DFT+DMFT with DCore

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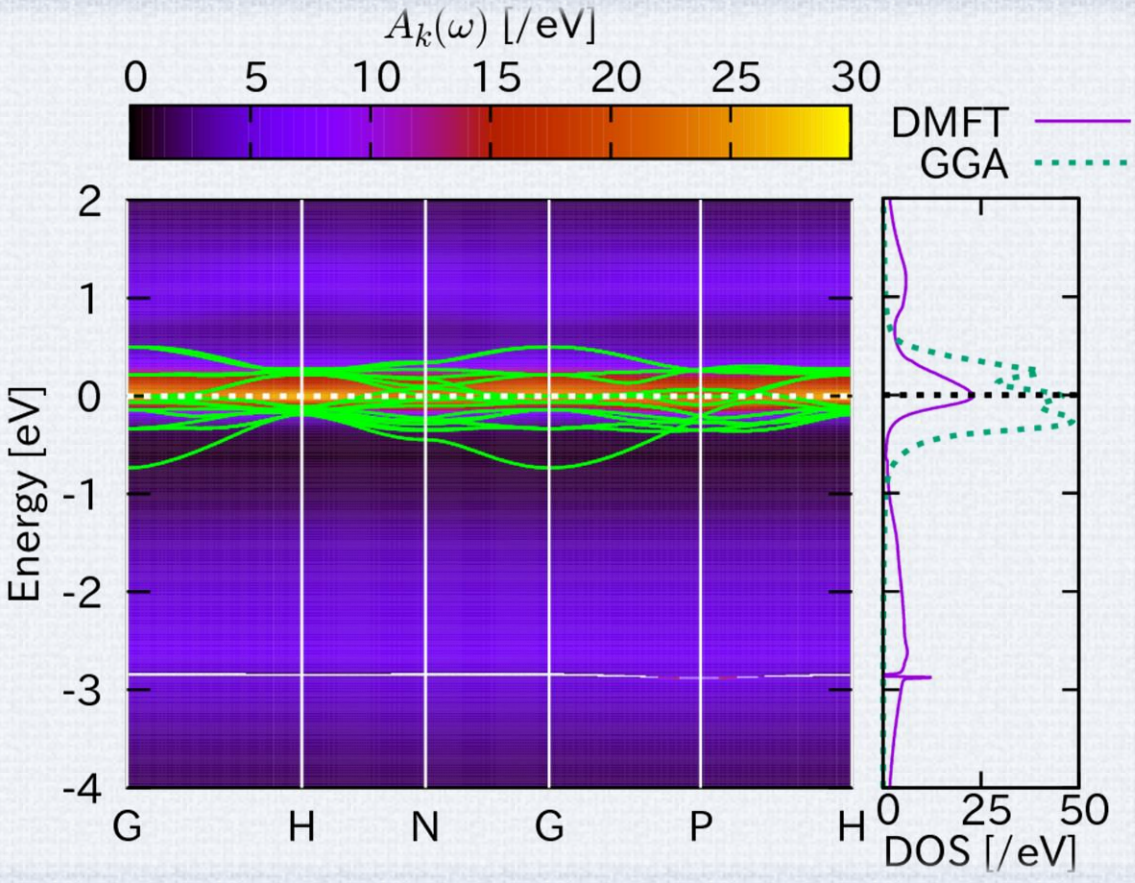

2018/7/30
DCore 講習会

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Intro

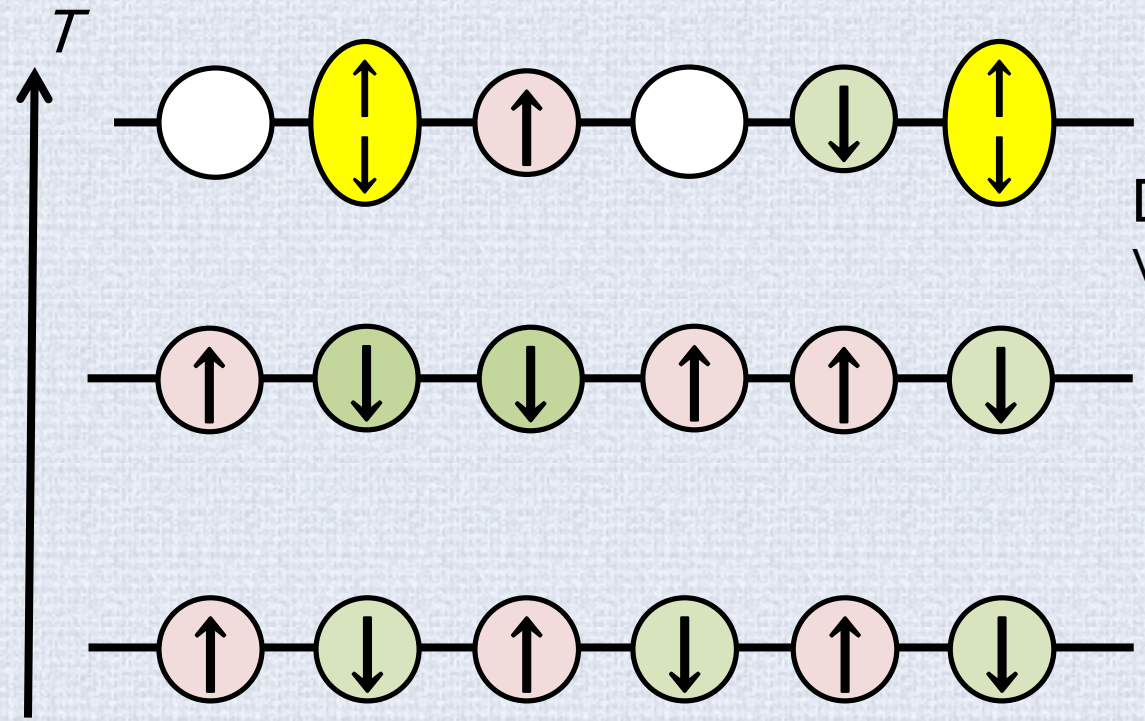
Motivation

Strongly correlated electrons – Field of exotic phenomena –

- High T_C superconductivity
- Quantum spin liquid
- Permanent magnet
- Multiferroicity
- Etc.

$$H = \sum_{RR'\sigma\alpha\beta} t_{RR'\alpha\beta} \hat{c}^{\dagger}_{\sigma R\alpha} \hat{c}_{\sigma R'\beta} + \frac{1}{2} \sum_{\sigma\sigma'} \sum_{\alpha\beta\gamma\delta} U_{\alpha\beta\gamma\delta} \hat{c}^{\dagger}_{\sigma R\alpha} \hat{c}^{\dagger}_{\sigma' R\beta} \hat{c}_{\sigma' R\gamma} \hat{c}_{\sigma R\delta}$$

Mott insulator

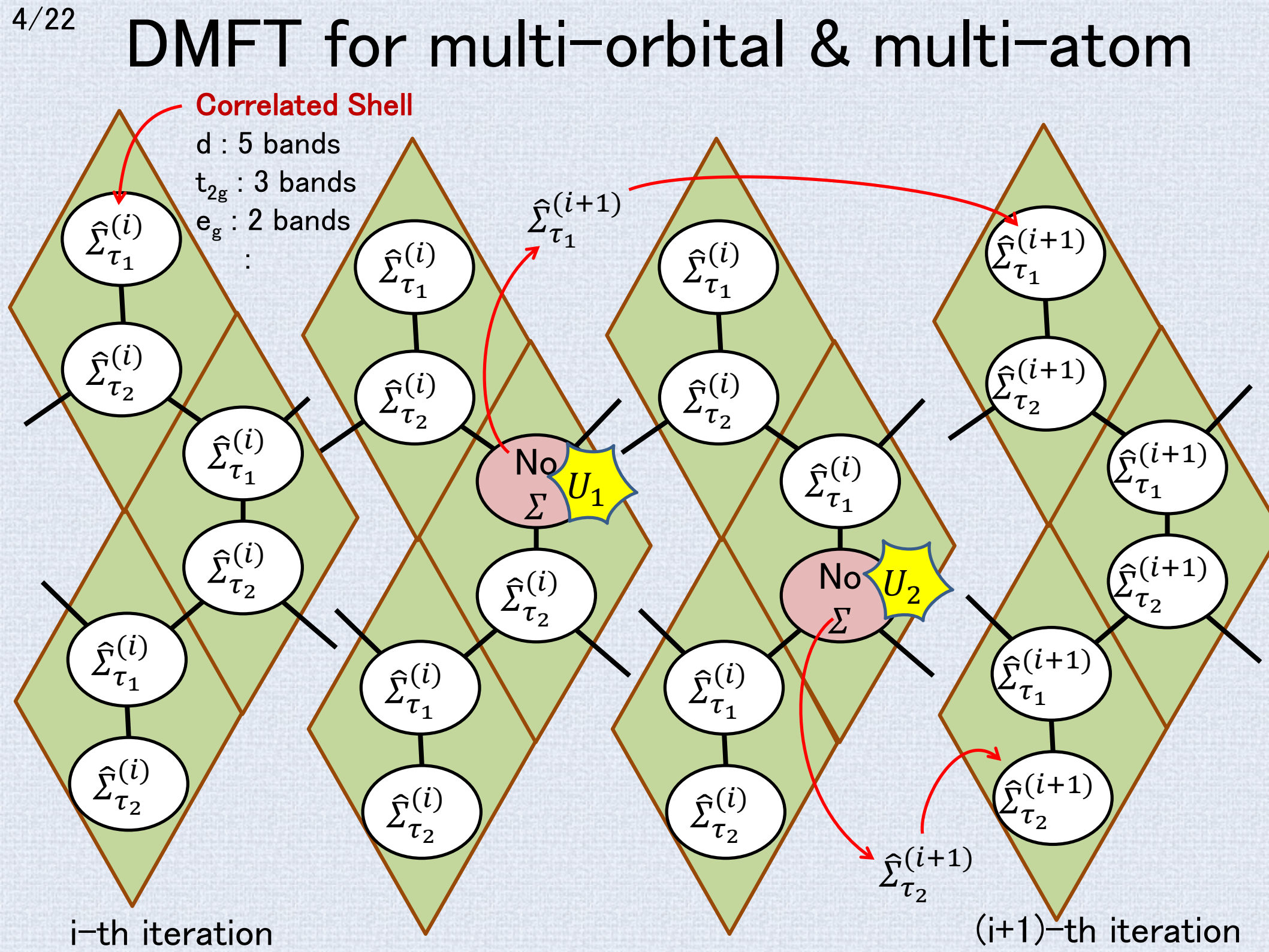


DFT+U

Vladimir I. Anisimov, *et al.* PRB **44**, 943 (1991).

cannot reproduce insulators without magnetic/charge order.

DMFT for multi-orbital & multi-atom



Method

DMFT loop

$$\hat{G}_k(i\omega) = \left(i\omega + \mu - \hat{H}_k^{\mathrm{KS}} - \sum_{\tau}^{N_\tau} \{\hat{\Sigma}_\tau(i\omega) - \hat{\Sigma}_\tau^{DC}\} \right)^{-1}$$

$$\hat{G}_\tau(i\omega) = \sum_k \hat{G}_k(i\omega)$$

$$\hat{G}_\tau^0(i\omega) = \left[\hat{G}_\tau^{-1}(i\omega) + \hat{\Sigma}_\tau(i\omega)\right]^{-1}$$

Impurity solver :
$\hat{G}_\tau^0(i\omega)$ and $H_{int} \rightarrow \hat{\Sigma}_\tau(i\omega)$

Double counting part (Estimate with Hartree–Fock)

$$\Sigma^{\mathrm{DC}}_{\tau,\sigma\alpha\sigma'\beta} = \delta_{\sigma\sigma'} \sum_{\gamma\delta\sigma_1} U_{\alpha\gamma\beta\delta} \left\langle c^\dagger_{\gamma\sigma}\, c_{\delta\sigma_1} \right\rangle_{KS} - \sum_{\gamma\delta} U_{\alpha\gamma\delta\beta} \left\langle c^\dagger_{\gamma\sigma'}\, c_{\delta\sigma} \right\rangle_{KS}$$

Method

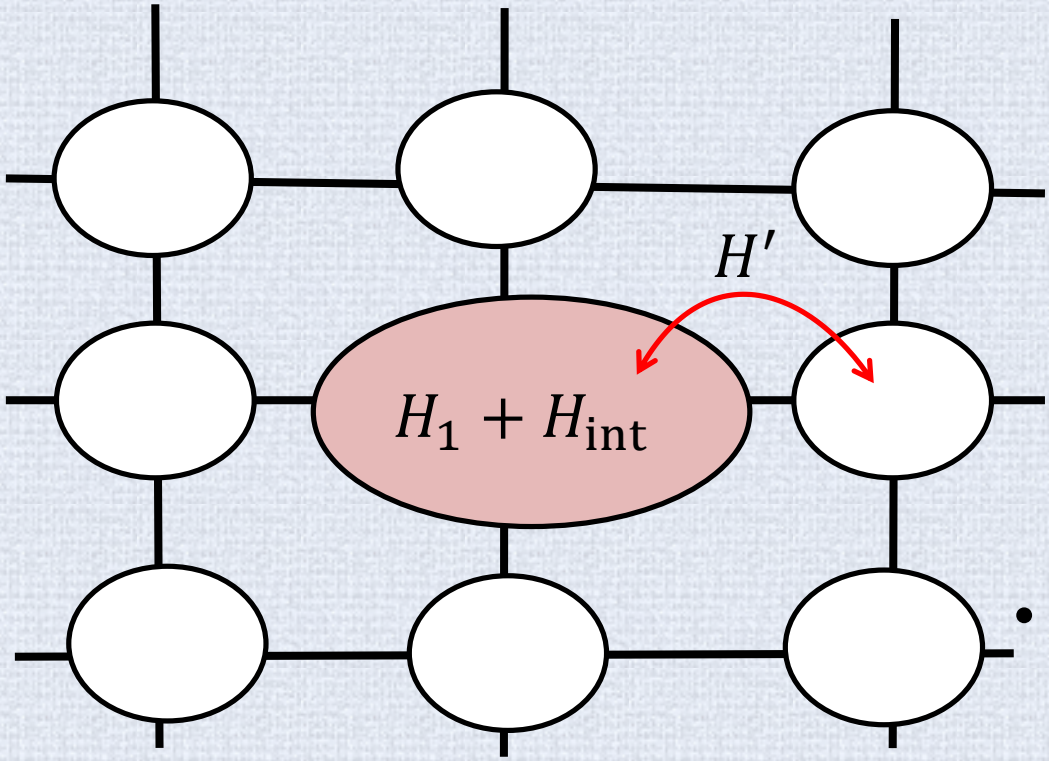
Impurity solver

Continuous-time Quantum Monte-Carlo method with Hybridization expansion

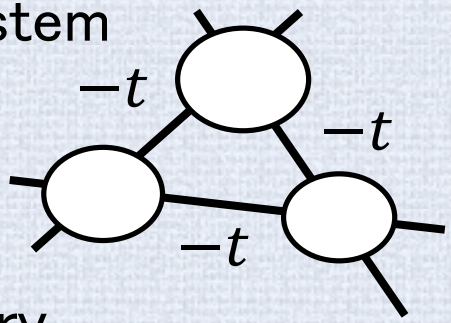
P. Werner, *et al.*, PRL **97**, 076405 (2006).

$$Z = 1 + \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \int d\tau_1 \cdots \int d\tau_n \langle P[H'(\tau_1) \cdots H'(\tau_n)] \rangle$$

$$G^0 \rightarrow H_1, H'$$



- Exact excepting Monte-Carlo error
- Negative-sign problem for multi band system



- CT-hyb QMC library
  - TRIQSP.
    Seth, *et al.*, CPC **200**, 274 (2016).
  - ALPSCore
    H. Shinaoka et al., CPC **215**, 128 (2017).

Numerical

Interaction

$$H_{int} = \frac{1}{2}\sum_{\sigma\sigma'}\sum_{\alpha\beta\gamma\delta} U_{\alpha\beta\gamma\delta}\hat{c}^{\dagger}_{\sigma\alpha}\hat{c}^{\dagger}_{\sigma'\beta}\hat{c}_{\sigma'\gamma}\hat{c}_{\sigma\delta}$$

Simplify

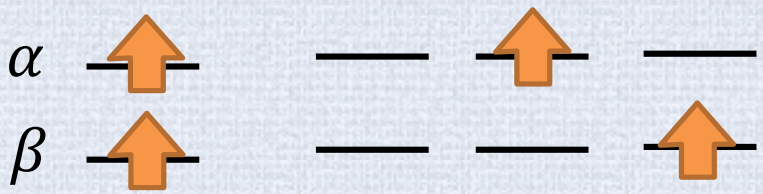
$$U_{\alpha\beta} \equiv U_{\alpha\beta\beta\alpha}$$

$$J_{\alpha\beta} \equiv U_{\alpha\beta\alpha\beta} = U_{\alpha\alpha\beta\beta}$$

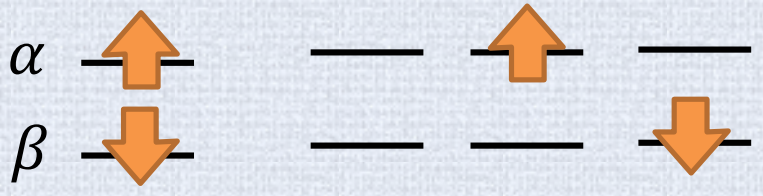
- **Constrained RPA (cRPA)** F. Aryasetiawan, *et al.*, PRB **70**, 195104 (2004).

$$U_{\alpha\beta\gamma\delta} = \int d^3r d^3r' V_{\mathrm{cRPA}}(r,r') w^*_{\alpha}(r) w^*_{\beta}(r') w_{\gamma}(r') w_{\delta}(r)$$

- **Constrained DFT** M. Cococcioni and S. Gironcoli, PRB 71, 035105 (2005). Etc.



$$U_{\alpha\beta} - J_{\alpha\beta} = E_{\alpha\uparrow,\beta\uparrow} + E_0 - E_{\alpha\uparrow} - E_{\beta\uparrow}$$



$$U_{\alpha\beta} = E_{\alpha\uparrow,\beta\downarrow} + E_0 - E_{\alpha\uparrow} - E_{\beta\downarrow}$$

- **Model-mapping RPA** H. Sakakibara, *et al.*, JPSJ **86**, 044714 (2017).

Solve $U^{\mathrm{model-RPA}}_{\alpha\beta}[U_{\alpha\beta}, J_{\alpha\beta}] = U^{\mathrm{original-RPA}}_{\alpha\beta}$

$J^{\mathrm{model-RPA}}_{\alpha\beta}[U_{\alpha\beta}, J_{\alpha\beta}] = J^{\mathrm{original-RPA}}_{\alpha\beta}$ w.r.t. $U_{\alpha\beta}, J_{\alpha\beta}$

Numerical

Calculation steps

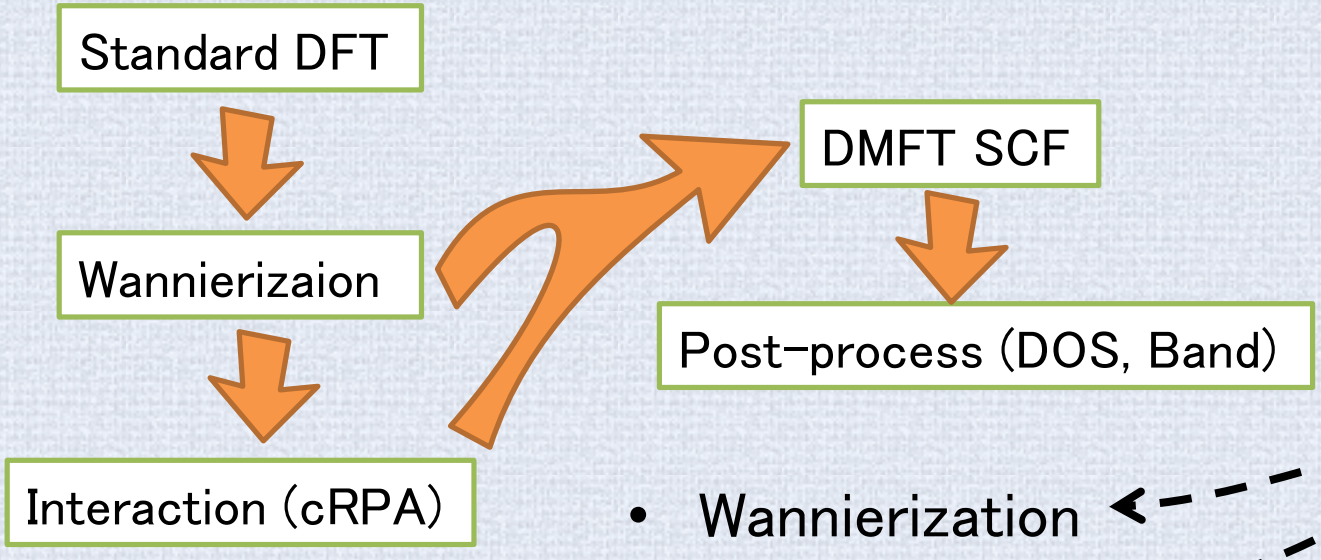
- Charge self-consistent method :

$$\rho^{(i+1)}(r) = \rho^{(i)}(r) + \sum_{\tau\alpha\beta} \left[\rho^{\mathrm{DMFT}}_{\tau,\alpha\beta} - \rho^{\mathrm{KS}}_{\tau,\alpha\beta}\right] w^{*}_{\tau\alpha}(r) w_{\tau\beta}(r)$$

- Charge non self-consistent method :

If there is no
- Magnetic order
- Orbital order

$$\rho^{\mathrm{DMFT}}_{\tau,\alpha\beta} \approx \rho^{\mathrm{KS}}_{\tau,\alpha\beta}$$



- Wannierization
- QMC statistics

Carefully check each steps

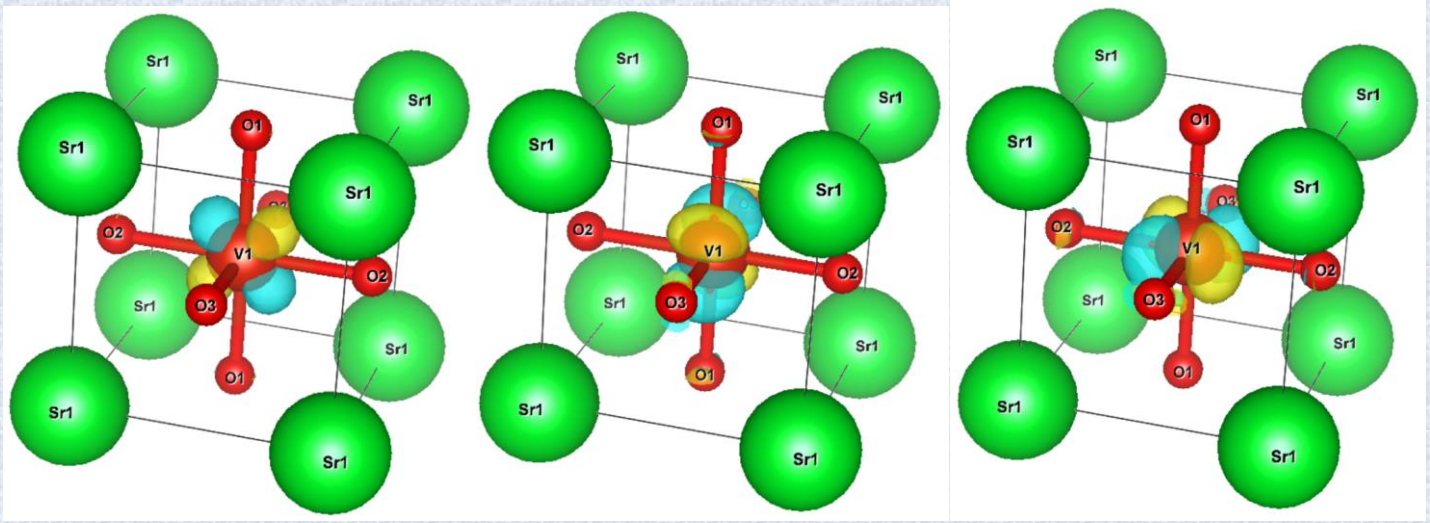
Numerical

Calculation steps

1. DFT calculation of charge density (QE: pw.x)
2. Band structure calculation (QE: pw.x, bands.x)
3. Non-SCF calculation for Wannierization (QE: pw.x, RESPACK: qe2respack.sh)
4. Wannierization (RESPACK: calc_wannier)
5. Dielectric function (RESPACK: calc_chiqw)
6. Effective interaction (RESPACK: calc_w3d, calc_j3d, DCore: respack2dcore.py)
7. DMFT calculation (DCore: dcore_pre, dcore, dcore_check)
8. DMFT post-process (DCore: dcore_post)

$SrVO_3$

- Correlated metal
- V^{4+}, t_{2g}^{1}



1, DFT calculation of charge density

$$\left(-\frac{1}{2}\nabla^2 + V_{KS}[\rho](r)\right)\varphi_{nk}(r) = \varepsilon_{nk}\varphi_{nk}(r) \qquad \rho(r) = \sum_{nk\sigma} |\varphi_{nk}(r)|^2$$

```
> cp -r /home/t0012/t001200/SrVO3 .
> cd SrVO3
> qsub 1_scf.sh
```

```
#!/bin/sh
#QSUB -queue ccms_i18cpu
#QSUB -node 1
#PBS -l walltime=0:30:00
source ~/.bashrc
export OMP_NUM_THREADS=1
cd $PBS_O_WORKDIR
mpijob pw.x -npool 24 -in scf.in > scf.out
```

```
> less scf.out
```

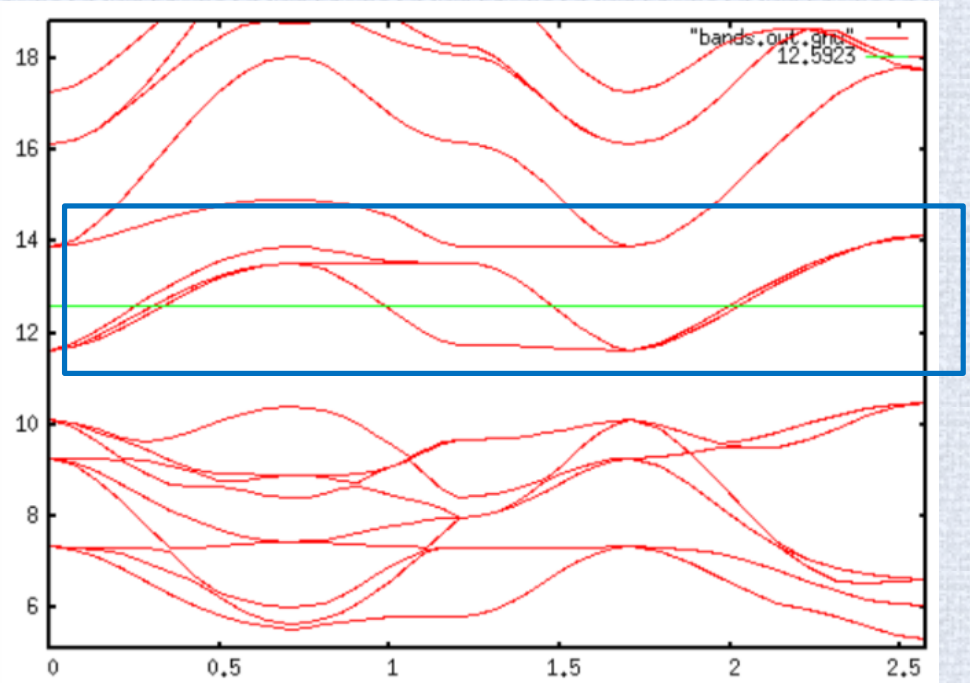
2, Band structure calculation

```
> qsub 2_band.sh
```

ε_{nk}

```
mpijob pw.x -npool 24 -in band.in > band.out
mpijob bands.x -npool 24 -in bandpp.in > bandpp.out
```

```
> grep Fermi scf.out
     the Fermi energy is    12.5923 ev
> gnuplot
  :
gnuplot> plot “bands.out.gnu”, 12.5923
```



Choose “outer energy window”

3, Non-SCF calculation for Wannierization

```
> qsub 3_nscf.sh
```

```
mpijob -n 20 pw.x -npool 10 -ntg 2 -in nscf.in > nscf.out
qe2respack.sh srvo3.save
```

```
&CONTROL
  ⋮
 wf_collect = .true.
/
&SYSTEM
  ⋮
 nbnd = 100
/
```

$$w_n(r-R) = \sum_m U_{knm}\, e^{-ikR} \varphi_{nk}(r)$$

4, Wannierization (1)

```
> qsub 4_wannier.sh
```

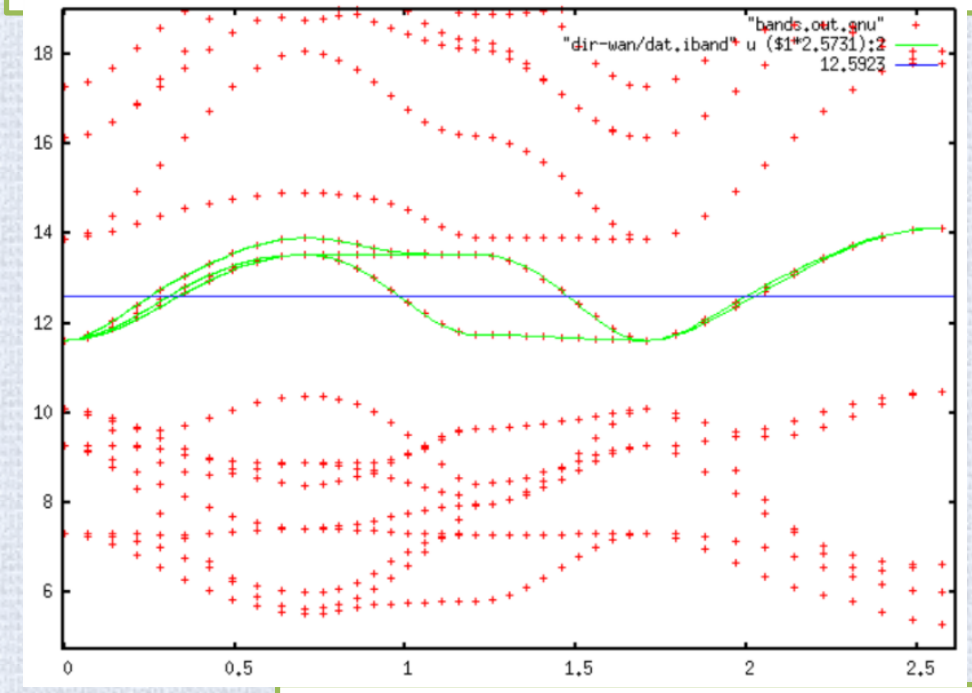
$$w_n(r-R) = \sum_m U_{knm}\, e^{-ikR} \varphi_{nk}(r)$$

```
  ⋮
#QSUB -node 1
#QSUB -mpi 1
#QSUB -omp 24
  ⋮
calc_wannier < respack.in
```

```
  ⋮
&PARAM_WANNIER
N_wannier = 3
Lower_energy_window = 11.0785
Upper_energy_window = 14.5440
N_initial_guess = 3
/
dxy 0.2 0.5 0.5 0.5
dyz 0.2 0.5 0.5 0.5
dzx 0.2 0.5 0.5 0.5
  ⋮
```

4, Wannierization (2)

```
> gnuplot
 ⋮
gnuplot> plot "bands.out.gnu" w p, ¥
"dir-wan/dat.iband" u ($1*2.5731):2 w l, 12.5923
```




```
> cat dir-wan/dat.wan-center
#Wannier center
#1:x, 2:y, 3:z (in xyz coord)
        3.6306363336        3.6306363302        3.6306363315
        3.6306363315        3.6306363351        3.6306363347
        3.6306363320        3.6306363315        3.6306363300
```

5, Dielectric function

$$\chi_q^0(\omega, r, r') = \sum_{knm} \frac{f(\varepsilon_{mk+q}) - f(\varepsilon_{nk})}{\varepsilon_{nk} - \varepsilon_{mk+q}} \varphi_{nk}^*(r)\varphi_{mk+q}^*(r')\varphi_{nk}(r')\varphi_{mk+q}(r)$$

$$\widehat{W}_q(i\omega) = \left(V_{q,\mathrm{bare}}^{-1} + \hat{\chi}_q\right)^{-1}$$

```
> qsub 5_chiqw.sh
```

```
 ⋮
#QSUB -node 3
 ⋮
mpijob -n 64 calc_chiqw < respack.in > chiqw.out
```

```
&PARAM_CHIQW
Num_freq_grid = 1
flg_cRPA = 1
/
 ⋮
```

6, Effective interaction

$$U_{mRnR'} = \iint d^3r\, d^3r' w_n^*(r-R) w_m^*(r'-R') W(\omega, r, r') w_m(r'-R') w_n(r-R)$$

$$J_{mRnR'} = \iint d^3r\, d^3r' w_n^*(r-R) w_m^*(r'-R') W(\omega, r, r') w_n(r'-R) w_m(r-R')$$

```
> qsub 6_calc_int.sh
```

```
 ⋮
#QSUB -node 1
#QSUB -mpi 1
#QSUB -omp 24
 ⋮
calc_w3d < respack.in > w3d.out
calc_j3d < respack.in > j3d.out
echo "">> dir-intJ/dat.Jmat
respack2wan90.py srvo3
```

```
 ⋮
&PARAM_CALC_INT
calc_ifreq = 1
ix_intJ_min = 0
ix_intJ_max = 0
iy_intJ_min = 0
iy_intJ_max = 0
iz_intJ_min = 0
iz_intJ_max = 0
/
```

7, DMFT calculation

```
> qsub 7_dcore.sh
```

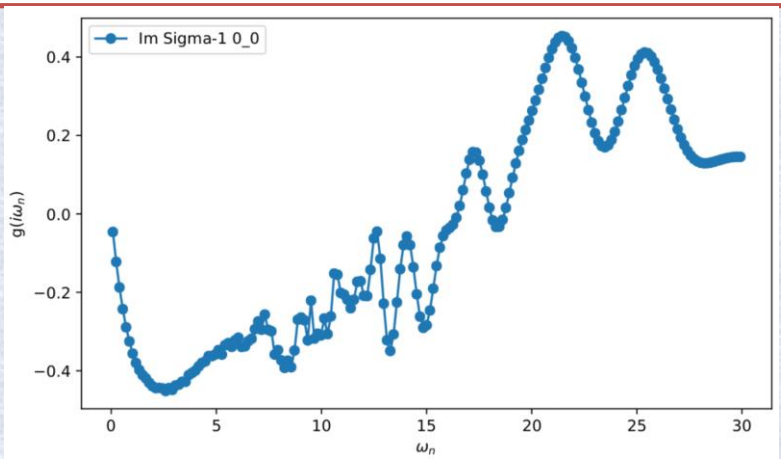
$\Sigma_{nm}(i\omega)$

```
mpijob -n 1 dcore_pre dcore.ini > pre.out
mpijob dcore dcore.ini > dcore.out
mpijob -n 1 dcore_check dcore.ini --output="dcore.pdf" > check.out
```

```
> less dcore.out
```

See Perturbation orders, Average sign, etc.

```
> evince dcore.pdf
```




```
⋮
[impurity_solver]
name = ALPS/cthyb
max_time{int} = 60
thermalization_time{int} = 30
verbosity{int} = 1
[control]
max_step = 1
restart = True
[tool]
omega_check = 30.0
```

```
> sed -i -e "/max_step/c max_step = 5" dcore.ini
> qsub 7_dcore.sh
```

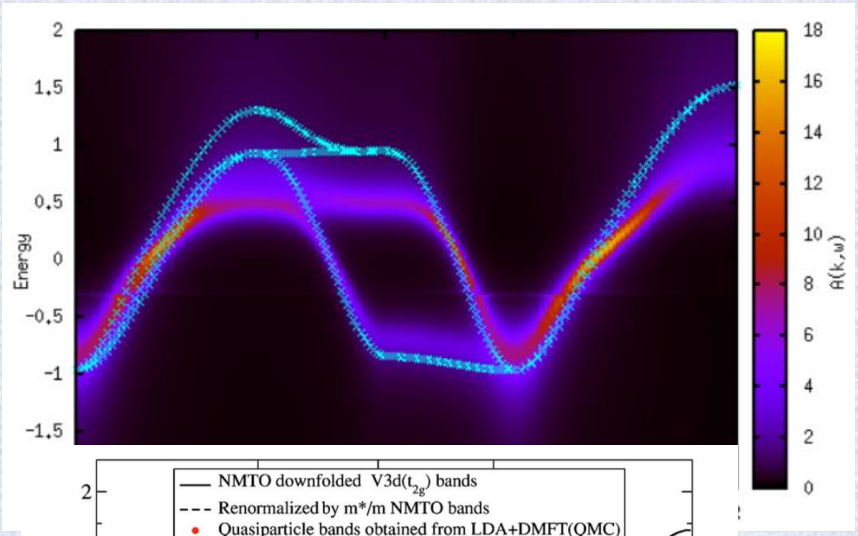
8, DMFT post-process

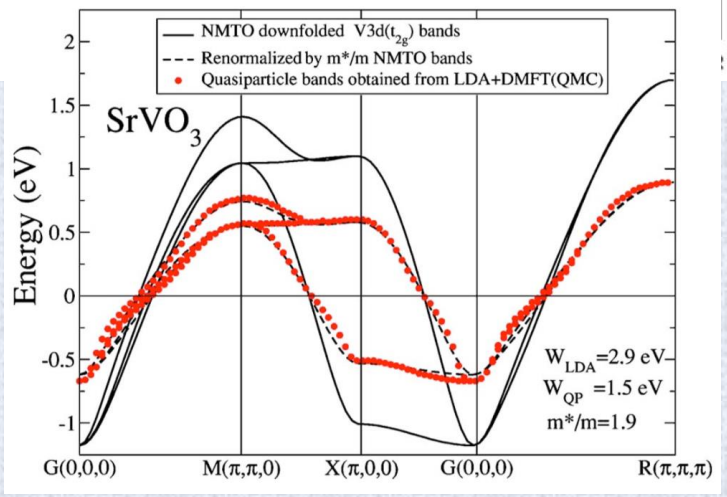
```
> qsub 8_post.sh
```

$A_k(\omega)$

```
mpijob -n 1 dcore_pre post.ini > post.out
mpijob dcore_post post.ini >> post.out
```

```
> sed -i -e "s/every 10//g" srvo3_akw.gp
```



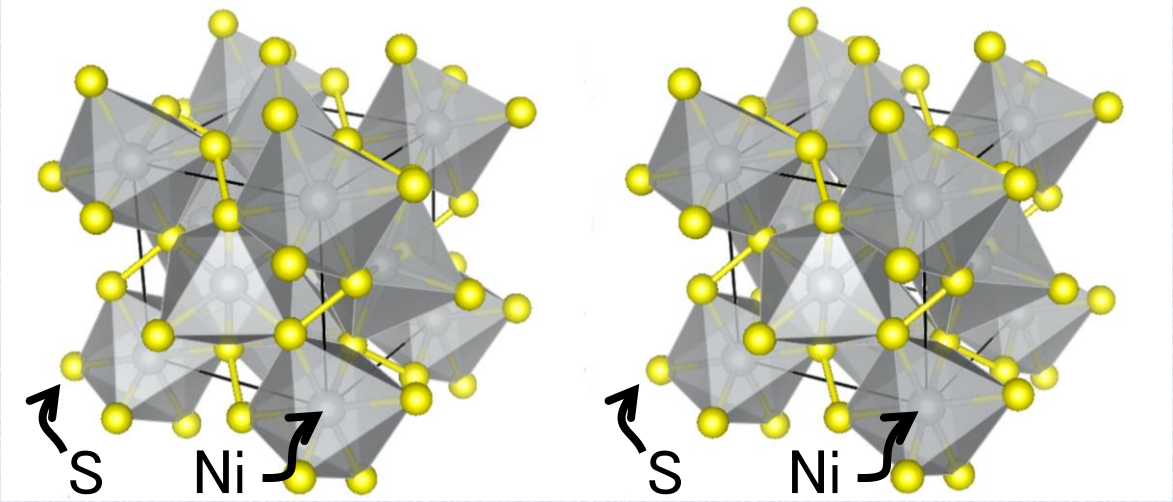



```
 ⋮
[system]
nk0 = 32
nk1 = 32
nk2 = 32
 ⋮
[tool]
broadening = 0.1
nk_line = 50
nnode = 5
knode=[(G,0.0,0.0,0.0),
(M,0.5,0.5,0.0), …]
omega_max =2.0
omega_min =-2.0
Nomega = 400
omega_pade = 6.0
```

I. A. Nekrasov, *et al.*, PRB 73, 155112 (2006).

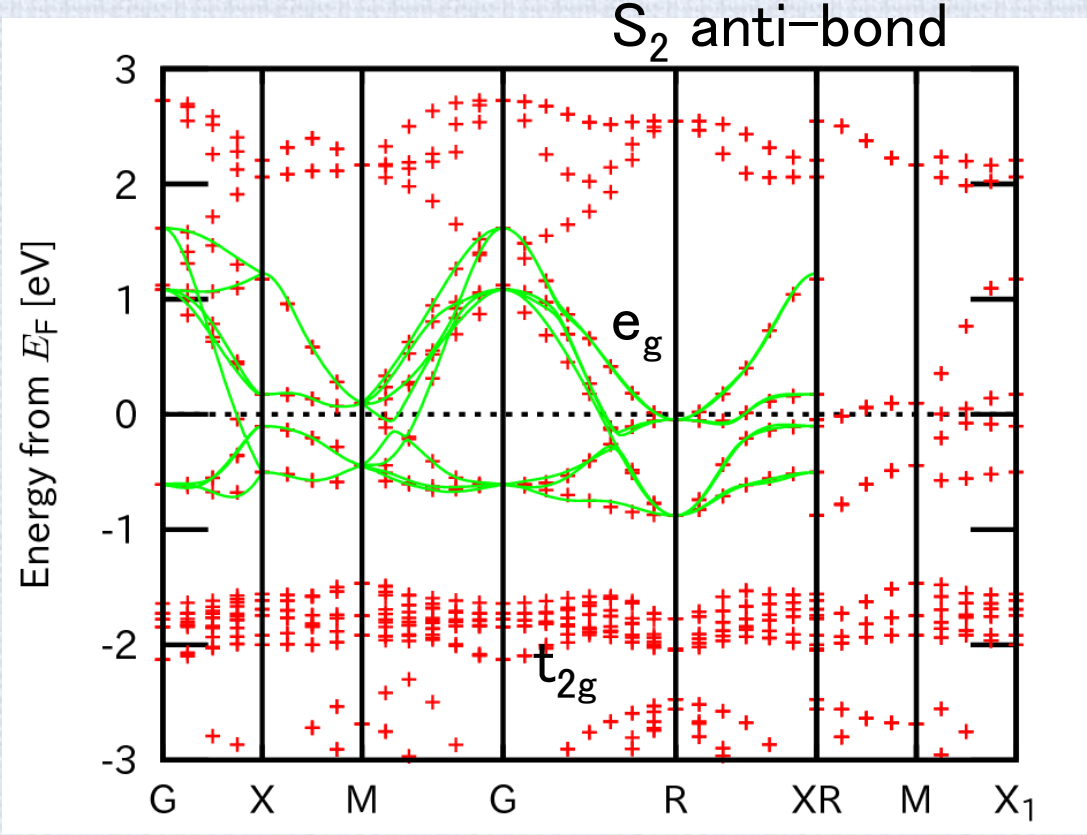
Result

TMS_2 (Pyrite structure)



- $TM^{2+}(S2)^{2-}$
- MnS_2 ($t_{2g}5$, e_g0)
- FeS_2 ($t_{2g}6$, e_g0)
- CoS_2 ($t_{2g}6$, e_g1)
- NiS_2 ($t_{2g}6$, e_g2), Mott ins.
- CuS_2 ($t_{2g}6$, e_g3)
- ZnS_2 ($t_{2g}6$, e_g4)

- Quantum ESPRESSO
- Norm-conserving PP (SG15)
- Cutoff(WFC): 60 Ry (CoS_2), 65 Ry(NiS_2), 90 Ry(CuS_2)
- GGA-PBE+DMFT
- cRPA(RESPACK)
- k-grid : 6×6×6 (Charge), 4×4×4 (Wannier, cRPA), 12×12×12 (DMFT), 32×32×32 (DOS)
- cRPA bands: 116(CoS_2), 120(NiS_2), 124(CuS_2)

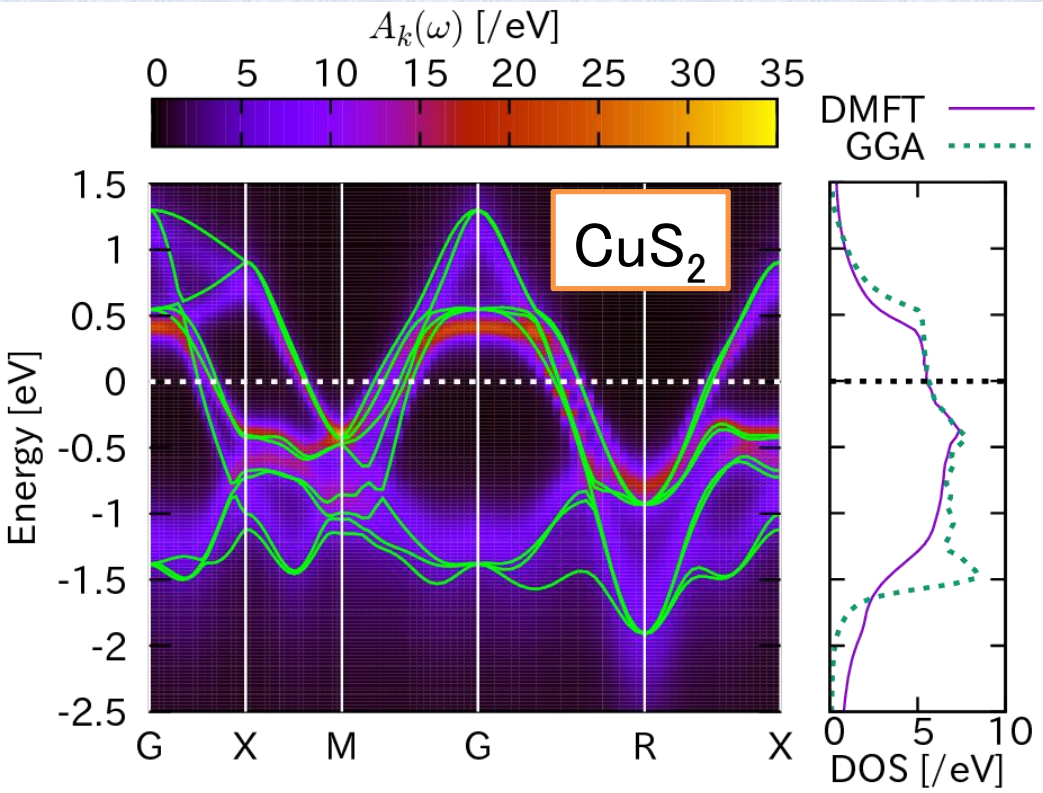


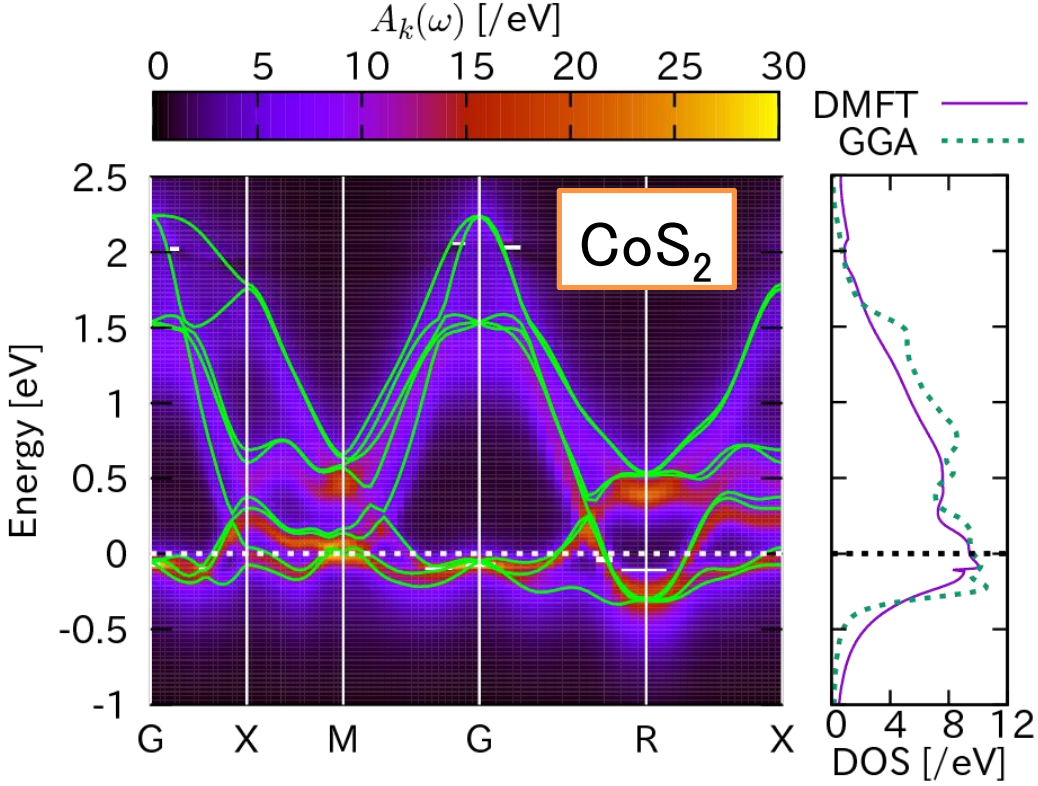
Result

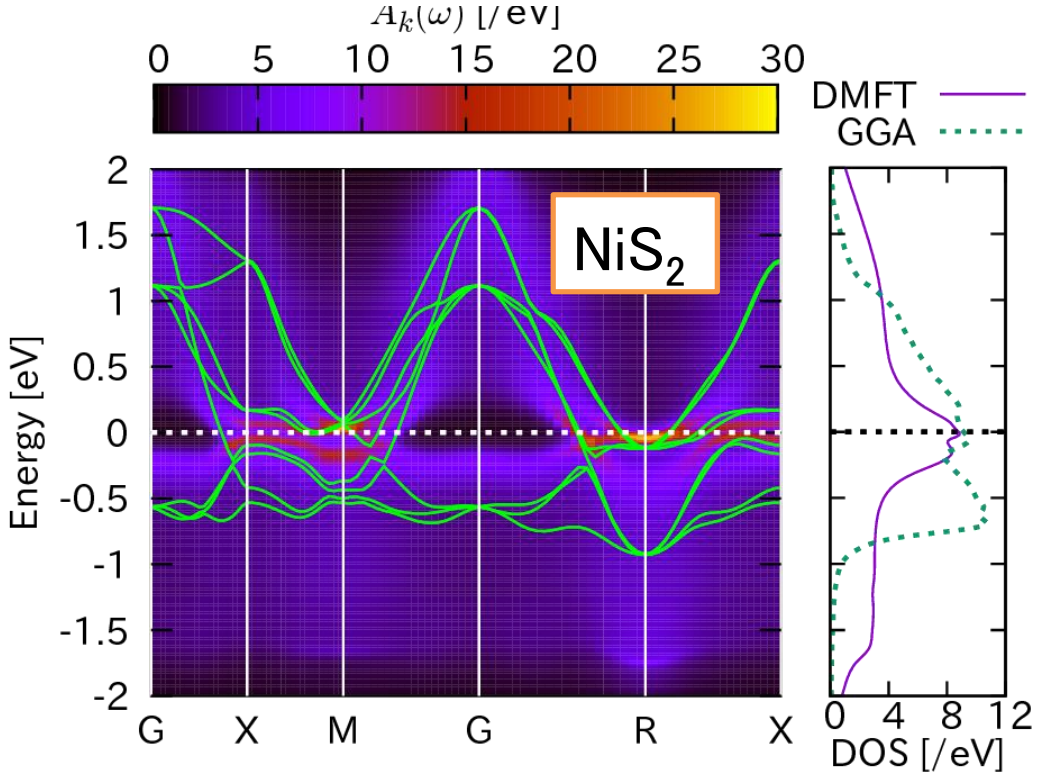
Result

[eV]	U	U'	J
CoS_2	1.24	0.45	0.39
NiS_2	1.43	0.69	0.37
CuS_2	1.61	1.05	0.28

$\beta = 40$ (290 K)

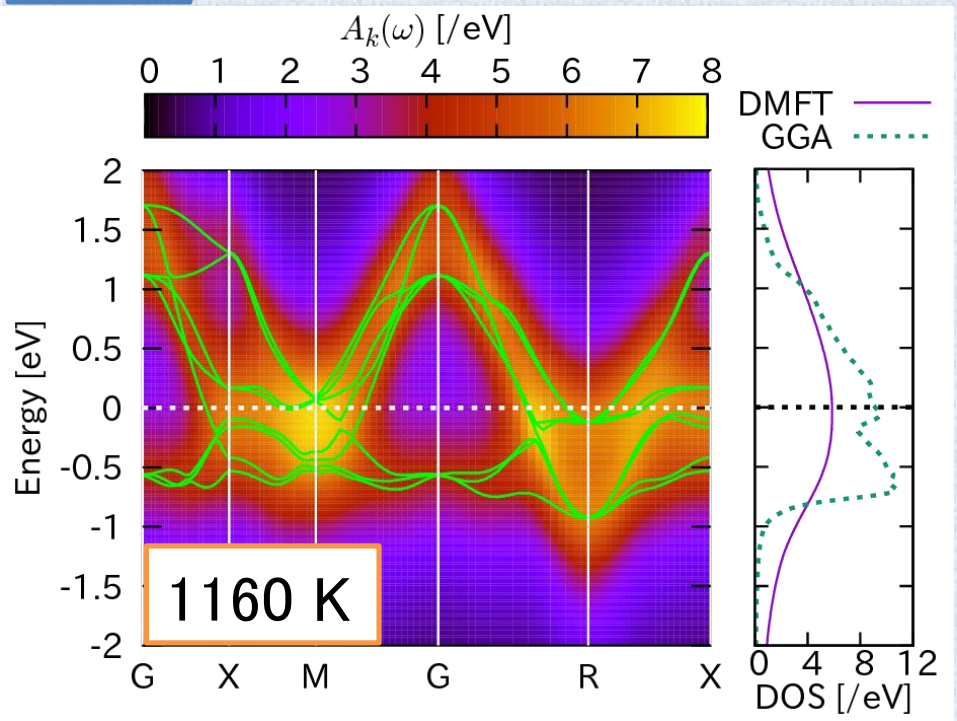


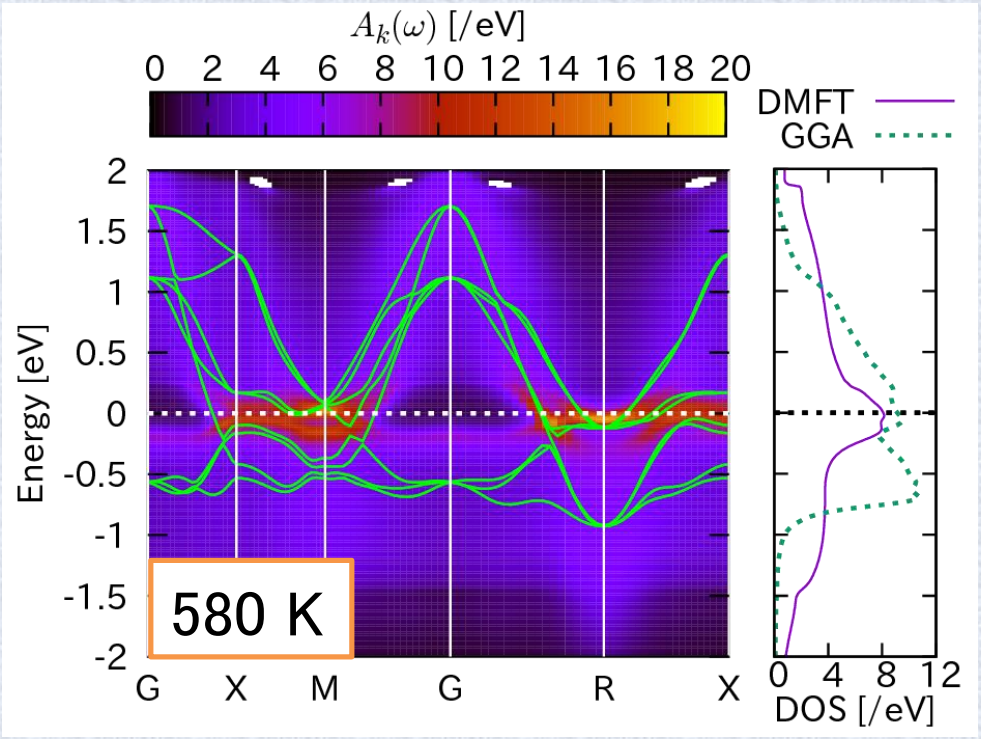


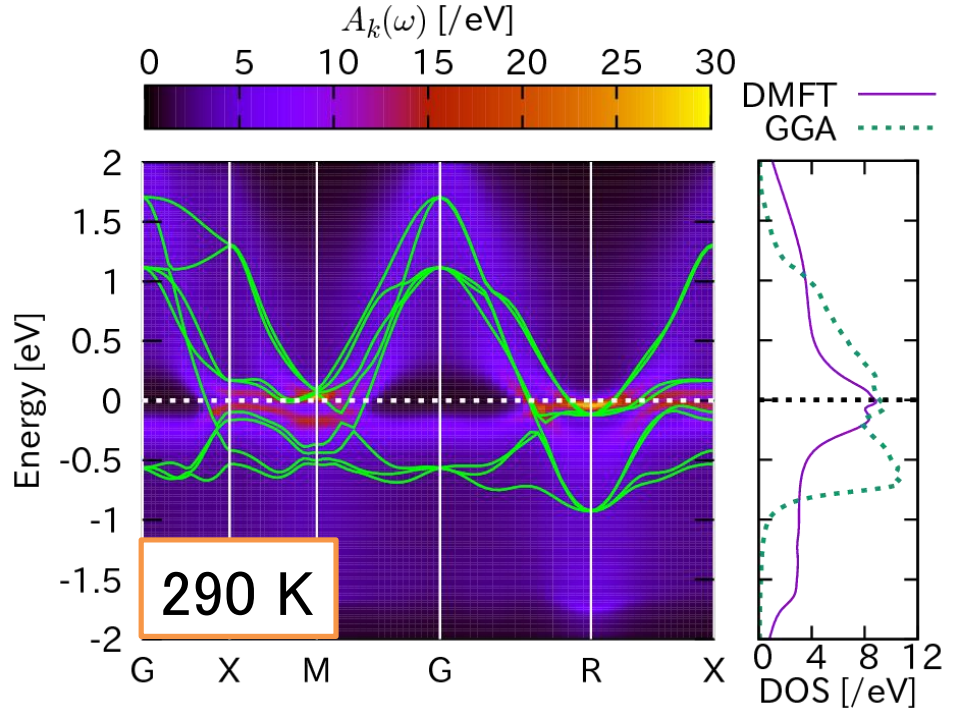


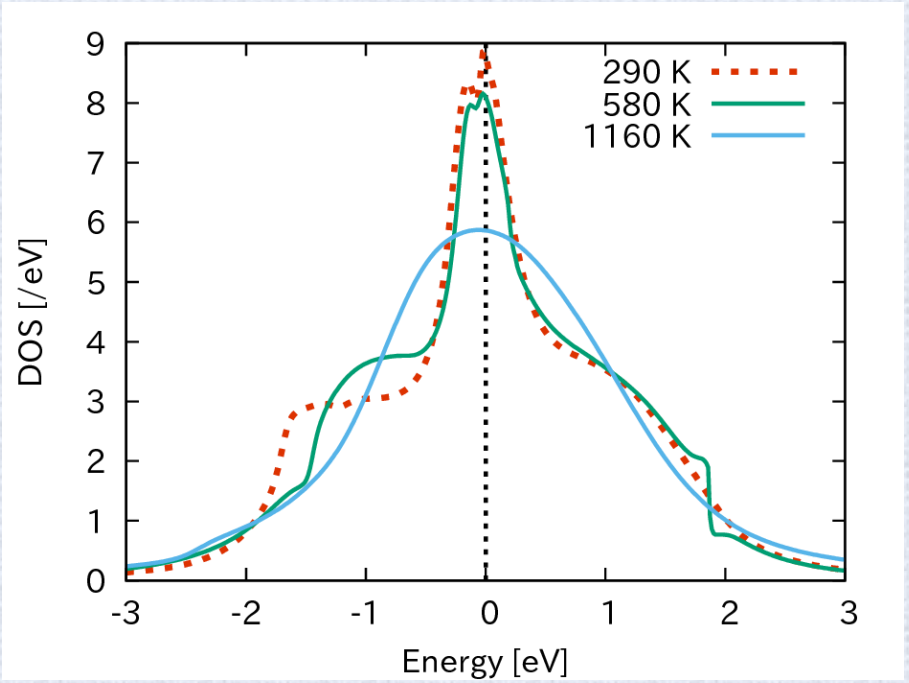
Result

Result









Summary

- We overview the theory and procedure of DFT+DMFT calculation with DCore.
  1. DFT calculation of charge density
  2. Band structure calculation
  3. Non-SCF calculation for Wannierization
  4. Wannierization
  5. Dielectric function
  6. Effective interaction
  7. DMFT calculation
  8. DMFT post-process