Orbital Order, Structural Transition and Superconductivity in Iron Pnictides

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We investigate the 16-band $d$-$p$ model for iron pnictide superconductors in the presence of the electron-phonon coupling $g$ with the orthorhombic mode which is crucial for reproducing the recently observed ultrasonic softening. Within the RPA, we obtain the ferro-orbital order below $T_Q$ which induces the tetragonal-orthorhombic structural transition at $T_\text{c}$, together with the stripe-type antiferromagnetic (AFM) transition at $T_N$. The carrier doping $x$ suppresses both of the transition temperatures $T_\text{c}$ and $T_N$ and induces the superconductivity. In RFePnO$_{1-x}$F$_x$ (R=Rare Earth, Pn=As, P) with a transition temperature $T_\text{c}$ exceeding 50K, they attracted much attention. The parent compounds with the transition temperatures $T_\text{s}$ and $T_N$ are considered to rapidly decrease with the nonmagnetic impurities. The small $T_\text{c}$ suppression against nonmagnetic impurities is not consistent with the experiments. As the elastic constant $C_{ij}$ with the orthorhombic mode which is crucial for reproducing the recently observed ultrasonic softening. Within the RPA, we obtain the ferro-orbital order below $T_Q$ which induces the tetragonal-orthorhombic structural transition at $T_\text{c}$, together with the stripe-type antiferromagnetic (AFM) transition at $T_N$. The carrier doping $x$ suppresses both of the transition temperatures $T_\text{c}$ and $T_N$. The parent compounds with the transition temperatures $T_\text{s}$ and $T_N$ are considered to rapidly decrease with the nonmagnetic impurities. Therefore, the $s$-wave state without sign change of the order parameter, so called $s_{\pm}$-wave, mediated by the orbital fluctuation which is enhanced due to the effects of the inter-orbital Coulomb interaction was proposed on the basis of the one-dimensional two-band Hubbard model and the two-dimensional 16-band $d$-$p$ model.

Remarkably, drastic softenings of the elastic constants have been observed in recent ultrasonic experiments. As the elastic constant $C_{ij}$ is given by the second derivative of the total energy w.r.t. the strain field $\varepsilon$, the strain field $\varepsilon$ and includes the contribution such as $-g_{ij}^{\text{el}}\chi_\eta$ with the susceptibility $\chi_\eta$ for the electric operator $\eta$ linearly coupled with the strain field as $g_{ij}^{\text{el}}\varepsilon$, the enhancement of $\chi_\eta$ is responsible for the softening of $C_{ij}$. The detailed ultrasonic measurements revealed that nondoped and underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ shows drastic softenings of the elastic constants with decreasing $T$ down to $T_\text{s}$.

for $C_E = (C_{11} - C_{12})/2$, $C_{44}$ and $C_{66}$ modes relevant to the strain fields $\varepsilon_{xx} - \varepsilon_{yy}$, $\varepsilon_{yz}$ ($\varepsilon_{zx}$) and $\varepsilon_{xy}$ shown in Fig. 1 (a), (b) and (c) which are linearly coupled with the orbital fluctuations shown in Figs. 1 (d), (e) and (f), respectively, where $x'$, $y'$ ($x$, $y$) axes are directed along the nearest (second nearest) Fe-Fe bonds.

Fig. 1. (Color online) Schematic figures of the strain fields for $C_E$, $C_{44}$ and $C_{66}$ modes (a), (b) and (c), the orbital fluctuations coupled with the corresponding strain fields (d), (e) and (f), and the phonons for $B_{1g}$, $E_g$ and orthorhombic modes which enhance the corresponding orbital fluctuations (g), (h) and (i), respectively. The $x'$, $y'$ ($x$, $y$) axes are directed along the nearest (second nearest) Fe-Fe bonds.

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transition at $T_a \sim T_Q$. In addition, optimally doped BaFe$_{1.84}$Co$_{1.6}$As$_2$ shows a significant softening of $C_{66}$ mode down to $T_c$. Therefore, the orbital order and its fluctuations relevant to $C_{66}$ mode are considered to play crucial roles in both the structural transition and the superconductivity.

The orbital fluctuation is known to be enhanced by the electron-phonon interaction in addition to the inter-orbital Coulomb interaction. Recently, the effects of the electron-phonon interaction with $B_{1g}$ and $E_g$ modes on the orbital fluctuation and its induced $s_{+ \pm}$-wave superconductivity have been investigated on the basis of the 5-band Hubbard model$^{17}$ and the 16-band $d$-$p$ model.$^{18}$ As shown in Fig. 1, the $B_{1g}$ phonon enhances the longitudinal $d_{yz}$-$d_{xz}$ and transverse $d_{3z^2-r^2}$-$d_{x^2-y^2}$ orbital fluctuations responsible for the softening of $E_g$ mode, while the $E_g$ phonon enhances the transverse $d_{x^2-y^2}$-$d_{yz}$, $d_{xy}$-$d_{xz}$ and $d_{3z^2-r^2}$-$d_{yg}$ orbital fluctuations responsible for the softening of $C_{4u}$ mode. However, the effect of the orthorhombic mode which enhances the longitudinal $d_{y'z'}$-$d_{z'x'}$ and transverse $d_{3z^2-r^2}$-$d_{xy}$ orbital fluctuations responsible for the most dominant softening of $C_{66}$ mode was not considered there.$^{17,18}$ The present paper is a straightforward extension of our previous work$^{18}$ to include the orthorhombic mode which enable us to reproduce the ultrasonic softening of $C_{66}$ and to obtain the $x$-$T$ phase diagram including the tetragonal-orthorhombic structural transition and the superconductivity.

Our Hamiltonian of the two-dimensional 16-band $d$-$p$ Holstein model, in which 3$d$ orbitals ($d_{3z^2-r^2}$, $d_{x^2-y^2}$, $d_{xy}$, $d_{yz}$, $d_{xz}$) of two Fe atoms (Fe$^1$-$A$, Fe$^2$-$B$) and 4$p$ orbitals ($p_x$, $p_y$, $p_z$) of two As atoms are explicitly included, is given by$^{18}$

$$H = H_0 + H_{\text{int}} + H_{\phi} + H_{\text{el-ph}} - T_c \rho \frac{\lambda_0}{\nu_m},$$

(1)

where $H_0$, $H_{\text{int}}$, $H_{\phi}$ and $H_{\text{el-ph}}$ are the kinetic, Coulomb interaction, phonon and electron-phonon interaction parts of the Hamiltonian, respectively. The kinetic part of the Hamiltonian $H_0$ includes the atomic energies and the transfer integrals which are determined so as to fit both the energy and the weights of orbitals for each band obtained from the tight-binding approximation to those from the density functional calculation for LaFeAsO and are listed in ref. 13. In the present model, the doping concentration $x$ corresponds to the number of electrons per unit cell $n = 24 + 2x$ and there are two hole FSs (FS1 and FS2) around the $\Gamma$ point and two electron FSs (FS3 and FS4) around the $M$ point for $x = 0.1$. The Coulomb interaction part $H_{\text{int}}$ includes the multi-orbital interaction on a Fe site: the intra- and inter-orbital direct terms $U$ and $U'$, Hund’s rule coupling $J$ and the pair transfer $J'$. For simplicity, we assume the relation $U = U' + 2J$ and $J = J'$ throughout this paper. Hereafter, we number the Fe-3$d$ orbitals as follows: $d_{3z^2-r^2}(1)$, $d_{x^2-y^2}(2)$, $d_{xy}(3)$, $d_{yz}(4)$, $d_{xz}(5)$.

Now we consider the effect of the phonon and the electron-phonon interaction parts of the Hamiltonian $H_{\phi}$ and $H_{\text{el-ph}}$ which includes the phonon energy $\omega_s$ and the electron-phonon coupling constant $g_{\ell\ell'}^s$ between the orbital $\ell$ and $\ell'$ (see Fig. 2 (a)), respectively, where $s$ represents the phonon mode. In the present paper, we consider the $B_{1g}$, $E_g$ and orthorhombic modes as shown in Figs. 1 (g), (h) and (i). We note that the orthorhombic mode is not a normal coordinate but a general coordinate which is given by a linear combination of normal coordinates including both optical and acoustic modes. To avoid the difficulty with many phonon modes, we treat the orthorhombic mode as a local phonon similar to the $B_{1g}$ and $E_g$ modes as a simplest first step in including the orthorhombic mode. As following refs. 17 and 18, we take the electron-phonon coupling into account as the atomic energy variance of the Fe-3$d$ electrons. The resulting electron-phonon coupling matrix elements are given as follows: $\sqrt{3}g_{35}^{g_{25}} = g_{45}^{g_{25}} = -\sqrt{3}g_{45}^{g_{12}} = g_{24}^{g_{12}}$, $g_{35}^{g_{12}} = g_{45}^{g_{12}}$, $g_{45}^{g_{12}} = -\sqrt{3}g_{45}^{g_{12}} = g_{24}^{g_{12}}$, $g_{35}^{g_{12}} = g_{45}^{g_{12}} = 0$ and 0 for otherwise, where $E_g$ and $E_g$ correspond to the oscillation of the Fe atom along the $x$- and $y$-axis, respectively, and $\theta$ denotes the orthorhombic mode (see Fig. 1).

Within the RPA,$^{19}$ the spin susceptibility $\chi^s(q)$ and the charge-orbital susceptibility $\chi^c(q)$ are given in the $50 \times 50$ matrix representation as follows$^{6,13,18}$

$$\chi^s(q) = [1 - \chi^{(0)}(q)\Sigma^{-1}\chi^{(0)}(q)],$$

(2)

$$\chi^c(q) = [1 + \chi^{(0)}(q)\Sigma^{-1}\chi^{(0)}(q)],$$

(3)

with the noninteracting susceptibility, $\chi^{(0)}(q)$ is given by

$$\chi^{(0)}(q) = \left\langle 0 \left| \sum_k \hat{G}_{t'\ell'1t\ell}^{\alpha\beta}(k) \hat{G}_{t\ell1t'\ell'}^{\beta\alpha}(k + q) \right| 0 \right\rangle,$$

(4)

where $\alpha, \beta = (A, B)$ represent two Fe sites, $\ell$ represents Fe 3$d$ orbitals, $\hat{G}(k) = [(i\epsilon_n + \mu) - H_0(k)]^{-1}$ is the noninteracting Fe-3$d$ electron Green’s function in the 10$\times$10 matrix representation, $\mu$ is the chemical potential, $H_0(k)$ is the kinetic part of the Hamiltonian with the momentum $k$ given in eq. (1), $k = (k, i\epsilon_n)$, $q = (q, i\nu_m)$ and $\epsilon_n = (2n + 1)\pi T$ and $\nu_m = 2m\pi T$ are the fermionic and bosonic Matsubara frequencies, respectively. It is noted that when the largest eigenvalue $\lambda_{\text{spin}}(\lambda_{\text{charge}})$ of $\chi^{(0)}(q)\bar{S}(-\chi^{(0)}(q)\bar{C})$ reaches unity, the magnetic (charge- orbital) instability occurs. In eqs. (2) and (3), bare vertices for the spin and charge- orbital susceptibilities $\bar{S}$ and $\bar{C}$ are given by$^{17,18}$

$$\langle \bar{S} \rangle = \left\langle \frac{\lambda_0}{\nu_m} \sum_k \bar{G}_{t'\ell'1t\ell}^{\alpha\beta}(k) \bar{G}_{t\ell1t'\ell'}^{\beta\alpha}(k + q) \right\rangle,$$

(5)

$$\langle \bar{C} \rangle = \left\langle \frac{\lambda_0}{\nu_m} \sum_k \bar{G}_{t'\ell'1t\ell}^{\alpha\beta}(k) \bar{G}_{t\ell1t'\ell'}^{\beta\alpha}(k + q) \right\rangle,$$

(6)

where $\bar{D}_s(i\nu_m) = 2\omega_s/(\nu_m^2 + \omega_s^2)$ is the local phonon Green’s function for the mode $s$ (see Fig. 2 (b)). The bare vertices due to the Coulomb interaction $\bar{U}(s \alpha)$ are given by $\bar{U}(\bar{S}(\alpha \ell))_{t'\ell, t\ell}^{\alpha\beta} = U \bar{U}(\bar{U}(s \alpha))_{t'\ell, t\ell}^{\alpha\beta} = U(2U' - J)$ and $\bar{U}(s \alpha)^{\alpha\beta}_{t'\ell, t\ell} = J' \left( J' \right)$, where $\ell \neq \ell'$ and the other...
Fig. 3. (Color online) Several components of the charge-orbital susceptibility $\chi_c(q,0)$ for $U' = 1.0$, $J = 0.2$ and $g = 0.065$ at $x = 0.1$ and $T = 0.036$, where we number the Fe-3d orbitals as follows: $d_{x^2-y^2}(1)$, $d_{xy}(2)$, $d_{yz}(3)$, $d_{yz}(4)$, $d_{x^2-y^2}(5)$.

The linearized Eliashberg equation is solved to obtain the gap function $\Delta(k)$ with the eigenvalue $\lambda_{sc}$. At $T = T_c$, the largest eigenvalue $\lambda_{sc}$ becomes unity. We use $32 \times 32 k$ point meshes and 512 Matsubara frequencies ($-511\pi T \leq \varepsilon_n \leq 511\pi T$) in the numerical calculations for eqs. (2)-(5). For simplicity, we set $\omega_{B_{1g}} = \omega_{E_g} = \omega_{E_2g} = \omega_0 = 0.02\text{eV}$ as done in the previous study. To reproduce the experimental results that the elastic softening is the largest for the $C_{66}$ mode, we assume $g_{B_{1g}} = g_{E_g} = 0.85g_\theta$ and put $g_\theta = g$. Here and hereafter, we measure the energy in units of eV.

Fig. 3 shows several components of the static charge-orbital susceptibility $\chi_c(q,0)$ for $U' = 1.0$, $J = 0.2$ and $g = 0.065$ at $x = 0.1$ and $T = 0.036$. In this case, the dimensionless electron-phonon coupling parameter is given by $\lambda = 2g^2\rho_0/\omega_0 \sim 2g^2/\omega_0 = 0.42$ with the density of states at the Fermi level $\rho_0 \sim 1\text{eV}$. We find that, when $T$ decreases, the transverse $d_{yz} - d_{xz}$ orbital susceptibility $[\chi_c(q,0)]_{45,45}^{A,A}$, which is equivalent to the longitudinal $d_{yz} - d_{xz}$ one, is most enhanced as compared to the other orbital and magnetic susceptibilities (not shown) due to the cooperative effects of the electron-phonon interaction with the orthorhombic mode and the inter-orbital Coulomb interaction $U'$. We note that the incommensurate peaks around $q = (0,0)$ largely depend on the electron-phonon coupling strength $g_{E_g}^\alpha$ and move to the commensurate peak at $q = (0,0)$ for a slightly different parameter set where the resulting pairing state and the phase diagram discussed below is essentially unchanged.

In Figs. 4(a)-(d), we show several components of the gap function $\Delta(k,\pi T)$ for $U' = 1.0$, $J = 0.2$ and $g = 0.065$ at $x = 0.1$ and $T = 0.036$ (a)-(d), and those for $U' = 1.48$, $J = 0.2$ and $g = 0.032$ at $x = 0.1$ and $T = 0.034$ (e)-(h).
and the AFM fluctuations on the superconductivity do not compete to each other as they are mainly responsible for the different \( q \) regions in \( V(q) \), in contrast to the case with the antiferro-orbital and the AFM fluctuations.

Fig. 5(a) shows the phase diagram on the \( x-T \) plane in the case with a large \( g \), \( U' = 1.0 \), \( J = 0.2 \) and \( g = 0.065 \). When \( T \) decreases, the orbital susceptibilities \( \chi^c(q, 0)|_{x, y} \) and \( \chi^c(q, 0)|_{x, z} \) with \( q \sim (0, 0) \) (see Fig. 3(c)) diverge at a critical temperature \( T_Q \). Below \( T_Q \), the ferro-orbital order with different occupations of the \( d_{y'z} \) and \( d_{zx'} \) orbitals occurs and induces the orthorhombic distortion resulting in the tetragonal-orthorhombic structural transition at \( T_s = T_Q \). When approaching \( T_Q \), the ferro-orbital fluctuation is largely enhanced and mediates the \( s_{++} \)-wave superconductivity (see Figs. 4(a)-(d)). We also investigate the same model within the Hartree-Fock approximation\(^{20}\) and obtain the phase diagram consistent with RFePnO\(_{1-x}\)F\(_x\) where the \( T_s = T_Q \) is always higher than that of \( T_Q \) and/or \( T_N \), where the orbital and/or the magnetic fluctuations diverge. With including the effects of the self-energy correction and the vertex correction neglected in the RPA, it is expected that the ferro-orbital order and/or the antiferromagnetic orders are realized for relatively large \( x \), while the superconductivity is realized for relatively large \( x \). The explicit calculation including such effects is a future problem.

The obtained \( s_{++} \)-wave superconductivity seems to be consistent with experimental results of iron pnictides including the impurity effects. The enhanced ferro-orbital fluctuation above \( T_Q \) might be observed by experiments with a kind of external field inducing the anisotropy of \( x' \), \( y' \) axes, similar to the case with the ferromagnetic fluctuation above the Curie temperature observed by experiments with the external magnetic field. In fact, a resistivity anisotropy for \( T > T_s \) is induced by uniaxial stress.\(^{22}\) In the present paper, we treated the orthorhombic mode as a optical phonon, as a simplest first step. More realistic model including acoustic phonons together with a suitable parameter set of the electron-phonon coupling strengths \( g_{\nu \xi}^{\nu} \) will be discussed in a subsequent paper.
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