Multiorbital Effects on Antiferromagnetism in Fe Pnictides

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We apply a Hartree-Fock approximation to a two-orbital model proposed for Fe pnictide superconductors. It is found that the antiferromagnetic (AFM) order with the ordering vector $Q = (\pi, 0)$ is realized. The AFM order appears simultaneously with ferro-orbital order, the latter leads to a secondary lattice distortion. We also investigate the influence of doping on the AFM order. The size of the AFM moment changes continuously for lightly doped cases, but when the amount of doped carriers exceeds a certain value the AFM state is suddenly destroyed. We also show that Fermi surfaces remain and change significantly on doping even in the AFM state. This behaviour is explained by considering the nesting due to the multi-sheet Fermi-surface structure and multiorbital nature of the electronic bands characteristic to Fe pnictides.

KEYWORDS: iron pnictides, magnetic order, orbital order, lattice distortion, Fermi surface, superconductivity

Since the discovery of superconductivity in LaFeAsO$_{1-x}$F$_x$ with a high transition temperature $T_c = 26$ K\textsuperscript{1} extensive studies have been done on Fe pnictides. The main interests on these materials are not only on the high transition temperatures such as $T_c = 55$ K in SmFeAsO$_{1-x}$F$_x$\textsuperscript{2} and $T_c = 56$ K in Gd$_{1-x}$Th$_x$FeAsO\textsuperscript{3} but also on the mechanism of the superconductivity. The electronic structure is quasi-two-dimensional\textsuperscript{4–9} and superconductivity occurs around the magnetic phase boundaries\textsuperscript{1,10–13} as in high-$T_c$ cuprates. Such similarities suggest that magnetism is probably playing an important role in the emergence of superconductivity, and it is highly desirable to unveil the microscopic origin of magnetism characteristic to Fe pnictides.

The magnetism in Fe pnictides is much different from that in cuprates. In the latter, ordering vector of the antiferromagnetism is $(\pi, \pi)$, while it is $(\pi, 0)$ in Fe pnictides in the unfolded Brillouin zone (BZ) with one Fe ion per unit cell.\textsuperscript{14–17} The undoped antiferromagnetic (AFM) states are metallic\textsuperscript{10–12,18–20} in Fe pnictides while insulating in cuprates. The AFM transition occurs at\textsuperscript{15–17,21,22} or near\textsuperscript{14} the structural transition temperature in Fe pnictides.

Such differences in magnetism may originate from the multiorbital electronic states and multi-sheet Fermi-surface (FS) structure in Fe pnictides.\textsuperscript{4–9} Indeed, the AFM order with $(\pi, 0)$ due to nesting between hole and electron pockets [see Fig. 1(a)] has been suggested by using tight-binding models\textsuperscript{23–25} and by band-structure calculations.\textsuperscript{6,7} Yildirim\textsuperscript{26} has shown that the tetragonal lattice distortion occurs in AFM state with $(\pi, 0)$, but not in the normal state. In addition, the lattice distortion occurs neither in an AFM state with $(\pi, \pi)$ nor in a ferromagnetic state. Ran \textit{et al.}\textsuperscript{27} have shown that a full band gap does not open and the system remains metallic even in the AFM state from a topological view point of the multiorbital system.

Fig. 1. (Color online) (a) Fermi surfaces of the two-orbital model\textsuperscript{24} in the unfolded BZ at electron number $n = 2$ per site. The arrows indicate the nesting vector $Q = (\pi, 0)$. The hole surfaces locate around $(0, 0)$ and $(\pi, \pi)$, and the electron surfaces locate around $(\pi, 0)$ and $(0, \pi)$. (b) Schematic views of the orbital states in the normal state (left) and the AFM state accompanying ferro-orbital order (right). The orbital stretched along $x$ ($y$) axis represents the $d_{zx}$ ($d_{yz}$) orbital. The arrows represent the spin state, and the sizes of the orbitals indicate the occupancies of those orbitals.

In this Letter, we show that such characteristic features on magnetism are explained even in the simplest model, i.e., by a two-orbital model, proposed for Fe pnictides\textsuperscript{24,25} by applying Hartree-Fock approximation. While the Hartree-Fock approximation has already been applied to the two-orbital model,\textsuperscript{28} possibility of orbital order and effects of doping have not been investigated yet. They are main topics of the present study. The two-

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orbital model cannot reproduce well the band structure obtained with the density functional theory, while a five-orbital model does well.\textsuperscript{23} In addition, \( d_{xy} \) orbital, which is not included in the two-orbital model, contributes some portions of the FSs.\textsuperscript{29} Thus, to describe some properties, all the five \( d \)-orbitals may be necessary. However, the two-orbital model can reproduce at least the characteristic FS topology in Fe pnictides in the folded BZ, and is enough for the purpose of the present Letter. In this study, we take into consideration orbital order on an equal footing with AFM order, since they are closely related to each other in Fe pnictides. Indeed, the AFM order with the ordering vector \( \mathbf{Q} = (\pi, 0) \) inevitably accompanies ferro-orbital (FO) order [schematically shown in Fig. 1(b)] which results in a secondary orthorhombic distortion. In addition, we investigate doping effects on the antiferromagnetism and FSs reconstructed by the AFM order. As shown in Fig. 1(a), there are two kinds of nesting with the same nesting vector \( \mathbf{Q} = (\pi, 0) \), i.e., between FSs around \((0, 0)\) and \((\pi, 0)\), and between FSs around \((0, \pi)\) and \((\pi, \pi)\). The existence of the two kinds of nesting is important for stabilization of the AFM state against doping. We find that the structure of FSs changes significantly with doping even in the ordered state due to the multi-sheet FS nesting.

In the two-orbital model, we consider a square lattice of Fe ions with \( d_{zx} \) and \( d_{yz} \) orbitals.\textsuperscript{24,25} The model Hamiltonian is given by

\[
H = \epsilon_i \sum_{\sigma} n_{i\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + U' \sum_{i} n_{i\uparrow} n_{i\downarrow} + J \sum_{i,\sigma} c_{i\sigma}^\dagger c_{i\sigma}^\dagger c_{i\sigma} c_{i\sigma},
\]

where \( c_{i\sigma} \) is the annihilation operator of the electron at site \( i \) with orbital \( \tau \) and spin \( \sigma \) (=\( \uparrow \) or \( \downarrow \)) and \( \epsilon_{k\tau\sigma} \) is the Fourier transform of \( c_{i\tau\sigma} \). \( \tau \) is \( x \) and \( y \) represent \( d_{zx} \) and \( d_{yz} \) orbitals, respectively. \( n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma} \) and \( n_{i\tau} = \sum_{\sigma} n_{i\sigma} \). The coupling constants \( U \), \( U' \), \( J \), and \( J' \) denote the intraorbital Coulomb, interorbital Coulomb, exchange, and pair-hopping interactions, respectively. For the \( t_{2g} \) orbitals, relations \( U = U' + J + J' \) and \( J = J' \) hold\textsuperscript{30} and we use them. For the kinetic energy term, we use the hopping parameters proposed by Raghu et al.,\textsuperscript{24} \( \epsilon_{k\tau x} = -2t_1 \cos k_x - 2t_2 \cos k_y - 4t_3 \cos k_x \cos k_y \), \( \epsilon_{k\tau y} = -2t_2 \cos k_z - 2t_1 \cos k_y - 4t_3 \cos k_x \cos k_y \), and \( \epsilon_{k\tau y} = -4t_4 \sin k_z \sin k_y \), where \( t_1 = -t_2 = 1.3t \), \( t_3 = t_4 = -0.85t \), and we have set the lattice constant unity.

In this study, we consider weakly correlated cases, e.g., \( U/W \simeq 0.29 \) for \( U/t = 3.5 \), where \( W = 12t \) is the bandwidth. Thus, it is reasonable to apply a Hartree-Fock approximation. We assume that the expectation value of the number \( n_{i\tau\sigma} \) is given by the following form:

\[
\langle n_{i\tau\sigma} \rangle = \left[ n_{\sigma} + m_{s} (\delta_{\sigma\uparrow} - \delta_{\sigma\downarrow}) + m_{o} (\delta_{\tau x} - \delta_{\tau y}) \right] + m_{so} (\delta_{\sigma\uparrow} - \delta_{\sigma\downarrow})(\delta_{\tau x} - \delta_{\tau y})
\]

\[
+ n_{q} + m_{sq} (\delta_{\sigma\uparrow} - \delta_{\sigma\downarrow}) + m_{soq} (\delta_{\tau x} - \delta_{\tau y})
\]

\[
+ m_{soq} (\delta_{\sigma\uparrow} - \delta_{\sigma\downarrow})(\delta_{\tau x} - \delta_{\tau y})[\mathbf{q} \cdot \mathbf{r}_i]/4,
\]

where \( \mathbf{q} = (\pi, \pi) \) or \( (\pi, 0) \equiv \mathbf{Q} \), \( \mathbf{r}_i \) denotes the position of site \( i \), and \( n \) is the number of electrons per site. The order parameters are \( m_{s}, m_{o}, m_{so}, m_{sq}, m_{soq}, \) and \( m_{soq} \). We determine the lowest energy state among the solutions of the Hartree-Fock approximation. In Eq. (2), we consider the \( z \) component for the orbital state, i.e., \( m_{q} = \frac{1}{N} \sum_{i,\tau,\sigma} \langle c_{i\tau\sigma}^\dagger c_{i\tau\sigma} \rangle \) with \( \alpha = z \), where \( \hat{\tau}^z \) is the Pauli matrix. We also considered order parameters with \( \alpha = x \) and \( y \), and we found that the \( z \)-component ordered state with \( q = (\pi, \pi) \) always has lower energy than the other ordered states within parameters we investigate here.

Figure 2(a) shows \( U \) dependence of the order parameters \( m_{o} \) (for FO order), \( m_{s} \) (for AFM order), and \( m_{so} \) (for antiferro-spin-orbital order) at \( n = 2 \) and \( J = 0.1U \). We find that the other order parameters are zero. As shown in the inset, \( m_{s} \) jumps to a finite value at \( U/t \simeq 2.97 \), and \( m_{o} \) and \( m_{so} \) also have jumps to finite values at the same point while they are small and not visible on the scale of Fig. 2(a). Thus, the transition to the AFM state is of first order.
Figure 2(b) shows doping dependence of the order parameters at $U/t = 3.5$ and $J = 0.1U$. We have chosen this value of $U$ so as to the AFM state is destabilized by the doping of $\sim 0.1$ as in experimental observations. The AFM moment changes continuously with doping at first, but suddenly disappears at $n \simeq 1.92$ and 2.1. We obtained small values of $m_{\bf Q}$, which are much smaller than the saturation value $n$ (for $n \leq 2$) or $4 - n$ (for $n > 2$), as in experimental observations ($m_{\bf Q} = 1$ is corresponding to $1 \mu_B$ of an AFM moment): $0.25 \mu_B$, $0.35 \mu_B$, or $0.36 \mu_B$ in LaFeAsO; $0.94 \mu_B$ or $1.01 \mu_B$ in SrFe$_2$As$_2$; $0.4 \mu_B$ or $0.87 \mu_B$ in BaFe$_2$As$_2$. The estimated values of the ordered moments depend on the experimental probes even for the same material, and the reason of this discrepancy is not clear at present. In the electron doped case, the AFM moment increases with doping, and the transition temperature is expected to become higher than the undoped case. It is in contradiction to experimental observations, and to resolve this discrepancy, we have to extend the model, e.g., by using the five-orbital basis.

Note that in the AFM state with $Q = (\pi, 0)$, $x$ and $y$ directions are not equivalent, and the occupancies of $d_{xz}$ and $d_{yz}$ become different. Thus, the AFM state in the multiorbital system inevitably accompanies FO order, i.e., finite $m_{\bf Q}$. Through an electron-lattice interaction, the FO order results in a lattice distortion from a tetragonal to orthorhombic structure. This is consistent with experimental observations that the AFM phase is always orthorhombic. The obtained small values of the order parameter $m_{\bf Q}$ for the FO order may be responsible for the weakness of anomaly in lattice distortion, e.g., small volume change in SrFe$_2$As$_2$ at the transition. Note that another scenario is proposed for the lattice distortion, in which the lattice distortion relaxes magnetic frustration and is necessary for occurrence of the AFM order.

On the contrary, in our theory, the lattice distortion is a secondary effect due to the FO order accompanied by the AFM order. Since the AFM state in Fe pnictides is metallic, we believe that our picture is more suitable for Fe pnictides. In the coexistent state of antiferromagnetism and FO order, $m_{\bf Q}$ also becomes finite as shown in Fig. 2. Note that we obtain similar results for $J = 0$ at least in a small-$U$ region and the choice of the value of $J$ does not change the present results qualitatively. In this model, at zero temperature, the AFM order and the FO order disappear at the same doping and it is consistent with the experimental observations. However, some extensions, e.g., inclusion of electron-lattice interaction, may be necessary to obtain different transition temperatures for the AFM order and for the FO order as in LaFeAsO.

To obtain further insights into the ordered states, we show FSs in Fig. 3 in the normal and ordered states at half-filling ($n = 2$) and at around phase boundaries. It is evident from the figure that the $x$ and $y$ directions are not equivalent in the ordered states. In the hole doped case, $n = 1.93$, nesting between FSs centered at $k = (0, 0)$ and $(\pi, 0)$ in the unfolded BZ is strong. Then, these FSs are reconstructed into small pockets around $(\pm \pi/4, 0)$, while the other FSs centered at $(0, \pi)$ and $(\pi, \pi)$ are almost unchanged. On the other hand, in the electron doped case, $n = 2.07$, nesting is strong between FSs centered at $k = (0, \pi)$ and $(\pi, \pi)$. Around zero doping, both types of nesting can contribute to stabilize the AFM state. As a result, the doping effect on the AFM moment is not so significant in the lightly doped cases, while the structure of the FSs changes very much. Thus, the multi-sheet FS nesting is important for the stabilization of the AFM state in this system. When we dope carriers further, the nesting becomes weak and the AFM state is destabilized suddenly.

Note that such a mechanism to stabilize the AFM state against doping is applicable as long as the sizes of the hole surfaces around $(0, 0)$ and $(\pi, \pi)$ are different and the undoped system is a compensated or nearly compensated metal. The smaller hole surface mainly contributes for the realization of antiferromagnetism for the hole doped case, and the larger hole surface mainly contributes for the electron doped case. Thus, this mechanism works irrespective of precise choice of the model parameters. We also note that in a more realistic model, i.e., five-orbital model, both the two hole surfaces locate around $(0, 0)$ even in the unfolded BZ. However, the nesting vector $Q = (\pi, 0)$ is the same as in the two-orbital model, and the present mechanism to stabilize the AFM state is applicable provided the two hole surfaces have different sizes.

At $n = 2$ only small pockets of FSs remain in the ordered states. The area of one pocket at $n = 2$ is 0.86% of the folded BZ for $Q = (\pi, 0)$ and of the normal state BZ folded due to the actual lattice structures of Fe pnictides.
(two Fe ions per unit cell). There are two electron pockets and two hole pockets, but the areas of them are the same, since the model is a compensated metal at \( n = 2 \) and the two electron (hole) pockets occupy the same amount of area due to symmetry. Experimentally observed volumes of FSs in the AFM state are small: 0.26%-1.38% in \( \text{SrFe}_2\text{As}_2 \) and 0.3%-1.7% in \( \text{BaFe}_2\text{As}_2 \) of the normal state folded BZ. These values are comparable with our theoretical ones. In the normal state at \( n = 2 \), the hole pocket around \((0, 0)\), hole pocket around \((\pi, \pi)\), and electron pocket around \((\pi, 0)\) occupy 7.13%, 13.24%, and 10.18% of the folded BZ, respectively. These values are also comparable with experimental ones, 2.8%-9% of BZ in \( \text{LaFePO} \) in the normal state.

Figure 4 shows the band structure and density of states in the normal and ordered states. In the ordered state, band gaps open at some points at the Fermi level, but not at \((\sim \pi/4, 0)\) and \((\sim \pi/4, \pi)\). At \( k_y = 0 \) and \( \pi \), the off-diagonal element \( \epsilon_{kxy} \) in the kinetic energy term is zero, and \( d_{xz} \) and \( d_{yz} \) orbitals do not mix. The mean field in the ordered state mixes electrons with \( k \) and \( k + Q \) in the same orbitals, and \( d_{xz} \)- and \( d_{yz} \)-orbital states are not mixed even in the ordered state at \( k_y = 0 \) and \( \pi \). The two bands crossing at around the Fermi level are different orbitals at both \((\sim \pi/4, 0)\) and \((\sim \pi/4, \pi)\), and a gap cannot open there. As a result, the FSs do not disappear on the lines \( k_y = 0 \) and \( \pi \) even in the AFM state as shown in Fig. 3. Note that in Fig. 3 the Fermi pockets at \( n = 2.07 \) on \( k_y = \pi \) are very small in the ordered state but have finite volumes. Thus, the system remains metallic in the AFM state as in experimental observations. The density of states in the ordered state has a gap-like structure around the Fermi level, but remains finite at the Fermi level. For larger \( U \) cases, the band structure changes very much, and the system can become insulating. By increasing \( U \), the band crossing points on \((0, 0)-(\pi/2, 0)\) and \((0, \pi)-(\pi/2, \pi)\) lines move to \((\pi/2, 0)\) and \((\pi/2, \pi)\), respectively, and finally the band crossing disappears. Then, the system can become insulating, e.g., at \( U/t \gtrsim 6.7 \) for \( n = 2 \) and \( J = 0.1U \). Thus, \( U \) should not be very large in Fe pnictides.

In conclusion, we have shown that characteristic features of the AFM state in Fe pnictides can be naturally understood within the two-orbital model. The stability of AFM phase is due to the multi-sheet FS nesting. The tetragonal to orthorhombic lattice distortion is a secondary effect due to the FO order but not a driving mechanism of antiferromagnetism. FSs remain in the ordered state due to the multiorbital character of the crossing bands. Our theory indicates that the FSs change significantly upon doping. In the doped AFM states around phase boundaries, some Fermi pockets become very small, while the other Fermi pockets have large volumes as in the normal state. Experimental observations of these FSs are highly desired, since we can know what kind of nesting is strong around the phase boundaries from the reconstructed FSs. Such a knowledge is important to unveil fluctuations which mediate the superconducting pairing.

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