A unified theory for the cuprates, iron-based and similar superconducting systems: application for spin and charge excitations in the hole-doped cuprates

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A unified theory for the cuprates and the iron-based superconductors is derived on the basis of common features in their electronic structures, including quasi-two-dimensionality, and the large-\(U\) nature of the electron orbitals close to \(E_F\) (smaller-\(U\) hybridized orbitals reside at bonding and antibonding states away from \(E_F\)). Consequently, low-energy excitations are described in terms of auxiliary particles, representing combinations of atomic-like electron configurations, rather than electron-like quasiparticles. The introduction of a Lagrange Bose field is necessary to enable the treatments of these auxiliary particles as bosons or fermions. The condensation of the bosons results in static or dynamical inhomogeneities, and consequently in a commensurate or an incommensurate resonance mode. The dynamics of the fermions determines the charge transport, and their strong coupling to the Lagrange-field bosons results in pairing and superconductivity. The calculated resonance mode in hole-doped cuprates agrees with the experimental results, and is shown to be correlated with the pairing gap on the Fermi arcs.

Keywords: superconductivity, cuprates, iron, pnictides, auxiliary particles

The recent discovery of high-temperature superconductivity (SC) in iron-based compounds, including pnictides \cite{1,4} and chalcogenides \cite{5} (referred to below as FeSCs), provides an opportunity to test the validity of high-\(T_c\) theories in correlated-electron systems. Similarly to the cuprates \cite{3}, the FeSCs are derived from an undoped “parent” compound which is generally magnetically ordered \cite{7,9} at low temperatures and becomes SC under electron- or hole-doping. Also, both systems are characterized by a layered structure and a quasi-two-dimensional electronic structure \cite{10,16}.

A variety of normal-state properties including, e.g., the transport properties (i.e. resistivity, Hall coefficient and thermoelectric power) of both the cuprates \cite{17,21} and the FeSCs \cite{3,22,23} are characterized by a remarkably similar anomalous behavior. Also, in both systems the suppression of SC by a high magnetic field results in a zero-temperature insulator-to-metal transition upon doping \cite{24,25}. Even though the pairing symmetry is different in the cuprates \cite{26,28} and the FeSCs \cite{29,32}, a resonant spin excitation, characterized by wave vectors around those of the magnetic order in the parent compound, exists in the SC state of both systems \cite{33,35}.

The approximate tetrahedral arrangement of the pnictogen/chalcogen atoms around the iron atoms in the FeSCs is typical of covalent bonding, and thus considerable hybridization is expected between orbitals corresponding to the two atoms. This is confirmed in electronic-structure calculations \cite{10,16}; however, such hybridization is found in antibonding and bonding states which lie at least \(\sim 1\) eV away from the Fermi level \(E_F\), while the states at the close vicinity of \(E_F\) are non-bonding and of almost a pure Fe(3\(d\)) nature \cite{12,16}.

Consequently, the intrasite Coulomb and exchange integrals, corresponding to Wannier functions of the hybridized orbitals of the entire bands which determine the Fermi-surface (FS), magnetic moments, \textit{etc}., may be not large in the FeSCs \cite{36}, resulting in itineracy and largely reduced magnetic moments \cite{7,9}. On the other hand, due to the dominantly Fe(3\(d\)) nature of the states at \(E_F\), their intrasite integrals are rather large \cite{36} and a large-\(U\) approach should be applied to study the physical properties (\textit{e.g.} transport and SC) derived from low-energy excitations.

This aspect of the electronic structure of the FeSCs is different from that of the cuprates, where an entire band around \(E_F\) is believed to correspond to the large-\(U\) physics \cite{28,37}, and an insulating state of large gaps and magnetic moments exists in the parent compounds. Low-energy carriers are present in the cuprates due to doping, and since such carriers in both the cuprates and the FeSCs correspond to the large-\(U\) regime, a unified theory could be worked out for both of them. This theory should be valid also for other quasi-two-dimensional SC systems which are close to a magnetic instability, and have large-\(U\) electrons at the vicinity of \(E_F\).

At the basis of this theory stands the observation that SC exists at stoichiometries where the dynamics of the low-energy carriers dominantly involves fluctuations between two adjacent occupation numbers \((n)\) of atomic-like configurations \((3d^n)\) around the copper or iron atoms. In the cuprates \cite{28,37} these are fluctuations between effective Cu\((3d^{0})\) (hybridized with O\((2p)\) orbitals) and Cu\((3d^{0})\) configurations for electron doping, and between effective Cu\((3d^{0})\) and Cu\((3d^{0})\) (obtained through Zhang-Rice-type hybridization with O\((2p)\) orbitals) configurations for hole doping. In the FeSCs these are fluctuations between Fe\((3d^{0})\) and Fe\((3d^{0})\) configu-
rations for electron doping, and between Fe(3d^5) and Fe(3d^6) configurations for hole doping.

Such dynamics of carriers could be treated through the auxiliary-particle approach [38]. A configuration corresponding to an occupation number n is denoted by α(n), and a combined orbital-spin index of an atomic-like electron by η. For notation simplicity, let α(n − 1(η)) be the configuration obtained by removing an η electron from α(n) ⊃ η. The operator \( a_{i\alpha(n)}^{\dagger} \) creates an auxiliary particle representing the configuration α(n) at site i (a two-dimensional approximation is applied of points \( R_i \) within a planar lattice which could be defined to contain one Cu or Fe atom per unit cell [11, 39]).

The creation operators of electrons of spin-orbitals \( \eta \) at sites i can be expressed as:

\[
d_{i\eta}^{\dagger} = \sum_{\alpha(n)\eta} a_{i\alpha(n)}^{\dagger} a_{i\alpha(n-1(\eta))}.
\]

They satisfy anticommutation relations of independent fermion operators under the following conditions: (i) the consequence of the large-U approximation that only the contribution of two adjacent values of n could be considered in rhs of Eq. (1) is valid; (ii) the auxiliary particles created by \( a_{i\alpha(n)}^{\dagger} \) are either bosons for even n and fermions for odd n, or fermions for even n and bosons for odd n; (iii) the following constraint is satisfied in every site i:

\[
\sum_{\alpha(n)} a_{i\alpha(n)}^{\dagger} a_{i\alpha(n)} = 1.
\]

As was discussed above, two occupation numbers (n) are considered, including \( n_0 \) (corresponding to the parent compound), and either \( n_0 + 1 \) (for electron doping) or \( n_0 - 1 \) (for hole doping). Let us denote by \( \alpha, \beta \) and \( \gamma \) the configurations corresponding to the occupation numbers \( n_0 + 1, n_0 \) and \( n_0 - 1 \), respectively. Their creation operators at site i are denoted by:

\[
e_{i\alpha}^{\dagger} \equiv a_{i\alpha(n_0+1)}^{\dagger}, \quad s_{i\beta}^{\dagger} \equiv a_{i\alpha(n_0)}^{\dagger}, \quad h_{i\gamma}^{\dagger} \equiv a_{i\alpha(n_0-1)}^{\dagger}.
\]

Auxiliary-particles created by \( s_{i\beta}^{\dagger} \) are chosen as bosons, and thus those created by \( e_{i\alpha}^{\dagger} \) and \( h_{i\gamma}^{\dagger} \) are fermions.

The Hamiltonian \( \mathcal{H} \), applied to study low-energy electron excitations, is based on intrasite one- and two-particle terms, and intersite one-particle terms. It is expressed in terms of the auxiliary-particle operators through Eqs. (13). A grand-canonical formalism is applied by including in the Hamiltonian terms corresponding to the chemical potential \( \mu \), and to a field of Lagrange multipliers \( \lambda_i \) (\( \lambda = \langle \lambda_i \rangle \)) associated with the auxiliary-particle constraint [Eq. (2)]. The values of \( \lambda_i \) and \( \mu \) should be determined to yield the correct charge and constraint in every site. \( \mathcal{H} \) could be, formally, expressed as (using constraint-preserving term):

\[
\mathcal{H} \cong \mathcal{H}^e + \mathcal{H}^c + \mathcal{H}^h + \mathcal{H}^{eh} + \Delta \mathcal{H},
\]

\[
\mathcal{H}^e = \sum_{i\beta} (e_{i\beta}^0 - \lambda) s_{i\beta}^{\dagger} s_{i\beta},
\]

\[
\mathcal{H}^c = \sum_{i\alpha} \left\{ (e_{i\alpha}^0 - \mu - \lambda) e_{i\alpha}^{\dagger} e_{i\alpha} + \sum_{j \neq i} \sum_{\alpha' \beta'} \left[ t_{j\alpha'\beta'}^{\dagger} (R_i - R_j) s_{i\beta}^{\dagger} s_{j\alpha'}^{\dagger} e_{j\alpha} + h.c. \right] \right\},
\]

\[
\mathcal{H}^h = \sum_{i\gamma} \left\{ (e_{i\gamma}^h + \mu - \lambda) h_{i\gamma}^{\dagger} h_{i\gamma} + \sum_{j \neq i} \sum_{\gamma' \beta'} [ t_{j\gamma'\beta'} s_{i\beta}^{\dagger} s_{j\gamma'} h_{i\gamma}^{\dagger} h_{j\gamma'} + h.c. ] \right\},
\]

\[
\mathcal{H}^{eh} = \sum_{i\alpha} \sum_{j \neq i} \sum_{\beta' \neq \beta} \left[ t_{j\alpha\beta'}^{\dagger} (R_i - R_j) e_{i\alpha}^{\dagger} h_{j\beta'}^{\dagger} s_{i\beta} s_{j\beta'} + h.c. \right],
\]

\[
\Delta \mathcal{H} = - \sum_{i} (\lambda_i - \lambda) \left[ \sum_{\alpha} e_{i\alpha}^{\dagger} e_{i\alpha} + \sum_{\beta} s_{i\beta}^{\dagger} s_{i\beta} + \sum_{\gamma} h_{i\gamma}^{\dagger} h_{i\gamma} \right].
\]

The \( \lambda_i - \lambda \) Lagrange field represents an effective fluctuating potential which prevents, through \( \Delta \mathcal{H} \), constraint-violating fluctuations in the auxiliary-particle site occupation (thus enabling the treatment of atomic-like electron configurations as bosons or fermions). The effect of such a fluctuating potential on these configurations is analogous to the effect of vibrating atoms on electrons. Consequently, similarly to lattice dynamics, the quantization of the \( \lambda_i - \lambda \) field yields bosons.

In the cuprates one often applies a one-orbital model [28, 67], under which there is one \( \alpha \) configuration, corresponding to a complete Cu(3d^10) shell, one \( \gamma \) configuration corresponding to a Zhang-Rice singlet, and two \( \beta \) configurations corresponding to the spin states of the orbital \( \sigma = +1 \) and \( \sigma = -1 \) (also presented here as \( \sigma = \pm 1 \)). The present auxiliary-particle method then becomes the “slave-fermion” method applied in previous works by the author [10, 41]. The parameters appearing in Eq. (4) are then simplified to the intrasite and transfer (hopping) integrals:

\[
e_{\alpha}^{\dagger} = e_{\alpha}^{\dagger} = e_d, \quad e_{\alpha} = 2e_d + U, \quad e_{\gamma}^{h} = 0,
\]

\[
t_{j\alpha\beta}^{\dagger}(R) = t_{j\gamma\beta}^{\dagger}(R) = t_{j\alpha\gamma}^{\dagger}(R) = t_{j\gamma\alpha}^{\dagger}(R) = t(R).
\]

In the FeSCs one needs at least three Fe(3d) orbitals (of the \( xx, yz \) and \( x^2 - y^2 \) symmetries) to study the electrons at the vicinity of \( E_F \), and there are numerous \( \alpha, \beta \) and \( \gamma \) configurations. The parameters appearing in Eq. (4) are then derived from intersite transfer, and intrasite one-particle, Coulomb and Hund’s-rule exchange integrals [42].

Within the large-U approximation, applied in the derivation of \( \mathcal{H} \), it could be approximated by omitting
The \( s_{ij} \)-field bosons are referred to as “svivons”. Their Bose condensation is manifested, at low doping levels, in AF order, in the cuprates [40], and in a structural distortion and magnetic order, characterized by a spin-density wave (SDW), in the FeSCs [7,9]. At higher doping levels the Bose condensation of svivons is manifested in static or dynamical inhomogeneities, based on modulations of the low-doping order.

When svivons are Bose condensed, an \( s_{ij} \) field operator can be expressed as a sum of its “condensed” part (i.e. the nonzero \( \langle s_{ij} \rangle \)) and fluctuating part \( s_{ij} - \langle s_{ij} \rangle \). Thus, the expression of an electron creation operator in terms of products of auxiliary-particle operators, through Eqs. [14], includes terms where either \( e_{i\alpha}^\dagger \) or \( h_{i\gamma} \) are multiplied by a condensed part of svivon operators, and terms where they are multiplied by their fluctuating part. A “quasi-electron” (QE) is defined as the fermion created by a normalized approximation to an electron creation operator, where only the terms in its expression which include condensed parts of svivon operators are maintained.

The QEs represent hypothetical approximate electrons which do not introduce fluctuations to the inhomogeneities resulting from the Bose condensation of the svivon field. Since QE states are expanded as combinations of auxiliary-particle fermion states created by either the \( e_{i\alpha}^\dagger \) or \( h_{i\gamma} \) operators, these auxiliary-particle states form a basis to the QE states, and could be referred to as QEs as well.

Thus, the problem of SC in strongly-interacting electron systems is treated in terms of an auxiliary space consisting of three types of coupled “particles”: \( i \) boson svivons which represent combinations of atomic-like electron configurations of the parent compounds, and their condensation results in static or dynamical inhomogeneities; \( ii \) fermion QEs which represent combinations of such configurations with an excess of an electron or a hole over those of the parent compounds, and their dynamics largely determines charge transport; \( iii \) boson lagrons which represent an effective fluctuating potential, enabling the treatment of the above configurations as bosons and fermions.

Within this auxiliary space the pairing between the fermions through the exchange of bosons could be rigorously worked out in terms of coupled \textit{independent} fields, in analogy to the electron and phonon fields within the BCS-Migdal-Eliashberg theory. The strong coupling between QEs and lagrons, necessary for the constraint [Eq. (2)] to be satisfied, results in high pairing temperatures. If the same scenario were worked out as the pairing between electrons through the exchange of spin or charge fluctuations, generated by \textit{the same} system of electrons, then two problems would have existed: \( i \) it is doubtful that such strongly-interacting electrons could be treated as quasiparticles; \( ii \) the coupled fermion and boson fields are \textit{not} independent of each other.

Svivon and QE spectra in hole-doped cuprates have been evaluated through a self-consistent second-order di-
FIG. 2: The absolute values of the svivon spectral functions below \( T_c \) for two typical hole-doped cuprates of different spin gaps; both the results for one condensate, and for their average over the four combined condensates are shown.

agrammatic expansion \([47]\), where a mean-field treatment of \( \mathcal{H} \) in Eq. (7) is applied at the zeroth order. The expansion is carried out on two Hamiltonian terms. One of them is \( \Delta \mathcal{H} \) which introduces svivon-lagron and QE-lagron coupling. Vertex corrections to it are negligible by a phase-space argument, as in Migdal’s theorem, since the dominant contribution of the fluctuating part of the constraint \([\text{Eq. (2)}]\) comes from a limited \( k \)-space range of the lagron spectrum around point \( Q \) (see Fig. 1). The other term, \( \mathcal{H}' \), introducing QE-svivon coupling, is the contribution of the fluctuating part of the svivon operators to \( \mathcal{H} \). It is treated as a perturbation, and approximated through a first-order expansion of the rhs of Eq. (7) in terms like \( s_{i\sigma} - \langle s_{i\sigma} \rangle \) \([47]\).

Lagron spectra of the type presented in Fig. 1 determine degenerate Bose-condensed svivon states, with energy minima at points \( \pm Q_m / 2 \), for one of the four values of \( m \) in Eq. (5). Since there are four inequivalent values of \( Q / 2 \) at \( \pm (\pi / 2 a)(\hat{x} \pm \hat{y}) \), the number of possible condensates is eight. In the absence of symmetry-breaking long-range order, the system is generally in a combination of these states (reflecting fluctuations between them). Tetragonal symmetry occurs when all the eight degenerate states are combined, while orthorhombic symmetry breaking results in the combination of four of the eight states. The resulting stripe-like inhomogeneities \([48, 51]\) (which resemble a checkerboard in the combination state) would be static or dynamical, depending on how close to zero are the spectrum minima.

As they occur in Bose fields, the svivon spectral functions are positive at positive energies, and negative at negative ones. Their absolute values for two typical cases, of different nonzero spin gaps, in hole-doped cuprates below \( T_c \) (where the low-energy svivon linewidths are small) are presented in Fig. 2. Shown are the results for the svivon condensate with energy minima at \( \pm (\pi / 2 a)(\hat{x} \pm \hat{y}) + \frac{1}{2} \delta q \hat{x} \hat{x} \), and the average of the results for the four condensates with minima at the vicinity of \( \pm (\pi / 2 a)(\hat{x} \pm \hat{y}) \) (representing their combination), in vertical and diagonal directions around this point.

The QE spectrum of hole-doped cuprates has been evaluated treating the fluctuations between the combined svivon condensates adiabatically, as is detailed in a separate paper \([52]\). By the definition of electron creation operators in Eq. (11), the electron Green’s functions are obtained at the zeroth-order as sums of products of QE and svivon Green’s functions. This results in the non-Fermi-liquid (non-FL) scenario of a distribution of con-
FIG. 3: The imaginary part of the spin susceptibility, corresponding to the svivon spectra for hole-doped cuprates below $T_c$ presented in Fig. 2; the small- and large-spin-gap results demonstrate, respectively, the existence of an incommensurate [34] or a commensurate [33] resonance mode.

The spin susceptibility (SS) of hole-doped cuprates has been evaluated, under an approximation where only the non-FL convoluted QE-svivon poles are considered [52]. Linear-response theory has been applied on the basis of spin-flip processes, expressed by constraint-preserving terms of the form $\langle s_{i\sigma}^\dagger s_{i\sigma}, s_{j\sigma'}^\dagger s_{j\sigma} \rangle$, and thus determined by the svivon spectrum. Results obtained for the imaginary part of the SS, in vertical and diagonal directions around $k = Q$, are presented in Fig. 3. They correspond to the two svivon spectra shown in Fig. 2 and since the svivon-system is in a combination state, the SS results are averaged over those of the four combined condensates. These results reproduce those observed in neutron-scattering measurements in different hole-doped cuprates. The larger spin-gap results correspond to the “commensurate resonance mode (RM)” [33], and the smaller spin-gap results correspond to the “incommensurate RM” [34].

If the constraint is imposed in any two sites $i$ and $j$, the following equation should be satisfied in these sites in hole-doped cuprates (see Eqs. 2,3):

$$\sum_{\sigma\sigma'} \langle s_{i\sigma}^\dagger s_{i\sigma}, s_{j\sigma'}^\dagger s_{j\sigma} \rangle \approx \langle h_i^\dagger h_i h_j^\dagger h_j \rangle.$$  (9)

The terms in the lhs of Eq. (9) are formally similar to the above spin-flip term applied for the derivation of the SS results presented in Fig. 3. Thus, a susceptibility-like function, referred to as the “constraint susceptibility” (CS) could be derived on the basis of either the svivon spectrum, through the lhs of Eq. (9), or the QE spectrum, through the rhs of Eq. (9). The results obtained for the CS on the basis of both spectra should agree with each other in order for the constraint to be satisfied, and this condition is the basis for the determination of the lagron spectrum, and of their coupling to the svivons and QEs. The CS represents the response of auxiliary particles, and not of electrons. However, it reflects, under certain conditions, an approximation to the response of the system to charge fluctuations which could be measured, e.g., by Raman spectroscopy [26, 53].
FIG. 4: The imaginary part of the constraint susceptibility corresponding to the svivon spectra for hole-doped cuprates below $T_c$ presented in Fig. 2; the low-energy peaks around $k = 0$, approximately, correspond to the integrated energies of the resonance-mode peaks shown in Fig. 3; these peaks also, approximately, correspond through Eq. (9) to the SC gap over the Fermi arcs (see discussion in the text).

Results obtained for the imaginary part of the CS, on the basis of the lhs of Eq. (9), in vertical and diagonal directions around $k = 0$, are presented in Fig. 4. They correspond to the two svivon spectra shown in Fig. 2 and evaluated similarly to the SS results presented in in Fig. 3. The major feature observed in CS results is a low-energy peak around $k = 0$ at energies which, approximately, correspond to the energies of the $k$-integrated low-energy features of the SS at the vicinity of $k = Q$ (thus the incommensurate or commensurate RM).

Since the same CS results, as those presented in Fig. 4, should be obtained also on the basis of the QE spectrum through the rhs of Eq. (9), and since they correspond to the SC state, the observed peak at $k = 0$ should represent some kind of an average value of the QE gap below $T_c$. As is explained elsewhere [52], this gap has two contributions: one of them originates from Brillouin zone (BZ) ranges around the antinodal points, where a narrow peak (of energy $\epsilon = 0$ at $T = 0$), lying between two humps, splits due to pairing below $T^*$; the other contribution to that gap opens below $T_c$ on the Fermi arcs (FAs) around the line of nodes.

In the SC state there are both “normal” and “anomalous” (pair-correlation) QE Green’s functions, and their contributions to the QE expression for the CS have opposite signs [47]. These contributions cancel each other for “gap-edge states”, where $\epsilon = 0$, $E = \sqrt{\epsilon^2 + \Delta^2} = \Delta$, and thus the fraction of both the particle and the hole states within the Bogoliubov states is $\frac{1}{2}[1 \pm \epsilon/E] = \frac{1}{2}$. So the QE-spectrum contributions to the CS peak at $k = 0$ come from states where $\epsilon \neq 0$.

Consequently [52], the averaged QE gap which determines the $k = 0$ CS peak is lowly weighted around the antinodal points, and represents a value somewhat larger than the averaged QE gap on the FAs. Since the averaged electron FA gap is also somewhat larger than the QE FA gap (due to convolution with svivon states), one expects a correlation between the values of this gap and the $k = 0$ CS peak, and as was discussed above (see Figs. 3 and 4), also with the averaged RM energy. The electron FA gap has been measured through, e.g., the $B_{2g}$ Raman mode, and its value has indeed been found to be correlated with the RM energy [20, 27, 33]. A correlation between the energies of the $A_{1g}$ Raman mode and the RM [52] has been
found to be partial. The observed correlation of the FA gap with $\sim 5k_{\perp}T_c$ is explained elsewhere.

Even though the electronic structure of low-energy states in the FeSCs is based on more orbitals than in the cuprates, important physical conclusions could be drawn from one system to the other due to the formally common Hamiltonian applied for both of them. Within a two-dimensional approximation, the lagron spectrum of the FeSCs is expected to differ from that of the cuprates, presented in Fig. 1 by replacing the minima positions from satellite points around $Q$, to satellite points around the two possible SDW wave vectors in the parent compounds: $Q_1 = (\pi/a)x$ and $Q_2 = (\pi/a)y$ (or $Q_1 = (\pi/2a)(x+y)$ and $Q_2 = (\pi/2a)(x-y)$), or points close to them. Similarly to the cuprates, stripe-like inhomogeneities characterized by modulations due to the differences between the satellite points and $Q_1$ or $Q_2$, could exist also in the FeSCs.

The svivon spectrum in the FeSCs is expected to have analogous features to those of the cuprates, presented in Fig. 2, resulting in a resonance mode in the vicinity of $Q_1$ and $Q_2$, below $T_c$, as has been observed. In a separate paper, it is explained that QE pairing requires a sign reversal of the order parameter upon a shift of $Q$ in the BZ, in the cuprates, and of $Q_1$ or $Q_2$ in the FeSCs. Due to their different FSs, this results in pairing symmetry of an approximate $d_{x^2-y^2}$ type in the cuprates, and of an $s_\pm$ type (thus with different signs on different FS pockets) in the FeSCs. Thus, it is predicted that there are no Fermi arcs in the FeSCs, and that their RM energy is correlated with an averaged value of the SC gap, as has been observed.

It could be concluded that high-$T_c$ SC occurs in quasi-two-dimensional strongly-interacting electron systems due to the fact that low-energy excitations in them are described in terms of auxiliary particles, representing combinations of atomic-like electron configurations, rather than electron-like quasiparticles. A Lagrange field which must be introduced to enable the treatments of these auxiliary particles as fermions or bosons, serves as the pairing glue between the fermions.

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