Pair correlation functions of FeAs-based superconductors: Quantum Monte Carlo study

V A Kashurnikov, A V Krasavin
National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), 31 Kashirskoye shosse, 115409, Moscow, Russia
avkrasavin@gmail.com

Abstract. The new generalized quantum continuous time world line Monte Carlo algorithm was developed to calculate pair correlation functions for two-dimensional FeAs-clusters modeling of iron-based superconductors within the framework of the two-orbital model. The analysis of pair correlations depending on the cluster size, temperature, interaction, and the type of symmetry of the order parameter is carried out. The data obtained for clusters with sizes up to 10 x 10 FeAs-cells favor the possibility of an effective charge carrier’s attraction that is corresponding the $A_{1g}$-symmetry, at some parameters of interaction.

1. Introduction
Recently new iron-based high-temperature superconductors (HTSC) [1], both pnictides and chalcogenides, came to be regarded as the subject of intense theoretical and experimental research [2, 3]. The reason is not only the similarity in structural, magnetic, electrophysical, and other properties and features to copper-oxide superconductors but also the presence of a complex band structure, and variety of magnetic and superconducting phases.

Iron pnictides have a layered structure of closely spaced atomic planes consisting of Fe and As atoms. As well as copper-oxide HTSC, they have a pronounced anisotropy which allows description of these systems on the basis of two-dimensional tight-binding models. However, these models have multiband structure and they are much more complex than, for example, the Emery model for copper-oxide HTSC [4]. Most appropriate tight-binding models for these compounds are two- [5, 6], three- [7–9] and the five-orbital ones [9, 10], which are typical generalized Hubbard multiband models. Strong correlations, a multi-gap band structure, features of magnetic and superconducting properties [11–13], the lack of expansion parameters for analytical approximations are the cause of to employ exact quantum methods for the study of iron-based HTSC.

The results known to date on exact calculations of FeAs clusters were obtained mainly using the methods based on the exact diagonalization (ED) of the Hamilton matrix of the model under consideration, and on Monte Carlo (MC) technique.

The properties of the two-orbital model for a cluster of size $\sqrt{8} \times \sqrt{8}$ (the so-called «tilted» cluster) containing eight iron atoms were studied in detail [6, 14] by ED technique. However, the ED method, although giving precise answers for the two-orbital model, is limited by the size of the clusters as the size of the Hamilton matrix grows exponentially with the system size. The largest cluster calculated by ED technique in the case of the two-orbital model took 10 iron atoms that correspond to cluster size $2 \times 5$. The power of Earth Simulator supercomputer (Japan) was required for such calculation [15]. The data on pairing correlations and the binding energy obtained in these studies were allowed to
identify areas on the phase diagram corresponding to the pairing symmetries $A_{1g}$ and $B_{2g}$ of the superconducting order parameter [6, 16].

As regards cluster calculations of iron-based HTSC by MC methods, it should be noted the following.

In [17] 8 × 8 and 10 × 10 clusters were calculated using the variational MC algorithm also in the framework of the two-orbital model. From our point of view the choice of a variational wave function with taking into account antiferromagnetic correlations only leads to results close to the Hartree-Fock (mean-field) approximation. Strictly speaking, this approach may be inadequate to do correct conclusions about such an important issue as the pair correlations in the system.

Calculations of FeAs clusters up to 16 × 16 size using the determinant MC method were implemented in [18, 19] in the framework of a simplified model that is referred to as the model with $S_4$-symmetry. In fact, this model is a two-band generalized Hubbard model with diagonal interaction between the bands. The data obtained on pair correlations, nevertheless, show the presence of $A_{1g}$ pairing symmetry.

The three-orbital model was studied in [20, 21] by MC method for clusters of size up to 16 × 16 in undoped state. On the one hand, this model is more complete in comparison with two-orbital one, and the results allow to describe correctly the experimentally observed photoemission spectra and the spectra of neutron scattering, as well as to obtain data on magnetic ordering and conductivity in the non-superconducting state. On the other hand, the pair correlations were not discussed in these works, and, according to [6], the two-orbital model is sufficient for the analysis of pair correlations in FeAs systems.

There were also approaches describing the features of pnictides by spin degrees of freedom [22] and focusing mainly on the study of antiferromagnetic correlations.

In this paper, the new generalized quantum continuous time world line MC algorithm (the generalized CTWL-algorithm) was developed and adapted to the full two-orbital model. An accurate calculation of non-diagonal terms of the form $a_{i\sigma}^+ a_{i\sigma}^+ a_{j\sigma}^- a_{j\sigma}^-$ within the full two-orbital model which may play a leading role for the pairing of charge carriers and, in our view, cannot be neglected, is the key feature of the presented algorithm. The new algorithm is based on the previously developed CTWL-algorithm [23, 24] which allowed a convenient coding of states for MC procedures.

2. Two-orbital model

In the crystal structure of iron pnictides each of Fe atoms is surrounded by a tetrahedron of As atoms, and vice versa. Band structure calculations [25] show that $3d$−states of iron atoms make the main contribution to the electronic density of states near the Fermi level. Therefore, a basic structural element can be defined as two Fe atoms located in a simple square lattice and two As atoms above and below the Fe-plane. One is responsible for conducting and superconducting properties of the system. Accordingly the tight-binding model of the system is defined [5, 6].

The two-orbital model is quite complex to implement algorithms based on quantum MC or ED technique for modeling of the iron based superconductors. The Hamiltonian of the two-orbital model is given by

$$H = H_{int} + H_{kin},$$

$$H_{int} = U \sum_{i,\alpha} n_{i,\alpha,\uparrow} n_{i,\alpha,\downarrow} + V \sum_{i} n_{i,x} n_{i,y} - \mu \sum_{i} n_{i} - J \sum_{i} (n_{i,x}\uparrow n_{i,y}\uparrow + n_{i,x}\downarrow n_{i,y}\downarrow),$$

$$H_{kin} = -t_1 \sum_{i,\sigma} (a_{i,x,\sigma}^+ a_{i+x,x,\sigma} + a_{i,y,\sigma}^+ a_{i+y,y,\sigma} + h.c.) - t_2 \sum_{i,\sigma} (a_{i,y,\sigma}^+ a_{i+x,y,\sigma} + a_{i,x,\sigma}^+ a_{i+y,x,\sigma} + h.c.)$$

$$-t_3 \sum_{i,\sigma} (a_{i,x,\sigma}^+ a_{i+x+y,x,\sigma} + a_{i,y,\sigma}^+ a_{i+x+y,y,\sigma} + a_{i,x,\sigma}^+ a_{i+y,x+y,\sigma} + a_{i,y,\sigma}^+ a_{i+x+y,y+y,\sigma} + h.c.)$$

The calculations of pair correlations in the system were performed using the MC method.
\[ -t_4 \sum_{i,\sigma} (a_{i,x,\sigma}^+ a_{i+x+y,y,\sigma} + a_{i,y,\sigma}^+ a_{i+x+y,x,\sigma} + a_{i,x,\sigma}^+ a_{i-x-y,y,\sigma} + a_{i,y,\sigma}^+ a_{i-x-y,x,\sigma} + h.c.) \]  

where operator \( a_{i,x(y),\sigma}^+ (a_{i,x(y),\sigma}) \) creates (annihilates) an electron with spin \( \sigma \) on site \( i \) and orbital \( x(y) \); \( t_i, i = 1, \ldots, A \) are the hopping amplitudes between \( d_{xz} \) and \( d_{yz} \) orbitals defined as \( x \) and \( y \), respectively; parameters \( U \) and \( V \) describe the Coulomb interaction within and between orbitals, respectively; \( J \) is the exchange integral; \( \mu \) is the chemical potential.

Experimental studies of iron-based HTSC, as well as band-structure calculations within the two-orbital model (see, e.g., Ref. 6 and references therein) give different possible sets of parameters of the Hamiltonian which allow to describe qualitatively the observed electronic properties of these compounds. The realistic parameters of hopping amplitudes were chosen in the present work (hereinafter all energy quantities are measured in 0.2 eV units):

\[ t_1 = 0.058; t_2 = 0.22; t_3 = -0.0208; t_4 = -0.079. \]  

The relationship between the parameters of interaction in most calculations was determined as

\[ V = 0.5U; J = 0.25U. \]  

3. Method

In this paper, the quantum world-line MC algorithm was modified so that the calculation of the Hamiltonian of general form

\[ H = \sum_{kln} {N_{n_1}^{n_2}} (A_{k_1}^{n_1} A_{k_2}^{n_2} + h.c.) + \sum_{kn} U_{k_1}^{n_1} N_{k_2}^{n_2} \]

\[ + \sum_{kln} B_{k_1}^{n_1} N_{k_2}^{n_2} (A_{k_1}^{n_1} + h.c.) + \sum_{kn} C_{k_1}^{n_1} N_{k_2}^{n_2} + \sum_{kln} D_{k_1}^{n_1} N_{k_2}^{n_2} (A_{k_1}^{n_1} + h.c.) \]

becomes possible. Here \( A_{k_1}^{n_1} \) are off-diagonal terms of transitions between states \( n_1 \) and \( n_2 \) on site \( k \); \( N_{k}^{n} \) is the particle number operator of a state \( n \) on site \( k \); \( t, B, C, D \) are the corresponding amplitudes of interactions. The two-orbital model enables the description in the framework of the specified general Hamiltonian [24].

In order for the analysis of the off-diagonal long-range order can be made, the pair correlation functions for different types of symmetry of the pairing of charge carriers should be calculated,

\[ Q_\delta = \sum_{ij} \int_0^\beta dt \langle \Delta_\delta (i, t) \Delta_\delta^+ (j, 0) \rangle. \]  

Here \( \delta \) denotes the pairing channel corresponding to the type of symmetry of the order parameter. According to the classification [6, 16], the correlator \( Q_2 \) is responsible for \( A_{1g} \) pairing, and \( Q_9 \) for \( B_{2g} \) pairing. The data on small clusters obtained by ED technique [6] showed that these types of symmetry appear to be the main ones for FeAs clusters; similar symmetry types were considered in the simplified \( S_4 \) model [18]. The corresponding order parameters are given by

\[ \Delta_\delta^0 (i) = \frac{1}{\sqrt{N}} \sum_{\alpha \mu} (a_{i,\alpha,\mu,\alpha,\mu}^+ a_{i,\alpha,\mu,\alpha,\mu} + a_{i,\alpha,\mu,\alpha,\mu}^+ a_{i,\alpha,\mu,\alpha,\mu}); \]

\[ \Delta_\delta^\pm (i) = \frac{1}{\sqrt{N}} \sum_{\alpha \mu} (a_{i,-\alpha,\mu,\alpha,\mu}^+ a_{i,\alpha,\mu,\alpha,\mu} - a_{i,\alpha,\mu,\alpha,\mu}^+ a_{i,-\alpha,\mu,\alpha,\mu}); \]

where \( N \) is the number of sites (Fe atoms), \( \alpha \) denotes the orbitals \( x \) and \( y \), and \( \mu \) here is a unit vector to one of four neighbor sites of the site \( i \).

For extracting the anomalous terms of the type \( \langle aa \rangle \) and \( \langle a^+ a^+ \rangle \) (5) has to be expanded according to Wick’s theorem. Each four-fermion term can be expressed in the following way [26]:

\[ \langle T_\tau a_{i\alpha} a_{i\alpha}^+ (\tau) a_{j\beta} a_{j\beta}^+ (0) a_{k\beta}^+ a_{k\beta} (0) a_{l\mu}^+ a_{l\mu} (0) \rangle \rightarrow \]

\[ \langle T_\tau a_{i\alpha} a_{i\alpha}^+ (\tau) a_{i\mu}^+ a_{i\mu}^+ (0) \rangle \langle T_\tau a_{j\beta} a_{j\beta}^+ (0) \rangle - \langle T_\tau a_{i\alpha} a_{i\alpha}^+ (\tau) a_{k\beta}^+ a_{k\beta}^+ (0) \rangle \langle T_\tau a_{j\beta} a_{j\beta}^+ (0) \rangle \langle T_\tau a_{l\mu} a_{l\mu}^+ (0) \rangle. \]
As a result, we have the following sequence of pairwise products of normal terms (Green's functions):

\[
G_2 = \frac{1}{N} \sum_{ij \alpha \beta} \int_0^\beta d\tau \left( \langle a_{j+\alpha \beta, \sigma}^+ (\tau) a_{i+\mu \alpha, \sigma} (0) \rangle - \langle a_{j+\alpha \beta, \sigma} (\tau) a_{i+\mu \alpha, \sigma}^+ (0) \rangle - \langle a_{j+\alpha \beta, \sigma} (\tau) a_{i+\mu \alpha, -\sigma}^+ (0) \rangle + \langle a_{j+\alpha \beta, -\sigma} (\tau) a_{i+\mu \alpha, \sigma}^+ (0) \rangle \right) - \\
G_9 = \frac{1}{N} \sum_{ij \alpha \beta} \int_0^\beta d\tau \left( \langle a_{j+\alpha \beta, \sigma} (\tau) a_{i+\mu \alpha, \sigma}^+ (0) \rangle - \langle a_{j+\alpha \beta, \sigma} (\tau) a_{i+\mu \alpha, -\sigma}^+ (0) \rangle - \langle a_{j+\alpha \beta, -\sigma} (\tau) a_{i+\mu \alpha, \sigma}^+ (0) \rangle + \langle a_{j+\alpha \beta, -\sigma} (\tau) a_{i+\mu \alpha, -\sigma}^+ (0) \rangle \right)
\]

where \( i \) and \( j \) denote the sites of the lattice (Fe atoms), \( \alpha \) and \( \beta \) denote the orbitals \( x \) and \( y \), \( \mu \) and \( \nu \) are unit vectors to one of neighbor sites of the site \( i \) and \( j \), respectively, \( \sigma = \uparrow, \downarrow \) is spin \( z \)-axis projection, \( s = 1 \) if \( \sigma = \uparrow \) and \( s = -1 \) otherwise. In this case the differences \( Q_2 - G_2 \) and \( Q_9 - G_9 \) give information about the anomalous terms \( \langle a_{i\alpha \beta}^+ a_{i\alpha \beta}^+ \rangle \) and about possible pairing of charge carriers in the given symmetry channel of the order parameter.

4. Correlation functions

Pair correlation functions (5) and normal terms (9, 10) were calculated for FeAs clusters of size \( 4 \times 4 \), \( 6 \times 6 \), \( 8 \times 8 \), and \( 10 \times 10 \) within the framework of the full two-orbital model (1) by the quantum MC method developed in section 3 in the temperature range \( \beta = 5 \div 20 \) and the range of the interaction \( U = 2 \div 16 \) while maintaining the relation (3). Results are presented for the half filling.

![Figure 1](image)

**Figure 1.** The dependence of the correlation functions \( Q_2 \) and \( Q_9 \), normal terms \( G_2 \) and \( G_9 \), and their differences on the interaction parameter for \( 8 \times 8 \) cluster, \( \beta = 5 \).

Figure 1 shows the dependence of the correlation functions \( Q_2 \) and \( Q_9 \), normal terms \( G_2 \) and \( G_9 \), and the difference between these values on the interaction parameter for \( 8 \times 8 \) cluster. The first thing to note is that in all our calculations \( G_2 \) and \( G_9 \) coincided within the error of the calculation which was \( \sim 10^{-2} \div 10^{-3} \) for all data. Hence, the normal terms (9, 10) were insensitive to the type of symmetry. Further, the differences \( Q_2 - G_2 \), \( Q_9 - G_9 \) points up the presence of anomalous terms, i.e. the pairing of charge carriers at \( U < 8 \) for both types of symmetries is possible. For larger values of the interaction the pair correlations were not observed.

The dependence of the same correlation functions on temperature is shown in figure 2 for the largest calculated cluster of size \( 10 \times 10 \) and two values of interaction \( U = 4; 8 \). The differences in
the behavior of the pair correlations are evident. At \( U = 4 \) correlations \( Q_2 \) take place right up to the smallest temperature \( \beta = 20 \), and the symmetry channel \( Q_9 \) disappears at \( \beta > 10 \). At larger value \( U = 8 \) pair correlations continuously decrease at \( \beta > 5 \).

**Figure 2.** The dependence of the correlation functions \( Q_2 \) and \( Q_9 \), normal terms \( G_2 \) and \( G_9 \), and their differences on temperature for \( 10 \times 10 \) cluster. (a) \( U = 4 \); (b) \( U = 8 \).

Let us consider now the dependence of the pair correlations on the system size. The results for the lowest temperature \( \beta = 20 \) are presented in figure 3. The scaling of the pair correlation functions shows pronounced \( A_{1g} \)-type correlations at \( U = 4 \) for clusters of size \( 8 \times 8 \) and larger; they weakly depend on the cluster size. The data indicate also the disappearance of \( B_{2g} \)-type correlations with increasing of system size. The same way as in the case of higher temperature, at greater interaction \( U = 8 \) the differences \( Q_2 - G_2 \) and \( Q_9 - G_9 \) are less than zero for both types of symmetries and clusters of any size.

**Figure 3.** The dependence of the correlation functions \( Q_2 \) and \( Q_9 \), normal terms \( G_2 \) and \( G_9 \), and their differences on the system size. (a) \( U = 4 \); (b) \( U = 8 \). \( \beta = 20 \).

Thus, the whole set of obtained data provides the evidence for the possibility of charge carriers pairing at the chosen values of the hopping amplitudes (2) and the interaction parameter \( U < 8 \) (with relations (3) between the \( U, V, \) and \( J \)). The type of realized symmetry is \( A_{1g} \). The results indicate also that the normal terms \( G_2 \) and \( G_9 \) depend on the interaction strength and temperature and almost linearly increase with \( U, \beta \) (for strong interactions), while the dependence of the correlation functions
on these parameters is much weaker. The increase of the normal terms leads to effective suppression of pair correlations with increasing interaction strength and inverse temperature at $U \geq 8$. For $6 \times 6$ cluster the values of correlation parameters reach saturation and slightly change in larger systems.

For the analysis of the relation between pair correlations and magnetic ordering, as the authors of [18], we investigated the magnetic correlations using diagonal part (along the $z$-axis) at zero frequency of the spin susceptibility

$$
\chi(q) = \int_0^\beta d\tau \sum_{\alpha\alpha'} \sum_{ij} e^{iq(t-j)} m_{\alpha\alpha}(\tau) m_{j\alpha'}(0); \quad m_{\alpha\alpha} = \frac{1}{2} (n_{\alpha\alpha} - n_{\alpha\alpha}).
$$

The data obtained show a clear trend for the displacement of the peak point from $(\pi, \pi)$ to $(\pi, 0)$ with increasing of the interaction. Thus, in the region of small $U$ where according to our data pair correlations are observed, a specific antiferromagnetic ordering emerges (spin density wave), which was also noted in [6, 14, 15, 18, 20].

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