

Pairing mechanism of iron pnictide superconductors

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By applying an *exact* unitary transformation to a two-band Hamiltonian which also includes the effects due to large pnictogen polarizabilities, we show that an attractive spin-mediated Hubbard term appears in the d_{xz} , d_{yz} nearest-neighbor channel. This pairing mechanism implies a singlet superconducting order parameter in iron pnictides.

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Recent observation of high-temperature superconductivity in Fe pnictide compounds¹ has generated a wide span interest and a heightened research effort. As a result four families of compounds have emerged having similar properties and exhibiting superconductivity. Namely, chalcogenides (FeSe and FeTe) and three families of pnictides (represented by LaOFeAs, BaFe₂As₂, and LiFeAs).

Band-structure calculations have revealed that (i) electron-phonon coupling alone cannot account for the high values of T_c ,² (ii) nesting between the hole Fermi surfaces at zone center and the electron Fermi surfaces around the zone corner may play a significant role both in magnetism and superconductivity.³ These results have prompted the idea that the pairing glue is provided by spin fluctuations exchange between electrons in different bands.^{3,4} However, the importance of spin fluctuations in iron-pnictide superconductivity is a subject of discussion, cf., conflicting conclusions drawn from temperature dependence of nuclear-spin-lattice relaxation rate ($1/T_1$) data, Ref. 5 vs Ref. 6; also Ref. 7 vs Ref. 8. Thus, exploring possibilities for other pairing mechanisms is needed.

In the following we will present the results of an *exact* unitary (canonical) transformation (UT) by which it can be shown rigorously that an attractive spin-mediated Hubbard term appears in the d_{xz} , d_{yz} nearest-neighbor (nn) channel. The mechanism is similar to a spin double-exchange type and hence has origins of a kinetic mechanism.

We start with the familiar two-band model description of the FeAs superconductors.⁹ We take a two-dimensional square lattice with d_{xz} and d_{yz} orbitals per site. For ease, we label the d_{xz} and d_{yz} orbitals by a and b , respectively. The kinetic-energy component can be expressed as

$$H_{\text{kin}} = -\frac{1}{2} \sum_{\mathbf{ij}, \sigma} \sum_{\alpha, \beta=a, b} t_{\alpha\beta} (\alpha_{\mathbf{i}, \sigma}^\dagger \beta_{\mathbf{j}, \sigma} + \text{H.c.}). \quad (1)$$

Here, the site indices, \mathbf{i} and \mathbf{j} , run over nn and next nn. The effective hoppings, $t_{\alpha\beta}$, have contributions from both direct Fe-Fe and Fe-As-Fe processes.

We use all five Fe 3d orbitals^{4,10} from which we extract a two-band model by focusing on the relevant 2×2 block with nn a - b hopping t_{ab} around 0.5 eV, cf., in Ref. 10 $t_{ab} = 0.54$ eV and the other hopping parameters enter into our

UT formulation through the bandwidth, ε , which we take as 2 eV.^{4,9-11}

$H_{\text{int}} = \sum_{\mathbf{i}} H_{\mathbf{i}}$ contains only on-site contributions with

$$H_{\mathbf{i}} = \sum_{\alpha} U_{\alpha} n_{\mathbf{i}, \alpha, \uparrow} n_{\mathbf{i}, \alpha, \downarrow} + (U' - J/2) \sum_{\mathbf{i}} n_{\mathbf{i}, a} n_{\mathbf{i}, b} - 2J \mathbf{S}_{\mathbf{i}, a} \cdot \mathbf{S}_{\mathbf{i}, b} + J' (a_{\mathbf{i}, \uparrow}^\dagger a_{\mathbf{i}, \downarrow}^\dagger b_{\mathbf{i}, \downarrow} b_{\mathbf{i}, \uparrow} + \text{H.c.}). \quad (2)$$

As in Eq. (1), $\alpha = a, b$ labels d_{xz}, d_{yz} orbitals. $\mathbf{S}_{\mathbf{i}, \alpha}$ ($n_{\mathbf{i}, \alpha, \sigma}$) is the spin (density) in orbital α at site \mathbf{i} . Following Ref. 9, we used $U' = U - 2J$ and the pair hopping term strength $J' = J$, where J is the Hund coupling.

The on-site Hubbard terms are obviously equal, $U_a = U_b = U$, with U chosen between 3.0–4.5 eV. While, there is debate on whether the iron pnictides should be considered as strongly correlated or, at most, moderately correlated materials, there is more consensus regarding the strength of U to be 4 eV,¹² 3.5 eV,¹³ or even smaller $U \leq 2$ eV.¹⁴ In our UT approach described below, U' and J give rise to no distinct physics, although they do render the transformation algebraically complicated. As such, their effect on the final result will be briefly discussed later.

The third and last contribution that we include in our starting Hamiltonian is the polarizability effect. As previously noted in Ref. 15 the polarizability effects in iron pnictides are much larger than in copper-based high T_c 's. It is well known that the ions As³⁻ and Se²⁻ have large polarizabilities due mainly to their large volume. Hence, whenever an iron site is charged, due to electrons hopping to or from it, the surrounding As or Se atoms will easily be polarized, an effect which needs to be captured.

The effect of As (or Se) polarizability on iron sites can be described by writing¹⁶ the Hamiltonian term first introduced in Ref. 17 per iron site as $gn(p^\dagger s + s^\dagger p)$. Here the possible excitations of an As electron from 4p to unoccupied 5s are taken into account with an effective coupling, denoted by g , due to a charge on Fe. The $n = n_\uparrow + n_\downarrow$ notation ($n = n_a$ or n_b) is identical to the one used in Eq. (2) and describes Fe 3d electrons.

The angle dependence between the As p and As s orbitals was extensively studied,¹⁸ hence we consider a simplified version of the Hamiltonian taking into account only the mean total polarizability per bond^{16,17} by a Hamiltonian term of the form: $H_{\text{pol}} = P \sum_{\langle \mathbf{i}, \mathbf{j} \rangle} (n_{\mathbf{i}} - n_{\mathbf{j}})$, where $P = g(p^\dagger s + s^\dagger p)$ will be the

measure of As average polarizability, with P estimated to be around 2 eV.¹⁸ In this way our starting Hamiltonian is $H_{\text{kin}} + H_{\text{int}} + H_{\text{pol}}$.

One of the most common tools used in theoretical physics is perturbation theory. Here, we present a perturbation theory which we solve *exactly* using a UT. There are two reasons for using UT (Ref. 19): (i) the belief that the transformed Hamiltonian is “simpler” in the sense that it is “more diagonal” and (ii) the desire to gain a deeper physical insight into the problem, given that the transformed Hamiltonian may reveal the appropriate independent subsystems. Our scope is to pursue the latter.

For the UT to work, we separate out from H_{kin} the off-diagonal hopping term, $H_{ab} = t_{ab} \sum_{(i,j),\sigma} (a_{i,\sigma}^\dagger b_{j,\sigma} + \text{H.c.})$. Hence we can write $H_{\text{kin}} = H_0 + H_{ab}$, where H_0 contains the remaining terms other than t_{ab} . For transparency, we rewrite our starting Hamiltonian as $H + H_{ab}$, where $H \equiv H_0 + H_{\text{int}} + H_{\text{pol}}$. H will be the zeroth-order Hamiltonian and t_{ab} the perturbation.

It is known^{20,21} that in fourth-order perturbation an attractive Hubbard-type carrier-carrier interaction appears, parallel to spin-carrier, spin-spin, carrier-carrier-spin, etc., terms.^{22,23} All these terms are spin mediated, of superexchange type,^{20,21} and hence with physical origins of a kinetic mechanism.²⁴ The attractive carrier-carrier terms that we are interested in appear either on-site or between nn sites.^{20,21} However, performing a perturbation up to fourth order is not enough to draw a final conclusion, as pointed out in Ref. 21. Depending upon the value of input parameters, the second, sixth, etc., order terms may well give repulsive interactions while the fourth, eighth, etc., offer attractive ones.²³ Thus, higher contributions need to be calculated in order to verify the convergence.

We began by checking for the convergence of higher-order terms for the particular case of Fe pnictides. In a standard UT (Refs. 19–23) the transformed Hamiltonian $e^{\mathcal{S}}(H + H_{ab})e^{-\mathcal{S}}$ is identical to $H + \sum_{n=1}^{\infty} [1/n! - 1/(n+1)!] \tilde{H}_n$ (henceforth, $\tilde{}$ denotes a UT result), where

$$\tilde{H}_n = \overbrace{[\mathcal{S}, [\mathcal{S}, [\mathcal{S}, \dots [\mathcal{S}, H_{ab}], \dots]]]}^{n \text{ times}} \quad (3)$$

for an \mathcal{S} which satisfies $H_{ab} + [\mathcal{S}, H] = 0$. Using the notations: $C_1 = 1/(\varepsilon + P)$, $C_2 = 1/(\varepsilon + P + U_b) - C_1$, $C_3 = 1/(\varepsilon + P - U_a) - C_1$, and $C_4 = 1/(\varepsilon + P + U_b - U_a) - 1/(\varepsilon + P + U_b) - 1/(\varepsilon + P - U_a) + C_1$, the unitary operator \mathcal{S} is

$$\mathcal{S} = -t_{ab} \sum_{(i,j),\sigma} (C_1 + C_2 n_{j,-\sigma}^b + C_3 n_{i,-\sigma}^a + C_4 n_{i,-\sigma}^a n_{j,-\sigma}^b) \times (a_{i,\sigma}^\dagger b_{j,\sigma} - \text{H.c.}) \quad (4)$$

The first-order UT (i.e., second-order perturbation theory²⁶) can be readily performed. This result is well known^{21,23} and consequently not pursued further. In the second and all other “even” order transformations, H_{ab} is recovered structurally with the addition of all possible correlated hopping terms:^{19,21} $H_{n=\text{even}} = \sum_{(i,j),\sigma} [\tilde{t}_{ab} + \tilde{t}_{ab}^a n_{i,-\sigma}^a + \tilde{t}_{ab}^b n_{j,-\sigma}^b + \tilde{t}_{ab}^{ab} n_{i,-\sigma}^a n_{j,-\sigma}^b] (a_{i,\sigma}^\dagger b_{j,\sigma} + \text{H.c.})$.

Consequently, the weight of the original t_{ab} is redistributed in every order of the transformation among \tilde{t}_{ab} , \tilde{t}_{ab}^a , \tilde{t}_{ab}^b ,

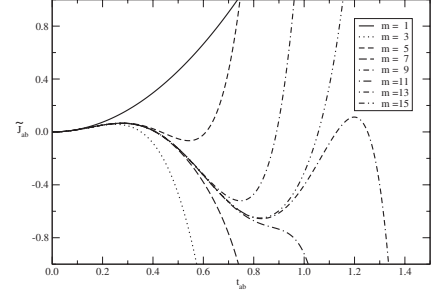


FIG. 1. The calculated superexchange interaction \tilde{J}_{ab} in different orders n , as defined in Eq. (3). We show the total contributions for each order, i.e., $\sum_{n=1}^m [1/n! - 1/(n+1)!] \tilde{H}_n$, see also Ref. 25. For input parameters we used $\varepsilon = 2$ eV, $P = 2$ eV, and $U = 4.5$ eV.

and \tilde{t}_{ab}^{ab} . Because of the smallness of U in Fe pnictides, as the UT is performed to higher-orders weight is shifted in and out of the correlated hopping terms generating an oscillatory behavior. These oscillations are well known^{19,21,23} in standard perturbation theory. As an example, the Heisenberg superexchange term \tilde{J}_{ab} is shown in Fig. 1 for high orders. Hence, the problem with applying standard UT is that unless U is very large, the perturbation series will not converge.

To overcome this challenge, a different approach is needed to handle Fe pnictide case, i.e., to perform an exact UT. We have chosen to perform such a transformation by eliminating three consecutive even order terms simultaneously (the $n=0$, $n=2$, and $n=4$ order terms) from Eq. (3) via a new unitary operator \mathcal{S} such that

$$H_{ab} + [\mathcal{S}, H] + \frac{1}{2!} [\mathcal{S}, [\mathcal{S}, H_{ab}]] = 0. \quad (5)$$

This guarantees that the transformation cannot be continued to higher orders since all $a \leftrightarrow b$ hopping processes are eliminated, as \tilde{t}_{ab} and the correlated hopping terms, \tilde{t}_{ab}^a , \tilde{t}_{ab}^b , \tilde{t}_{ab}^{ab} are strictly zero. Accordingly, the transformed Hamiltonian

$$e^{\mathcal{S}}(H + H_{ab})e^{-\mathcal{S}} = H + [\mathcal{S}, H_{ab}] + \frac{1}{2!} [\mathcal{S}, [\mathcal{S}, H]], \quad (6)$$

is exact in a strict mathematical sense.¹⁹ Equation (6) is merely a self-consistent mixing of the $n=1$ and $n=3$ order standard UT,¹⁹ i.e., of a standard second- and fourth-order perturbation theory.²⁶ This guarantees that we can capture the whole spectrum of the carrier-carrier interaction terms.^{20,21}

The solution of Eq. (5) has the same algebraic form as Eq. (4) with new coefficients C_1 , C_2 , C_3 , and C_4 as unknown parameters. These are determined from Eq. (5), which gives the following systems of equations: $\tilde{t}_{ab} = t_{ab} [\cos(2t_{ab}C_1) + (\varepsilon + P)C_1 \sin(2t_{ab}C_1)/(2t_{ab}C_1)]$,

$$\begin{aligned} \tilde{t}_{ab}^a &= 8t_{ab}^3 \mu [(\varepsilon + P)(\mu^2 - \nu^2) - U_a \mu^2 - U_b \nu^2] \phi(2\tau) \\ &+ 8t_{ab}^3 \mu (\mu - \nu) \psi(2\tau) + 4t_{ab}^3 \mu \nu^2 (\varepsilon + P) \phi(\tau) + 2t_{ab}^3 \nu (\mu \\ &+ \nu) \psi(\tau) - t_{ab} \cos(2t_{ab}C_1) - (1/2)(\varepsilon + P) \sin(2t_{ab}C_1), \end{aligned}$$

where $\mu = C_1 + C_2$, $\nu = C_1 + C_3$, $\tau = t_{ab} [2(\mu^2 + \nu^2)]^{1/2}$, and the notations $\phi(x) = \sin(x)/x^3$, $\psi(x) = \cos(x)/x^3$ were used. For \tilde{t}_{ab}^b

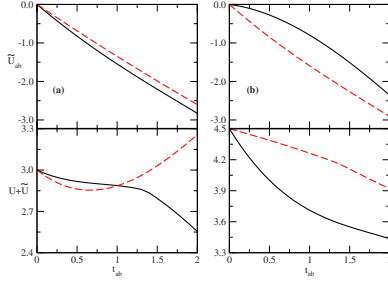


FIG. 2. (Color online) The calculated nn, \tilde{U}_{ab} (top), and the total on-site, U (initial value)+ \tilde{U} (calculated value) (bottom), Hubbard coefficients as a function of t_{ab} . Input parameters are $\varepsilon=2$ eV, $U=3$ eV [part (a)] and $U=4.5$ eV [part (b)]. Continuous (black) curves are $P=1.5$ eV while long-dashed (red) curves are for $P=3$ eV.

we have to exchange U_a with U_b . Finally, the last of the correlated hopping terms is:

$$\begin{aligned} \tilde{t}_{ab}^{ab} = & -8t_{ab}^3(\mu - \nu)[(\varepsilon + P)(\mu^2 - \nu^2) - U_a\mu^2 - U_b\nu^2]\phi(2\tau) \\ & - 8t_{ab}^3(\mu - \nu)^2\psi(2\tau) - 2t_{ab}^3\mu\nu(\mu + \nu)[(\varepsilon + P) - U_a \\ & + U_b]\phi(\tau) - 2t_{ab}^3(\mu + \nu)^2\psi(\tau) + t_{ab}(C_1^2 + \rho^2)\cos(2t_{ab}\rho) \\ & + \frac{1}{2}(\varepsilon + P) \\ & \times (1 + C_1^3/\rho^3)\sin(2t_{ab}\rho) + t_{ab}(1 - C_1^2/\rho^2)[1 + (\varepsilon + P)C_1] \end{aligned}$$

where $\rho=C_1+C_2+C_3+C_4$ and note that these equations can also be used if $U_a \neq U_b$.

We calculated numerically the parameters C_1 , C_2 , C_3 , and C_4 from the above system of equations by setting \tilde{t}_{ab} , \tilde{t}_{ab}^a , \tilde{t}_{ab}^b , and \tilde{t}_{ab}^{ab} to zero. The new UT, as any standard perturbation theory, gives several spin-spin, carrier-carrier, spin-carrier on-site, nn, next-nn, etc., terms. Much attention has been given to the spin-spin terms²⁷ while none to the charge-charge terms. Hence, the focus of our study: the on-site, $\tilde{U}\sum_{i,\alpha,\uparrow}n_{i,\alpha,\uparrow}n_{i,\alpha,\downarrow}$ ($\alpha=a,b$) and nn $\tilde{U}_{ab}\sum_{(i,j)}n_{i,a,\uparrow}n_{j,b,\downarrow}$ Hubbard interactions. Longer range contributions exhibit an Ruderman-Kittel-Kasuya-Yoshida-type behavior with rapidly decreasing strength.²² The results are plotted in Fig. 2, as a function of t_{ab} and in Fig. 3 as a function of the polarizability, P .

Figure 2 shows a nn interorbital attractive Hubbard interaction \tilde{U}_{ab} , which appears due to a spin-mediated superexchange-type mechanism.^{20,21} Being of kinetic origin, the spins are not excited,²⁴ only virtual excitations of on-site singlets occur. The situation is similar to the phonon-mediated attraction: at temperatures well below the Debye scale real phonons are never excited, yet they provide an attraction mechanism between electrons.

The values of \tilde{U}_{ab} in Fig. 2 can be well approximated for small t_{ab} with a line: $\tilde{U}_{ab} \approx -\text{const } t_{ab}$ with the constant being $4\xi(\xi_+ - \xi_-)\{\sin\{2[2\xi^2(\xi_+^2 + \xi_-^2)]^{1/2}\}/\{2[2\xi^2(\xi_+^2 + \xi_-^2)]^{1/2}\} + [\cos\{2 \times [2\xi^2(\xi_+^2 + \xi_-^2)]^{1/2}\} - 1]/\{2[2\xi^2(\xi_+^2 + \xi_-^2)]^{1/2}\}^2\}$, where $\xi_{\pm} = 1/[\varepsilon + P \pm U]$, $\xi = \eta/[(\varepsilon + P)^2 - U^2]/[(\varepsilon + P)^2 + U^2]^{1/2}$, and the numerical coefficient η is 0.392 for $\varepsilon + P < U$, or 1.178 otherwise.

The new Hamiltonian term, H_{Pol} , which gives a measure

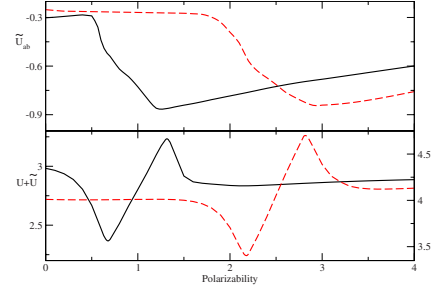


FIG. 3. (Color online) The same Hubbard coefficients as in Fig. 2 as a function of the polarizability, P , for $\varepsilon=2$ eV and $t_{ab}=0.5$ eV. $U=3$ eV, continuous (black) curves and 4.5 eV long-dashed (red) curves, respectively. For the lower curves, the left (right) axis is for $U=3$ eV (4.5 eV).

of the polarizability is not crucial in obtaining this attractive interaction. Any finite value of bandwidth alone suffices for attraction. In fact, the UT expressions depend only on the sum $\varepsilon + P$ and not separately on either. Thus, polarizability effectively acts as a *bandwidth*. So, Fig. 3 covers a wider range for P to allow for cases with different bandwidth and P values. From Fig. 3, upper panel we observe that the attractive \tilde{U}_{ab} is enhanced rather strongly by P for $\varepsilon + P \gtrsim U$. The strongest attraction occurs around the values of P satisfying $\varepsilon + P \approx U$. Note that, this regime is not accessible by standard perturbation theory but it works well in an exact UT approach.

An additional effect due to H_{Pol} is the modified screening of the on-site Coulomb repulsion U in the presence of P . In the lower panel of Fig. 3 the net repulsion, $U + \tilde{U}$, is seen to deviate significantly from a roughly constant value (≤ 3 eV for $U=3$ eV, ≈ 4 eV for $U=4.5$ eV) only in an interval given by $|\varepsilon + P - U| \leq 0.5$ eV = t_{ab} . Normally, ligand polarizabilities are expected to increase the screening (smaller net U values), however, when the double exchange is the strongest, it restricts the hopping and as such screening is reduced, similarly to manganites.

We have also checked smaller U values, namely, for U ($\varepsilon=2$, $t_{ab}=0.5$)=2.5, 1.5, and 0.5, we obtain \tilde{U}_{ab} ($P=1.5$) = -0.71, -0.48, and -0.17 or \tilde{U}_{ab} ($P=3$) = -0.54, -0.35, and -0.12, respectively, depending on polarizability. This shows that our results also hold firm in the weakly correlated limit.

While U' and J from Eq. (2), have been neglected up to now, their effects in our UT can be accounted for substituting U with $U + J(1 - U'/U)/2$ in all instances. Using the known U' and J values^{4,9-14} this only causes a less than 5% change in U and hence their effect is minor.

In conclusion we have shown that the Fe pnictide family of alloys exhibits a rather strong pairing interaction due to a spin-mediated superexchange-type mechanism. There is also a strong enhancement of the attraction due to the polarizability. However we do not observe polaron and bipolaron formation as in Ref. 18 because we are using realistic on-site Hubbard values, instead of $U > 8$ eV as used in Ref. 18. Our pairing mechanism will give rise to a singlet superconducting order parameter. In a two-band system, the self-consistent solution of the gap equations always has a sym-

metric (recently denoted as s_{++}) and asymmetric (s_{+-}) solution.²⁸ In the parameter regime of superconducting Cr alloys, the asymmetric solution wins.²⁹ However, for Fe pnictides a standard multiband mean-field calculation^{28,29} with an attractive \tilde{U}_{ab} will show a tendency toward a s_{++} state³⁰ due to the enhancement of interband coupling.

In closing we note that nonphononic mechanisms support³¹ negative isotope effect.³² The normal isotope effect measured³³ for both superconductivity and itinerant antiferromagnetism in Fe pnictides would require phonon input, which may contradict an electronic mechanism. However,

this may not necessarily be the case, as phonons can act as random impurity potentials in certain cases for systems exhibiting electron and hole Fermi surfaces.³⁴ The pair-breaking effect originating from this also explains why Cr has an isotope effect in T_N . This actually can be very large; the phonons can decrease T_N by as much as 70% from its value in their absence.³⁵ Another interesting consequence of the pair breaking by virtual or thermal phonons³⁵ in a multiband superconductor, is that the ratio $2\Delta/k_B T_c$ can be much larger than the BCS value of 3.53.

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