Sufaces & Interfaces (III) [Millis]

- Theoretical problem: need to solve inhomogenous strongly correlated systems.
  - 1D : numerical / exact solutions ; otherwise it's generally some kind of mean field, e.g. band theory, Hartree Fock, etc.
  - Part of the issue is to find a good system to study — simple enough to model (or to grow stablily), but complicated enough to show "new physics".

- Density Functional Theory : there is a functional of density

  $$\Gamma[\rho] = \Gamma_{univ}[\rho] + \int V_{lattice}(\vec{r})\,\rho(\vec{r})\,d\vec{r}$$

  minimized at physical density, and its minimum gives ground state energy
  - have uncontrolled (but good) approximation to Γ, and efficient ways to calculate
  - LDA + U : add Hartree energy to LDA energy
  - It may be useful to use DFT to define the Hamiltonian

- Charge profile: dipole layers, strong Coulomb, lattice distortion/screening
  - For $SrTiO_3$, $\epsilon > 10^3$

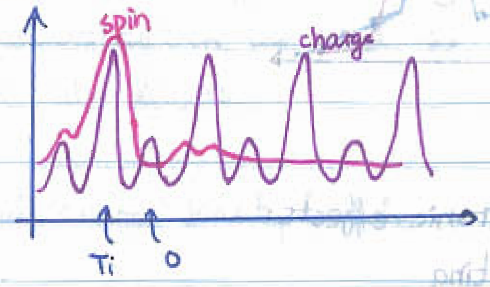
- More generally, proximity of interface $\Rightarrow$ symmetry broken $\Rightarrow$ level spliting & changes in hopping.

- Example : $(LaTiO_3)_1/(SrTiO_3)_8$ — LDA + U
  - Allow atoms to relax $\perp$ to the planes
  - Found O moves towards La by 10% of original bond length, for nearest O.
  - Relaxation negligible in far regions.
  - The O—Ti—O bond becomes buckled.
    (Ti—Ti bond length in $\hat{z}$ also changes)

- ▲ The near La layers shows ferroelectric distortion.

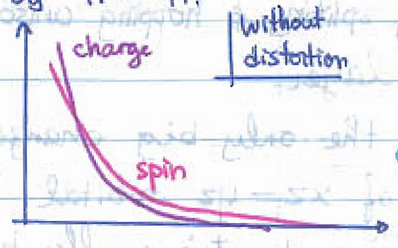
◦ Understanding e^--density calculated from band calculations

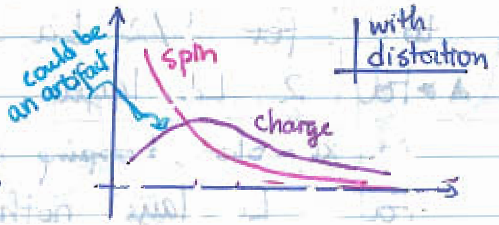
- ▲ outputs: atomic positions, charge density, many bands.... difficult to interpret
- ▲ calculations do not use all e^-, core e^- treated by (e.g.) pseudopotential, but what "core" amounts to depend on situation
- ▲ what concern us is conduction e^- density. Either check bands that cross Fermi energy (too many bands to be useful) or just consider charge "around" Ti (to determine "around", draw sphere of radius r around Ti, but result depends crucial r)
- ▲ Another possibility is to focus on spin density & consider it as the desired correlated conduction e^-. This works in this case only because the ground state is fully polarized ferromagnet



- ▲ charge near Ti
- ▲ spin near Ti
- ▲ spin decay fast

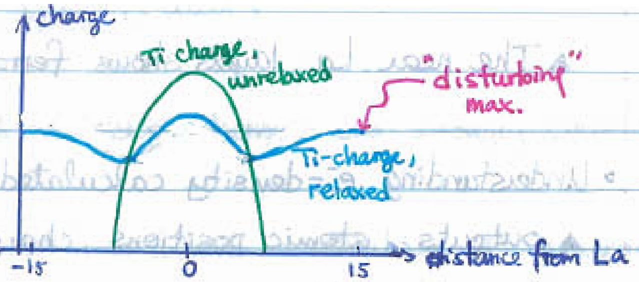
- ▲ To interpret charge density, use the unit cell furthest away from La as reference pt ("∞-far away" pt)
  - ▸ But with distortion, "unit cell" not of same size. Define unit cell by Ti — Ti.





- ▲ with distortion, charge decay is much slower (lattice distortion tends to create uniform density (screening)

- If we include GGA:

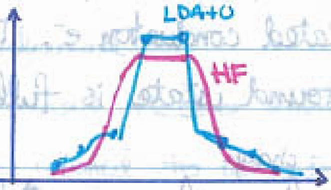


  - ▲ Shows maximum outside Ti ion (!)

- Consider toy model — tight-binding + self-consistent potential

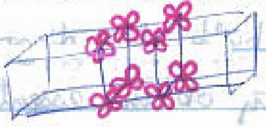
$$\mathcal{H} = \underbrace{-\sum_{ijab\sigma} t_{ij}^{ab} d_{ia\sigma}^{\dagger} d_{jb\sigma}}_{\text{tight-binding}} \underbrace{-\sum_{j} \frac{e^2}{\epsilon |\vec{R}_i - \vec{R}_j^A|}}_{\text{La charge}} \underbrace{+ \sum_{j\sigma} \frac{e^2 \langle n_{j\sigma} \rangle}{|\vec{R}_i - \vec{R}_j|}}_{\text{hartree}}$$

  - ▲ For unrelaxed structure, $\underset{\equiv \text{Hartree-Fock}}{\underline{\text{HF}}}$ with $\epsilon = 4$ in above model fit well to LDA+U.
  - ▲ For relaxed structure, HF with $\epsilon = 15$ in above model fit okay to LDA+U (fail to get sharp / fat tail):



- Other possible electronic effects:
  - ▲ t2g level spliting
  - ▲ hopping anisotropy
  - ▲ Consider Ti ion, take lattice distortion from LDA+U, fit tight binding parameter to LDA+U bands.
  - ▲ Calculation shows that t2g spliting & hopping anisotropy are small for 1 or 2 La layer.
  - ▲ For 2 La layer, the only big change occur in xy-plane hopping of xz-yz orbital ✗.
    For 1 La layer nothing drastic really happens

- For large U & small La layer, LDA+U predicts ferromagnet with Oxy orbital ordering
  - ▲ But experiments so far does not strongly support this proposal



- Approach so far is approximating Coulomb by Hartree-Fock. now consider dynamical mean-field

$$F[\Sigma(r,r',\omega)] = \underbrace{F_{univ}[\Sigma(r,r',\omega)]}_{\text{depend only on form of interaction}} + \mathrm{Tr}[\ln(G_0^{-1}(r,r',\omega) - \Sigma(r,r',\omega))]$$

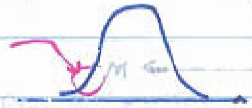
F extremize at correct self-energy & from which ALL response functions can be extracted

- ▲ Then we may try to find accurate & effecient way to approximate F

$$F[\ .(r,r',\omega)] \rightarrow \sum_{r_i} F_{approx}[\{\Sigma(\vec{r}_i,\omega)\}] + \mathrm{Tr}[\ln(G_0^{-1}(r,r',\omega) - \Sigma(r,\omega))]$$

- ▲ This leads to self-consistency on atom—bath system
- ▲ For hetrostructure we need to do self-consistency that include all layers.

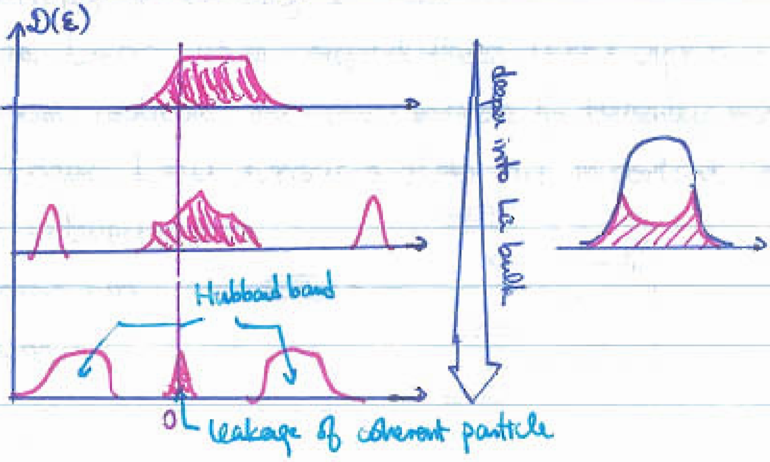
- The total charge density profile is essentially independant of methods.



- Consider tail region

$A(z,z';k,\omega)$

$\sim \mathcal{D}(\epsilon)$ on layer.



- Competing order — $(LaMnO_3)_8/(SrMnO_3)_4$

  $LaMnO_3$: insulator, orbital order, A-type AF

  $SrMnO_3$: insulator, no orbital order, G-type AF

  - ▲ Phases can be understood by solving charge density & go back to bulk phase diagram
  - ▲ charge density obtain essentially from electrostatic.
  - ▲ Orbital order weakly coupled between layer
  - ▲ Conductitivity obtain by calculating each layer conductivity, & assemble them as in parallel circuit.
  - ▲ The theoretically results do not agree with expriment, where $\ln \underset{\text{resistivity}}{\rho}$ changes fast with n (theory predict $\rho \sim n$)
    - ▸ because large interface ⇒ bad things happen to sample?

- A possible interesting system: V_nO_{2n-1}