RAPID COMMUNICATION

Variational Monte Carlo study of the nematic state in iron-pnictide superconductors with a five-orbital model*

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(Received 5 November 2014; revised manuscript received 10 November 2014; published online 25 November 2014)

We perform a variational Monte Carlo study of the nematic state in iron-pnictide superconductors within a realistic five-orbital model. Our numerical results show that the nematic state, formed by introducing an anisotropic hopping order into the projected wave function, is not stable unless the off-site Coulomb interaction V exceeds a critical value. This demonstrates that V plays a key role in forming the nematic state in iron-pnictide superconductors. In the nematic state, the orbital order and the anisotropic spin correlations are consistent with the experimental observations. We argue that the experimentally observed anisotropic magnetic couplings and structural transition are associated with the nematic state and can be understood in a unified framework.

Keywords: iron-pnictide superconductors, nematic state, off-site Coulomb interaction, variational Monte Carlo simulation

PACS: 74.70.Xa, 71.10.Fd, 74.25.Jb **DOI:** 10.1088/1674-1056/24/1/017404

1. Introduction

Understanding the properties of normal state is believed to be key to unveiling the pairing mechanism in high-T_c superconductors. In both high-T_c cuprates and iron-pnictides, superconductivity emerges when the magnetic ordering is suppressed by additional charge doping or by an applied pressure.[1–3] This phenomenon has stimulated considerable studies on the magnetic property and its relation to superconductivity. Recently, another intriguing property of iron-pnictides was observed in the high temperature regime, namely the electronic nematic phase, in which the discrete lattice rotational symmetry is broken while the translational symmetry is retained. Experiments showed that iron-pnictides suffer a structural transition[4–8] at certain temperature T_S, across which the crystal symmetry changes from tetragonal to orthorhombic. Although the lattice deformation is very small ($(a-b)/(a+b) < 0.01$, with a and b denoting the in-plane lattice constants), sizeable electronic anisotropy has been observed in the in-plane resistivity measurements.[9,10] This so-called nematic character has also been identified in the optical conductivity spectrum,[11–13] scanning tunnelling microscopy,[14,15] and angle-resolved photoemission studies[16] at a temperature even above T_S.

Currently, the nature of the nematic state as well as its relation to anisotropic magnetic couplings[17] and the structural transition is a hot topic. Based on a spin-nematic scenario,[18–20] anisotropic spin fluctuations above T_S have been taken as the driving force for the nematic state, and they induce the orbital polarization and drive a structural transition at a lower temperature T_S. On the other hand, in an orbital order scenario,[21–24] a spontaneous breaking of the degeneracy between $3d_{xz}$ and $3d_{yz}$ orbitals is considered to be responsible for the nematic instability. The resulting unequal orbital occupations lead to anisotropic magnetic couplings and trigger a magnetic transition at a lower temperature T_N. While quite a number of experimental phenomena have been explained by either of the two scenarios,[20,25] some important issues still need to be addressed. For instance, in the orbital order scenario, both calculated anisotropy in Drude weight[22] and anisotropic orbital occupations[26] showed the opposite sign to the experimental observations. Therefore, a unified physical picture is needed to clarify these anisotropic properties in the nematic state.

In our previous variational Monte Carlo (VMC) calculations,[27] we demonstrated that the nematic state can arise in a three-orbital model when a realistic off-site Coulomb interaction V is considered, which is the first time that the role of V in the formation of nematic state in iron-pnictides is identified. Here, we extend our work to a more realistic five-orbital model.[28] In this five-orbital model, two iron 3d e_g orbitals

*Project supported by the National Natural Science Foundation of China (Grant Nos. 11274310, 11474287, 11174072, and 91221103) and the Specialized Research Fund for the Doctoral Program of Higher Education of China (Grant No. 20104208110001).

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are included in addition to the three iron 3d t_{2g} orbitals. Although these two e_g orbitals do not contribute significantly to the density of states close to the Fermi level, their strong hybridizations with the three t_{2g} orbitals give a better fitting to the bands obtained by the density functional theory. Moreover, in this five-orbital model, the π-bond like nearest-neighbor (NN) hopping is much stronger than the σ-bond one, leading to distinct NN hoppings between five- and three-orbital models (see Fig. 3). This is consistent with the first-principles Wannier function analysis[21] and we will show that it is important for providing a unified physical picture in which the nematic state, the orbital order, and the anisotropic magnetic couplings are all associated.

The rest of the paper is organized as follows. In Section 2, the model and the variational Monte Carlo method are introduced. In Section 3, we present the main results and discuss the relations between the orbital, spin, and structure degrees of freedom. Conclusions of this paper are given in Section 4.

2. Model and method

We start from the following five-orbital model on a two-dimensional square lattice:

$$\begin{aligned} H &= H_{\text{band}} + U_1 \sum_{i\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} \\ &+ \sum_{i,\alpha<\beta,\sigma,\sigma'} \left[(U_2 - J\delta_{\sigma\sigma'}) n_{i\alpha\sigma} n_{i\beta\sigma'} \right] \\ &+ J \sum_{i,\alpha<\beta} \left(c^{\dagger}_{i\alpha\uparrow} c^{\dagger}_{i\beta\downarrow} c_{i\alpha\downarrow} c_{i\beta\uparrow} + c^{\dagger}_{i\alpha\uparrow} c^{\dagger}_{i\alpha\downarrow} c_{i\beta\downarrow} c_{i\beta\uparrow} + \text{H.c.} \right) \\ &+ V \sum_{\langle ij \rangle} n_i n_j . \end{aligned} \tag{1}$$

The first term of the Hamiltonian reads

$$H_{\text{band}} = \sum_{ij,\alpha\beta,\sigma} t_{\alpha\beta}[x_j - x_i, y_j - y_i] c^{\dagger}_{i\alpha\sigma} c_{j\beta\sigma} . \tag{2}$$

The hopping integrals used here are from the Graser *et al.* paper,[28] in which the orbital basis is aligned parallel to the NN Fe–Fe direction rather than the Fe–As direction; this is convenient for investigating the nematic state in which the two orthogonal Fe–Fe directions are non-equivalent. Another important feature is that the σ-bond like NN d_{xz}–d_{xz} hopping along the x direction is much weaker than the π-bond like one along the y direction, e.g., $|t_{xz,xz}[1,0]| \ll |t_{xz,xz}[0,1]|$, and for the d_{yz} orbital. This is due to the strong deformation of the orbitals induced by the hybridization between Fe 3d and As 4p orbitals.[21]

The interaction part of the model includes the intraorbital and interorbital Coulomb interactions U_1 and U_2, the Hund coupling J, as well as the Coulomb interaction V between the NN sites. The interacting parameters are $U_1 = 2.0$, $U_2 = 1.0$, and $J = 0.5$ in units of eV in this paper unless otherwise stated, which correspond to the typical values of iron-pnictide superconductors.[29–31] In our previous work, we demonstrated that the nematic state is not stable unless V is larger than a critical value $V_c = 0.33$.[27] In this paper, we vary V from 0.0 to 0.4.

We describe the nematic state by introducing an anisotropic hopping order (AHO) with order parameter δ_{var}. This kind of introducing the nematic state has successfully captured the nature of the nematic state in high-T_c cuprates.[32,33] A non-interacting variational Hamiltonian H_{MF} is then obtained by substituting the hopping integrals $t_{\alpha\beta}[\Delta x, 0]$ and $t_{\alpha\beta}[0, \Delta y]$ in H_{band} by the effective hopping integrals

$$\begin{aligned} t^{\text{eff}}_{\alpha\beta,\Delta_x} &= (1 + \delta_{\text{var}}) t_{\alpha\beta}[\Delta x, 0] , \\ t^{\text{eff}}_{\alpha\beta,\Delta_y} &= (1 - \delta_{\text{var}}) t_{\alpha\beta}[0, \Delta y] . \end{aligned} \tag{3}$$

The wave function $|\psi_{\text{MF}}\rangle$ is obtained by diagonalizing the quadratic Hamiltonian H_{MF}. The projected wave function we use in VMC is then constructed as follows:

$$|\psi\rangle = P_{\text{G}} |\psi_{\text{MF}}\rangle = g_1^{\hat{N}_1} g_2^{\hat{N}_2} g_V^{\hat{N}_V} g_J^{\hat{N}_J} |\psi_{\text{MF}}\rangle , \tag{4}$$

where

$$\begin{aligned} \hat{N}_1 &= \sum_{i,\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow}, \quad \hat{N}_2 = \sum_{i,\alpha<\beta} n_{i\alpha} n_{i\beta}, \\ \hat{N}_V &= \sum_{\langle ij \rangle} n_i n_j, \quad \hat{N}_J = \sum_{i,\alpha<\beta,\sigma} n_{i\alpha\sigma} n_{i\beta\sigma}. \end{aligned} \tag{5}$$

Here g_1, g_2, and g_J are the variational parameters controlling the number of electrons residing in the same and the different on-site orbitals, and g_V controls the number of electrons on the NN sites. It is well-known that the ground state of iron-pnictides is an antiferromagnetic (AFM) state at low energy, while in this paper, we will focus on the nematic state in the high temperature paramagnetic regime. We are interested in the microscopic mechanism of the unusual anisotropy in the tetragonal crystal structure.

The VMC simulations are carried out on 10×10 and 14×14 lattices with periodic boundary conditions along the x and the y directions. We define N as the number of sites and n as the average number of electrons per site. In this paper, we investigate the undoped case with $n = 6$. The ground-state energy $\langle\psi|H|\psi\rangle$ is calculated using a standard Markovian chain Monte Carlo approach with the Metropolis update algorithm,[34–37] and it is optimized with respect to the variational parameters. During the optimization, a quasi-Newton method combined with a fixed sampling method[38,39] is used. In the figures presented below, the statistical errors are smaller than the symbol sizes unless otherwise stated.

3. Results and discussion

Firstly, the condensation energy per unit cell $E_{\rm cond} = \left[E\left(\delta_{\rm var}^{\rm opt}\right) - E(0)\right]/N$ and the optimized AHO parameter $\delta_{\rm var}^{\rm opt}$ as a function of V are presented in Fig. 1. One can see that $E_{\rm cond}$ and $\delta_{\rm var}^{\rm opt}$ are zero when V is less than a critical value $V_{\rm c} = 0.30$ in the 10×10 lattice (0.32 in the 14×14 lattice). For the three-orbital model, the critical value $V_{\rm c}$ is estimated to be about 0.33,[27] which is close to the value obtained for the five-orbital model. These results provide conclusive evidence that the off-site Coulomb interaction V plays a key role in forming the nematic state in iron-pnictides. The error bar (not shown) in Fig. 1(b) is about 0.05, which is due to the finite size effect, manifested as a discontinuous change of the Fermi surface when the variational parameter $\delta_{\rm var}$ changes continuously.[27]

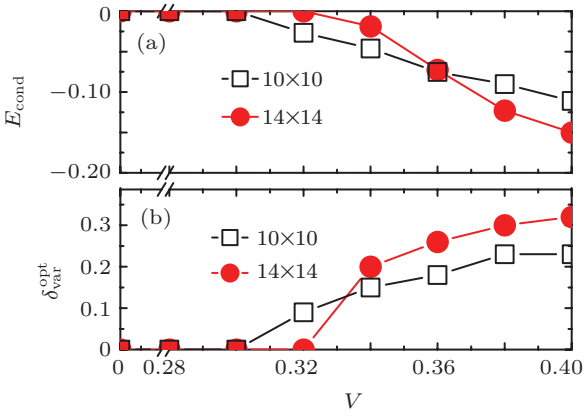


Fig. 1. (color online) (a) Condensation energy $E_{\rm cond}$ and (b) optimized $\delta_{\rm var}$ as a function of V.

Due to the development of AHO, the effective hopping integrals $t^{\rm eff}_{\alpha\beta,\Delta_x}$ and $t^{\rm eff}_{\alpha\beta,\Delta_y}$ defined in Eq. (3) become anisotropic. As a result, the kinetic energies along the x and the y directions, which are defined as

$$E_{\rm kin}x = \left\langle \sum_{i\alpha\beta\sigma,\Delta x} t_{\alpha\beta}\left[\Delta x, 0\right] c^{\dagger}_{i,\alpha\sigma} c_{i+\Delta x,\beta\sigma} \right\rangle, \quad (6)$$

$$E_{\rm kin}y = \left\langle \sum_{i\alpha\beta\sigma,\Delta y} t_{\alpha\beta}\left[0, \Delta y\right] c^{\dagger}_{i,\alpha\sigma} c_{i+\Delta y,\beta\sigma} \right\rangle, \quad (7)$$

become anisotropic, as shown in Fig. 2(a). The decrease (increase) of $E_{\rm kin}x$ ($E_{\rm kin}y$) as a function of $\delta_{\rm var}$ will result in a stronger anisotropy of the kinetic energy as the nematicity is enhanced. This behavior is very similar to the one observed in the three-orbital model.[27] As already discussed in our previous work,[27] the derived optical conductivity and Drude weight contain the terms that are closely related to the hopping integrals and the hopping processes of electrons. The anisotropic kinetic energy, which is obtained from the kinetic (hopping) terms in Eq. (2), suggests that the calculated x and y components of the optical conductivity and the Drude weight in the nematic phase are unequal, resulting in anisotropic optical and dc conductivities as observed in $Ba(Fe_{1-x}Co_x)_2As$.[13]

The anisotropic effective hopping integrals also lead to the anisotropy of electron occupations in the d_{xz} and d_{yz} orbitals, as shown in Fig. 2(b). While the d_{yz} orbital occupation n_{yz} decreases and approaches half-filling, the d_{xz} orbital occupation n_{xz} increases monotonically as $\delta_{\rm var}$ is increased. This kind of anisotropic electron occupation is opposite to the one in the three-orbital model.[27] As shown in Fig. 3(a), the leading d_{yz}–d_{yz} and d_{xz}–d_{xz} hopping integrals in the five-orbital model are along the x and y directions, respectively, while the two leading hopping integrals are reversed in the three-orbital model (see Fig. 3(b)). In the nematic state with $\left|t^{\rm eff}_{\alpha\beta,\Delta_x}\right| > \left|t^{\rm eff}_{\alpha\beta,\Delta_y}\right|$, $n_{xz} > n_{yz}$ or $n_{xz} < n_{yz}$ is developed to benefit the kinetic energy in the five- or three-orbital model due to the different leading hopping processes. The orbital order obtained in this work is in agreement with the experiment,[16] where it was found that as the temperature is lowered toward $T_{\rm N}$, the shift up and shift down of the d_{yz} and d_{xz} bands lead to larger occupation of the d_{xz} orbital.

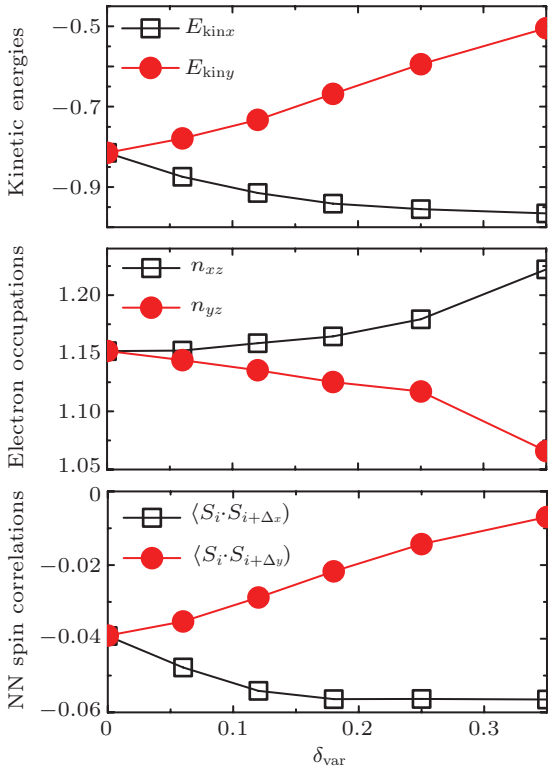


Fig. 2. (color online) (a) Kinetic energies, (b) electron occupations in the d_{xz} and d_{yz} orbitals, and (c) NN spin correlations as a function of $\delta_{\rm var}$ at $V = 0.36$ on the 14×14 lattice.

The physical picture of anisotropic NN magnetic couplings[17] driven by the orbital order has already been proposed by Lee *et al.*[21] In their picture, the d_{xz} orbital is almost doubly-occupied, while the d_{yz} orbital is almost singly-occupied and spin-polarized. As the d_{yz}–d_{yz} hopping along the x direction is much stronger than that along the y direction,

anisotropic spin superexchanges with $J_x > J_y$ are naturally obtained. Our results shown in Figs. 2(b) and 3(a) are in qualitative agreement with Lee *et al.*'s analysis, therefore anisotropic NN magnetic couplings can be realized in the nematic state, which leads to the anisotropic magnetic properties shown in Fig. 2(c). One can see that in the normal state ($\delta_{\rm var} = 0$), the NN spin correlations along the two directions have the same negative value. When the nematicity develops ($\delta_{\rm var} > 0$), the spin correlation along the y direction approaches 0, while the one along the x direction is enhanced.

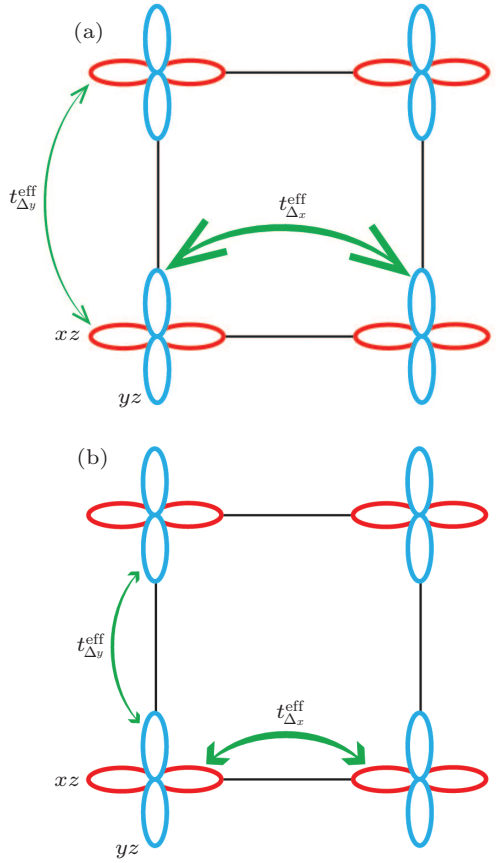


Fig. 3. (color online) The leading hopping integrals along the x and y directions in (a) the five-orbital model and (b) the three-orbital model.

Having identified the microscopic origin of the nematic state and the induced orbital order, we now turn to the instability of the structural transition with $a > b$, which was observed by experiments.[4–6] As shown in Fig. 4(a), the shape of the d_{xz} orbital combined with the dominant d_{xz} electron occupation prefers a lattice distortion with $a > b$, since the repulsive energy caused by the NN d_{xz}–d_{xz} off-site Coulomb interaction can be dramatically reduced. On the other hand, for dominant d_{yz} electron occupation, a lattice distortion with $a < b$ is preferred, as displayed in Fig. 4(b). In the nematic state, we have unequal electron occupations with $n_{xz} > n_{yz}$ as shown in Fig. 2(b), so the experimentally observed lattice distortion with $a > b$ is obtained. From this physical picture, one can see that the off-site Coulomb interaction V not only plays a key role in the formation of the nematic state, but also contributes to the structural transition, across which the crystal symmetry changes from tetragonal to orthorhombic.

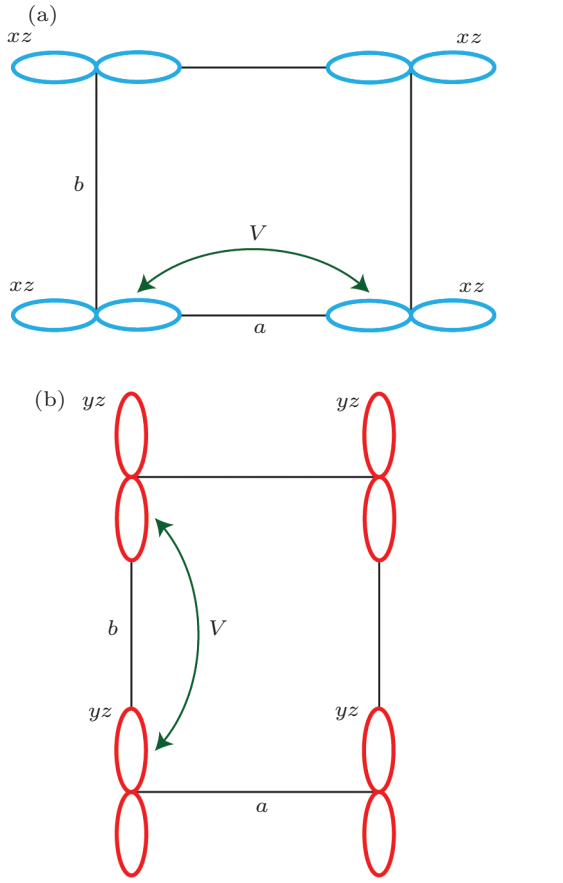


Fig. 4. (color online) Schematic diagram of the contributions of the orbital order to the structural distortion: (a) dominant d_{xz} occupation prefers a structural distortion with $a > b$; (b) dominant d_{yz} occupation prefers $b > a$.

4. Conclusion

Our VMC study demonstrates that the nematic state is stable in the realistic five-orbital model when the off-site Coulomb interaction V is larger than a critical value $V_{\rm c} \approx 0.30$. Miyake *et al.*'s *ab initio* calculations[40] indicated that the typical value of V is about $0.50 \sim 0.90$ in iron-pnictides, which is much larger than $V_{\rm c}$, therefore, the nematic state can be realized in actual materials. The development of nematicity results in unequal electron occupations with $n_{xz} > n_{yz}$ and anisotropic magnetic couplings with $J_x > J_y$, which are consistent with the ones observed in experiments. The resulting anisotropic kinetic energies are closely related to the experimentally observed anisotropic optical and dc conductivities. In addition, we find that the inclusion of the off-site Coulomb interaction V is crucial for the structural distortion with $a > b$.

In our work, the nematic state is introduced by AHO, and the orbital order and the anisotropic spin correlations are the consequents of the development of AHO. Thus, we do not disentangle the roles of orbital degree of freedom and spin degree of freedom in the formation of the nematic state. Further

works that directly describe the nematic state in the form of orbital order or spin order are needed. The question of which one is the dominant degree of freedom will be answered when comparing the calculated results to the experimental measurements.

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