Strength of correlations in pnictides and its assessment by theoretical calculations and spectroscopy experiments

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Abstract

LDA+DMFT (Local Density Approximation combined with Dynamical Mean-Field Theory) computation scheme has been used to calculate spectral properties of LaFeAsO – the parent compound of the new high-$T_c$ iron oxypnictides. The average Coulomb repulsion $\bar{U} = 3 \div 4$ eV and Hund’s exchange $J = 0.8$ eV parameters for iron $3d$ electrons were calculated using the \textit{first principles} constrained density functional theory scheme in the Wannier functions formalism. DMFT calculations using these parameters result in moderately correlated electronic structure with effective electron mass enhancement $m^* \approx 2$ that is in agreement with the experimental X-ray and photoemission spectra. Conclusion of moderate correlations strength is confirmed by the observation that pnictides experimental spectra agree well with corresponding spectra for metallic iron while being very different with Mott insulator FeO spectra.

Key words: PACS: 74.25.Jb Electronic structure

Recent discovery of high-$T_c$ superconductivity in iron oxypnictides LaO$_{1-x}$F$_x$FeAs\textsuperscript{[1]} has stimulated an intense experimental and theoretical activity. In a striking similarity with the high-$T_c$ cuprates, the undoped LaFeAsO is not superconducting, but exhibits an antiferromagnetic commensurate spin-density wave below 150 K\textsuperscript{[2]}. Only when electrons (or holes) are added to the system via doping, antiferromagnetism is suppressed and superconductivity appears. As it is generally accepted that the Coulomb correlations between the copper $3d$ electrons are responsible for the anomalous properties of cuprates,
it is tempting to suggest that the same is true for the iron 3d electrons in LaFeAsO.

The ratio of the Coulomb interaction $U$ and the band width $W$ determines the correlation strength. For $U/W < 1$ the system is weakly correlated and the results of Density Functional Theory (DFT) calculations are enough to explain its electronic and magnetic properties. However, if the $U$ value is comparable with $W$ or even larger the system is in an intermediate or a strongly correlated regime and the Coulomb interactions must be treated explicitly in the electronic structure calculations. The partially filled bands formed by Fe-3d states in LaFeAsO have a width of $\approx 4$ eV (see the shaded area in the lower panel of Fig. 1), so the interaction parameter $U$ should be compared with this value.

In practice, $U$ is often considered a free parameter to achieve the best agreement between the calculated and measured properties of the investigated system. However, the most attractive approach is to determine the Coulomb interaction parameter $U$ from the first principles. Two methods are generally used for this purpose: constrained DFT scheme [3, 4], where the $d$-orbital occupancies in DFT calculations are fixed to the certain values and $U$ is determined as a numerical derivative of the $d$-orbital energy over its occupancy, and Random Phase Approximation (RPA) [5], where the screened Coulomb interaction between the $d$-electrons is calculated using a perturbation theory. Recently, RPA calculations of the interaction parameter $U$ in LaFeAsO were reported [6], estimating putting the $U$ value in the range $1.8 \div 2.7$ eV. In Ref. [7] it was proposed to use the $U$ of 4 eV obtained in RPA calculations for metallic iron [8]. This value of the Coulomb parameter (with Hund’s exchange parameter $J=0.7$ eV) was used in Dynamical Mean-Field Theory (DMFT) [9] calculations for LaFeAsO [7, 10, 11]. These studies find the iron 3d electrons to be in an intermediate or a strongly correlated regime, as can be expected for the Coulomb parameter $U=4$ eV and the Fe-3d band width of $\approx 4$ eV.

To estimate the correlation strength one can compare experimental spectra with the densities of states (DOS) obtained in DFT calculations. For strongly correlated materials additional features in the experimental photoemission, X-ray absorption and optical spectra appear that are absent in the DFT DOS. These features are interpreted as lower and upper Hubbard bands. If no Hubbard bands are observed and the DOS obtained in DFT calculations describes the experimental spectra satisfactorily the material is considered to be in a weakly correlated regime. LaFeAsO was studied by
soft X-ray absorption and emission spectroscopy \cite{12}, X-ray absorption (O K-edge) spectroscopy \cite{13}, and photoemission spectroscopy \cite{14}. All these studies conclude that DOS obtained in DFT calculations agrees well with the experimental spectra and the estimated value of the Coulomb parameter is less than 1 eV \cite{13}. Such a contradiction with the DMFT results \cite{7, 10, 11} using $U=4$ eV shows that the first principles calculation of the Coulomb parameter $U$ for LaFeAsO is needed to assess the strength of correlations in this material. Results of such calculations using the constrained DFT methods are reported in the present work.

A source of uncertainty in the constrained DFT scheme is the definition of atomic orbitals whose occupancies are fixed and energies are calculated. In some DFT methods, like Linearized Muffin-Tin Orbitals (LMTO), these orbitals could be identified with LMTOs. However, for other basis sets for example plane waves in the pseudopotential methods one should use a more general definition of the localized atomic-like orbitals such as Wannier functions \cite{15} (WFs). A practical way to calculate WFs for specific materials using projection of atomic orbitals on the Bloch functions was developed in Ref. \cite{16}.

In Fig. 1 the total and partial DOS for LaFeAsO obtained in LMTO basis are shown. The crystal field splitting of the Fe-3$d$ states in this material is rather weak ($\Delta_{\text{cf}}=0.25$ eV) and all five $d$ orbitals of iron form a common band in the energy range ($-2$, $+2$) eV relative to the Fermi level (see the gray region in the bottom panel of Fig. 1). There is a strong hybridization of the iron $t_{2g}$ orbitals with the $p$ orbitals of arsenic, the effect of which becomes apparent in the energy interval ($-3$, $-2$) eV (the white region in the bottom panel of Fig. 1) where the As-$p$ band is situated. A weaker hybridization with the oxygen $p$ states can be seen in ($-5.5$, $-3$) eV energy window (the black region in the bottom panel of Fig. 1).

We have calculated the average Coulomb interaction $\bar{U}$ and Hund’s exchange $J$ parameters in WFs basis using the constrained DFT procedure with fixed occupancies of WFs of $d$ symmetry. For this purpose we have used two computational schemes based on two different DFT methods. The first involves linearized muffin-tin orbitals produced by the TB-LMTO-ASA code \cite{17}; the corresponding WFs calculation procedure is described in details in Ref. \cite{19}. The second, based on the pseudopotential plane-wave method PWSCF, as implemented in the Quantum ESPRESSO package \cite{18}, is described in Ref. \cite{20}. The difference between results obtained with the two schemes gives an estimate of the uncertainty of $\bar{U}$ and $J$ determination.
The WFs are defined by the choice of Bloch functions Hilbert space and by a set of trial localized orbitals that will be projected onto these Bloch functions \[19\]. We have performed calculations for all bands in energy window \((-5.5, +2)\) eV that are formed by O-2\(p\), As-4\(p\) and Fe-3\(d\) states and correspondingly full set of O-2\(p\), As-4\(p\) and Fe-3\(d\) atomic orbitals to be projected on the Bloch functions for these bands. This corresponds to an extended model where in addition to the Fe-\(d\) orbitals all the \(p\)-orbitals are included as well.

Constrain DFT calculations using TB-LMTO-ASA method gave values \(\bar{U} = 3.10, J = 0.81\) while calculations with PWSF gave \(\bar{U} = 4.00, J = 1.02\). The difference between those two sets of parameter values shows the ”error bar” in definition of Coulomb interaction parameters. In the following we have used results obtained in TB-LMTO-ASA calculations.

In the constrained DFT calculations an average Coulomb interaction \(\bar{U}\) is obtained which can be approximated\[4\] as \(\bar{U} = F^0 - J/2\), where \(F^0\) is 0th Slater integral. Hence \(F^0\) can be calculated as \(F^0 = \bar{U} + J/2\) and this gives \(F^0 = 3.5\) eV and \(J = 0.81\) eV. With this set of parameters Coulomb interaction matrix \(U_{mm'}\) was calculated and used in LDA+DMFT \[21\] calculations (for a detailed description of the present computation scheme see Ref. \[19\]). The DFT band structure was calculated within the TB-LMTO-ASA method \[17\]. Crystal structure parameters of Ref. \[1\] were used.

A double-counting term \(\bar{U}(n_{\text{DMFT}} - \frac{1}{2})\) was used to obtain the noninteracting Hamiltonian \[20\]. Here \(n_{\text{DMFT}}\) is the total number of \(d\)-electrons obtained selfconsistently within the LDA+DMFT scheme. The effective impurity model within the DMFT was solved by Hirsch-Fye QMC method \[22\]. In the present implementation of the QMC impurity solver the Coulomb interaction between different orbitals on the same atom is limited to density-density terms, i.e. the form \(\sum_{m,m',\sigma\sigma'} U_{mm'}^{\sigma\sigma'}(F^0, J)\hat{n}_{m\sigma}\hat{n}_{m'\sigma'}\). In particular, this means that the coupling between the local spins is of Ising and not Heisenberg type. Since this is a significant approximation a few comments are in order especially concerning the question whether this approximation underestimates or overestimates the many-body renormalization of quasi-particle bands. We argue quite generally that introducing the spin-flip exchange (and other interaction terms beyond density-density) allows electrons to avoid each other more efficiently and thus the electron propagation through the crystal is inhibited less than with Ising exchange. As an example one can imagine two electrons with opposite spin in different orbitals. Allowing the spin-flip exchange the Ising-only interaction energy \(U'\) can be reduced to \(U' - J\) by
forming the triplet state with $S_z = 0$. This argument is also supported by recent numerical studies of two-band model comparing full Coulomb and Ising-only interaction terms \cite{23}. Therefore we expect our results rather to overestimate than underestimate the quasi-particle renormalization.

Calculations were performed at the inverse temperature $\beta = 10 \text{ eV}^{-1}$. The interval $0 < \tau < \beta$ was divided into 100 slices. $6 \cdot 10^6$ QMC sweeps were used in self-consistency loop within the LDA+DMFT scheme and $12 \cdot 10^6$ of QMC sweeps were used to calculate the spectral functions.

The effect of correlations on the electronic structure is minimal: there are relatively small changes of peak positions for $3z^2 - r^2$, $xy$ and $x^2 - y^2$ orbitals (a shift toward the Fermi energy) and practically unchanged spectral functions for the $yz$, $zx$ bands. There is no appearance of either Kondo resonance peak on the Fermi level or Hubbard bands in the spectrum, the features in Fe-$d$ spectral functions below -2 eV correspond to hybridization with As-$p$ and O-$p$ bands. The reason for such weak correlation effects is very strong hybridization of the Fe-$d$ orbitals with As-$p$ states (see the peaks in the Fe-$d$ spectral function in the -2÷-3 eV range corresponding to mixing with As-$p$ bands). The hybridization provides an additional very efficient channel for screening of the Coulomb interaction between Fe-$d$ electrons.

These observations agree with the results of soft X-ray absorption and emission spectroscopy study \cite{12}. It was concluded there that LaFeAsO does not represent a strongly correlated system since the Fe $L_3$ X-ray emission spectra do not show any features that would indicate presence of the lower Hubbard band or a sharp quasiparticle peak that were predicted by the LDA+DMFT analysis \cite{7,10,11}. The comparison of the X-ray absorption spectra (O K-edge) with the LDA calculations gave \cite{13} an upper limit of the effective on-site Hubbard $U \approx 1$ eV. The photoemission study of LaFeAsO suggests \cite{14} that the line shapes of Fe $2p$ core-level spectra correspond to an itinerant character of Fe 3$d$ electrons. It was demonstrated there that the valence-band spectra are generally consistent with the band-structure calculations except for shifts of the Fe 3$d$-derived peaks towards the Fermi level. Such a shift is indeed observed in our LDA+DMFT spectra (Fig. 2).

The resonant inelastic X-ray scattering (RIXS) measurements of the LaOFeAs \cite{12}, LiFeAs and NaFeAs \cite{27}, CaFe$_2$As$_2$ \cite{28} were performed at the soft X-ray fluorescence endstation at Beamline 8.0.1 of the Advanced Light Source at Lawrence Berkeley National Laboratory \cite{29}. We have measured the res-
onant and non-resonant Fe $L_{2,3}$ ($3d4s \to 2p_{1/2,3/2}$ transition) X-ray emission spectra (XES). The instrument resolution for Fe $L_{2,3}$ X-ray emission spectra was 0.8 eV. X-ray absorption spectra (XAS) were measured in the total fluorescence yield mode with a resolving power $E/\Delta E=5000$.

Resonantly excited Fe $L_3$ XES spectra of LaOFeAs, CaFe$_2$As$_2$, NaFeAs and LiFeAs which directly probe the distribution of occupied Fe $3d$-states are presented in Fig. 3. The position of the Fermi level on the spectral curves is determined using the XPS Fe $2p$ binding energy for CaFe$_2$As$_2$ ($E_b=706.7$ eV) [28]. One can see that the intensity maximum of Fe $L_3$ XES is located within 0.9-1.25 eV with respect to the Fermi level which demonstrates that the Fe $3d$-states dominate at the top of the valence band for both one-layered and two-layered FeAs-systems. For all investigated compounds Fe $L_3$ X-ray emission spectra do not show any features that would indicate the presence of the lower Hubbard band or a sharp quasiparticle peak.

The XPS Fe $2p$ core level spectra of LaOFeAs [14], CaFe$_2$As$_2$ [28], LaOFeP [30] and FeO [31] (see Fig. 4) don’t show any presence of satellites typical for correlated systems (such as FeO) and very similar to that of metallic Fe [32]. Therefore the line shapes of XPS Fe $2p$ core-level spectra of iron pnictides correspond to an itinerant character of Fe $3d$ electrons.

This conclusion is supported by comparison of non-resonant Fe $L_{2,3}$ XES of LaOFeAs, CaFe$_2$As$_2$, NaFeAs and LiFeAs (Fig. 5). The two main bands located around 705 and 718 eV correspond to Fe $L_3$ ($3d4s \to 2p_{3/2}$ transition) and Fe $L_2$ ($3d4s \to 2p_{1/2}$ transition) normal emission lines, respectively, separated by spin-orbital splitting of Fe $2p$. For free atoms the relative intensity ratio of $L_2$ and $L_3$ XES lines, $I(L_2)/I(L_3)$, is determined only by the statistical population of $2p_{1/2}$ and $2p_{3/2}$ levels and therefore should be equal to 1/2. In metals the radiationless $L_2L_3M_{4,5}$ Coster-Kronig (C-K) transitions strongly reduce the intensity ratio $I(L_2)/I(L_3)$ [33]. One can see from the Fig. 3 that the intensity ratio $I(L_2)/I(L_3)$ is almost identical for all above mentioned FeAs-compounds and more close to that of Fe metal than to FeO.

All available photoemission measurements of iron pnictides [14, 28, 30, 26, 34, 35] as well as FeSe [36] which probe a total density of states show a very similar fine structure which is consistent with LDA calculations. The Fe $3d$ partial density of states of LaOFeAs determined by indirect way as a difference of photoemission spectra measured at and below Fe$3p \to 3d$ resonance is found to be different in Refs. [14] and [35]. Authors [35] have revealed in difference spectrum a broad peak centered at 7 eV and satellite at 12 eV which are attributed to the incoherent part of Fe $3d$ states and
contribution of super-Coster-Kronig Fe $3p - 3d$ Auger transition (similar to that of FeO [37]), respectively, which are considered as an indication of relatively strong correlation effects in this material. On the other hand, in difference spectrum obtained in [14] no broad peak at 7 eV is detected and spectral feature at 12 eV is found to be consistent with calculated As $4s$ low energy subband.

The behavior of the real part of self energy near zero frequency $\Sigma(\omega)|_{\omega=0}$ provides an important information about band narrowing and renormalization of the electron mass. Padé approximant [24] was used to obtain the self energy on the real frequency axis. The results are presented in Fig. 6. The calculated values of the quasiparticle renormalization factor $Z = (1 - \frac{\partial \Sigma(\omega)}{\partial \omega}|_{\omega=0})^{-1}$ are found to be 0.56, 0.54, 0.45, 0.56 for $d_{xy}$, $d_{yz}$ (or $d_{zx}$), $d_{3z^2-r^2}$, $d_{x^2-y^2}$ orbitals, respectively. These values agree well with the effective narrowing of the LDA+DMFT spectral functions relative to LDA DOS (Fig. 2). The effective mass enhancement $m^* = Z^{-1}$ are 1.78, 1.85, 2.22, 1.95 for $d_{xy}$, $d_{yz}$ (or $d_{zx}$), $d_{3z^2-r^2}$, $d_{x^2-y^2}$ orbitals, respectively, agrees well with the mass enhancement factor between 1.7 and 2.1 reported in the dHvA study [25] and also with the results of Angle-Resolved Photoemission Spectroscopy (ARPES) for pnictides [35, 30] where overall bandwidth was found to be reduced by a factor of 2. The $d_{x^2-y^2}$ orbital has the largest effective mass and exhibits the most evident narrowing of LDA spectrum (see Fig. 2). This orbital has its lobes directed into the empty space between nearest iron neighbors in the Fe plane. Hence it has the weakest overlap, the smallest band width, and the largest $U/W$ ratio.

The small effective mass enhancement shows that LaFeAsO is a moderately correlated system in contrast to the results of LDA+DMFT calculation [7] by Haule et al where a strongly renormalized low energy band with a fraction of the original width ($Z \approx 0.2-0.3$) was found while most of the spectral weight was transferred into a broad Hubbard band at the binding energy $\approx 4$ eV. Authors of Ref. [7] report that ”slightly enhanced Coulomb repulsion ($U = 4.5$ eV) opens the gap” so that the system is in strongly correlated regime on the edge of a metal-insulator transition. It is difficult to understand why two LDA+DMFT calculations gave so different results because descriptions of calculation details in Haule et al paper are very short. The only decisive way solve this problem would be third party independent calculation.

In LaFeAsO the iron ion is tetrahedrally coordinated with four As ions exhibiting a slight tetragonal distortion. In the tetrahedral symmetry group
the five \(d\)-orbitals should be split by the crystal field into a low-energy doublet of \(3z^2-r^2, xy\) corresponding to the \(e_g\) irreducible representation and a high-energy triplet of \(x^2-y^2, xz, yz\) belonging to the \(t_{2g}\) representation. We have calculated the WF orbital energies and have found that the \(t_{2g}-e_g\) crystal field splitting is very small \(\Delta_{cf} \approx 0.25\) eV. The slight tetragonal distortion of the tetrahedron leads to an additional splitting of the \(t_{2g}\) and \(e_g\) levels with the following orbital energies (the energy of the lowest \(3z^2-r^2\) orbital is set to zero):

\[
\varepsilon_{3z^2-r^2} = 0.00\text{ eV}, \quad \varepsilon_{xy} = 0.03\text{ eV}, \quad \varepsilon_{xz, yz} = 0.26\text{ eV}, \quad \varepsilon_{x^2-y^2} = 0.41\text{ eV}.
\]

The correlation leads not only to narrowing of the bands but also to substantial shifts of the Fe-\(d\) orbital energies. Adding the \(Re(\Sigma(0))\) to the LDA orbital energies results in \(\varepsilon_{3z^2-r^2} = 0.00\) eV, \(\varepsilon_{xy} = -0.37\) eV, \(\varepsilon_{xz, yz} = 0.10\) eV, \(\varepsilon_{x^2-y^2} = 0.20\) eV (see Fig. 7). Note that the actual band shifts are smaller due to the \(p-d\) hybridization.

Comparison of the LDA+DMFT single-particle spectral functions to various experimental spectra is presented in Figs. 8-9. Taking into account the selection rules for XES (X-ray emission spectroscopy) (neglecting the energy dependence of matrix elements) we compare the Fe L3 XES spectrum of Ref. [12], which corresponds to \(2p \rightarrow 3d\) transitions, with the calculated LDA+DMFT Fe-3d spectral function (see Fig. 8) to find a good agreement between the two. The shoulder in the experimental curve near -2.5 eV corresponds to the low energy peak in the calculated spectrum originating from strong hybridization between the Fe-\(d\) and As-\(p\) states (see also Fig. 1). In Fig. 9 we present the total LDA+DMFT spectral function together with the experimental photoemission data of Ref. [26]. Again we find a very good agreement between the theory and experiment. The sharp peak at the Fermi energy corresponds to a partially filled Fe-\(d\) band while the broad feature between -2 and 6 eV corresponds to the oxygen and arsenic \(p\) bands.

In conclusion, we have calculated the average Coulomb interaction \(U\) and \(J\) within the Fe \(d\)-shell in LaFeAsO using the constrained DFT procedure in the basis of Wannier functions and have obtained the Coulomb parameters values \(F^0 = 3.5\) eV, \(J = 0.8\) eV. The LDA+DMFT calculations yield moderately correlated iron \(d\) bands in this compound. This conclusion is supported by spectroscopic studies of this material and other pnictides.

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Figure 1: Total and partial densities of states for LaFeAsO obtained in DFT calculation in frame of LMTO method.
Figure 2: Partial densities of states for Fe-3$d$, As-4$p$ and O-2$p$ states obtained within the DFT (filled areas) and LDA+DMFT orbitally resolved spectral functions (bold lines).
Figure 3: $\text{Fe L}_3$ XES of LaOFeAs \cite{12}, CaFe$_2$As$_2$ \cite{28}, LiFeAs and NaFeAs \cite{27}.

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Figure 4: XPS Fe 2p of LaOFeAs [14], CaFe2As2 [28], LaOFeP [30], FeO [31] and Fe metal [32].
Figure 5: Fe L$_{2,3}$ NXES of FeAs-systems and reference samples.
Figure 6: (Colour online) Real (upper panel) and imaginary (lower panel) parts of LDA+DMFT self energy interpolated on real axis with the use of Padé approximant.
Figure 7: (Colour online) Splitting of Fe-\(d\) orbitals obtained in LDA and LDA+DMFT.
Figure 8: (Colour online) Calculated Fe-$d$ LDA+DMFT spectral function (solid line) and experimental Fe $L_3$ XES spectrum (circles) from Ref. [12].
Figure 9: (Colour online) Calculated total LDA+DMFT spectral function (solid line) and experimental LaFeAsO PES spectrum (circles) from Ref. [26].