Introduction to LDA + DMFT (II) [Kotliar]

- Example: Building Kohm–Sham Eqns:
  - $S_0 + \int J\rho = \int \bar{\psi}\left(\frac{\partial}{\partial\tau} + V_{cryst} - \nabla^2 + J_0\right)\psi = \tilde{S}_0$

    $\Longrightarrow \Gamma_0[\rho] = \mathrm{Tr}\ln\left(\frac{\partial}{\partial\tau} + g_0 - \nabla^2 + V_{cryst}\right) - \int J_0(x)\rho(x)$ $\quad [x = (r,\tau)$
  - $\Delta\Gamma[\rho] = \frac{\beta e^2}{2}\int_0^1\iint \frac{\langle\bar{\psi}(r)\psi(r)\,\bar{\psi}(r')\psi(r')\rangle_{(J_0(\rho),\lambda)}}{|r-r'|}\,dr\,dr'\,d\lambda$
  - Now define V_{KS} via $V_{KS} = V_{cryst} + J_0$ (connected part)
  - By factorization, $\Delta\Gamma(\rho) = \underbrace{\frac{\beta e^2}{2}\int_0^1\iint \frac{\langle\bar{\psi}(r)\psi(r)\,\bar{\psi}(r')\psi(r')\rangle_c(J_0(\rho),\lambda)}{|r-r'|}\,dr\,dr'\,d\lambda}_{=\Delta\Gamma_{xc}(\rho)} + \underbrace{\frac{\beta e^2}{2}\iint\frac{\rho(r)\rho(r')}{|r-r'|}\,dr\,dr'}_{=\Delta\Gamma_{Hartree}(\rho)}$
  - In LDA, $\Delta\Gamma_{xc}(\rho) \approx \int \underbrace{\varepsilon_{xc}(\rho(r))}_{\propto\rho^{1/3}}\,dr$
  - Technically, we may solve numerically the interacting e^- problem and extract $\varepsilon_{xc}(\rho(r))$, & use it for all problems.

- Example: Building DMFT for Hubbard
  - $\mathcal{H} = \sum (t_{ij} - \mu\delta_{ij})(c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + U\sum_i n_{i\uparrow}n_{i\downarrow}$
  - $\Gamma_0[G_{ii}, M_{ii}] = -\mathrm{Tr}\ln(i\omega + \mu - t_{ij} - M_{ii}) - \mathrm{Tr}[MG]$

    $\Delta\Gamma_{xc} \approx \sum_i \Delta\Gamma_{xc\text{-}atom}[G_{ii}]$

    $G_{ii} = \sum_k \frac{1}{i\omega+\mu-\varepsilon(k)-M_{ii}}$

    $M_{ii} = \frac{\delta}{\delta G_{ii}}\Delta\Gamma_{atom}[G_{ii}]$
  - We start with atomic problem as S_0. Thus diagramatically the above corresponds to summing diagrams to all order
  - Self-consistency: $G_0 \rightarrow$ impurity problem $\rightarrow G_c, M$; $G_0 \leftarrow$ self-consistency

    where
    $$\begin{cases} G_c(\tau) = -\langle T c(\tau) c^\dagger(0)\rangle_{S_{eff}} \\ S_{eff} = -\int_0^\beta c_\sigma^\dagger(\tau) G_0^{-1}(\tau-\tau') c_\sigma(\tau') + \int_0^\beta d\tau\, U n_\uparrow(\tau) n_\downarrow(\tau) \\ \mathcal{M} = G_0^{-1} - G_c^{-1} \\ G_0^{-1}(i\omega) = \left(\sum_k \frac{1}{i\omega_n + \mu - \varepsilon(k) - M(i\omega_n)}\right)^{-1} + \mathcal{M}(i\omega_n) \end{cases}$$

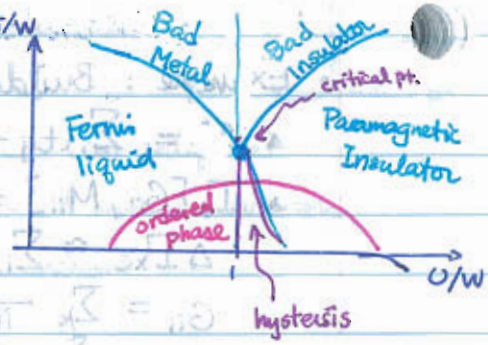
▲ In doing so, we need efficient numerical method to solve the one-site interacting problem. i.e., a "black box":

where $G_0^{-1} = i\omega_n + \mu - \Delta$

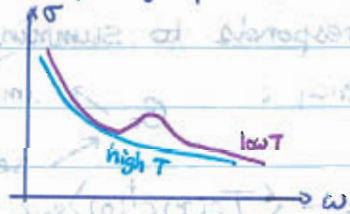
• The Weiss field $\Delta(\omega)$ quantifies degree of localization
  ▲ $\Delta(\omega)$ small $\Longrightarrow$ localized, $\Delta(\omega)$ large $\Longrightarrow$ delocalize
  ▲ Note that localization of not is freq. dependent
  ▲ $\Delta(\omega)^{ab} \sim \sum_\alpha \frac{V_\alpha^{a*} V_\alpha^{b}}{\omega - \varepsilon_\alpha}$

• From the calculation we can also extract valence histogram

• Phase diagram — frustrated Hubbard
  - (Rozenberg, PRL, 75, 105 (1995)



• Example : V_2O_3
  ▲ DMFT suggests transfer of spectral weight, which is observed in experiment



  ▲ The (Ising) critical point & hysteresis are also observed.

• To solve for strip order, etc., need multi-impurity model
  ▲ But each impurity

- The hunt for strip pattern, need multi-impurity model
  - ▲ each impurity interacts with each other only through self-consistency.

- For 5f, Pu is close to Mott transition. Traditional (~DFT) methods have problem getting good results, but LDA + DMFT get reasonable results
  - ▲ e.g. total energy as function of volume for Pu
  - ▲ Allowing coupling to lattice, rich phase diagram can be obtained (lattice structure, etc.)
  - ▲ DMFT also match well to phonon spectrum of Pu
    - ▶ Phonon spectrum of Pu is hard to measure by neutron, since single crystal is hard to get & Pu absorb neutron. Problem can be circumvent by X-ray.
  - ▲ The high temperature (δ-phase) phase is more localized than the low temperature (α-phase) phase for 5f electrons.
    $\Longrightarrow$ transfer of spectrual weight in (e.g.) photoemission
  - ▲ Pu also shows very different valence diagram than usual (many valence states are important, particular between 5+ & 6+)
    - ▶ The "conjugate" element Curium does not show the same mixed valance.

- Idea of LDA + DMFT —— treat delocalized e^- (s, p, sometimes d) by DFT & localized e^- (f, sometimes d) by DMFT
  - ▲ LDA + U amounts to treating the localized problem by Hartree-Fock.
  - ▲ LDA + U & LDA + DMFT are "2nd principle" calculations, since some parameters ("J", "U") have to be put in. (LDA itself is parameter free)