Coexistence of itinerant electrons and local moments in iron-based superconductors

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Abstract – In view of the recent experimental facts in the iron-pnictides, we make a proposal that the itinerant electrons and local moments are simultaneously present in such multiband materials. We study a minimal model composed of coupled itinerant electrons and local moments to illustrate how a consistent explanation of the experimental measurements can be obtained in the leading-order approximation. In this mean-field approach, the spin-density-wave (SDW) order and superconducting pairing of the itinerant electrons are not directly driven by the Fermi surface nesting, but are mainly induced by their coupling to the local moments with momentum match. The presence of the local moments as independent degrees of freedom naturally provides strong pairing strength for superconductivity and also explains the normal-state linear-temperature magnetic susceptibility above the SDW transition temperature. We show that this simple model is supported by various anomalous magnetic properties which are in quantitative agreement with experiments.

Since the discovery of superconductivity (SC) in iron pnictides [1–5], with \(T_c\) being quickly raised to 55 K [5], intensive attention has been focused on possible underlying mechanisms. With the neutron scattering measurement [6,7] subsequently establishing the fact that an SDW order exists in the undoped LaOFeAs compound below \(T_{SDW} \approx 134\) K, which has been later generically found in other iron pnictides [8–11], the interplay between SC and antiferromagnetism (AF) has become a central issue.

The iron 3d-electrons are believed to be quite itinerant with their hybridized multiorbitals forming multiple Fermi pockets at the Fermi level [12]. Many theoretical efforts [12–21] are based on itinerant approaches in searching for possible SDW and SC mechanisms responsible for the iron pnictides. This kind of theory is generally sensitive to the detailed band structure where the Fermi surface nesting effect is important. As shown by a renormalization group analysis [21], such an itinerant model does possess the instabilities towards the SDW and SC orderings. However, how to reach high-\(T_c\) in the SC phase and at the same time have a self-consistent description of the magnetic phase within a unified framework remains a challenge.

Alternatively local-moment descriptions have been also promoted [22–27] in view of the d-electrons, local Coulomb and Hund’s rule interactions in the iron pnictides, as opposed to the itinerant RPA-type treatment. Of them the so-called \(J_1-J_2\) model which emphasizes the As-bridged superexchange couplings [22,28,29] between the nearest-neighboring (NN) and next-nearest-neighboring (NNN) local moments of the irons has been used due to its natural tendency to form the collinear AF order at low temperature. The local-moment approach is especially appealing over the itinerant one in explaining the anomalous large linear-temperature susceptibility in the normal state over a wide temperature regime [27]. However, how this localized spin picture can be meaningfully applied to a metallic material (albeit a bad metal in the undoped case of the

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The density of states for the itinerant bands (\(\tilde{\varepsilon}\)) remains unclear. Whether the doping effect is similar to that of the cuprates as described by a multiband (t-J1-J2)-like model is also controversial.

In this paper, we point out that if both itinerant electrons and local moments are allowed to simultaneously present in the system, then many basic properties of the iron pnictides can be naturally accommodated by a single framework in the leading-order approximation. To illustrate the point, we study a highly simplified model with the local moments and itinerant electrons coupled together by a Hund’s rule coupling as schematically illustrated in fig. 1. We show that an SDW order of the itinerant electrons can take place simultaneously with a collinear AF order of the local moments, while the ordering would be absent in either degrees of freedom if they do not couple, clearly different from the Fermi surface nesting mechanism or the AF ordering in a J1-J2 model. Furthermore, the superconducting pairing of the itinerant electrons is also driven by the same coupling with the strength reaching strong coupling. It is predicted that in order to consistently account for \(T_{\text{SDW}}\), magnetization, spin gap, uniform susceptibility, as well as the competition between the AF and SC phases, the (high-temperature) normal state of local moments should be close to a critical regime of quantum magnets, which can be tested by a neutron scattering experiment.

**Minimal model.** – Our model Hamiltonian is composed of three terms,

\[
H = H_{\text{it}} + H_{J_0} + H_{J_2}.
\]

The first term \(H_{\text{it}} = \sum_{\mathbf{k}, \sigma} (\varepsilon_{\mathbf{k}} - \mu) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}\) describes the itinerant electrons forming the hole and electron pockets near the Fermi energy as illustrated in fig. 1(a), which are located at the \(\Gamma\)-point and the \(M\)-point, respectively, and separated by momenta \(\mathbf{Q}_s = (\pi, 0)\) and \((0, \pi)\) (only the former is shown in fig. 1(a)) in an extended Brillouin zone (BZ). For simplicity we shall assume the symmetric dispersions for the hole and electron bands, with \(\varepsilon_{\mathbf{k}} = -\varepsilon_{\mathbf{k}+\mathbf{Q}_s}\) such that the hole and electron pockets are exactly nested at \(\varepsilon_{\mathbf{k}} = \mu = 0\), where \(\mu\) is the chemical potential. Such nesting of the Fermi pockets will be lifted as the increase (decrease) of \(\mu\), which effectively controls the electron (hole) doping (the undoped case in iron pnictides may correspond to some small but finite \(|\mu|\) here).

In contrast to the conventional itinerant approach, we shall omit the Coulomb interaction between the itinerant electrons gets enhanced via the Fermi surface nesting effect. Instead, we emphasize the importance of the coupling between the itinerant electrons and some preformed local moments via the second term \(H_{J_0}\) in (1), by a renormalized Hund’s rule coupling \(J_0\) as follows:

\[
H_{J_0} = -J_0 \sum_i \mathbf{M}_i \cdot \mathbf{S}_i,
\]

where \(\mathbf{S}_i\) is the spin operator for the itinerant electrons and \(\mathbf{M}_i\) denotes the local moment at site \(i\). In the following we shall focus on the weak \(J_0\) case where \(\mathbf{M}_i\) behaves like an independent degree of freedom. In the strong-coupling limit of \(J_0\), by contrast, \(\mathbf{S}_i\) and \(\mathbf{M}_i\) should be locked together in strongly correlated regime as has been previously discussed in ref. [23].

An essential assumption of this model will be that besides the itinerant electrons described above, there are some \(d\)-electrons sitting below the Fermi energy that can also contribute to the low-energy physics by forming effective local magnetic moments [23]. Namely, some of the \(d\)-electron multibands can open up a Mott-Hubbard gap crossing the Fermi energy, due to the on-site Coulomb interaction and the Hund’s rule ferromagnetic (FM) coupling, with the filled lower Hubbard bands giving rise to an effective local moment as illustrated in fig. 1(b). These localized \(d\)-electrons may be different from the itinerant electrons as mainly coming from more isolated [30] \(d_{x^2-y^2}\) and \(d_{z^2}\) orbitals. In the following we shall simply assume their existence and explore the consequences of it.

The third term in (1) describes the predominant interaction between these local moments by a Heisenberg-like
model,

\[ H_{J_2} = J_2 \sum_{\langle i,j \rangle \in A} M_i \cdot M_j + J_2 \sum_{\langle i,j \rangle \in B} M_i \cdot M_j, \]

where the NNN superexchange coupling \( J_2 \) is bridged by the As ions between the diagonal iron sites, with \( A \) and \( B \) referring to two sublattices of the square Fe ion lattice, according to the LDA calculation [28,29] and analysis [22]. Note that the NN exchange \( J_1 \), bridged by the As ions, can be either AF or FM in Nature and much weaker than \( J_2 \) for the isolated \( d_{x^2-y^2} \) and \( d_{z^2} \) orbitals due to the symmetry reason [22,30,31]. Furthermore, the itinerant electrons can effectively induce an additional NN FM interaction between the local moments, which is assumed to be predominant (to be consistent with the lattice distortion induced by the SDW ordering observed in the neutron scattering measurement [6,8], see below). Thus we shall neglect the effect of \( J_1 \) to the leading-order approximation and focus on the minimal model (1) in the following study.

**Effective description of the local moments.** According to (3), the local moments \( M_i \) will antiferromagnetically fluctuate in each sublattice of the iron square lattice, and thus may be redefined by \( M_i \equiv M_p n_i \) with \( n_i \equiv e^{iQ \cdot r_i} \), with \( Q_0 = (\pi, 0) \) and \( Q = (\pi, \pi) \) such that the unit vector \( n_i \) will fluctuate smoothly in each sublattice. Thus the itinerant-electron wavefunction is not expected to be very large (\( \sim 0.6 \) eV as indicated in the optical experiment [32]). It is enough to protect the local moments from amplitude fluctuations over a wide temperature regime presumably much higher than the SDW ordering temperature \( T_{SDW} \) as well as \( T_c \). But it also means that in reality the local moment \( M \) is not quantized and described by a Heisenberg-like Hamiltonian (with \( S = 2 \) for instance).

Thus it would be more suitable to use a nonlinear \( \sigma \)-model [33,34] to characterize the low-energy fluctuations of local moments in replace of (3):

\[ L_{J_2} = \sum_{a=A,B} \left\{ \frac{1}{2g_0} \left[ (\partial_\tau n_a) \cdot (\partial_\tau n_a) + c^2 (\nabla n_a) \cdot (\nabla n_a) + i\lambda_0 (n_a^2 - 1) \right] \right\}, \]

with \( c \simeq 4M_2 J_2 \) (with the lattice constant of the Fe square lattice taking as the unit) and \( g_0 \simeq 16 J_2 \). (Note that \( n_a \) \((a = A, B)\) here denotes the unit vector in a given sublattice taking two separated Neel orders would emerge, if \( \lambda_0 = 0 \) at \( T = 0 \).) Denoting \( n_\alpha \equiv (n_a) \), the fluctuations of \( \delta n_a \equiv n_a - n_\alpha \) are described by the propagator \( D_0 (q, \tau) = -\langle T_0 \delta n_a (q, \tau) \cdot \delta n_a (-q, 0) \rangle \) with \( D_0 (q, i\omega_n) = -\frac{\lambda_0}{\omega_n + \Omega_\alpha} \) where the spin wave spectrum \( \Omega_\alpha = \sqrt{c^2 q^2 + \eta^2} \) and \( \eta^2 \equiv i\lambda_0 \) with the subscript \( a \) being dropped, which is determined by the condition \((n_\alpha^2) = 1 \)

with \( \beta = 1/k_B T \) and \( \omega_n = 2\pi n\beta [33,34] \). Hence, without coupling to the itinerant electrons, \( n_\alpha \) \((a = A, B)\) do not couple to one another, and the local moment \( M_\alpha \) governed by (4) will intrinsically fluctuate around any two possible \( \Omega_\alpha \)'s.

**Mean-field theory.** It is important to note that such fluctuations will strongly couple to itinerant electrons via \( H_{J_2} \), for the hole Fermi pockets around the \( \Gamma \)-point and electron Fermi pockets around the \( M \)-point are approximately connected by the momentum \( Q_0 \) at small \( \mu \), as shown in fig. 1(a). In particular, due to the momentum match of the itinerant electrons and local moments, driven by \( H_{J_k} \), the local moments and particle-hole pairs can simultaneously condense at a specific wave vector \( Q_0 \), giving rise to an AF order at a finite mean-field temperature \( T_{SDW} \), which can be stabilized presumably by a weak interlayer coupling.

Assume an SDW order parameter for the itinerant electrons \( \langle S_i \rangle = m = m_0 \neq 0 \) with a specific wave vector \( Q_0 = (\pi, 0) \) in \( m_0 \equiv e^{iQ \cdot r} \). Then a staggered “easy-axis field” from \( H_{J_2} \) should be added to the nonlinear \( \sigma \)-model in (4):

\[ -J_0 M_n \cdot \sum_i n_i. \]

The resulting Euclidean action is still quadratic in \( n_i \) and can be integrated out in a standard way to give rise to the same expression of (5), except that now \( n_\alpha \) is determined by \( n_\alpha = (J_0 M_0 \eta^2/m_0) \). Thus, no matter how weak \( m \) is, it can always induce a collinear AF order of the local moments at the same \( Q_0 \), with \( n_\alpha \neq 0 \). In particular, the independent spin wave spectrum \( \Omega_\alpha \) of the local moment gains a finite gap \( \eta \neq 0 \), in contrast to the limit of \( m = 0 \) where \( m_0 = 0 \) can only occur at \( \eta = 0 \) in a pure 2 nonlinear \( \sigma \)-model [33,34].

Self-consistently, with \( n_\alpha \neq 0 \) a finite \( m \) will always be induced in the itinerant electrons. At the mean-field level, it is governed by \( H_{J_2} - J_0 M_n \cdot \sum_i p_i S_i \), leading to the following band folding and reconstruction:

\[ \sum_{k, \sigma} \left[ (-E_k - \mu) \alpha_\sigma (\alpha_\sigma \sigma_\sigma + \beta_\alpha \beta_\sigma) \right], \]

with the summation over \( k \) is restricted within the reduced magnetic BZ defined by \( Q_0 \) and the itinerant-electron bands are split into \( \alpha \) and \( \beta \) bands, with the Bogoliubov transformation \( \alpha_\sigma \equiv \alpha_\sigma \cdot \sigma_\sigma \), \( \beta_\alpha \equiv \beta_\alpha \cdot \sigma_\sigma \). Here \( u_k = [(1 - E_k/E_0^\alpha)^2]^{1/2} \) and the electron excitation spectrum becomes \( E_k = \sqrt{\epsilon_k^2 + \Delta_{SDW}^2} \) in which the SDW gap \( \Delta_{SDW} = \frac{k_B T}{2} |n_\alpha| \). Finally, the self-consistent mean-field equation reads \( \langle m \rangle = \sum_{k, \sigma} n_k \frac{1}{\epsilon_k} (\alpha_\sigma \sigma_\sigma + \beta_\alpha \beta_\sigma) \), where \( n_k = 1/(e^{\beta(E_k - \mu)} + 1) \) and \( n_k = 1/(e^{\beta(E_k - \mu)} + 1) \).

Solving the above mean-field equations, one can determine \( T_{SDW} \) for the collinear AF order and the total magnetization defined by \( M_{tot} \equiv \mu_B \sum (\alpha + \beta) \). The results are shown in fig. 2. Here we have fixed the parameters \( M = 0.8 \) and \( J_0 = J_2 \) throughout the paper, with \( J_2 \) tunable. The hole dispersion of the itinerant electrons is parameterized as \( E_k = -\alpha(k^2 - k_0^2) \) with \( \alpha = 7J_2 \) and

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$k_0 = 0.1\pi$ based on the ARPES measurement for the so-called “122-type” BaFe$_2$As$_2$ [35–38].

It is noted that the above choice of the parameters is not necessarily optimized. But such a set of parameters can give rise to a quantitative account of a series of important experimental results. The calculated $|M_{\text{tot}}| \approx 0.85\mu_B$ at $T=0$ (which is independent of $J_2$ as shown in fig. 2) is close to $0.87\mu_B$ for BaFe$_2$As$_2$ [8,11]. By taking $J_2 = 30\text{meV}$ (incidentally it is comparable to the LDA estimation [29] for BaFe$_2$As$_2$), one obtains $T_{\text{SDW}} \approx 0.415J_2 = 144\text{K}$ (compared to the experimental value $143\text{K}$ [8]), and in the inset of fig. 2, a gap $\eta = 0.31J_2 = 9.3\text{meV}$ opened up in the spin wave spectrum of the local moments is also fairly close to the experimental value $9.8\text{meV}$ [11] (a similar fit for SrFe$_2$As$_2$ [10] can also be obtained). Note that a proper cut-off momentum $\Lambda = 0.225\pi$ is taken in solving (5) and its role in general will be discussed later together with the comparison with other materials.

**Uniform magnetic susceptibility.** – Using the same set of parameters, the uniform susceptibility $\chi_u = \chi_{\text{lo}} + \chi_{\text{it}}$ is calculated and presented in fig. 3, which exhibits a pseudogap behavior below $T_{\text{SDW}}$ and a rough linear-temperature dependence in the normal state, mainly due to the contribution $\chi_{\text{lo}}$ from the local moments as shown in the inset. As indicated in the latter, both the magnitude and slope of $\chi_{\text{lo}}$ are also quantitatively comparable to the experimental measurements [27,39,40]. Here $\chi_{\text{lo}}$ is given by [34]

$$\chi_{\text{lo}} = \frac{2}{3} \chi_0 n_0^2 + 2\beta^{-1} \sum_{\omega_n,\mathbf{q}} \frac{-\omega_n^2 + c^2 \mathbf{q}^2 + \eta^2}{(\omega_n^2 + c^2 \mathbf{q}^2 + \eta^2)^2}$$

in units of $(g\mu_B)^2$, where $\chi_0 = 1/g_0$.

The contribution from the itinerant electrons is given by

$$\chi_{\text{it}} = \frac{\beta}{2N} \sum_{\mathbf{k}} [n_{\mathbf{k}\alpha} (1-n_{\mathbf{k}\alpha}) + n_{\mathbf{k}\beta} (1-n_{\mathbf{k}\beta})]$$

in units of $(g\mu_B)^2$. $\chi_{\text{it}}$ is mostly Pauli-like, except for an upturn at low temperature as shown in the inset of fig. 3, which is due to the enhanced density of states in the induced SDW state where the SDW gap is near but not right at the Fermi level. It results in the upturn of the total susceptibility at low temperature as shown in fig. 3. Thus, the present theory not only gives rise to the large, linear-temperature–dependent susceptibility above $T_{\text{SDW}}$, but also naturally explains the quick drop of the susceptibility below $T_{\text{SDW}}$ due to the gap opening and its upturn at even lower temperatures generically found in the iron pnictides [39,40].

**Pairing strength.** – The itinerant electrons can also exchange the quantum fluctuations of the local moments to form Cooper pairs, which resonantly hop between the electron and hole pockets with large momentum transfers around $\mathbf{Q}_s$. The pairing amplitude is the $s$-wave within each Fermi pocket, but has to change sign between the two pockets due to exchanging the spin fluctuation $D_0$, consistent with other approaches [15,16,21,25] and measurement [37].

In fig. 4, an effective dimensionless pairing strength $\lambda$ as a function of $\mu$ is calculated based on the same set of parameters with fixing $\mathbf{m} = 0$, i.e., in the absence of the SDW order. Here $\lambda$ is given by

$$\lambda = 2(J_0 M^2) N_F \langle (-1)D_0(k-k'-\mathbf{Q}_s,\omega = 0) \rangle_{FS}$$

where $N_F = 1/(4\pi\alpha)$ denotes the density of states at the Fermi energy and the average is over the Fermi pockets. Note that to properly estimate the strength we have
considered two hole pockets at $\Gamma$ and two electron pockets at two $M$'s in consistency with ARPES experiments [37], as marked in the left bottom part in fig. 4, where the sign change of the pairing amplitude is also indicated. The main panel of fig. 4 shows that $\lambda$ is fairly large in a wide regime (it even diverges at $\mu = 0$ due to the artificial nesting effect of the Fermi pockets). Thus, the superconducting phase is expected to strongly compete with the SDW state at low doping.

We further find that $M_{\text{tot}}$ and $T_{\text{SDW}}$ are quite flat as a function of $\mu$, insensitive to doping. But the SDW order is very sensitive to the aforementioned momentum cut-off $\Lambda$, which decides the critical coupling constant $\Lambda 4\pi\sigma/3\Lambda$ in the nonlinear $\sigma$-model (4) (see text).

![Illustration of the pairing strength $\lambda$ vs. $\mu$, with the relative sign change of the pair amplitude shown at the left bottom. Inset: variations of $\lambda$, $M_{\text{tot}}$, and $T_{\text{SDW}}$ vs. the parameter $g_0/g_c$ of the nonlinear $\sigma$-model (4) (see text).](image)

This is a multiband model, composed of two independent and distinct components, i.e., the itinerant electrons and local moments, respectively. These two independent degrees of freedom are coupled together by the local Hund’s rule interaction at each iron site. Since the imperfect “nesting” momentum of the hole and electron pockets is close to $Q_s$ at small $\mu$, which matches the wave vector of the short-range AF correlation of the local moments, the Hund’s rule coupling term (2) can result in an locking of the SDW ordering of the itinerant electrons and local moments together at $Q_s$. One finds that mean-field transition temperature $T_{\text{SDW}}$, the magnitude of the total magnetization moment, the small spin gap due to the out-of-phase relative fluctuations of the SDW moments between the itinerant electrons and local moments, can be quantitatively determined, in good agreement with the experimental results.

Compared to the conventional Fermi surface nesting mechanism for the itinerant electrons, the present model naturally predicts the presence of pre-formed magnetic moments above the SDW ordering temperature. The calculated normal-state magnetic susceptibility above $T_{\text{SDW}}$ shows a linear-temperature behavior consistent with the experiment in magnitude and slope, under the same set of parameters, which is difficult to understand by the itinerant electrons alone. Furthermore, while the Fermi surface nesting mechanism is difficult to explain why $T_c$ is so high in the iron pnictides, the computed pairing strength between the itinerant electrons via exchanging the AF fluctuations of the local moments is found to be easily in strong-coupling regime in the present work, using the same parameters. The situation is difference to that of the pure itinerant RPA-type treatment. For example, one cannot get large pairing strength under an imperfect “nesting” condition of the itinerant electrons.

Of course, one may further consider the Coulomb interaction in the itinerant bands which may result in an SDW instability without involving the local moments. Or one can further introduce the NN coupling $J_1$ for the local moments and obtain a collinear AF state independent of the itinerant electrons. But the key assumption in this approach is that these effects are negligible, to the leading-order approximation, as compared to the Hund’s rule coupling term and thus are omitted for simplicity. One can always make the model more realistic by adding more perturbative terms later. As a matter of fact, in the experimentally observed lattice distortion [6,8] accompanying the collinear AF order, the NN spins along the shorter lattice constant direction are always FM parallel while the antiparallel NN spins usually correspond to the longer lattice constant [8], in contrast to a $J_1$-driven mechanism which would prefer an opposite lattice distortion [24]. This provides a further support for
the mechanism of the Hund’s rule coupling, with the FM spins at the shorter NN sites in favor of the kinetic energy of the itinerant electrons.

As a matter of fact, in order to provide a consistent explanation of some generic phenomena found in the iron pnictides including the SDW and SC orders within a single framework, the present model suggests that in the normal state the local moments may be close to a critical regime of quantum magnets with the dominant $J_2$-type superexchange interaction, which is consistent with the recent experimental measurement in ref. [41] and can be critically tested by further neutron scattering experiment. The magnetic state of the local moments is also expected to be close to the critical regime in all types of pnictide superconductors. How the present mechanism can be realized microscopically is beyond the scope of the present work, which may involve detailed and realistic local interactions (including $J_1$), and will be left for a future study.

Finally we point out that in the present mean-field approach, the ordered moment of the itinerant electron is chosen to be fixed along the $z$-axis with the spin rotational symmetry broken by hand, and then the induced total magnetization is explored self-consistently. So there is no gapless Goldstone mode at this level of approximation (the spin gap shown in fig. 3 represents the out-of-phase fluctuation between the on-site local moment and the itinerant-electron moment). We shall consider the Goldstone modes and their efforts in the present model by an RPA approach elsewhere [43].

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