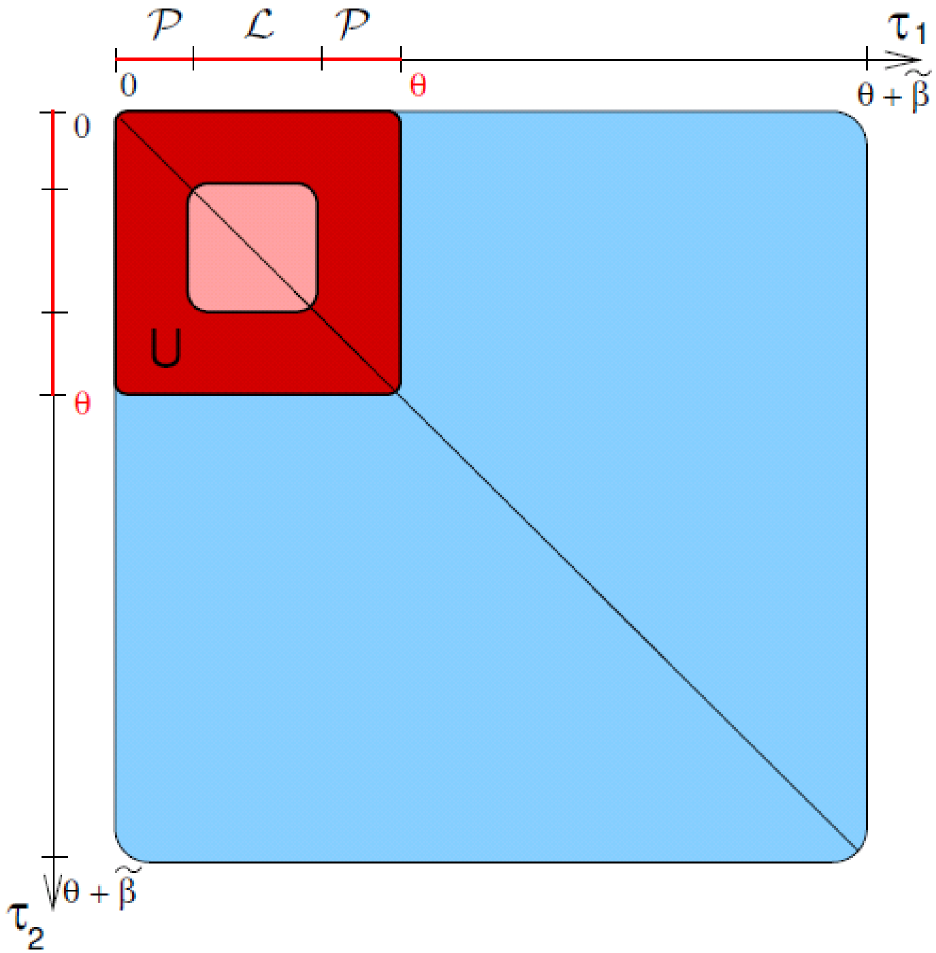


Projective QMC

We need Green function, i.e., $\mathcal{O} = -c(\tau_1)c^\dagger(\tau_2) = -e^{\tau_1/2\,H} c e^{-(\tau_1-\tau_2)/2\,H} c^\dagger e^{-\tau_2/2\,H}$

Green function matrix

$$G(\tau_1,\tau_2) = -\lim_{\tilde\beta\to\infty} \frac{\mathrm{Tr} e^{-\tilde\beta H_0} e^{-(\theta_{\mathcal{P}}-\tau_1)/2\,H} c e^{-(\tau_1-\tau_2)/2} c^\dagger e^{-(\theta_{\mathcal{P}}+\tau_2)/2\,H}}{\mathrm{Tr} e^{-\tilde\beta H_0} e^{-\theta_{\mathcal{P}} H}}$$



Interaction *U* only in red part

for sufficiently large *P*:
Accurate information on
***G* for light red part**

DMFT(PQMC)

- DMFT self-consistent loop

PQMC

$G_0(\tau_1,\tau_2)$ *(T=0)*

$G(\tau_1,\tau_2)$ *(T=0)*

$$\Sigma(i\omega) = G_0^{-1}(i\omega) - G^{-1}(i\omega)$$

$$G(i\omega) = \int d\varepsilon \frac{D(\varepsilon)}{i\omega_n + \mu - \Sigma(i\omega_n) - \varepsilon}$$

$$G_0^{-1}(i\omega) = G^{-1}(i\omega) + \Sigma(i\omega)$$

<u>Problem</u>

G(τ)→FT→G(iω)? <u>No</u>

only G(τ),τ<θ obtained by PQMC

<u>Maximum Entropy Method</u>

$$G(\tau) = \frac{1}{\pi}\int A(\omega)\exp(-\omega\tau)d\omega$$

$$G(i\omega_n) = \frac{1}{\pi}\int \frac{A(\omega)}{i\omega_n - \omega}d\omega$$

DMFT(PQMC)

- DMFT self-consistent loop

PQMC

$G_0(\tau_1,\tau_2)$ *(T=0)*

$G(\tau_1,\tau_2)$ *(T=0)*

$$\Sigma(i\omega) = G_0^{-1}(i\omega) - G^{-1}(i\omega)$$

$$G(i\omega) = \int d\varepsilon \frac{D(\varepsilon)}{i\omega_n + \mu - \Sigma(i\omega_n) - \varepsilon}$$

$$G_0^{-1}(i\omega) = G^{-1}(i\omega) + \Sigma(i\omega)$$

<u>Problem</u>

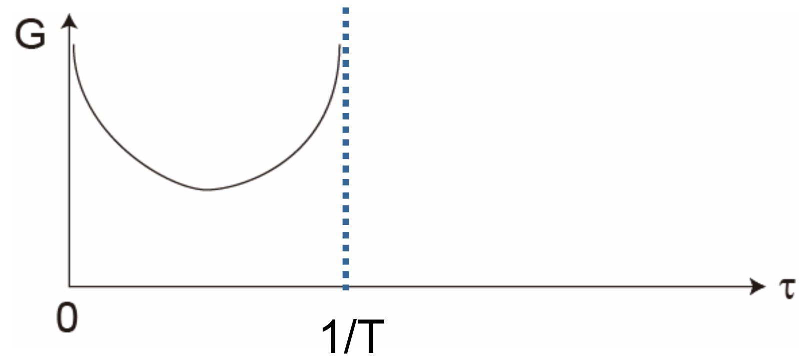
G(τ)→FT→G(iω)? <u>No</u>

only G(τ),$\tau<\theta$ obtained by PQMC

<u>Maximum Entropy Method</u>

$$G(\tau) = \frac{1}{\pi}\int A(\omega)\exp(-\omega\tau)d\omega$$

$$G(i\omega_n) = \frac{1}{\pi}\int \frac{A(\omega)}{i\omega_n - \omega}d\omega$$



DMFT(PQMC)

- DMFT self-consistent loop

PQMC

$G_0(\tau_1,\tau_2)$ *(T=0)*

$G(\tau_1,\tau_2)$ *(T=0)*

$$\Sigma(i\omega) = G_0^{-1}(i\omega) - G^{-1}(i\omega)$$

$$G(i\omega) = \int d\varepsilon \frac{D(\varepsilon)}{i\omega_n + \mu - \Sigma(i\omega_n) - \varepsilon}$$

$$G_0^{-1}(i\omega) = G^{-1}(i\omega) + \Sigma(i\omega)$$

<ins>Problem</ins>

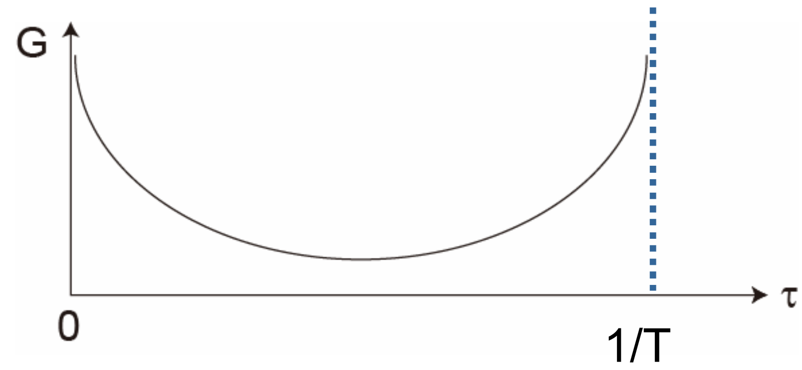
G(τ)→FT→G(iω)? <ins>No</ins>

only G(τ),$\tau<\theta_P$ obtained by PQMC

<ins>Maximum Entropy Method</ins>

$$G(\tau) = \frac{1}{\pi}\int A(\omega)\exp(-\omega\tau)d\omega$$

$$G(i\omega_n) = \frac{1}{\pi}\int \frac{A(\omega)}{i\omega_n - \omega}d\omega$$



DMFT(PQMC)

- DMFT self-consistent loop

PQMC

$G_0(\tau_1,\tau_2)$ *(T=0)*

$G(\tau_1,\tau_2)$ *(T=0)*

$$\Sigma(i\omega)=G_0^{-1}(i\omega)-G^{-1}(i\omega)$$

$$G(i\omega)=\int d\varepsilon \frac{D(\varepsilon)}{i\omega_n+\mu-\Sigma(i\omega_n)-\varepsilon}$$

$$G_0^{-1}(i\omega)=G^{-1}(i\omega)+\Sigma(i\omega)$$

Problem

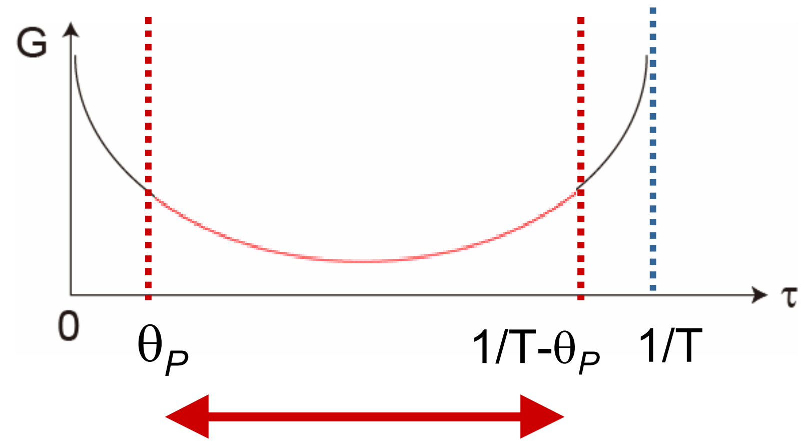
G(τ)→FT→G(iω)? No

only G(τ),τ<θ obtained by PQMC

Maximum Entropy Method

$$G(\tau)=\frac{1}{\pi}\int A(\omega)\exp(-\omega\tau)d\omega$$

$$G(i\omega_n)=\frac{1}{\pi}\int \frac{A(\omega)}{i\omega_n-\omega}d\omega$$



Calculate G only for $\tau<\theta_P$
Large τ: Extrapolation by
Maximum Entropy Method

DMFT(PQMC)

- DMFT self-consistent loop

PQMC

$G(\tau_1,\tau_2)$ *(T=0)*

$$\Sigma(i\omega) = G_0^{-1}(i\omega) - G^{-1}(i\omega)$$

$$G(i\omega) = \int d\varepsilon \frac{D(\varepsilon)}{i\omega_n + \mu - \Sigma(i\omega_n) - \varepsilon}$$

$$G_0^{-1}(i\omega) = G^{-1}(i\omega) + \Sigma(i\omega)$$

$G_0(\tau_1,\tau_2)$ *(T=0)*

<u>Problem</u>

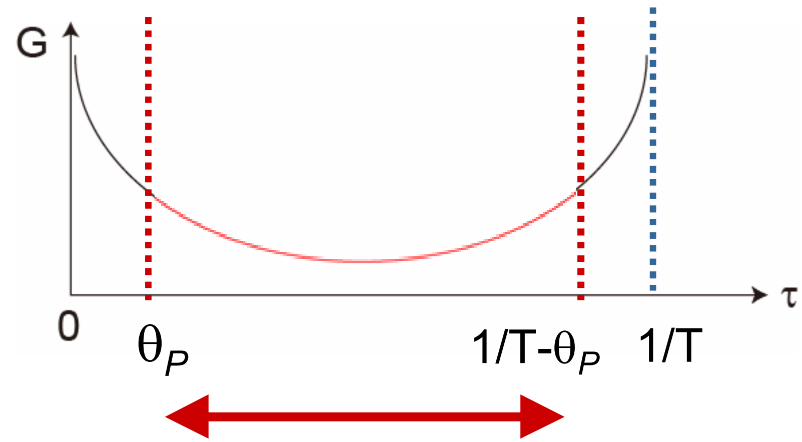
G(τ)→FT→G(iω)? <u>No</u>

only G(τ),τ<θ obtained by PQMC

<u>Maximum Entropy Method</u>

$$G(\tau) = \frac{1}{\pi}\int A(\omega)\exp(-\omega\tau)d\omega$$

$$G(i\omega_n) = \frac{1}{\pi}\int \frac{A(\omega)}{i\omega_n - \omega}d\omega$$



Calculate G only for $\tau<\theta_P$

Large τ: Extrapolation by

Maximum Entropy Method

FAQ:

Why can we discuss *A*(ω→0) even if we do not calculate *G*(τ→∞) explicitly?

DMFT(PQMC)

- DMFT self-consistent loop

PQMC

$G_0(\tau_1,\tau_2)$ *(T=0)*

$G(\tau_1,\tau_2)$ *(T=0)*

$$\Sigma(i\omega) = G_0^{-1}(i\omega) - G^{-1}(i\omega)$$

$$G(i\omega) = \int d\varepsilon \frac{D(\varepsilon)}{i\omega_n + \mu - \Sigma(i\omega_n) - \varepsilon}$$

$$G_0^{-1}(i\omega) = G^{-1}(i\omega) + \Sigma(i\omega)$$

<u>Problem</u>

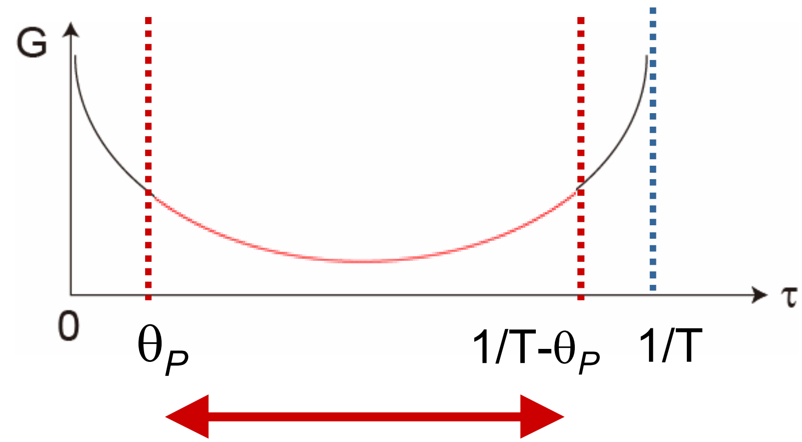
G(τ)→FT→G(iω)? <u>No</u>

only G(τ),τ<θ obtained by PQMC

<u>Maximum Entropy Method</u>

$$G(\tau) = \frac{1}{\pi}\int A(\omega)\exp(-\omega\tau)d\omega$$

$$G(i\omega_n) = \frac{1}{\pi}\int \frac{A(\omega)}{i\omega_n - \omega}d\omega$$



Calculate G only for $\tau<\theta_P$

Large τ: Extrapolation by

Maximum Entropy Method

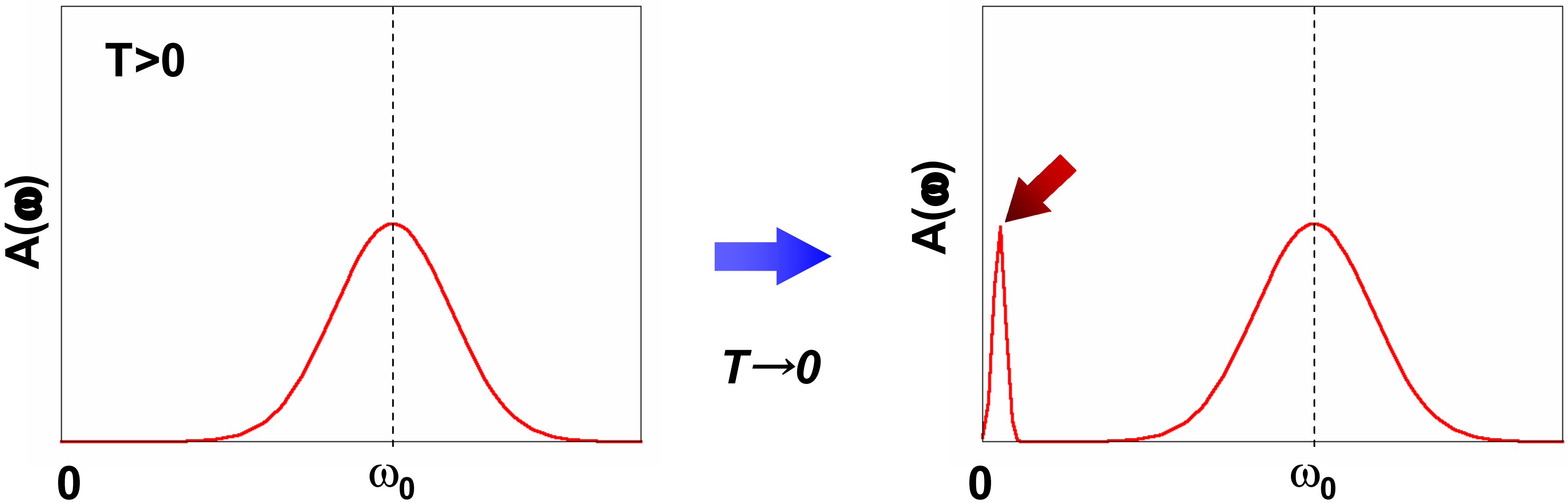
FAQ:

Why can we discuss *A*(ω→0) even if we do not calculate *G*(τ→∞) explicitly?

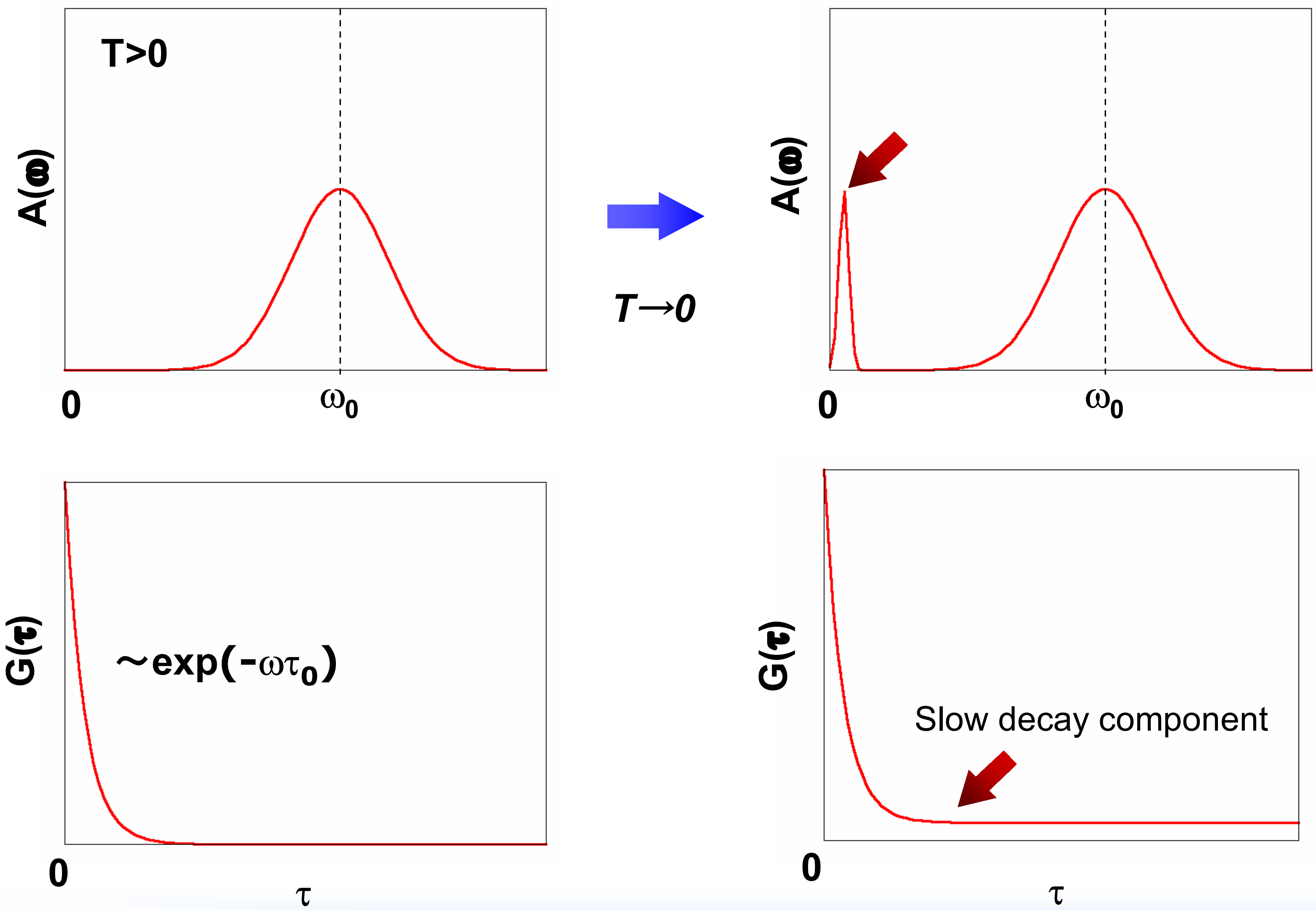
<u>**Sufficiently large θ_P needed**</u>

R. Arita

DMFT(PQMC)

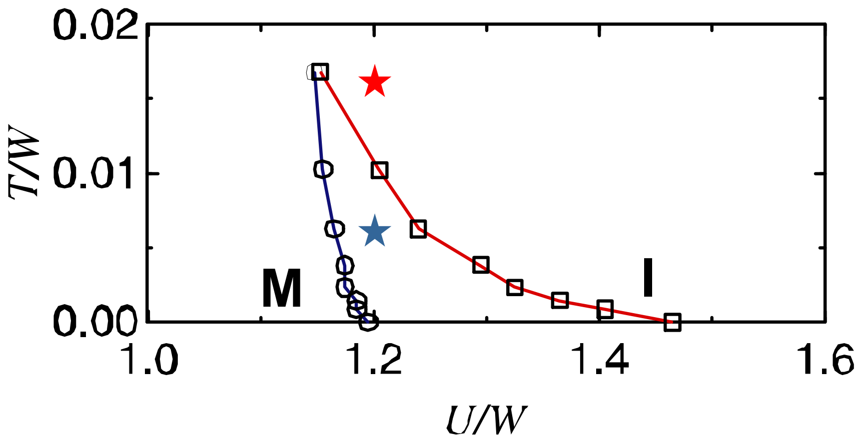


DMFT(PQMC)

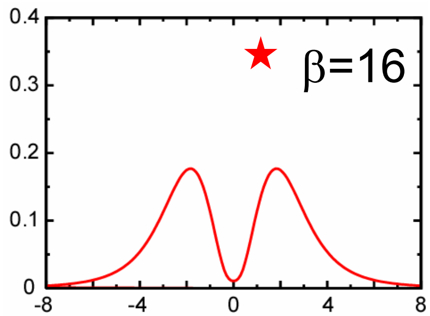


DMFT(PQMC)

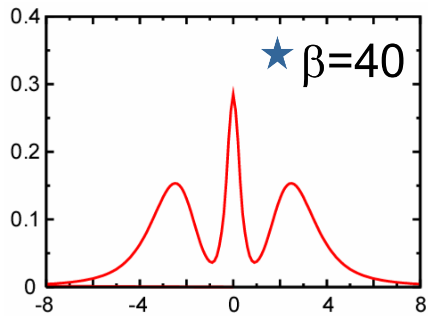
- Single-band Hubbard model
  HF-QMC vs. PQMC



HF-QMC



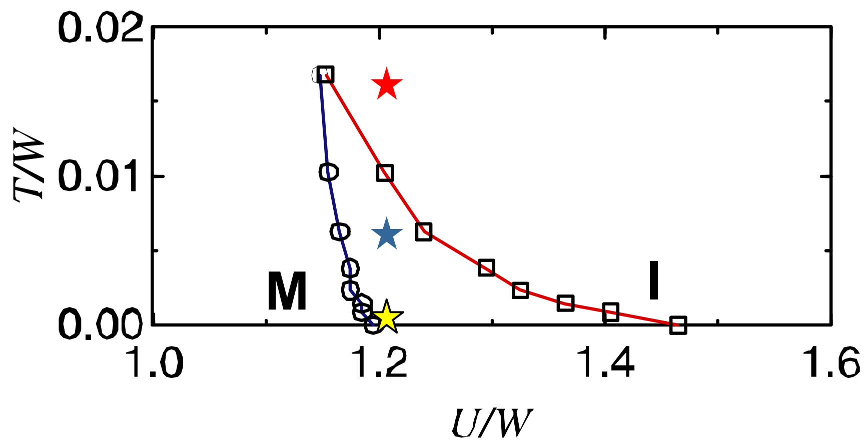
insulating



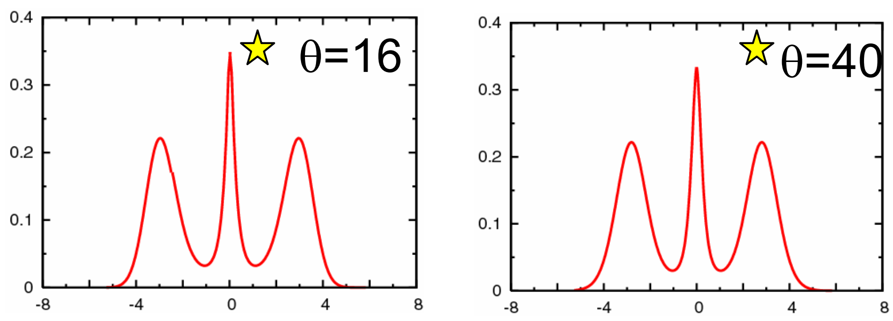
metallic

DMFT(PQMC)

- Single-band Hubbard model
  HF-QMC vs. PQMC



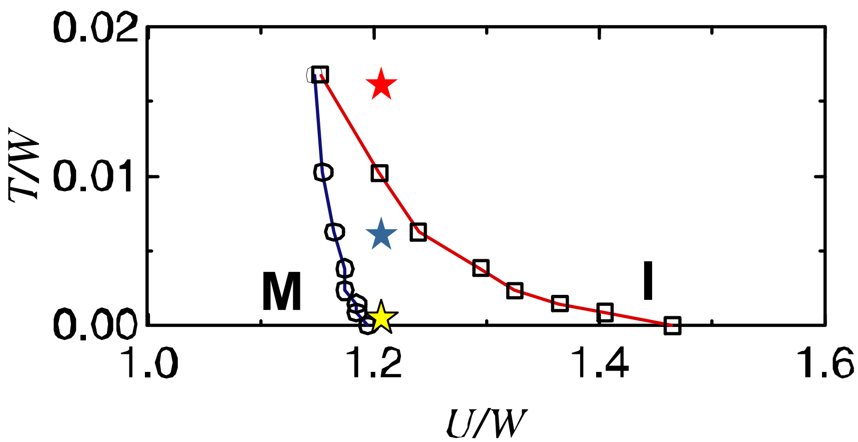
PQMC



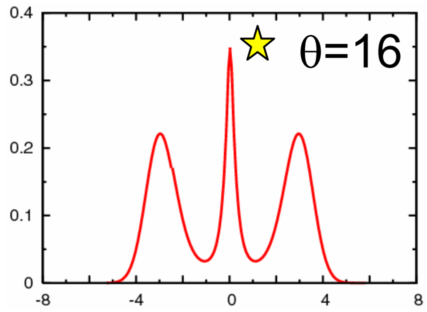
Convergence w.r.t θ is much better than β

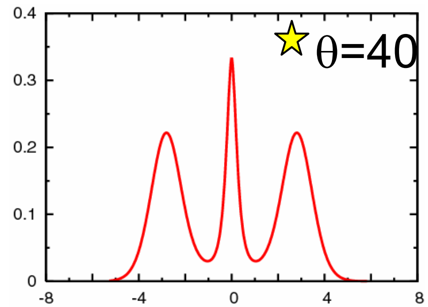
DMFT(PQMC)

- Single-band Hubbard model
  HF-QMC vs. PQMC



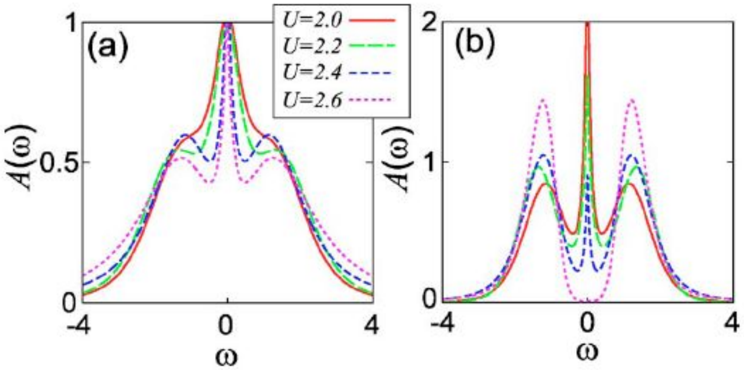
PQMC





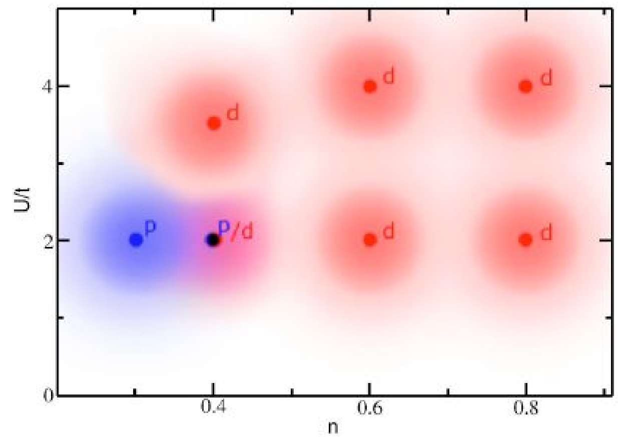
Convergence w.r.t θ is much better than β

- Orbital selective Mott transition in the two-orbital Hubbard model



RA and KH, PRB 72 201102(2005)

- DCA(PQMC) study for anisotropic pairing in the t-t' Hubbard model

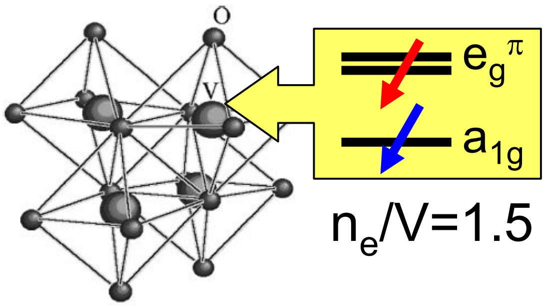


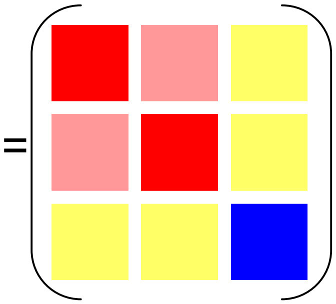
RA and KH, PRB 73 064515(2006)

R. Arita

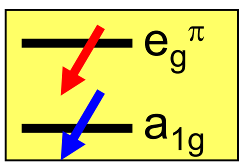
Effective low energy Hamiltonian

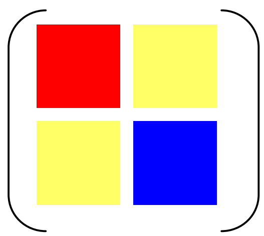
12 (=4x3) band Hamiltonian

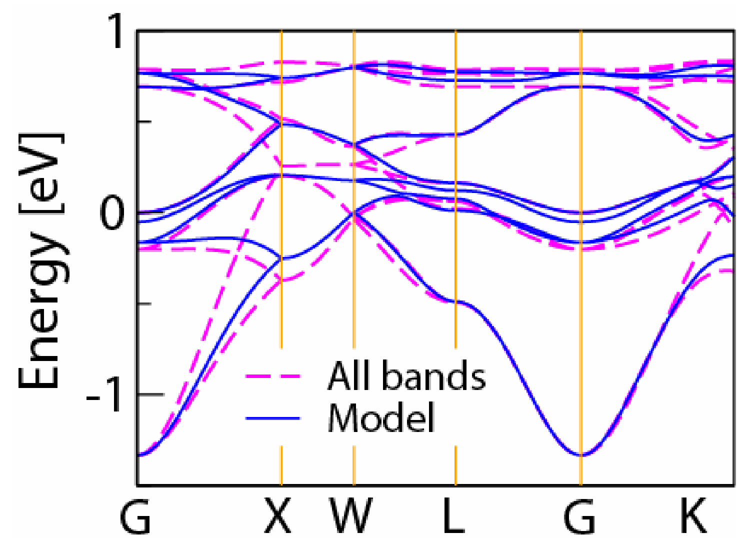


$H_{ij}(\boldsymbol{k})=$ 

8 (=4x2) band model



$H_{ij}(\boldsymbol{k})=$ 



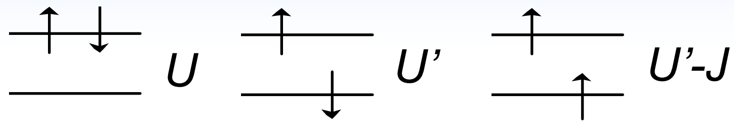
<u>DMFT self-consistent eq.</u>

$$G_i = \sum_{\mathbf{k}} \frac{1}{i\omega_n - H(\mathbf{k}) - \Sigma_i}$$

$$\mathcal{G}_0^{-1} = G^{-1} + \Sigma$$

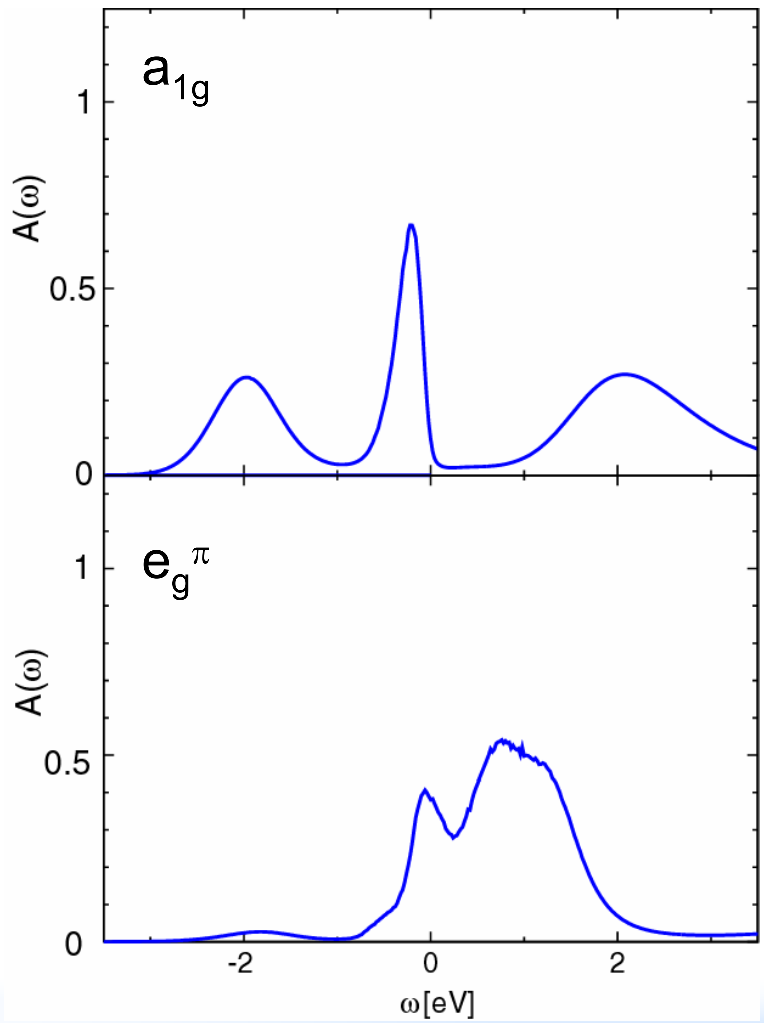
hybridization ($H_{ij,}$ $i \neq j$) taken into account explicitly

LDA+DMFT Result (T=1200K, HF-QMC)

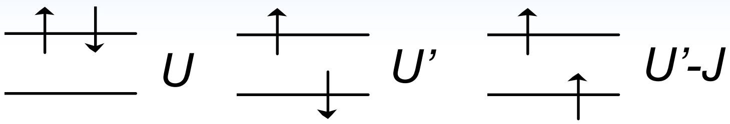


Hund coupling = Ising

U=3.6, U'=2.4, J=0.6

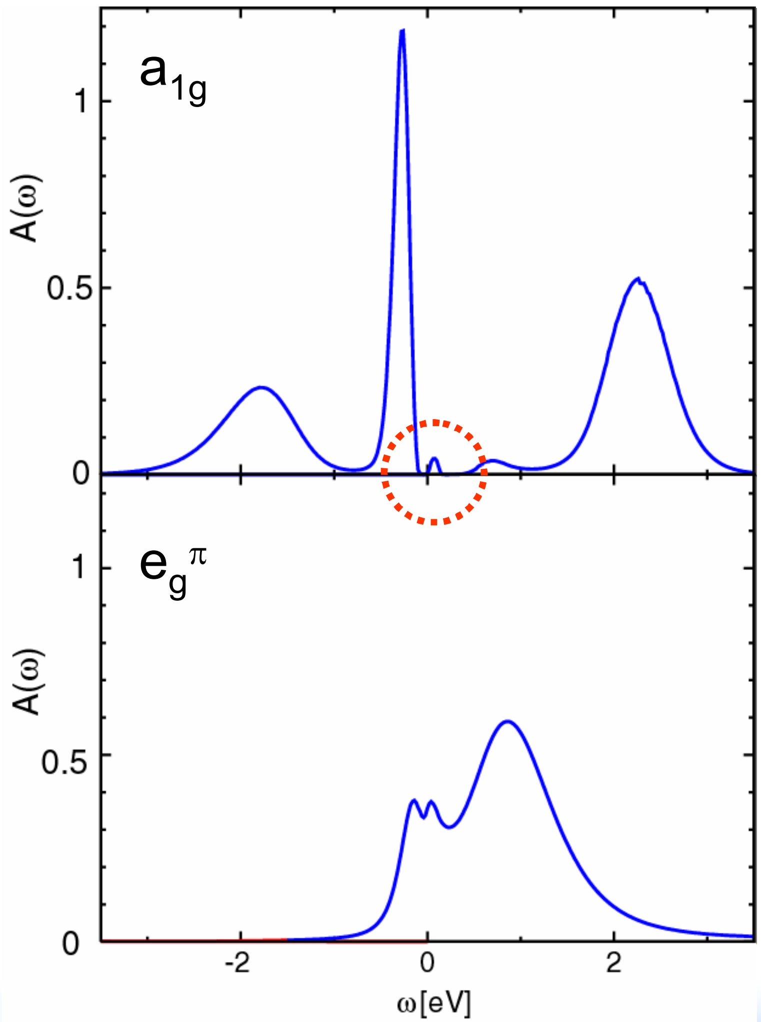


LDA+DMFT Result (T=300K, HF-QMC)

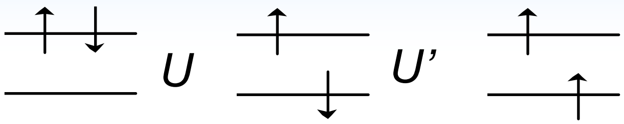


Hund coupling = Ising

U=3.6, U'=2.4, J=0.6

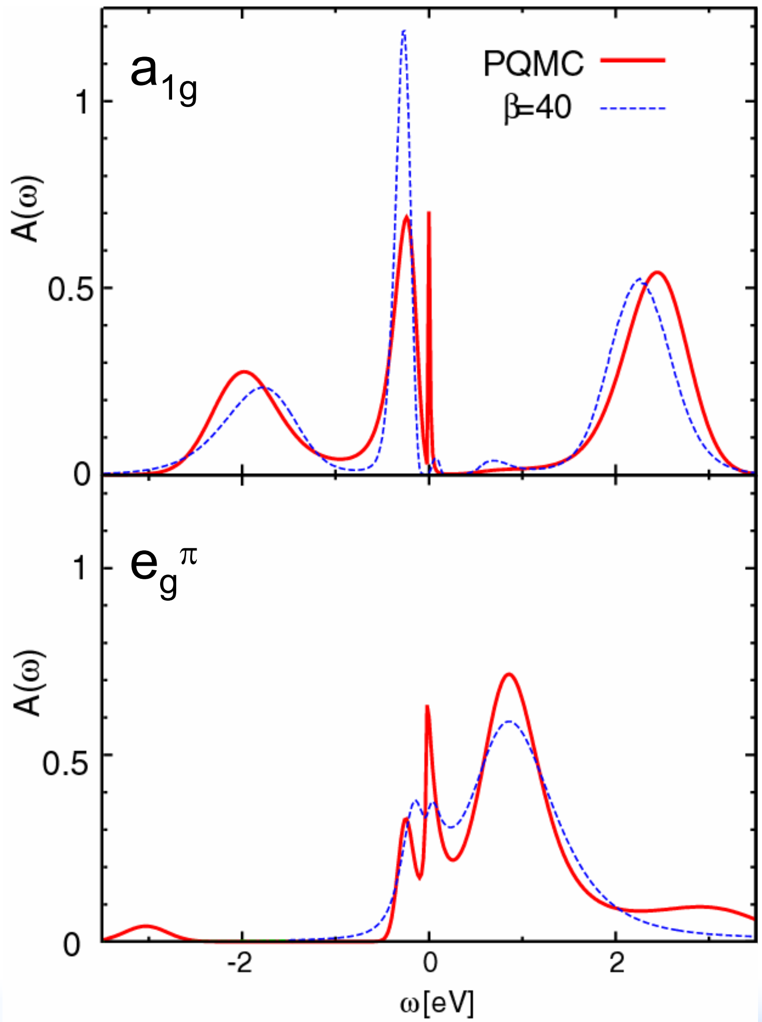


LDA+DMFT Result (T=0, PQMC)



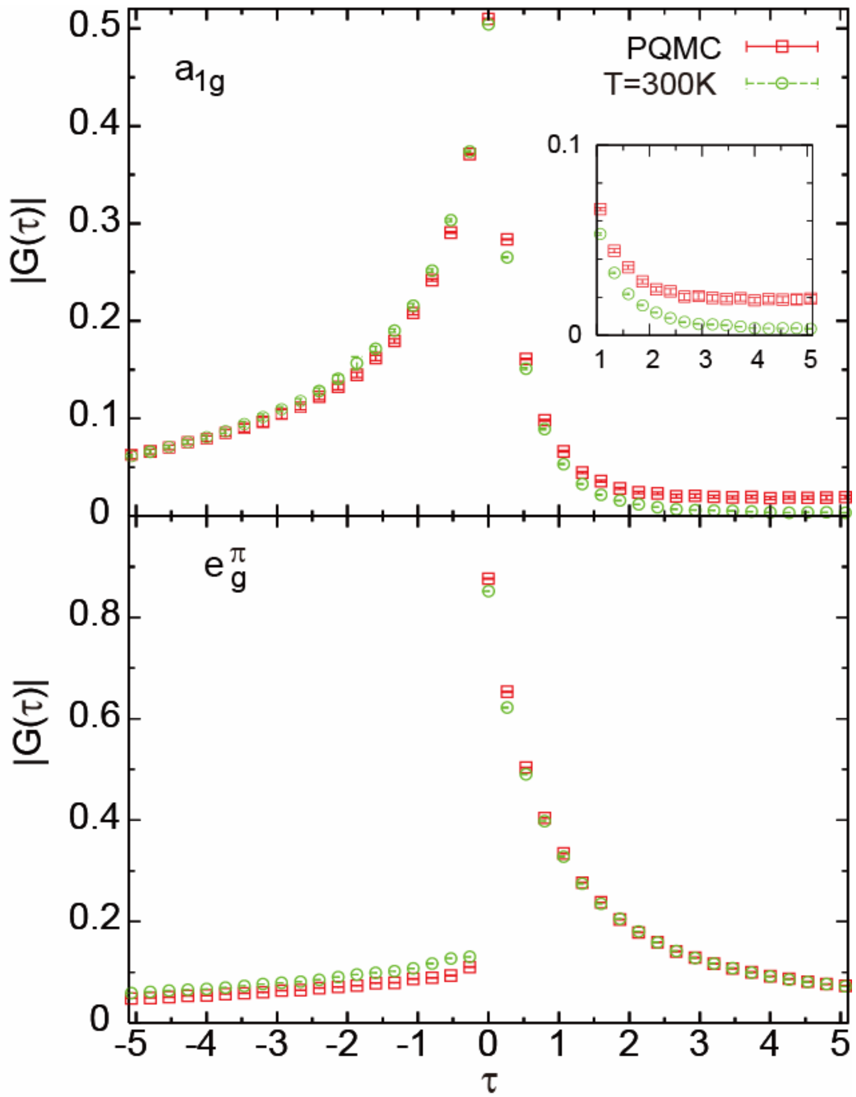
Hund coupling = Ising

U=3.6, U'=2.4, J=0.6



R. Arita

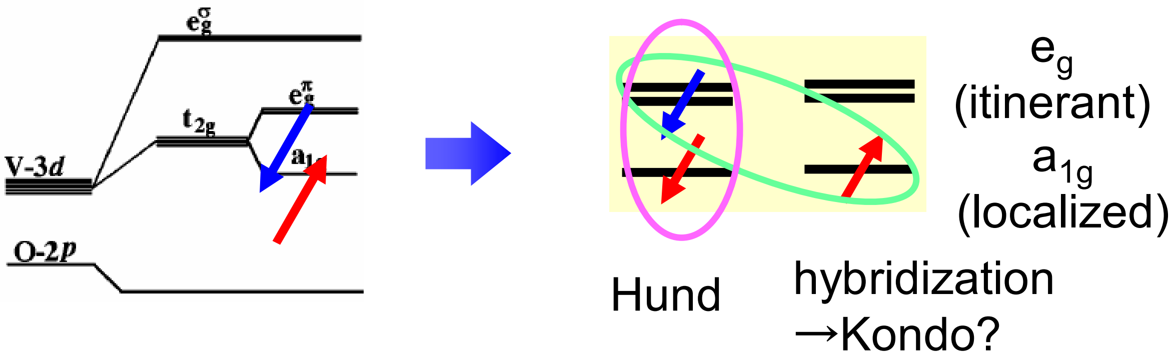
LDA+DMFT Result (T=0, PQMC)



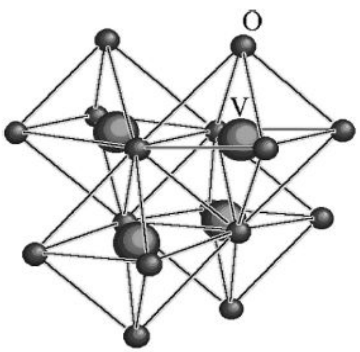
Theoretical studies so far

- Anisimov et al, 99
- Eyert et al, 99
- Matsuno et al, 99
- Kusunose et al, 00
- Lacroix, 01
- Shannon, 01
- Fulde et al, 01
- Burdin et al, 02
- Hopkinson et al, 02
- Fujimoto, 02
- Tsunetsugu, 02
- Yamashita et al, 03
- Laad et al, 03
- Nekrasov et al, 03
- Yushankhai et al, 07

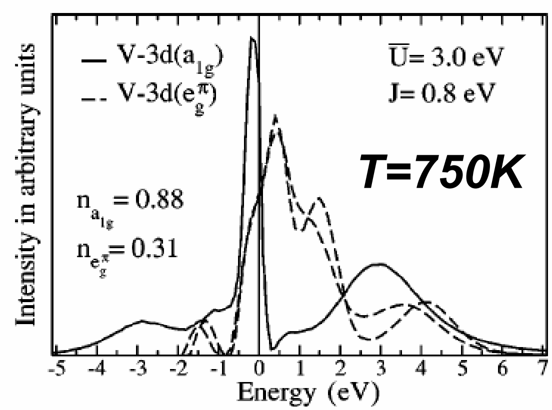
- Kondo scenario   Anisimov et al, PRL 83 364(1999)



- Geometrical Frustration



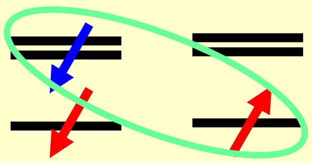
Short range (local) correlations become dominant
⇒ **DMFT** expected to be a good approx.



LDA+DMFT by Nekrasov et al, PRB 67 085111 (2003)

Calculation for T→0

Effect of a_{1g}-e_g^π hybridization

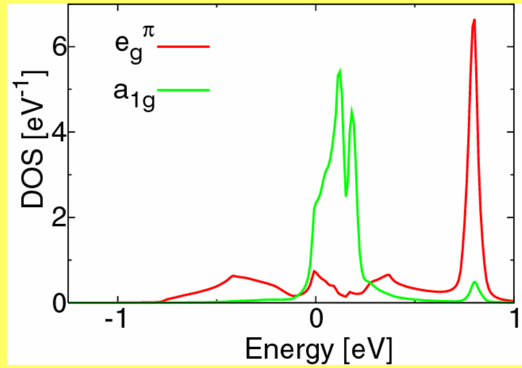


DMFT self-consistent eq.

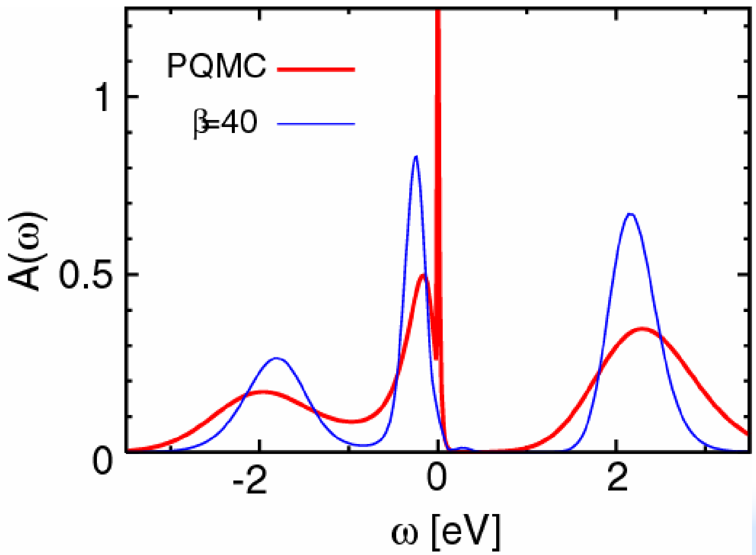
$$G_i = \sum_{\mathbf{k}} \frac{1}{i\omega_n - H(\mathbf{k}) - \Sigma_i}$$

$$\mathcal{G}_0^{-1} = G^{-1} + \Sigma$$

$$G_i = \int d\epsilon \frac{D_i(\epsilon)}{i\omega_n - \epsilon - \Sigma_i}$$



a_{1g}-e_g^π hybridization **not considered explicitly**

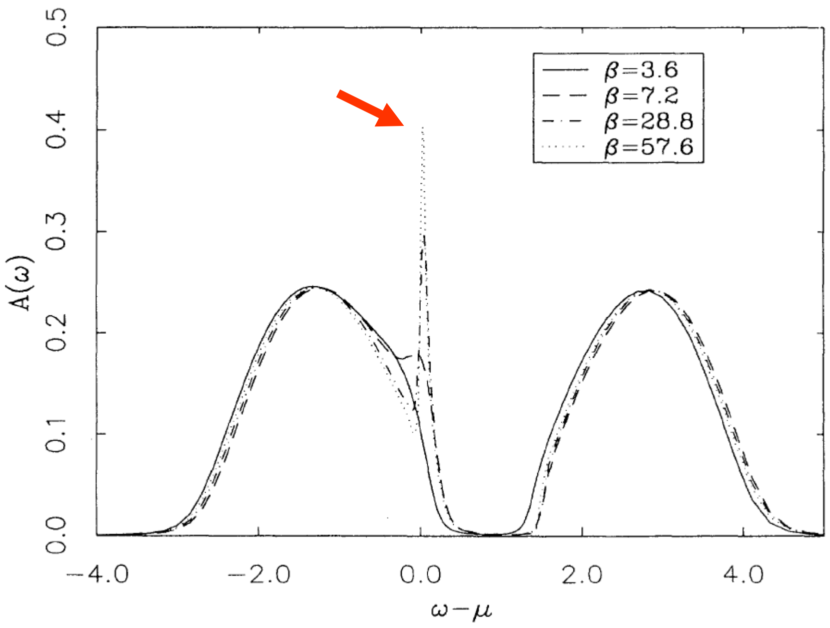


Kondo coupling between a_{1g} and e_g^π
not reason for peak above E_F

Origin of the peak: a_{1g} =slightly doped Mott insulator ?

LDA		LDA+DMFT
$n_{a1g} \sim n_{eg}$	→	$n_{a1g} \sim 0.95$

DMFT for the single band Hubbard
n=0.97 (U=4)

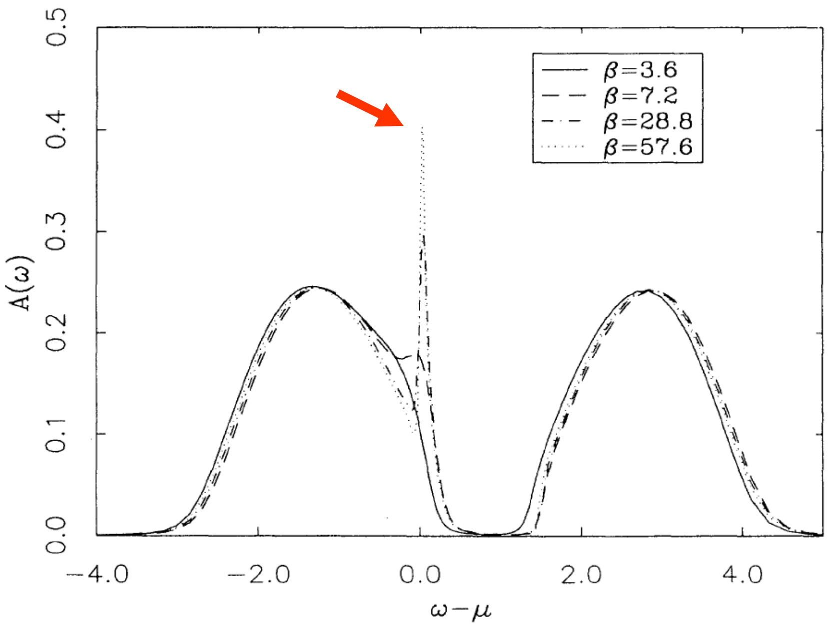


Th. Pruschke et al, PRB 47 3553 (1993)

Origin of the peak: a_{1g} =slightly doped Mott insulator ?

<u>LDA</u>	→	<u>LDA+DMFT</u>
$n_{a1g} \sim n_{eg}$		$n_{a1g} \sim 0.95$

DMFT for the single band Hubbard
n=0.97 (U=4)



Th. Pruschke et al, PRB 47 3553 (1993)

Question:
Strong renormalization can survive the presence of short-range correlation beyond DMFT?

cf) A.Toschi, A. Katanin, K. Held
(PRB 75 045118 (2007))
DGA study for cubic lattice

Damping of the peak:
Irrelevant for 3D frustrated lattice(?)

Conclusions

- Development of **PQMC** and its application to **LDA+DMFT** calculations for **LiV_2O_4**
- Origin of the sharp peak just above E_F
  - **a_{1g} = slightly doped Mott Insulator**
- Future problems
  - beyond-DMFT