

Ab-initio theory of Iron based superconductors

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We report the first-principles study of superconducting critical temperature and superconducting properties of Fe-based superconductors taking into account on the same footing phonon, charge and spin-fluctuation mediated Cooper pairing. We show that in FeSe this leads to a modulated $s\pm$ gap symmetry, and that the antiferromagnetic paramagnons are the leading mechanism for superconductivity in FeSe, overcoming the strong repulsive effect of both phonons and charge pairing.

The discovery of the iron based superconductors (FeSC) in the last decade[1–4] has been a crucial event in the history of superconductivity (SC). Showing that high temperature (hi- T_c) SC is not a unique property of the cuprates[5], and that it could occur in remarkably different classes of systems[6–10]. This discovery renewed the hope to find a room temperature superconductor, probably the most desired system in solid state physics.

In order to facilitate the search for new hi- T_c materials, it is highly desirable to achieve a theoretical understanding of the physical mechanism of hi- T_c SC. On a microscopic level, a SC state is created by the pairing of electrons to form Cooper pairs [11, 12]. Hence, the essential question that theorists try to answer is what causes this attractive pairing interaction.

A coupling provided by phonons has been ruled out quickly after the discovery of Fe based superconductors because the electron-phonon (el-ph) coupling is much too weak to explain the observed high transition temperatures[13, 14] and no clear isotope effect has been measured [15–19]. A large number of different theories have been proposed: resonating valence bond [20, 21], fluctuation exchange[22, 23], functional renormalization group[24, 25], orbital fluctuations[26, 27], charge-fluctuations[28–32], spin-fluctuations[33–35] (SF). The scientific community remains far from a general consensus on which is the dominant coupling mechanism.

Among the different theories the ones based on magnetism are, in our opinion, the most promising ones, since in both cuprates and FeSC the superconductivity appears close to an antiferromagnetic (AFM) phase[7, 8]. Approaching the AFM phase, if the transition is of second order, the magnetic susceptibility will become large, and eventually diverge. This implies that spin-fluctuations could become strong enough to overcompensate for the direct electron-electron repulsion and trigger the SC condensation. This idea is supported by strong experimental[3] and theoretical[36] arguments.

SF mediated pairing has been extensively investigated in the realm of Hubbard like models[35, 37–40]. Since this approach necessarily involves a set of parameters, it does not allow genuine predictions of the critical temperature.

Therefore, albeit very useful as a tool for a general physical understanding, it does not directly help in the search for new superconducting systems with desired properties.

The only way for theory to take the lead in the search for new and better hi- T_c materials is to develop a quantitatively predictive *ab-initio* theory and solve the dilemma of the pairing mechanism.

In this work we take a step in this direction by constructing a many-body perturbation based effective interaction, solely from first-principles calculations, and use it within density functional theory for superconductors (SCDFT) [41–44]. While SCDFT proved to be highly reliable in the description of el-ph superconductors[31, 45–50], its spin-fluctuation extension has been tested, until now, just on a simple electron gas model[51]. As neither the functional, nor the theoretical framework, contain any adjustable parameters, this scheme promises the prediction of T_c , symmetry of the order parameter and excitation spectrum, just from knowledge of the chemical structure of the material.

In the present work we apply the scheme to the FeSC. We choose this family because the metallic parent state can be better described than the Mott-insulator state in the cuprates. However we will observe that an unsatisfactory description of the parent compound affects the predictive power of our theoretical approach.

The first conceptual step in the SCDFT scheme currently in use is the assumption of a second order phase transition between the SC phase and its metallic parent compound. This implies that T_c can be estimated by taking the electronic structure of the metallic non-superconducting phase as a starting point to act-on with a pairing field computed from first principles. So, the very starting point of the theory is an approximation for the quasi-particle states of the parent metal, that we do by taking the DFT Kohn-Sham (KS) band structure [52, 53].

The calculated Fermi surface (FS) for FeSe, LiFeAs and LaOFeAs (representatives of the 11, 111 and 1111 family of FeSC) are shown in Fig. 1. All systems feature the same characteristics: Hole FS around the Γ -point forming a barrel and electron FM around the M -point

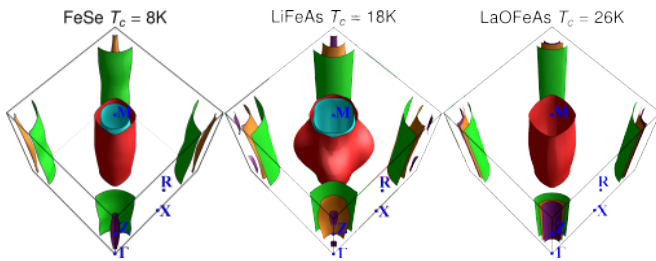


Fig. 1. (color online) Fermi surface for three Iron superconductors in the tetragonal unit cell.

which may not extend along the whole k_z direction forming the pockets. The calculations have been done using a state of the art plane wave code [54] and have been cross-checked with an all-electron linearized augmented plane wave code [55]. Since the results are very sensitive to the atomic positions a full lattice relaxation is performed [56, 57]. The calculated FS are in reasonable agreement with the results of the ARPES measurements [8, 9].

However, the pronounced nesting between the two parts of the FS with a nesting vector of $\mathbf{q}_M = (\frac{\pi}{a}, \frac{\pi}{a}, 0)$ leads to an instability with respect to the formation of the stripe ordered AFM state. All compounds converge to an AFM ground state with a large moment of about $1 \mu_B$ per iron atom. This is in striking contrast with the experimental observation [58], and a well known problem of DFT calculations for this class of systems [33, 57]. It has been suggested that the reason for this disagreement is that conventional DFT functionals like LSDA or GGA do not account for dynamic zero-point spin fluctuations that are strong here. This leads to the overestimation of the stability of the long range magnetic order [58].

In the construction of the SCDFT SF functional [51] an important role is played by the spin susceptibility χ_{zz} . The standard calculation of this quantity cannot be performed if the system has an artificial instability with respect to the formation of a long range magnetic order.

The solution of this problem could come from the use an improved DFT functional perhaps going beyond standard Kohn-Sham scheme. However, such a calculational scheme is not yet developed within ab-initio methods, and we have to opt for another solution. We will consider two different ways to deal with the problem that will help us to examine the dependence of the superconducting transition temperature on the details of the accepted approach. The first method we consider is to compute the magnetic susceptibility by scaling down the exchange-correlation (xc) field, as has been proposed in Ref. 14. By introducing a scaling parameter α the magnetic response function can be written as [59, 60]:

$$\chi_{zz}(\mathbf{q}\omega) = \frac{\chi^{\text{KS}}(\mathbf{q}\omega)}{1 - \alpha f_{\text{xc}}(\mathbf{q}\omega) \chi^{\text{KS}}(\mathbf{q}\omega)}. \quad (1)$$

By using a sufficiently small α (< 1) one avoids the sin-

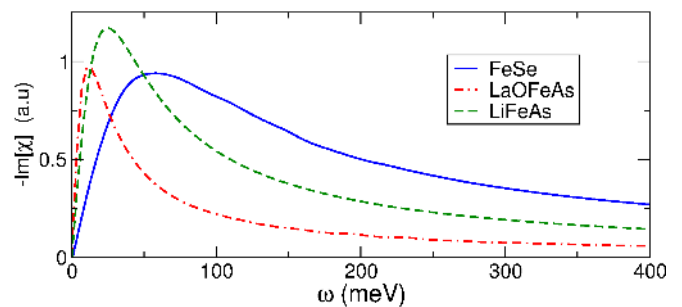


Fig. 2. (color online) Spin susceptibilities $\text{Im}[\chi_{zz}(\mathbf{q} = M, \omega, \mathbf{G} = 0, \mathbf{G}' = 0)]$ for LiFeAs, LaOFeAs and FeSe, computed at $\frac{\alpha}{\alpha_c} = 0.95$.

gularity of the susceptibility corresponding to the phase transition to the magnetic ordered state. Instead the susceptibility features finite-high peaks corresponding to paramagnons. The energy, lifetime and intensity of the paramagnons depend on the value of α and all materials feature a critical value α_c for which the susceptibility $\chi_{zz}(\mathbf{q}_M)$ diverges.

In Fig. 2 we show the $\text{Im}[\chi_{zz}(\mathbf{q}_M)]$ of representatives of the 11,111 and 1111 families using $\frac{\alpha}{\alpha_c} = 0.95$. All three compounds show a peak in the low-energy region featuring the presence of paramagnon-type fluctuations. In the rest of the paper we will focus on one of them: FeSe.

The SCDFT theoretical framework taking into account spin-fluctuation effects has been discussed in Ref. 51. This approach considers SF, el-ph and Coulomb (charge) pairing on the same footing. First, to keep the formalism complexity at minimum, we consider a multiband isotropic approximation, meaning that states and pairings are averaged over Brillouin zone volumes on isoenergy surfaces. For example, the Kohn-Sham potential [42] of the SCDFT system $\Delta_{n\mathbf{k}}$ is averaged as:

$$\Delta_{n\mathbf{k}} \approx \Delta_n(E) := \sum_{\mathbf{k}} \frac{\delta(\epsilon_{n\mathbf{k}} - E)}{N_n(E)} \Delta_{n\mathbf{k}} \quad (2)$$

This way we can group together the holes (see Fig. 1) around the Γ -point (labeled with $n = +$) and the electrons at the M -point (labeled with $n = -$). Within this approximation only s -wave pairing symmetries are possible, including the s_{\pm} symmetry suggested in Ref. 4. We will release this averaging approximation at the end of the paper.

Under this approximation we account for: 1) the phononic interaction by a band-resolved Eliashberg function $\alpha^2 F_{ij}$ (detailed definitions can be found in Ref. 45 and references therein); 2) the Coulomb repulsion by the average of the RPA screened matrix elements (as it was already done in several previous applications of SCDFT, more details can be found in Ref. 48 and 49); 3) the SF term containing an effective interaction mediated by paramagnon excitations Λ^{SF} as introduced in Ref. 51, and

averaged as $\Lambda_{n\mathbf{k}n'\mathbf{k}'}(\omega) \rightarrow \Lambda_{ij}(E, E', \omega)$ with $i, j \in \{+, -\}$. The structure of phononic, SF and Coulomb contributions is very different in nature. For the Coulomb interaction we use a static screening that proved to be highly reliable in phononic SCDFT[49, 50, 61–63]. The SF and phonon contributions, on the other hand, have a strong structure in ω with a low energy characteristic frequency. Therefore it is essential to take frequency dependence into account. In particular these interactions are negligible at energy scales large with respect to its characteristic one. This allows to safely disregard the E dependence of the interaction focusing on its behavior near the Fermi level [64, 65]. These considerations then translate into the following approximation scheme:

$$\begin{aligned} \alpha^2 F_{ij}(E, E', \omega) &\approx \alpha^2 F_{ij}(\epsilon_F, \epsilon_F, \omega) \quad (\text{attractive}) \\ w_{ij}(E, E', \omega) &\approx w_{ij}(E, E', 0) \quad (\text{repulsive}) \\ \Lambda_{ij}^{\text{SF}}(E, E', \omega) &\approx \Lambda_{ij}^{\text{SF}}(\epsilon_F, \epsilon_F, \omega) \quad (\text{repulsive}) \end{aligned}$$

Calculations show [66] the el-ph coupling (see Fig. 3a) to be very small, in agreement with previous works[67, 68]. With the integrated coupling λ being less than 0.1, T_c would be exponentially small if this was the only pairing channel. The Coulomb pairing (reported Fig. 3b) is as expected, diagonally dominated ($++$ and $--$ components). Within the static approximation, this cannot lead to any pairing, therefore no superconductivity can be sustained at any temperature by the combined effect of Coulomb forces and phonons.

The only possible superconductivity pairing must lay in the SF term. By the reasons discussed above this term depends critically on the parameter α . This can be appreciated with the help of Fig. 3c). Here Λ_{ij}^{SF} is shown as a function of the α/α_c ratio. At $\alpha/\alpha_c = 0.9$ the pairing is negligible. The maximum eigenvalue[69] of the λ_{ij} matrix being $\lambda^{\text{max}} = 0.07$, comparable with the phononic pairing. In the limit of $\alpha \rightarrow \alpha_c$ and $\mathbf{q} = M$ this value is essentially the highest limit that can be reached after integrating over the Brillouin zone. In combination with the very high SF characteristic frequencies, such a coupling leads to a sizable critical temperature and, since (in this limit) SF are dominated by off diagonal components, to the expected s_{\pm} symmetry[33].

If only SF coupling is considered a T_c as high as 11 K is found. However this critical temperature is reduced both by the inclusion of Coulomb terms (11 K \rightarrow 4 K) and phonons [70] (11 K \rightarrow 10 K), leading to an estimated maximum T_c of 3 K.

This estimation for T_c is in reasonable agreement with the experimentally observed 8 K[71]. This is an important success of the theory, showing that the SF are indeed the origin of the superconductivity in FeSe and giving T_c of the same order of magnitude as the experimental one. However, the inability of the standard DFT to de-

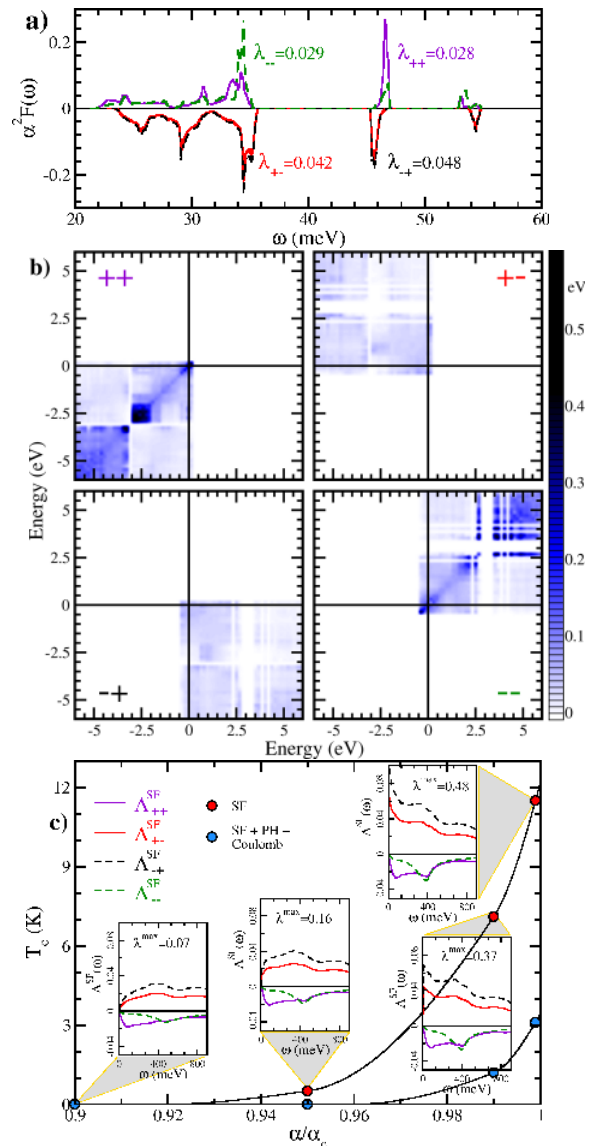


Fig. 3. (color online) Two bands resolved superconducting pairing functions evaluated at the theoretically optimized z_{Se} . **a)** electron-phonon Eliashberg function (PH) ; **b)** Screened Coulomb matrix elements averaged on iso-energy surfaces (Coulomb); **c)** Spin fluctuation Eliashberg function (SF) and SCDFT critical temperatures (T_c) as a function of the scaling parameter α .

scribe the ground state of FeSe enforced us to introduce parameter α that we cannot determine from the first-principles. The estimation of T_c also depends sensitively on the underlying electronic system, both via the KS electronic structure (computed at the Se Wyckoff position $z_{\text{Se}} = 0.25$ [72]) and the calculated spin susceptibility.

To enforce our findings we perform the SCDFT calculations using an alternative way to overcome the problem of the description of the magnetic ground state of FeSe. As shown in Fig. d the magnetic properties of FeSe strongly depend on the Wyckoff positions of the Se atoms, z_{Se} .

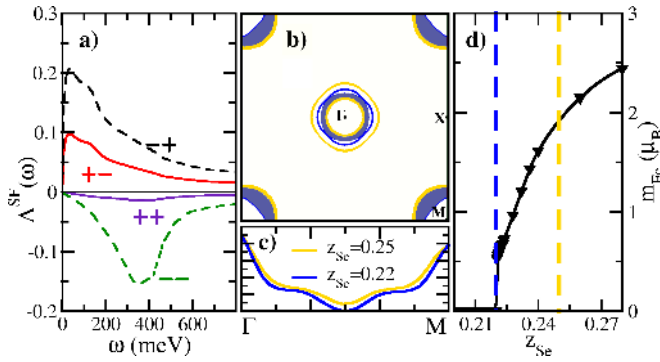


Fig. 4. Effect of the variation of z_{Se} position. **a)** Spin fluctuation spectral function Λ^{SF} computed at $z_{\text{Se}}=0.22$, leading to $\lambda^{\text{max}} = 1.3$. **b)** projection of the Fermi surface (FS) on the xy -plane. Blue (dark) computed at $z_{\text{Se}}=0.22$, and Yellow (gray) at $z_{\text{Se}}=0.25$. **c)** Nesting function (normalized) along the $\Gamma - M$ line. **d)** magnetic moment at the Iron site as a function of the z_{Se} parameter. The values used in this work are marked by vertical dashed lines.

The calculations show that the system is magnetic for $z_{\text{Se}} > 0.22$. We fix $z_{\text{Se}}=0.22$ in the paramagnetic region close to the transition to the AFM state. The proximity to the phase boundary results in intense paramagnon fluctuations. This leads in our theory to large SF pairing functions (see Fig. a) and to an estimated T_c of 24 K (by including all three pairing channels, the SF-only calculation gives T_c of 32K). The detailed analysis shows that the large difference in the value of T_c obtained in our two approaches is not due to the form of the spin susceptibility but due to a different electronic structure near the Fermi energy. This second case provides a sharper nesting for the SF pairing (see Fig. c) and leads to an increased interband pairing.

This large sensitivity of the predicted T_c to the lattice properties is consistent with experimental observations [3, 73–76], in particular with the observed correlation of T_c with the anion position[77].

We conclude the paper with an investigation of the gap function in k -space, going beyond the two-band isotropic approximation used so far (Eq. 2). We divide the xy -plane of the Brillouin zone into sectors as shown in Fig. 5, and compute the superconducting gap corresponding to each sub-band. The use of this more accurate approach does not significantly affect the value of the critical temperature (T_c increases by a few percent), it leads to a significantly modulated gap function within the s_{\pm} symmetry. The large dip in the Δ_- is related to the CB fluctuation ($M_1 - M_2$) in this band and is consequently not seen in Δ_+ ($\Gamma_1 - \Gamma_2$). The smaller oscillations are related to intraband scattering, where the interaction is smaller between two minima of the gap ($\Gamma_2 - M_2$) and larger between two maxima ($\Gamma_{1,3} = M_2$).

In this work we report the first application of the SCDF theory taking into account the SF. In particu-

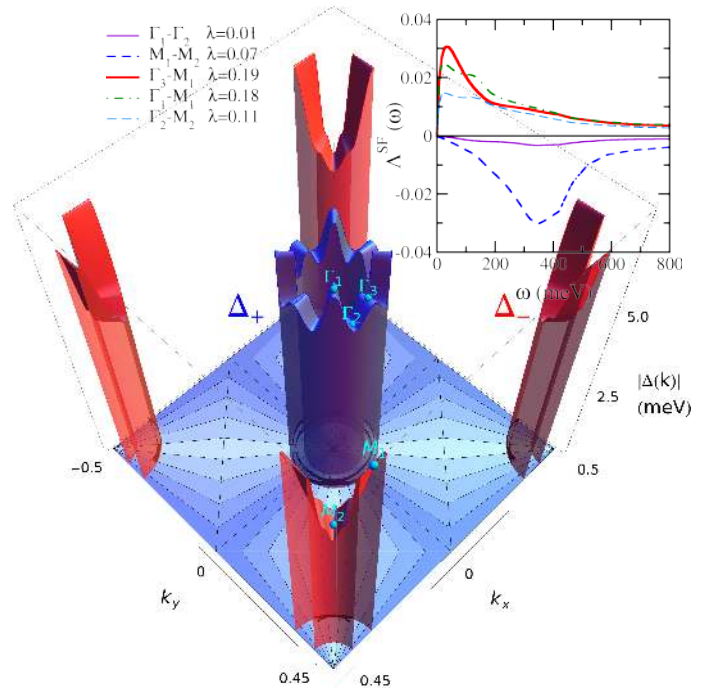


Fig. 5. Many body gap function in k -resolved calculation at $T = 0$ and $\omega = 0$. The black lines are the FS at $k_z = -\frac{\pi}{2c}$.

lar we consider the case of FeSe. We demonstrate that the SF are indeed the physical mechanism leading to the formation of the Cooper pairs and superconductivity of the system. We demonstrate that the pairing symmetry is of the s_{\pm} type. To overcome the problem of the standard DFT theory with the description of the FeSe ground state we adopt to different approaches that allow us to reveal strong sensitivity to the details of the electronic structure. The estimated T_c varies between 3 K and 24 K that is in reasonable correlation with experimental value of 8 K. We believe that as soon as the problem of the DFT description of the magnetic ground state of FeSe will be solved the suggested machinery will provide a completely ab-initio estimation of the superconducting T_c in this compound and will open an avenue for the first-principle design of the systems with high- T_c superconductivity.

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