In the Bardeen-Cooper-Schrieffer theory of superconductivity, electrons form Cooper pairs through an interaction mediated by vibrations of the crystal. Like lattice vibrations, antiferromagnetic fluctuations can also produce an attractive interaction creating Cooper pairs, though with spin and angular momentum properties different from those of conventional superconductors. Such interactions were implicated for class of heavy fermion materials based on Ce, U, and Pu with rather low transition temperatures, and cuprate superconductors with the highest known transition temperatures. Recently a surprising discovery of superconductivity in the iron-based compound LaO$_{1-x}$F$_x$FeP \cite{1} with $T_c \sim 7$ K sparked a new direction to explore superconductivity in a completely new class of materials. Very recently a substitution of P by As raised $T_c$ to 26 K \cite{2} becoming already one of the superconductors among noncuprate based materials. Exploring superconductivity in similar iron-based compounds holds a lot of promise for increasing $T_c$.

In this Letter we explore the electronic structure and optical properties of LaO$_{1-x}$F$_x$FeAs within density functional theory (DFT) and dynamical mean field theory (DMFT).

LaOFeAs has a layered tetragonal crystal structure shown in Fig. 1. Layers of La and O are sandwiched between layers of Fe and As. The Fe atoms form a square two dimensional lattice with Fe-Fe lattice spacing of 2.853 Å. To understand the material properties, it is important to identify the character of dominant bands near the Fermi level, their energy and momentum distribution. For this purpose, the first principles density functional theory is the invaluable tool. We used the full-potential augmented plane-wave method as implemented in the WIEN2K code \cite{3}. The lattice parameters and internal atomic positions have been determine experimentally ($a = 4.035$, $c = 8.741$, $z_{La} = 0.142$, $z_{As} = 0.651$). For the exchange correlation potential we used the generalized gradient approximation \cite{4} (GGA) in the Perdew-Burke-Ernzerhof variant, and $12 \times 12 \times 5$ $k$ points.

This method predicts that the dominant states at the Fermi level come from Fe 3$d$ atomic states extending roughly between $-2$ eV and 2 eV as shown in Fig. 2.

The important feature of LaOFeAs compound is that DFT predicts a very steep and negative slope of the density of states (DOS) at the Fermi level. In the rigid band approximation, neglecting the many body effects, the hole doping will lead to increases of the DOS at the Fermi level, while the electron doping will be accompanied by a decrease of DOS. Hence, for the conventional phonon mediated superconductor, one would expect decrease (increase) of $T_c$ when doping electrons (holes), contrary to what is observed in experiment \cite{2}. This gives a clear hint that the superconductivity in this compound might not be phonon mediated. Indeed an explicit calculation of the phonon coupling constants within the DFT, using the code of Ref. \cite{5}, gives a value too small to explain the observed critical temperature ($T_c < 1$ K).

While the dominant electronic character near the Fermi level is due to Fe, a strong mixing with As is apparent at $-2.7$ eV, where the As 4$p$ band is strongly peaked. There
level. This interplay of crystal field splitting and correlation effects was addressed in many model Hamiltonian studies [7–9] and it was shown how a bad metal or bad semiconductor can appear on the metallic side at finite temperature. In LaOFeAs we checked that a slightly enhanced Coulomb repulsion ($U = 4.5$ eV) leads to a finite gap in the $d$ band. For typical Coulomb repulsion of Fe ($U = 4$ eV) the system is still metallic, but a bad metal having some characteristics of a bad semiconductor. The metallic state is however still very correlated with a quasiparticle renormalization amplitude between $Z \sim 0.2$–0.3.

To describe this type of system, one needs to go beyond the traditional band structure methods and concentrate on the spectral function $A(k, \omega) = (G^\dagger(k, \omega) - G(k, \omega))/(2\pi i)$ where the latter takes the form

$$G(k, \omega) = \frac{1}{O_k(\omega + \mu) - H_k - \Sigma(\omega)}.$$  

The one electron part of the Hamiltonian $H_k$ and overlap matrix $O_k$ is obtained by the LDA method [12] while the self-energy is computed by solving an auxiliary quantum impurity problem embedded in a self-consistent medium, for which we used the numerically exact continuous time quantum Monte Carlo method [13]. The on-site Coulomb repulsion on Fe-3$d$ bands is estimated to be 4 eV [6] and the Hund’s coupling of $J = 0.7$ eV. The temperature is fixed at 116 K.

The local spectral function $A(\omega) = \sum_k A(k, \omega)$ at temperature $T = 116$ K is shown in Fig. 3(a) together with the corresponding LDA density of states. The DMFT approach predicts a renormalized low energy band with a fraction of the original width ($Z \sim 0.2$–0.3) while most of the weight is transferred into a broad Hubbard band at the binding energy $\sim -4$ eV. The system remains metallic at finite temperatures but a very bad metal with a scattering rate at the Fermi level as high as 0.4 eV at 116 K. With decreasing temperature the Fermi surface shrinks and the semiconducting gap is likely to open at zero temperature. Indeed slightly enhanced Coulomb repulsion ($U = 4.5$ eV) opens the gap even at room temperature. The correlation enhanced splitting between different orbitals leads to separation of bands into those that act as fully filled or a fully
empty bands at low energy. The high energy Hubbard bands are only weakly affected by such splitting. The Coulomb repulsion thus strongly reduces the carrier density and pushes the parent compound on the verge of the transition between a bad metal and a bad semiconductor. At the same time, the localization of electrons leads to local moment formation and enhancement of the spin susceptibility in the doped compound.

Many of the unconventional superconductors are known to have a very simple low energy band structure. For example, in the copper oxides, a single band is crossing the Fermi level. Similarly, the Fermi surface of the Na doped cobaltates [14] has primarily a $a_1g$ single sheet Fermi surface. The situation is very different in LaOFeAs within LDA. As can be seen in Fig. 3(b) LDA predicts that all five Fe 3d orbitals have finite weight close to or at the Fermi level. The situation is not simplified when the Coulomb correlation is accounted for. The spectral weight splits into a high energy incoherent part and a low energy part, which is very asymmetric and considerably reduced due to the proximity to the semiconducting state. Upon doping, the quasiparticle peaks move to the Fermi level, the scattering rate is reduced, and the system becomes a better conductor. Experimentally, doping leads to the superconducting ground state at low temperature, which is likely to be of unconventional origin. The cooper pairs are likely to be formed out of composite singlets of spin and orbital degrees of freedom.

Figure 4(a) shows the momentum resolved spectral function $\sum_l A(k, \omega)_{LL}$ in color coding together with the LDA bands in the energy range between $-5$ and $2.5$ eV and momentum dispersion in the high symmetry directions of the first Brillouin zone. The higher energy band structure at $\omega > 2$ eV and $\omega < -5$ eV is not considerably different in the two approaches. In the intermediate frequency region between $-1.5$ and $1.5$ eV a depletion of the spectral weight is apparent in the DMFT approach. While LDA predicts a large number of bands in this region, DMFT redistributes most of this weight further away from the Fermi level to the range between $-2$ and $-4$ eV. Consequently, a set of states with a large scattering rate and short lifetime is predicted by DMFT. Finally, the low energy part of the spectra is considerably modified when Coulomb correlation is taken into account. The hole pockets around the $\Gamma$
mostly due to transitions within the rate still remains large. Finally, the hole pockets around considerably enhanced (3–5 times) while the scattering Fermi level. The band velocity and effective mass are Fig. 4(b) where the electron pockets clearly cross the a bad metal at finite temperatures.

The situation is different in the doped compound [see Fig. 4(b)] where the electron pockets clearly cross the Fermi level. The band velocity and effective mass are considerably enhanced (3–5 times) while the scattering rate still remains large. Finally, the hole pockets around Γ remain highly scattered.

We also compute the optical conductivity of LaOFeAs within DMFT in both the xy plane and along the z direction. Considerable anisotropy can be identified in Fig. 5. The Drude peak, which is a hallmark of metallicity and Fermi liquid state, is absent because of a proximity to the semiconducting state. Our calculations predict a small number of carriers and large scattering rate, due to partial localization of carriers in this compound, and consequently low conductivity. One can identify a shoulder in optical conductivity close to 2 eV. It corresponds to transitions between Fe-3d and As 4p states. The large peak at 4 eV is mostly due to transitions within the d shell from the lower Hubbard band to the quasiparticle peak above the Fermi level.

We studied the band structure of the newly discovered superconductor LaO$_{1-x}$F$_x$FeAs, and we predict the orbital and momentum resolved spectral function and optical conductivity of the compound. Density functional theory predicts that a set of Fe 3d bands are crossing the Fermi level with no clear splitting into the $e_g$ and $t_{2g}$ manifold. The Coulomb correlations among the six electrons in the set of five Fe-3d orbitals is strong enough to push the compound close to the metal insulator transition. The correlation enhances the crystal field splitting among the d orbitals, which in turn increase correlations. At temperature $T = 116$ K studied here, the parent compound LaOFeAs is still metallic, although a bad metal with a very large scattering rate and a strongly reduced number of carriers. Doping the parent compound leads to electron pockets centered at the $M$ and $A$ point in the momentum-resolved spectral function.

In conclusion, we studied the band structure of the newly discovered superconductor LaO$_{1-x}$F$_x$FeAs, and we predict the orbital and momentum resolved spectral function and optical conductivity of the compound. Density functional theory predicts that a set of Fe 3d bands are crossing the Fermi level with no clear splitting into the $e_g$ and $t_{2g}$ manifold. The Coulomb correlations among the six electrons in the set of five Fe-3d orbitals is strong enough to push the compound close to the metal insulator transition. The correlation enhances the crystal field splitting among the d orbitals, which in turn increase correlations. At temperature $T = 116$ K studied here, the parent compound LaOFeAs is still metallic, although a bad metal with a very large scattering rate and a strongly reduced number of carriers. Doping the parent compound leads to electron pockets centered at the $M$ and $A$ point in the momentum-resolved spectral function.

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