The Role of Dynamic U in La2CuO4

F. Aryasetiawan,
Mathematical Physics, Lund University

Collaborators:
Philipp Werner (Fribourg)
Rei Sakuma (Lund)
Fredrik Nilsson (Lund)
Consider $La_2CuO_4$ as an example

Method for calculating $U(\omega)$

Importance of dynamic screening:
- Gap opening
- Satellite features

Reinterpretation of lower Hubbard band in charge-transfer insulators

Frequency-dependent $U$ vs $k$-dependent self-energy
$La_2CuO_4$(undoped)

CuO plane seen from above

Wannier bands from LDA

Werner et al, PRB87 165118 (2015)
Three-band model: Emery PRL58 2794 (1987)

\[
H_{3b} = - t_{pd} \sum_{<ij>} p_i^\dagger (d_i + \text{H.c.}) - t_{pp} \sum_{<jj>} p_j^\dagger (p_{j\uparrow} + \text{H.c.}) \\
+ \epsilon_d \sum_i n_i^d + \epsilon_p \sum_i n_i^p + U_d \sum_i n_i^d n_i^d \\
+ U_p \sum_j n_j^p n_j^p + U_{pd} \sum_{<ij>} n_i^d n_j^p
\]

In actual calculations, p-d screening is usually neglected

Constrained LDA

<table>
<thead>
<tr>
<th>$\epsilon_p - \epsilon_d$</th>
<th>$t_{pd}$</th>
<th>$t_{pp}$</th>
<th>$U_d$</th>
<th>$U_p$</th>
<th>$U_{pd}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.6 eV</td>
<td>1.3 eV</td>
<td>0.65 eV</td>
<td>10.5 eV</td>
<td>4 eV</td>
<td>1.2 eV</td>
</tr>
</tbody>
</table>

Three-band model

One-band model

From Akihiro Ino’s Thesis (University of Tokyo, 1999)
Three-band model with static $U$.

(No p-d screening in the model)

Figure 2: (color online) DMFT spectra for the 0MTO basis with interaction parameters $U_{dd} = 10 \text{eV}$, $U_{pp} = 5 \text{eV}$, and varying $U_{pd}$.

Hansmann et al, arXiv:1312.2757v1
Constrained Random-Phase Approximation (cRPA): A method for calculating the Hubbard $U$

\[ P = P_d + P_r \]

Polarisation: $P = P_d + P_r$

PRB 70, 195104 (2004)
Constrained RPA (cRPA)

Real system: \[ W = \nu + \nu PW \]

Model system: require that
\[ W = U + UP_d W \rightarrow \text{Determines } U \text{ from } W \]

Can be shown that
\[ U = \nu + \nu P_r U \]
\[ P_r = P - P_d \]

\[
P(r, r'; \omega) = \sum_{i}^{\text{occ}} \sum_{j}^{\text{unocc}} \psi_i(r)\psi_j^*(r)\psi_i^*(r')\psi_j(r') \frac{\omega - \varepsilon_j + \varepsilon_i \pm i\delta}{\omega - \varepsilon_j + \varepsilon_i \pm i\delta}
\]

\[
P_d(r, r'; \omega) = \sum_{i \in d}^{\text{occ}} \sum_{j \in d}^{\text{unocc}} \psi_i(r)\psi_j^*(r)\psi_i^*(r')\psi_j(r') \frac{\omega - \varepsilon_j + \varepsilon_i \pm i\delta}{\omega - \varepsilon_j + \varepsilon_i \pm i\delta}
\]

\(\rightarrow\) can study the effects of screening at microscopic level.
Example: 3-level $\rightarrow$ 2-level model

\[ P_r(r, r'; \omega) = 2 \frac{\varphi_0(r)\varphi_2^*(r)\varphi_0^*(r')\varphi_2(r')}{\omega - \varepsilon_2 + \varepsilon_0 \pm i\delta} \]

\[ P_d(r, r'; \omega) = 2 \frac{\varphi_0(r)\varphi_1^*(r)\varphi_0^*(r')\varphi_1(r')}{\omega - \varepsilon_1 + \varepsilon_0 \pm i\delta} \]

\[ U = \nu + \nu P_r U \]

\[ U_{00,00}(\omega) = \nu_{00,00} + \frac{\alpha^2}{\omega^2 - \Delta^2} \]

\[ \alpha^2 = 4(\varepsilon_2 - \varepsilon_0)\nu_{02,00}^2 \]

\[ \Delta^2 = (\varepsilon_2 - \varepsilon_0)^2 \left(1 + \frac{4\nu_{02,02}}{\varepsilon_2 - \varepsilon_0}\right) \]
Dynamic $U$ for a three-band model of $La_2CuO_4$

$$U(\omega) = \int d^3r d^3r' \phi(r) \phi_d(r) U(r, r'; \omega) \phi_d(r') \phi_d(r')$$

$$U(0) = v + \frac{2}{\pi} \int_0^\infty d\omega \frac{\text{Im}U(\omega)}{\omega}$$

The strong satellite feature at $\sim 9$ eV is due to subplasmon excitation arising from p-d screening.

(The red curves are obtained by including p-d transitions)
The static $U$ from cRPA is considerably smaller than commonly used value of $\sim 10^{-12}$ eV.

The strong excitation at $\sim 9$ eV is localised on the copper site.

A static $U$ can be used for a three-band model but p-d screening should be taken into account in the model.

A one-band model should use a dynamic $U$. 

Dynamic $U$ for a “three-band model” of $La_2CuO_4$ (with p-d screening included in $U$)

Werner et al, PRB91, 125142 (2015)
$U$ of the four-band model is almost indistinguishable from that of the three-band model. (not always the case with other copper oxide compounds)
DMFT in picture

Taken from Vollhardt
Effective dynamics for an impurity problem

The Coulomb interaction is fully taken into account in one site (impurity), the rest of the sites (medium) is treated as an effective field

\[
S_{\text{eff}} = -\int d\tau dt \sum_{\sigma} c_{\sigma}^{+} G_{0}^{-1}(\tau - \tau') c_{\sigma}(\tau') + \int d\tau d\tau' n_{\uparrow}(\tau) U(\tau, \tau') n_{\downarrow}(\tau')
\]

with the dynamical mean-field \( G_{0}^{-1} \)

Solve impurity problem using continuous-time QMC
Werner and Millis (PRL 2007, 2010)
Importance of dynamic U in gap opening

Werner et al, PHYSICAL REVIEW B 91, 125142 (2015)

$La_2CuO_4$

One-band model

No gap opening with static $U$.
Too small gap with dynamic $U$
La$_2$CuO$_4$

Three-band model
(p-d screening included in U)

Extracting the satellite contribution

\[ A(k, \omega \leq \mu) = A_{QP}(k, \omega) + A_{QP}(k, \omega)A^S(k, \omega), \]

\[ A^S(k, \omega) = \frac{n_k}{2\pi} \int_{-\infty}^{\infty} dt e^{i\omega t} [e^{C^S(k,t)} - 1] \]

Bosonic part (satellite)

Cumulant expansion
PRL77 2268 (2006)
Fully screened interaction $W$

$La_2CuO_4$

How is d-d plasmon related to lower-upper Huuband bands?
Figure 2: (color online) DMFT spectra for the 0MTO basis with interaction parameters $U_{dd} = 10\text{eV}$, $U_{pp} = 5\text{eV}$, and varying $U_{pd}$.

Hansmann et al, arXiv:1312.2757v1
k-resolved spectral functions for the three-band model

$La_2CuO_4$

- "Zhang-Rice singlet"
- p band well-described in LDA

Upper Hubbard band
- "Zhang-Rice singlet"
- and lower Hubbard band
- p-d plasmon
NiO

J. Kuneš, V. I. Anisimov, A. V. Lukoyanov, and D. Vollhardt
PHYSICAL REVIEW B 75, 165115 (2007)

S. Hüfner, P. Steiner, I. Sander, M. Neumann, and S. Witzel

Fig. 1. Combined XPS and XPS/BIS spectrum of NiO. The main excitation features are: at 8.5 eV the $d^8 \rightarrow d^7$ excitation, at 2.0 eV the $d^8 \rightarrow d^8 L^{-1}$ excitation and at $-3.7$ eV the $3d^8 \rightarrow 3d^9$ excitation.
Dynamic $U$ in TMO

S. Hübner$^1$, P. Steiner$^1$, I. Sander$^1$, M. Neumann$^2$, and S. Witzel$^2$


Therefore we can use Fig. 1 to get in first order $U = 12 \text{ eV}$, a number which is slightly too large because it contains some hybridisation contributions and if they are taken care of, a value of $U_{\text{eff}} \approx 10 \text{ eV}$ is obtained [15]. The large peak observed at slightly less than 2 eV below the Fermi energy is a transition $d^8 \rightarrow d^8L^{-1}$. The final state can be interpreted as being created by the excitation of a $d$-elec-

![Graph showing Re U(ω) and Im U(ω) for various compounds in the d-model and dp-model.](image1)

![Graph showing counts per channel vs binding energy for NiO in XPS and BIS.](image2)

Fig. 1. Combined XPS and XPS/BIS spectrum of NiO. The main excitation features are: at 8.5 eV the $d^8 \rightarrow d^7$ excitation, at 2.0 eV the $d^8 \rightarrow d^8L^{-1}$ excitation and at $-3.7$ eV the $3d^8 \rightarrow 3d^9$ excitation.
Dynamic $U$ vs $k$-dependent self-energy in SrVO$_3$

\[ \text{Im}\, U \]

\[ \text{Re}\, U \]

\[ \text{Re} U(\omega) = v - \frac{2}{\pi} \int_0^\infty d\omega' \frac{\omega'\text{Im} U(\omega')}{\omega^2 - \omega'^2} \]

Miyake et al, unpublished
Effects of dynamic $U$ on the self-energy of SrVO$_3$

The $Z$-factor is significantly smaller in the dynamic $U$ than in the static $U$ case.

Expect a larger band renormalisation in the dynamic than the static $U$ case.

Miyake et al, PRB 87, 115110 (2013)
Effects of dynamic $U$ (within $GW$)

Cancellation between the effects of the $k$- and $\omega$-dependence of the self-energy on the QP dispersion

The dynamic model tends to overestimate band narrowing. $k$-dependent self-energy widens the QP band.

Sakuma et al PRB 2014
Dynamic $U$ enhances correlation effects
$\rightarrow$ needed for gap opening in La2CuO4

New interpretation of the lower Hubbard band in charge-transfer insulators:
$\rightarrow$ a subplasmon satellite arising from p-d transitions.
$\rightarrow$ the subplasmon is localised on the copper site.

A static $U$ can be used in a three-band model
but $p-d$ screening must be taken into account in the model

The effects of $k$- and $\omega$-dependent self-energy tend to cancel each other.
$\rightarrow$ Local self-energy with dynamic $U$ may overestimate effective mass.
$\rightarrow$ Band narrowing is not synonymous with Z-factor.