Ab initio theory of iron-based superconductors

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We report a first-principles study of the superconducting critical temperature and other properties of Fe-based superconductors taking into account, on equal footing, phonon, charge, and spin-fluctuation mediated Cooper pairing. We show that in FeSe this leads to a modulated s± 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.

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I. INTRODUCTION

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 materials [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 crucial to achieve a theoretical understanding of the physical mechanism of hi-T_c SC. On the 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 pairing interaction [13].

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 [14,15] and no clear isotope effect has been measured [16–20]. A large number of different theories have been proposed: resonating valence bond [21,22], fluctuation exchange [23,24], functional renormalization group [25,26], orbital fluctuations [27,28], charge-fluctuations [29–33], spin-fluctuations [34–36] (SF). The scientific community remains far from a general consensus on which is the dominant coupling mechanism [37].

Among the different theories the ones based on magnetism are, in our opinion, the most promising, since in both cuprates and FeSC the superconductivity appears close to an antiferromagnetic (AFM) phase [7,8,37]. 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 scenario is supported by strong experimental [3,38,39] and theoretical [40,41] arguments.

SF mediated pairing has been extensively investigated in the realm of Hubbard-like models [36,42–45]. 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.

II. OVERVIEW

In this paper we take a step in this direction by computing a many-body perturbation based effective interaction, solely from first-principles calculations and use it within density functional theory for superconductors (SCDFT) [46–53]. While SCDFT proved to be highly reliable in the description of el-ph superconductors [32,54–65], its spin-fluctuation extension has been tested, until now, just on a simple electron gas model [46]. 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 the knowledge of the chemical structure of the material. The superiority of such an *ab-initio* scheme with respect to the many existing models is that, being derived from first principles in Ref. [46] within well defined approximations, any success would be an ultimate proof that the interactions accounted for within the approximations made are the relevant ones. On the other hand a failure would prove the opposite, i.e., that the leading interactions are not all included: leaving no space for speculation and pointing to the need of improving the approximations made in Ref. [46].

In the present paper we apply the scheme to the FeSC. We choose this family because the metallic parent state can be better described by *ab initio* methods than the Mott-insulator state in the cuprates. While, with few exceptions [66–68], the general features of their electronic properties are captured by state of the art theoretical methods and are compatible with the spin-fluctuation scenario, we observe that the description of the parent compounds is lacking in quantitative accuracy, affecting the predictive power of our theoretical approach.

III. ELECTRONIC STRUCTURE AND SPIN-SUSCEPTIBILITY

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 nonsuperconducting 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

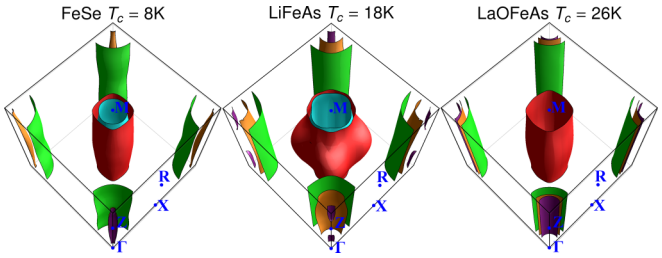

FIG. 1. Fermi surface for three iron superconductors in the tetragonal unit cell.

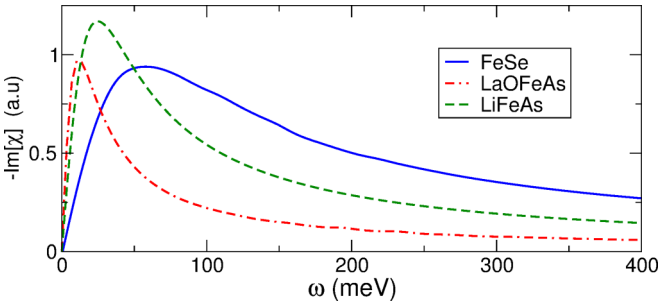

FIG. 2. Spin susceptibilities $\mathrm{Im}[\chi_{zz}(\mathbf{q}=\mathrm{M},\omega)]$ for LiFeAs, LaOFeAs, and FeSe, computed for $\frac{\gamma}{\gamma_c}=0.95$.

approximation for the quasiparticle states of the parent metal, that we do by taking the DFT Kohn-Sham (KS) band structure [69,70].

The calculated Fermi surfaces (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: a hole FS around the Γ point forming a barrel and an electron FS around the M point 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 [71] and have been cross-checked with an all-electron linearized augmented plane wave code [72]. Since the results are very sensitive to the atomic positions a full lattice relaxation is performed [73,74]. 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 $\mathbf{q}_\mathrm{M}=(\frac{\pi}{a},\frac{\pi}{a},0)$ 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 μ_B per iron atom. This is in striking contrast to the experimental observation [75] and a well known problem of DFT calculations for this class of systems [34,74]. 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 in this class of materials. This leads to the overestimation of the stability of the long range magnetic order [75].

In the construction of the SCDFT SF functional [46] 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 using an improved DFT functional, or, perhaps, from going beyond the standard Kohn-Sham scheme. However, such a calculational scheme does not yet exist 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 adopted approach. The first method we consider is to compute the magnetic susceptibility by scaling down the exchange-correlation (xc) kernel, as has been proposed in Ref. [15]. By introducing a scaling parameter γ the magnetic response function can be written as [76,77]:

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

By using a sufficiently small γ (<1) one avoids the singularity of the susceptibility corresponding to the phase transition to the magnetic ordered state. Instead the susceptibility features finite-height peaks corresponding to paramagnons. The energy, lifetime, and intensity of the paramagnons depend on the value of γ, and all materials feature a critical value γ_c for which the susceptibility $\chi_{zz}(\mathbf{q}_\mathrm{M})$ diverges. γ_c is 0.69 in FeSe, 0.91 in LiFeAs, 0.70 in LaOFeAs.

In Fig. 2 we show the $\mathrm{Im}[\chi_{zz}(\mathbf{q}_\mathrm{M})]$ of representatives of the 11, 111, and 1111 families using $\frac{\gamma}{\gamma_c}=0.95$. 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.

IV. SCDFT AND PAIRING INTERACTIONS

The SCDFT theoretical framework taking into account spin-fluctuation effects has been discussed in Ref. [46]. This approach considers SF, el-ph, and Coulomb (charge, within RPA) pairing on the same footing (more details on the properties of these interactions can be found in Ref. [46]). First, to keep the formalism complexity at a minimum, we consider a multiband isotropic approximation. This means that band and momentum ($n\mathbf{k}$) dependence in the gap equation and kernels is simplified by an averaging on isoenergy surfaces and Brillouin zone volumes (see also the Supplemental Material [78]). As an example, the Kohn-Sham potential [48] of the SCDFT system $\Delta^{\mathrm{xc}}_{n\mathbf{k}}$ is averaged as:

$$\Delta^{\mathrm{xc}}_{n\mathbf{k}}\approx\Delta^{\mathrm{xc}}_{j}(E):=\sum_{n\mathbf{k}\in j}\frac{\delta(\epsilon_{n\mathbf{k}}-E)}{N_j(E)}\Delta^{\mathrm{xc}}_{n\mathbf{k}}, \quad (2)$$

where $\epsilon_{n\mathbf{k}}$ are the normal state Kohn-Sham eigenvalues. This way we can group together the holes (Fig. 1) around the Γ point (labeled with $j=+$) and the electrons at the M point (labeled with $j=-$), N_j being their density of states at the Fermi energy. 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 α^2F_{ij} (detailed definitions can be found in Ref. [54]and references therein);

(2) the Coulomb repulsion by the average of the RPA screened matrix elements (as was already done in several previous applications of SCDFT as Refs. [57,58]);

(3) the SF term containing an effective interaction mediated by paramagnon excitations Λ^{SF} as introduced in Ref. [46], and averaged [78] as $\Lambda_{n\mathbf{k}n'\mathbf{k}'}(\omega) \longrightarrow \Lambda_{ij}(E,E',\omega)$ 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 [58,59,79–81]. The SF and phonon contributions, on the other hand, are dominated by low frequency modes (Figs. 2 and 3). This implies [82] that these interactions are negligible at energy (E) large with respect to these frequencies and allows us to safely disregard their E dependence, focusing only on the behavior near the Fermi level [82]. These considerations translate into the following approximation scheme:

$$\alpha^2 F_{ij}\left(E,E',\omega\right) \approx \alpha^2 F_{ij}(\epsilon_{\mathrm{F}},\epsilon_{\mathrm{F}},\omega) \text{ (attractive)}$$
$$w_{ij}\left(E,E',\omega\right) \approx w_{ij}\left(E,E',0\right) \text{ (repulsive)}$$
$$\Lambda^{\mathrm{SF}}_{ij}\left(E,E',\omega\right) \approx \Lambda^{\mathrm{SF}}_{ij}(\epsilon_{\mathrm{F}},\epsilon_{\mathrm{F}},\omega) \text{ (repulsive).}$$

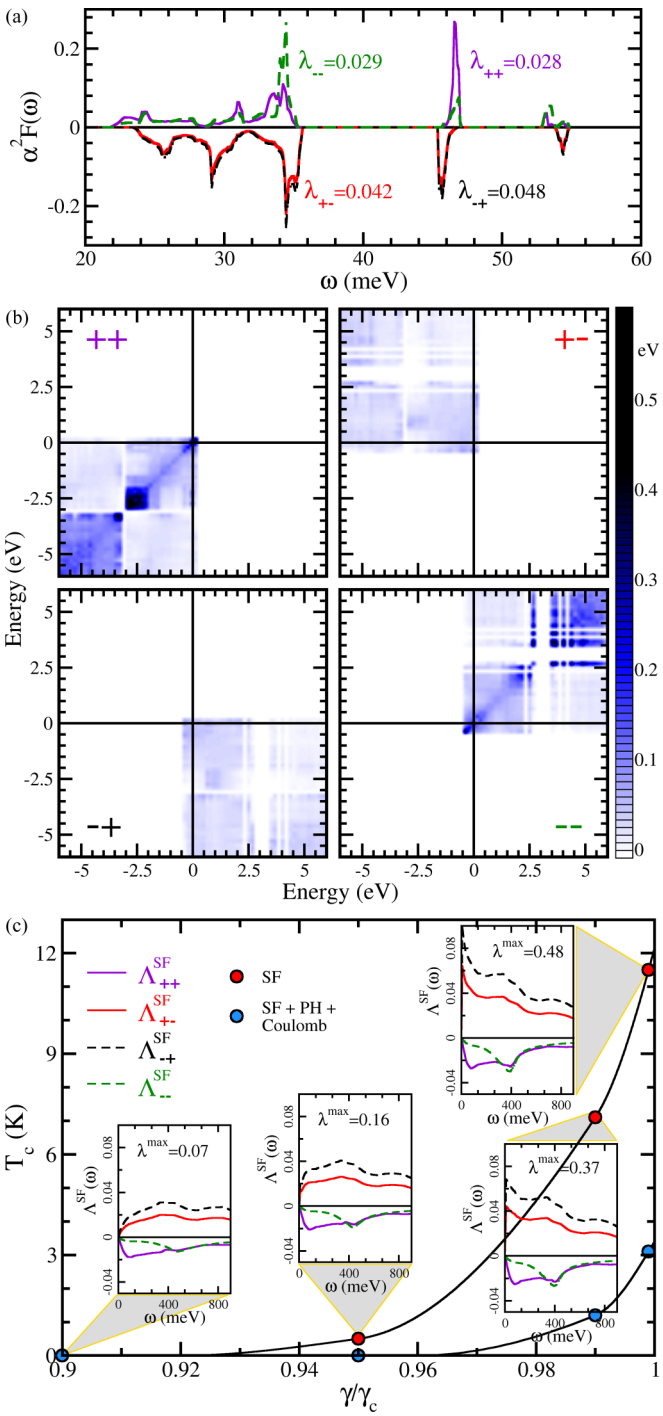

FIG. 3. Two bands resolved superconducting pairing functions evaluated at the theoretically optimized z_{Se}. (a) Electron-phonon Eliashberg function (PH); (b) screened Coulomb matrix elements averaged on isoenergy surfaces (Coulomb); (c) spin fluctuation Eliashberg function (SF) and SCDFT critical temperatures (T_c) as a function of the scaling parameter γ.

Calculations show [83] the el-ph coupling [Fig. 3(a)}to be very small, in agreement with previous works [84,85]. With the integrated coupling [86] λ being less that 0.1, T_c would be exponentially small if this was the only pairing channel. The Coulomb pairing [Fig. 3(b) and Ref. [78]] is, as expected, diagonally dominated ($++$ and $--$ components). Within the static approximation, this structure cannot lead to any pairing, therefore no superconductivity can be sustained at any temperature by the combined effect of Coulomb forces and phonons.

By contrast, the SF pairing term $\Lambda^{\mathrm{SF}}_{ij}$ [shown in the insets of Fig. 3(c)] is very large in proximity of the magnetic phase transition. Its effective coupling $\lambda^{\max}$ [86] rises from the negligible value of 0.07 at $\gamma/\gamma_c = 0.9$ up to 0.48 as $\gamma \to \gamma_c$. This limiting value of $\lambda^{\max}$, in combination with the very high SF characteristic frequencies (as compared to phononic ones), leads to a sizable critical temperature in the $s\pm$ symmetry channel [34].

V. CRITICAL TEMPERATURES AND DISCUSSION

If just SF coupling is considered a T_c as high as 11 K is obtained. However this critical temperature is reduced both by the inclusion of Coulomb terms (11 K $\to$ 4 K) and phonons [87] (11 K $\to$ 10 K), leading to an estimated maximum T_c of 3 K.

This estimation of T_c is in reasonable agreement with the experimentally observed 8 K [88]. This is an important success of the theory, strongly suggesting 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 standard DFT to describe the ground state of FeSe forced us to introduce the parameter γ that we cannot determine from first principles. The estimation of T_c also depends sensitively on the underlying electronic structure, both directly via the FS and density of states [89] and indirectly via the KS response used to calculate the spin susceptibility.

To further strengthen our findings we perform SCDFT calculations using an alternative way to overcome the problem of the description of the magnetic ground state of FeSe. As shown in Fig. 4(d) the magnetic properties of FeSe strongly depend on the positions of the Se atoms z_{Se}. The calculations show that the system is magnetic for $z_{Se} \geqslant 0.22$. We fix

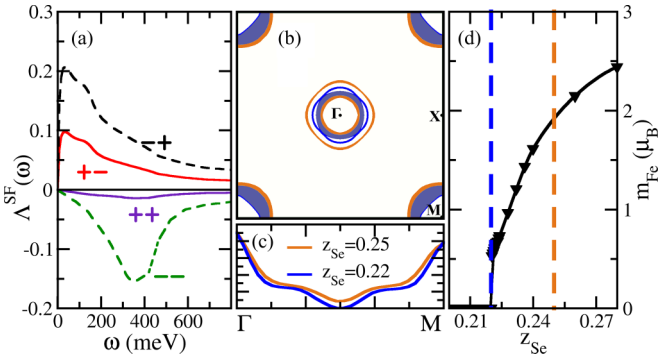

FIG. 4. Effect of the z_{Se} position. (a) Spin fluctuation spectral function Λ^{SF} computed at $z_{Se} = 0.22$, leading to $\lambda^{max} = 1.3$. (b) Projection of the Fermi surface on the xy plane. Blue computed at $z_{Se} = 0.22$, and orange at $z_{Se} = 0.25$. (c) Nesting function (normalized) along the Γ-M line. (d) Magnetic moment at the iron site as a function of the z_{Se} parameter. The values used in this paper are marked by vertical dashed lines.

$z_{Se} = 0.22$ in the paramagnetic region close to the transition to the AFM state. The proximity to the phase transition results in intense paramagnon fluctuations. This leads in our theory to large SF pairing functions [Fig. 4(a)] and to an estimated T_c of 24 K (by including all three pairing channels, the SF-only calculation gives a T_c of 32 K). A 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 nesting for the SF pairing, slightly sharper in the second case [Fig. 4(c)], this leads to an increased interband pairing. This large sensitivity of the predicted T_c to the lattice properties is consistent with experimental observations [3,90–93], in particular with the observed correlation of T_c with the anion position [94].

VI. ANISOTROPY

We conclude the paper reporting the superconducting gap 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 gap corresponding to each sector. 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, however, to a significantly modulated gap function within the $s_\pm$ symmetry. This modulation can be traced back to the properties of the magnetic susceptibility. In particular its most prominent feature is a large gap oscillation in the electronlike FS [see points labeled M_1 and M_2 in Fig. 5(b)]. This is induced by the repulsive effect of the high energy peak in the pairing function [blue dashed line in Fig. 5(a)] caused by the presence of the checkerboard type AFM instability.

VII. CONCLUSIONS

Summarizing, we report the application of SCDFT taking into account the SF. We mostly focus on bulk FeSe. The results strongly suggest that the SF are indeed the physical mechanism leading to the formation of the Cooper pairs in the system, with a pairing symmetry of $s\pm$ type, in agreement with the work

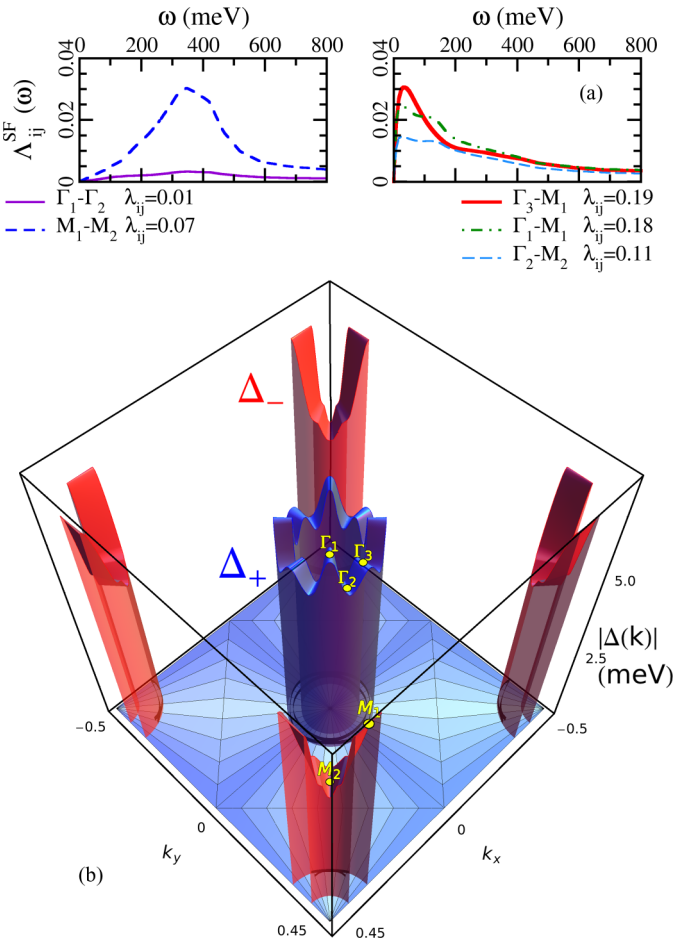

FIG. 5. Bottom: Modulus of the $T = 0$ superconducting gap at the Fermi surface on a $k_z = 0$ cut of the Brillouin zone. The gap sign is indicated by the red (−) and blue (+) colors. Triangular sectors (light blue) mark the areas in reciprocal space where the coupling has been averaged in the calculation. Top: Coupling spectral functions between five regions labeled as Γ_1, Γ_2, Γ_3 (on the hole pocket near Γ) and M_1, M_2 (on the electron pocket around M). Interactions on the left act as repulsive in the $s\pm$ channel (as they connect portions of the Fermi surface with the same gap phase), while those on the right act as attractive (by connecting regions with a + phase with those with a − phase).

of Lischner and co-worker [95], published recently. Also we show that spin fluctuations are strong enough to overcome the contribution of both the phononic and the spin-conserving part of the Coulomb interaction that in this gap symmetry both are repulsive for the Cooper pairing.

To overcome the problem of standard DFT in the description of the FeSe ground state, we adopt two different approaches that allow us to deal with the strong sensitivity to the details of the electronic structure. The estimated T_c varies between 3 and 24 K that is in reasonable correlation with the 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 approach will provide a reliable and completely *ab initio* estimation of the superconducting T_c in this and related compounds. This opens an avenue for the first-principle design of systems with high-T_c superconductivity.

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