Constrained RPA

Jinyuan Wu

April 6, 2023

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Important papers about cRPA

- F Aryasetiawan et al. "Frequency-dependent local interactions and low-energy effective models from electronic structure calculations". In: *Physical Review B* 70.19 (2004). Fig. 3 and Fig. 4 in the article are swapped in the published version: compare them with Fig 2, p. 195104
- F Aryasetiawan, T Miyake, and R Sakuma. "The constrained RPA method for calculating the Hubbard U from first-principles". In: *The LDA+ DMFT approach to strongly correlated materials*. 2011

Motivation

Strongly correlated electrons are ...

- Hard to treat *ab initio*
- Should be described by a lattice model

$$H = -t \sum_{\langle \boldsymbol{i},\boldsymbol{j} \rangle,\sigma} c^{\dagger}_{\boldsymbol{i}\sigma} c_{\boldsymbol{j}\sigma} + \text{h.c.} + U \sum_{\boldsymbol{i}} n_{\boldsymbol{i}\uparrow} n_{\boldsymbol{i}\downarrow}. \tag{1}$$

But should all electrons be included?

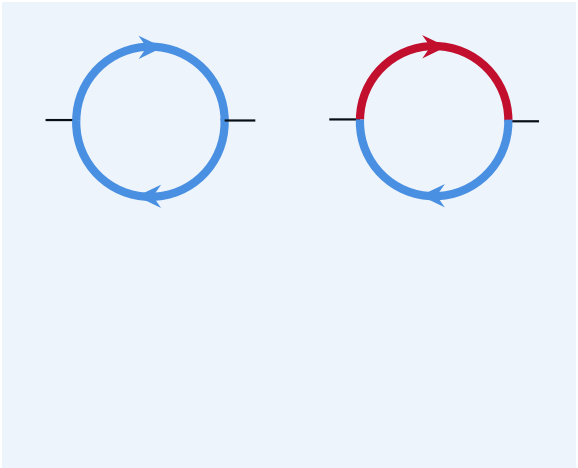
- d electron: correlated ones
- r electron: "trivial" ones

Diagrammatics

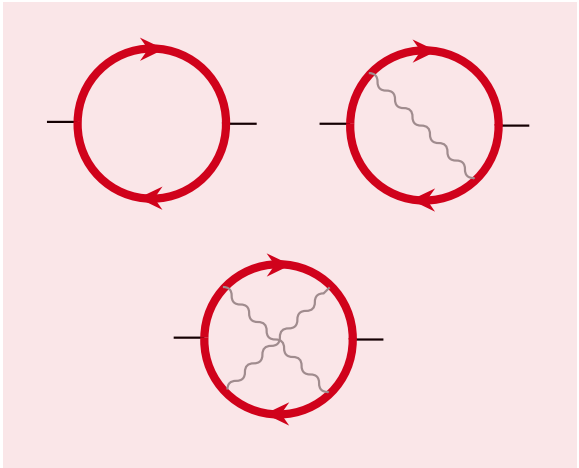
Screening by d and r = first screened by r and then d

$$W = (1 - vP)^{-1}v = (1 - W_r P_d)^{-1} W_r, \quad W_r = (1 - vP_r)^{-1}v, \qquad (2)$$

- W is effective interaction
- where P_d is the polarization within d subspace,
- and $P_r = P - P_d$



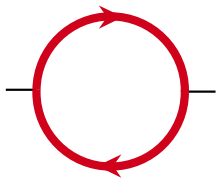
P_r
RPA works



P_d
RPA doesn't work

Diagrammatics

- P_d hard to obtain $\Rightarrow$ we left it to an effective interaction without doing resummation for it
- P_r can be done by `epsilon` $\Rightarrow$ W_r can be obtained by `epsilon`
- But



  is not in P_r, or otherwise we have double counting
- That's what we call **constrained RPA (cRPA)**
- A program to calculate a single ring diagram is needed

Band part

In GW:

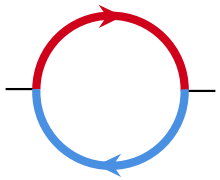
$$\begin{aligned} \varepsilon^{GW} &= \varepsilon^{0} + \Sigma^{\text{Hartree}} + \Sigma^{GW} \\ &= \underbrace{\varepsilon^{0} + \Sigma^{\text{Hartree}} + V^{\text{xc}}}_{\texttt{WFN}} + \underbrace{\Sigma^{GW}}_{\texttt{sigma}} - \underbrace{V^{\text{xc}}}_{\texttt{vxc.dat}} \end{aligned} \tag{3}$$

In the effective model ...

- We want ε^{0}
- So the same $-V^{\text{xc}}$ procedure is needed(?)

Risks in accuracy

Uncontrolled approximation: does RPA work for



The hopping between d electrons is small $\Rightarrow$ it's safe to do so[1]

[1] F Aryasetiawan et al. "Frequency-dependent local interactions and low-energy effective models from electronic structure calculations". In: *Physical Review B* 70.19 (2004). Fig. 3 and Fig. 4 in the article are swapped in the published version: compare them with Fig 2, p. 195104.

Dynamic interaction in Hubbard model

Recall that $W = W(\boldsymbol{r}, \boldsymbol{r}, \omega) \Rightarrow U_{\boldsymbol{ijkl}}(\omega) \Rightarrow$ retarded interaction!

- Hamiltonian form: interactions are always immediate (bosonic auxiliary field required to create retardation)
- . . . or path integral formalism is to be used
- In either cases, it's slow!!!

Comment: frequency dependence in effective models

This actually reflects something deep about downfolding ...

- **Hamiltonian effective theory; Rayleigh-Schrodinger perturbation theory.** In $1/(E-H)$, E is unperturbed energy
- **Feynman diagram resummation**. After splitting G into G^+ and G^-, it's equivalent to Brillouin-Wigner perturbation theory; In $1/(E-H)$, E is the *perturbed* energy
- An item in BWPT splits into an infinite series in RSPT![2]

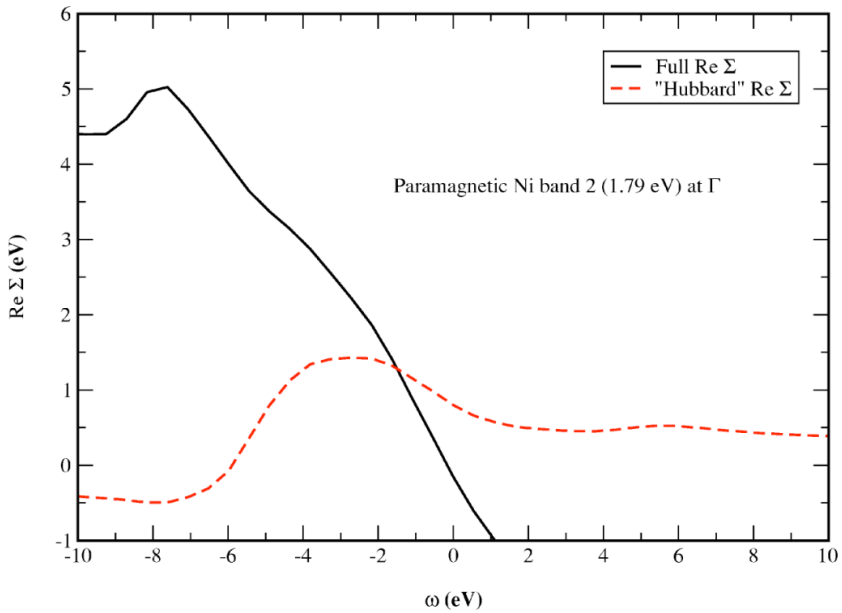
$$\begin{aligned} E^{GW} &\simeq E^{\mathrm{DFT}} + \mathrm{i}\, W \frac{1}{E^{GW} - E^{\mathrm{DFT}}} \\ &= E^{\mathrm{DFT}} + \mathrm{i}\, W \frac{1}{E^{\mathrm{DFT}} - E^{\mathrm{DFT\ (other\ bands)}}} + \cdots \end{aligned} \tag{4}$$

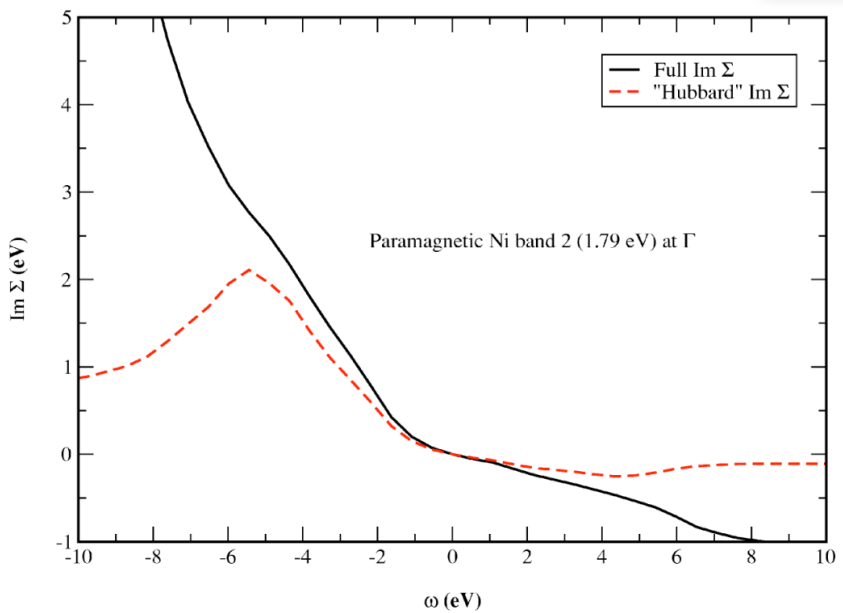
$\Rightarrow$ why we need eigenvalue self-consistency

[2] KA Brueckner. "Many-body problem for strongly interacting particles. II. Linked cluster expansion". In: *Physical Review* 100.1 (1955), p. 36.

Dynamic interaction in Hubbard model

Is it possible to just enforce $\omega = 0$ . . . But it's not accurate![3]





Ni self-energy, from static Hubbard $U = W_r(\omega = 0)$, and from full $W(\omega)$

[3] Aryasetiawan et al., "Frequency-dependent local interactions and low-energy effective models from electronic structure calculations".

Dynamic interaction in Hubbard model

Why $\mathrm{Im}\,\Sigma$ is better captured by static U than $\mathrm{Re}\,\Sigma$?

Because $\mathrm{Im}\,\Sigma \propto$ DOS

And then high-frequency electrons contribute to low-frequency $\mathrm{Re}\,\Sigma$

Making the Hubbard model static: method 1

Key point: correct the single-electron Hamiltonian using the retarded interaction

- Find static non-interactive $\tilde{G}_d$ such that –
- With the same static U, $\tilde{G}_d$ leads to an interactive Green function close enough to the real one

Making the Hubbard model static: method 1

Method 1: correct the single-electron Hamiltonian using the retarded interaction

- Def: G_d = Green function for d electrons; $\tilde{G}_d$ = Green function for d electrons corrected by $W - W_d$; W = RPA screened Coulomb interaction; W_d = static Hubbard U screened by G_d; $\tilde{W}_d$ = static Hubbard U screened by $\tilde{G}_d$;
- Ignoring hopping between d and r subspaces caused by W (so the $\mathrm{i}\, W(G - G_d)$ term is irrelevant for d electrons), we have

$$\tilde{G}_d^{-1} - \mathrm{i}\, \tilde{G}_d \tilde{W}_d = G_d^{-1} - \mathrm{i}\, G_d W_d - \mathrm{i}\, G_d (W - W_d). \tag{5}$$

- From

$$W = (1 - UP)^{-1} U, \quad P = -\mathrm{i}\, GG \Rightarrow G^{-1} - \mathrm{i}\, GW = (1 - UP)^{-1} G^{-1} \tag{6}$$

we get the final equation: (here $\tilde{P}_d = -\mathrm{i}\, \tilde{G}_d \tilde{G}_d$, $P_d = -\mathrm{i}\, G_d G_d$)

$$(1 - U\tilde{P}_d)^{-1} \tilde{G}_d^{-1} = (1 - UP_d^{-1})^{-1} G_d^{-1} - \mathrm{i}\, G_d (W - W_d). \tag{7}$$

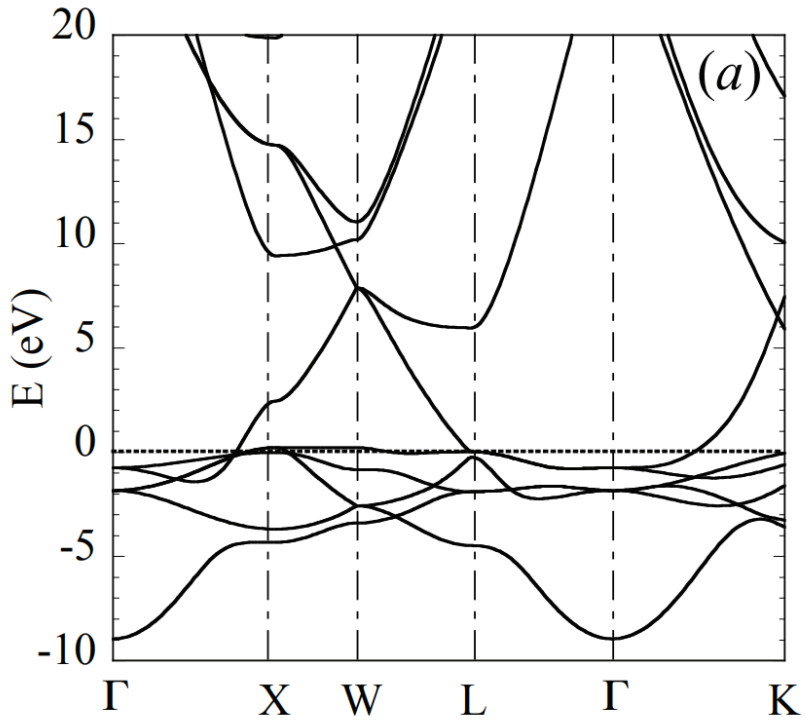
Making the Hubbard model static: method 2

Key point: introducing a bosonic field to "carry" the frequency-dependent $W(\omega)$.[4]
(Similar techniques are used in DMFT)

[4] M. Casula et al. "Low-Energy Models for Correlated Materials: Bandwidth Renormalization from Coulombic Screening". In: *Phys. Rev. Lett.* 109 (12 2012), p. 126408. DOI: 10.1103/PhysRevLett.109.126408. URL: https://link.aps.org/doi/10.1103/PhysRevLett.109.126408.

cRPA for entangled bands

Entangled band problem A band consisting mostly of *s* electrons and a band consisting mainly of *d* electrons may cross each other



And an effective theory solely targeting *d* electrons is not well-defined

cRPA for entangled bands

Solution Sacrifice some inter-orbital hopping terms for disentangled band structure

1. Standard DFT+Wannier run.
2. Wannier and fake Bloch functions for the d subspace:

$$\psi^d_{n\boldsymbol{i}} = \frac{1}{N}\sum_{\boldsymbol{k}} e^{-\boldsymbol{k}\cdot\boldsymbol{R_i}} \underbrace{\sum_m U_{mn}(\boldsymbol{k})\psi_{m\boldsymbol{k}}}_{\psi^d_{n\boldsymbol{k}}}, \tag{8}$$

3. Getting the r-subspace

$$\psi^r_{n\boldsymbol{k}} = \left(1 - \sum_m |\psi^d_{m\boldsymbol{k}}\rangle\langle\psi^d_{m\boldsymbol{k}}|\right)\psi_{n\boldsymbol{k}}. \tag{9}$$

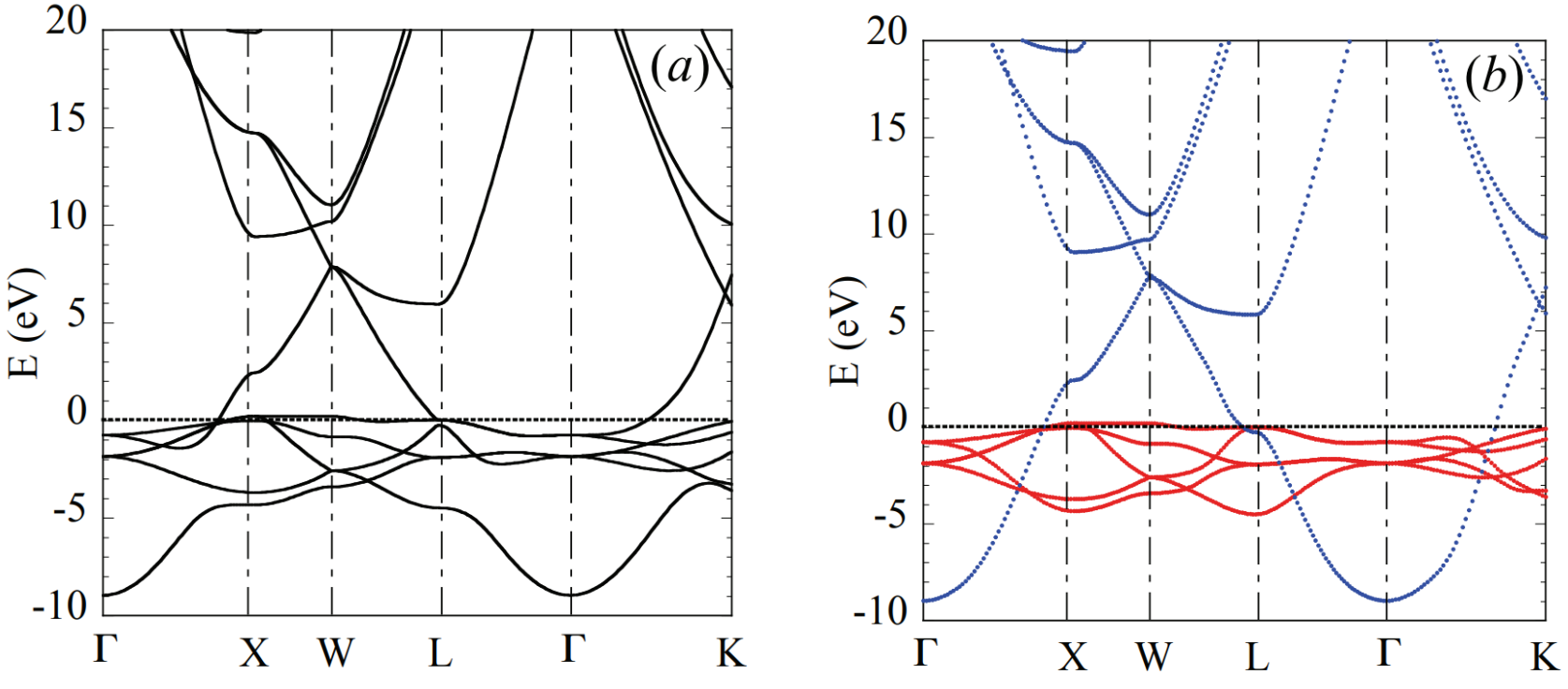
4. Calculating

$$H^{dd} = [\langle\psi^d_{m\boldsymbol{k}}|H(\boldsymbol{k})|\psi^d_{n\boldsymbol{k}}\rangle], \quad H^{rr} = [\langle\psi^r_{m\boldsymbol{k}}|H(\boldsymbol{k})|\psi^r_{n\boldsymbol{k}}\rangle] \tag{10}$$

H^{dr} is thrown away.

cRPA for entangled bands

Left: original bands



Right: H^{dd} and H^{rr}; after throwing away some *s*-*d* hopping (and *closing* several band gaps), the energy of H^{dd} is now bounded

Existing implementations

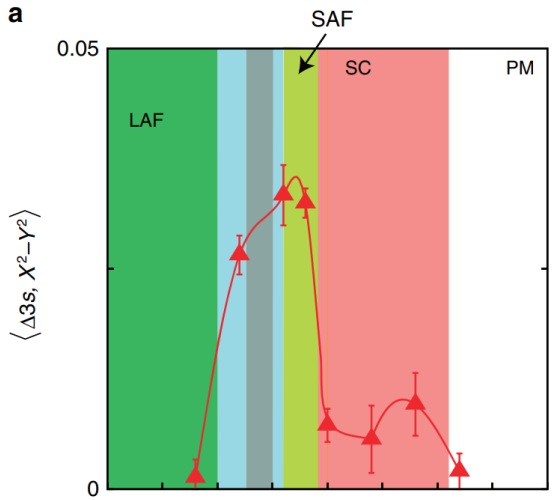
- **VASP** `ALGO=CRPA` selects constrained RPA calculations, available as of VASP.6.4
- **ABINIT** `ucrpa` before version 9, as an option in the RPA module
- **BerkeleyGW** ✗Unfortunately . . .
- **RESPACK**[5]: from Quantum ESPRESSO

[5] Kazuma Nakamura et al. "RESPACK: An ab initio tool for derivation of effective low-energy model of material". In: *Computer Physics Communications* 261 (2021), p. 107781.

Example: Fe-based superconductor

The cRPA-to-Hubbard methodology has already been applied to Fe-based SC[67]

Some claim that they can obtain the correct doping concentration required for SC[8]



[6] Kazuma Nakamura, Ryotaro Arita, and Masatoshi Imada. "Ab initio derivation of low-energy model for iron-based superconductors LaFeAsO and LaFePO". In: *Journal of the Physical Society of Japan* 77.9 (2008), p. 093711.

[7] Takashi Miyake et al. "Comparison of ab initio Low-Energy Models for LaFePO, LaFeAsO, BaFe, LiFeAs, FeSe, and FeTe: Electron Correlation and Covalency". In: *Journal of the Physical Society of Japan* 79.4 (2010), p. 044705. DOI: 10.1143/jpsj.79.044705. URL: https://doi.org/10.1143%2Fjpsj.79.044705.

[8] Takahiro Misawa and Masatoshi Imada. "Superconductivity and its mechanism in an ab