Two types of $s$-wave pairing due to magnetic and orbital fluctuations in the two-dimensional 16-band $d$-$p$ model for iron-based superconductors

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We study superconductivity in the two-dimensional 16-band $d$-$p$ model extracted from a tight-binding fit to the band structure of LaFeAsO, using the random phase approximation. When the intraorbital repulsion $U$ is larger than the interorbital one $U'$, an extended $s$-wave ($s_d$-wave) pairing with sign reversal of order parameter is mediated by antiferromagnetic spin fluctuations, while when $U < U'$ another kind of $s$-wave ($s_{++}$-wave) pairing without sign reversal is mediated by ferro-orbital fluctuations. The $s_{++}$-wave pairing is enhanced due to the electron-phonon coupling and then can be expanded over the realistic parameter region with $U > U'$.

I. INTRODUCTION

The recently discovered iron-based superconductors1,2 RFePnO$_{1-x}$F$_x$ (R=Rare Earth, Pn=As, P) with a transition temperature $T_c$ exceeding 50K have attracted much attention. The F nondoped compound LaFeAsO exhibits the structural transition from tetragonal (P4/nmm) to orthorhombic (Cmca) phase at a transition temperature $T = 155K$ and stripe-type antiferromagnetic order at $T = 134K$ with a magnetic moment $\sim 0.36\mu_B$ at low temperature. With increasing F doping, the system becomes metallic and the antiferromagnetic order disappears, and then, the superconductivity emerges for $x \sim 0.1$ with $T_c \sim 26K$. Rare-earth substitution compounds exhibit superconducting transition with higher $T_c$. Specific features of the systems are twodimensionality of the conducting Fe$_2$As$_2$ plane and the orbital degrees of freedom in Fe$^{2+}$ (3d$^8$)1,2. The pairing symmetry together with the mechanism of the superconductivity is one of the most significant issues.

The NMR Knight shift measurements revealed that the superconductivity of the systems is the spin-singlet pairing. Fully gapped superconducting states have been predicted by various experiments such as the penetration depth, the specific heat, the angle resolved photoemission spectroscopy (ARPES) and the impurity effect on $T_c$. In contrast to the above mentioned experiments, the NMR relaxation rate shows the power low behavior $1/T_1 \propto T^6$ below $T_c$, suggesting the nodal or highly anisotropic gap structure. The other NMR measurements, however, revealed $1/T_1 \propto T^3$ below $T_c$ and there is still controversy.

Theoretically, the first principle calculations have predicted that the nondoped system is metallic with two or three concentric hole Fermi surfaces around the $\Gamma$ point $(k = (0,0))$ and two elliptical electron Fermi surfaces around the $M$ point $(k = (\pi, \pi))$. Mazin et al. suggested that the spin-singlet extended $s$-wave pairing whose order parameter changes its sign between the hole pockets and the electron pockets ($s_d$-wave) is favored due to the antiferromagnetic spin fluctuations. According to the weak coupling approaches based on multi-orbital Hubbard models, the $s_{++}$-wave pairing or the $d_{xy}$-wave pairing is expected to emerge. It is shown that the $s_{++}$-wave pairing is realized also in the strong coupling region by the mean field study based on the $t$-$J_1$-$J_2$ model and the exact diagonalization study based on the one-dimensional two-band Hubbard model.

Generally speaking, the details of the band structure and the Fermi surface are crucial for determining the pairing symmetry. In the 5-band Hubbard model originally introduced by Kuroki et al., the energy bands obtained by reproduce those obtained by the density functional calculation very well. In this model, however, the spatial extensions of the Fe 3d like Wannier orbitals are different from each other and the resulting intra-orbital terms of the on-site Coulomb interaction are strongly orbital dependent. In addition, since the model explicitly includes the transfer integrals up to the fifth nearest neighbor sites, one should take the off-site Coulomb interaction, which is considered to be about 0.5eV between the nearest neighbor sites, into account to ensure the consistency of the model. On the other hand, in the effective model which includes both the Fe 3d orbitals and the As 4p orbitals, so called $d$-$p$ model, the spatial extensions and the differences of those between the orbitals will be considerably reduced. Due to these facts, in the $d$-$p$ model, it is expected that the intra-orbital terms of the on-site Coulomb interaction for each orbitals have almost the same values and the off-site Coulomb interaction are negligible. Therefore, theoretical studies based on the $d$-$p$ model, are highly desired.

In the previous papers, we have investigated the electronic states of the Fe$_2$As$_2$ plane in iron-based superconductors on the basis of the two-dimensional 16-band $d$-$p$ model which includes the Coulomb interaction on a Fe site: the intra- and inter-orbital direct terms $U$ and $U'$, the Hund’s coupling $J$ and the pair-transfer $J'$. Using the random phase approximation (RPA), we have found that, for a larger value of $J$, the most favorable pairing symmetry is $s_{++}$-wave, while, for a smaller value of $J$, it is $d_{xy}$-wave.

The present paper is a full paper to our previous papers with some numerical improvements. In the
present paper, we investigate the superconductivity in the wider parameter space by treating $U$, $U'$, $J$ and $J'$ as independent parameters in contrast to the previous study under the condition that $U = U' + 2J$ and $J = J'$ based on the two-dimensional 16-band $d$-$p$ model. Solving the superconducting gap equation with the pairing interaction obtained by using the RPA, we find that two kinds of the $s$-wave superconducting states appear. As above mentioned, the $s_{±}$-wave superconducting state emerges near the incommensurate spin density wave (ISDW) with $q \sim (\pi, \pi)$ phase. In addition, for $U < U'$, the $s$-wave superconducting state appears around the ferro-orbital ordered phase. The order parameter for this $s$-wave state does not change its sign in $k$ space. We refer to this $s$-wave state as the $s_{±}$-wave state, hereafter.

II. MODEL AND FORMULATION

First of all, we perform the density functional calculation for LaFeAsO with the generalized gradient approximation of Perdew, Burke and Ernzerhof by using the WIEN2k package, where the lattice parameters ($a = 4.03268\text{Å}$, $c = 8.74111\text{Å}$) and the internal coordinates ($z_{La} = 0.14134$, $z_{As} = 0.65166$) are experimentally determined. The crystal structure of Fe$_2$As$_2$ layer is shown in Fig. 1 (a). Since As atoms are tetrahedrally arranged around a Fe atom, there are two distinct Fe and As sites in the crystallographic unit cell (see Figs. 1 (a), (b)). Considering these facts, we then derive the two-dimensional 16-band $d$-$p$ model, where 3$d$ orbitals ($d_{3z^2-r^2}$, $d_{x^2-y^2}$, $d_{yz}$, $d_{zx}$) 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. We note that $x$, $y$ axes are directed along second nearest Fe-Fe bonds (see Fig. 1 (b)).

The total Hamiltonian of the $d$-$p$ model is given by

$$H = H_0 + H_{\text{int}},$$  \hspace{1cm} (1)

where $H_0$ and $H_{\text{int}}$ are the noninteracting and interacting parts of the Hamiltonian, respectively. The noninteracting part of the $d$-$p$ model is given by the following tight-binding Hamiltonian,

$$H_0 = \sum_{i,\ell,\sigma} \varepsilon_{\ell}^d d_{i\ell\sigma}^d d_{i\ell\bar{\sigma}}^d + \sum_{i,m,\sigma} \varepsilon_{m}^p p_{i\sigma m\sigma}^p$$

$$+ \sum_{i,j,\ell,\ell',\sigma} t_{ij,\ell,\ell'}^{dd} d_{i\ell\sigma}^d d_{j\ell'\bar{\sigma}}^d + \sum_{i,j,m,m',\sigma} t_{ij,m,m'}^{pp} p_{i\sigma m\sigma}^p p_{j\sigma m'\sigma}$$

$$+ \sum_{i,j,\ell,\ell',\sigma} t_{ij,\ell,\ell'}^{dp} d_{i\ell\sigma}^d p_{j\ell'\sigma}^m + h.c.,$$  \hspace{1cm} (2)

where $d_{i\ell\sigma}$ is the annihilation operator for Fe-3$d$ electrons with spin $\sigma$ in the orbital $\ell$ at the site $i$ and $p_{i\sigma m\sigma}$ is the annihilation operator for As-4$p$ electrons with spin $\sigma$ in the orbital $m$ at the site $i$. In eq. (2), the transfer integrals $t_{ij,\ell,\ell'}^{dd}, t_{ij,m,m'}^{pp}, t_{ij,\ell,\ell'}^{dp}$ and the atomic energies $\varepsilon_{\ell}^d, \varepsilon_{m}^p$ 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. Similar models have been used by the other authors but the model parameters are different from ours. The doping concentration $x$ corresponds to the number of electrons per unit cell $n = 24 + 2x$ in the present model.

Now we consider the effect of the Coulomb interaction on Fe site. The interacting part of the Hamiltonian is given as follows,

$$H_{\text{int}} = \frac{1}{2} U \sum_{i} \sum_{\ell \neq \bar{\ell}} \sum_{\sigma \neq \bar{\sigma}} d_{i\ell\sigma}^d d_{i\bar{\ell}\bar{\sigma}}^d d_{i\bar{\ell}\sigma}^d d_{i\ell\bar{\sigma}}^d$$

$$+ \frac{1}{2} U' \sum_{i} \sum_{\ell \neq \bar{\ell}} \sum_{\sigma, \sigma'} d_{i\ell\sigma}^d d_{i\bar{\ell}\bar{\sigma}}^d d_{i\bar{\ell}\sigma'}^d d_{i\ell\bar{\sigma'}}^d$$

$$+ \frac{1}{2} J \sum_{i} \sum_{\ell \neq \ell'} \sum_{\sigma \neq \sigma'} d_{i\ell\sigma}^d d_{i\ell'\bar{\sigma}}^d d_{i\ell'\bar{\sigma'}}^d d_{i\ell\sigma'}^d$$

$$+ \frac{1}{2} J' \sum_{i} \sum_{\ell \neq \ell'} \sum_{\sigma \neq \bar{\sigma}} d_{i\ell\sigma}^d d_{i\ell'\bar{\sigma}}^d d_{i\ell'\sigma}^d d_{i\ell\bar{\sigma}}^d,$$  \hspace{1cm} (3)

where $U$ and $U'$ are the intra- and inter-orbital direct terms, respectively, and $J$ and $J'$ are the Hund’s coupling and the pair-transfer, respectively. For the isolated atoms, the relations between Coulomb matrix elements...

FIG. 1. (Color online) Crystal structure of Fe$_2$As$_2$ layer. Small and large balls represent Fe and As atoms, respectively. The solid line represents the unit cell. It is noted that As$^1$ and As$^2$ denote the As atoms on the upper side and on the lower side of the Fe$_2$As$_2$ layer, respectively.
\[ U = U' + 2J \] and \( J = J' \) are derived due to the rotational invariance of the Coulomb interaction and the reality of the wave functions, respectively. For the atoms in the crystal, however, the relation is not satisfied generally due to the crystallographic effects and the many-body effects due to the Coulomb interaction and the electron-phonon coupling which will be discussed later. Therefore, we treat \( U, U', J \) and \( J' \) as independent parameters in the present paper.

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

\[
\chi^s(\mathbf{q}) = (\hat{1} - \chi^{(0)}(\mathbf{q})\hat{S})^{-1}\chi^{(0)}(\mathbf{q}),
\]
\[
\chi^c(\mathbf{q}) = (\hat{1} + \chi^{(0)}(\mathbf{q})\hat{C})^{-1}\chi^{(0)}(\mathbf{q})
\]

with the noninteracting susceptibility

\[
\chi^{(0)}_{\alpha,\beta}(\mathbf{q}) = -\frac{1}{N} \sum_{\mathbf{k},\mu,\nu} \sum_{\ell,\ell'} f(\varepsilon_{\mathbf{k} + \mathbf{q},\mu}) - f(\varepsilon_{\mathbf{k},\nu}) \varepsilon_{\mathbf{k} + \mathbf{q},\mu} - \varepsilon_{\mathbf{k},\nu} \\
\times u_{\ell_1,\ell_2}(\mathbf{k})^* u_{\ell,\ell'}(\mathbf{k} + \mathbf{q}) u_{\ell,\ell'}(\mathbf{k}) u_{\ell',\ell_2}(\mathbf{k} + \mathbf{q})^*,
\]

where \( \mu, \nu (=1-16) \) are band indexes, \( \alpha, \beta (=A, B) \) represent two Fe sites, \( \ell \) represents Fe 3d orbitals, \( u_{\ell_1,\ell_2}(\mathbf{k}) \) is the eigenvector which diagonalizes \( H_0 \) eq. (2), \( \varepsilon_{\mathbf{k},\mu} \) is the corresponding eigenenergy of band \( \mu \) with wave vector \( \mathbf{k} \) and \( f(\varepsilon) \) is the Fermi distribution function. In eqs. (4) and (5), the interaction matrix \( \hat{S}(\hat{C}) \) is given by

\[
\hat{S}(\hat{C}) = \begin{cases} 
U \langle U \rangle & (\alpha = \beta, \ell_1 = \ell_2 = \ell_3 = \ell_4) \\
U' \langle -U' + 2J \rangle & (\alpha = \beta, \ell_1 = \ell_2 \neq \ell_3 = \ell_4) \\
J \langle 2U' - J \rangle & (\alpha = \beta, \ell_1 = \ell_2 \neq \ell_3 \neq \ell_4) \\
J' \langle J' \rangle & (\alpha = \beta, \ell_1 = \ell_4 \neq \ell_2 = \ell_3) \\
0 & \text{(otherwise)}
\end{cases}
\]

In the weak coupling regime, the superconducting gap equation is given by

\[
\lambda \Delta_{\ell_1,\ell_2}^{\alpha,\beta}(\mathbf{k}) = \frac{1}{N} \sum_{\mathbf{k}'} \sum_{\ell_1,\ell_2,\ell_3,\ell_4} \sum_{\alpha',\beta',\mu,\nu} \\
\times f(\varepsilon_{-\mathbf{k}',\mu}) + f(\varepsilon_{\mathbf{k}',\nu}) - \frac{1}{2} V_{\ell_1,\ell_2}^{\alpha,\beta}(\mathbf{k} - \mathbf{k}') \Delta_{\ell_3,\ell_4}^{\alpha',\beta'}(\mathbf{k}') \\
\times u_{\ell_3,\ell_4}(\mathbf{k}')^* u_{\ell_1,\mu}(\mathbf{k}) u_{\ell_2,\mu}(\mathbf{k}) u_{\ell_3,\nu}(\mathbf{k}'),
\]

where \( \Delta_{\ell_1,\ell_2}^{\alpha,\beta}(\mathbf{k}) \) is the gap function and \( V_{\ell_1,\ell_2,\ell_3,\ell_4}^{\alpha,\beta}(\mathbf{q}) \) is the effective pairing interaction. Within the RPA, \( V_{\ell_1,\ell_2,\ell_3,\ell_4}^{\alpha,\beta}(\mathbf{q}) \) is given in the 50 \( \times \) 50 matrix,

\[
\hat{V}(\mathbf{q}) = \eta \left( \hat{S} \hat{\chi}^s(\mathbf{q}) \hat{S} + \frac{1}{2} \hat{S} \right) - \frac{1}{2} \left( \hat{C} \hat{\chi}^c(\mathbf{q}) \hat{C} - \frac{1}{2} \hat{C} \right)
\]

where \( \eta = \frac{3}{2} \) for the spin-singlet state and \( \eta = -1/2 \) for the spin-triplet state. The gap equation is solved to obtain the gap function \( \Delta_{\ell_1,\ell_2}^{\alpha,\beta}(\mathbf{k}) \) with the eigenvalue \( \lambda \). At \( T = T_c \), the largest eigenvalue \( \lambda \) becomes unity. In the present paper, we only focus on the case with \( x = 0.1 \), where the superconductivity is observed in the compound. For simplicity, we set \( x = 0.1 \) and \( T = 0.02 \) in the present study. We use \( 32 \times 32 \) points in the numerical calculations for eqs. (1)-(7), and also use the fast Fourier transformation (FFT) to solve the gap equation eq. (8). Here and hereafter, we measure the energy in units of eV.

### TABLE I. Tight-binding parameters (in units of eV) for the d-p Hamiltonian eq. (2). It is noted that we define the d-p hopping and the in-plane p-p hopping parameters along x-axis.

| Type          | Nearest | Next Nearest |
|---------------|---------|--------------|
| d-d hopping   |         |              |
| \( d_{3z^2-r^2} \) | -0.008  | -0.024       |
| \( d_{z^2-y^2} \) | 0.143   | -0.023       |
| \( d_{xy} \)    | 0.328   | 0.073        |
| \( d_{yz} \)    | 0.109   | -0.012       |
| \( d_{zx} \)    | 0.109   | 0.012        |
| \( d_{xy} \)    | 0.078   |              |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | -0.184 |
| \( d_{xy} \)    | 0.184   |              |
| p-p hopping    |         |              |
| \( p_x \)      | 0.650   | 0.311        |
| \( p_y \)      | 0.027   | 0.311        |
| \( p_z \)      | 0.048   | 0.389        |
| \( p_x \)      | 0.111   |              |
| \( p_y \)      | 0.297   |              |
| d-p hopping    |         |              |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | 0.646 |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | -0.291 |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | 0.276 |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | 0.563 |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | 0.694 |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | 0.319 |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | 0.783 |
| \( d_{d_{z^2-r^2}d_{z^2-r^2}} \) | 0.164 |

### III. CALCULATED RESULTS

#### A. Band Structure

We show the band structure obtained from the d-p tight-binding Hamiltonian eq. (2), where the tight-binding parameters are listed in Table I, together with that obtained from the density functional calculation in...
energy bands have very complicated structure.

The Fermi surface for the $d$-$p$ tight-binding Hamiltonian is shown in Fig. 2 (b), where we can see nearly circular hole pockets around the $\Gamma$ point and elliptical electron pockets around the $M$ point. These results are consistent with the previous first principle calculations.\cite{6, 12-23}

The density of states (DOS) obtained by the $d$-$p$ tight-binding Hamiltonian eq. (2) is shown in Fig. 2 (c). It is found that the dominant contribution near the Fermi level comes from Fe 3$d$ orbitals and the contribution of As 4$p$ orbitals is small but is not negligible. We show the partial DOS of Fe 3$d$ orbitals and that of As 4$p$ orbitals in the middle panel and the lower panel of Fig. 2 (c), respectively. The $d_{yz}$, $d_{zx}$ and $d_{z^2-y^2}$ states comprise the large part of the DOS near the Fermi level, while, the $d_{3z^2-r^2}$, $d_{xy}$ states occupy the small one and are comparable with the $p_x$, $p_y$ and $p_z$ states. The $d_{yz}$ and $d_{zx}$ states at the Fermi level are larger than the $d_{z^2-y^2}$ ones and this corresponds to the fact that the electron pockets have $d_{yz}$, $d_{zx}$ and $d_{z^2-y^2}$ orbital characters, while, the hole pockets have only $d_{yz}$ and $d_{zx}$ orbital characters. However, the $d_{z^2-y^2}$ states have large values just below the Fermi level as shown in the inset of the middle panel of Fig. 2 (c). This is due to the hole band near the $\Gamma$-point just below the Fermi level. Therefore, it is anticipated that the $d_{yz}$, $d_{zx}$ and $d_{z^2-y^2}$ orbitals play significant roles to determine the magnetic, orbital and superconducting properties.

B. RPA Results for $U > U'$

In this subsection, we concentrate our attention on the case with $U > U'$. We set the typical parameters as $U = 1.71$, $U' = 1.4$ and $J = J' = 0.1$, where the condition for the superconducting transition $\lambda = 1$ is satisfied as mentioned below (see Fig. 3 (d)).

The several components of the spin susceptibility $\chi^s(\mathbf{q})$ given in eq. (1) are plotted in Fig. 3 (a). The spin susceptibility is enhanced due to the effect of the Coulomb interaction. It is found that the most dominant component is the $d_{z^2-y^2}$ diagonal component and the incommensurate peaks around the $M$ point are observed as reflecting the nesting between the hole pockets and the electron pockets. As mentioned before, the hole band which has mainly $d_{z^2-y^2}$ orbital character exists just below ($\sim 0.01eV$) the Fermi level and contributes to the large value of the DOS (see Fig. 2 (c)). Therefore, the $d_{z^2-y^2}$ diagonal component of $\chi^s(\mathbf{q})$ becomes most dominant at finite temperature $T = 0.02eV$ ($> 0.01eV$). The result is consistent with the RPA results based on the 5-band Hubbard model.\cite{25}

The several components of the charge-orbital susceptibility $\chi^c(\mathbf{q})$ given in eq. (5) are plotted in Fig. 3 (b). In contrast to the case with the spin susceptibility, the off-diagonal component of $d_{z^2-y^2} - d_{yz}$ which corresponds to the transverse orbital susceptibility becomes most dominant and shows peaks around the $M$ point together with
finally becomes unity at a critical value $U$ at which the superconducting state is realized. For $J = J' = 0.1$ eV, it is noted that we number the orbitals as follows: $d_{3z^2-r^2}$, $d_{xz}$, $d_{yz}$, and $d_{xy}$. (d) $U$ dependence of the eigenvalues of the gap equation for several symmetries.

In contrast to the case with the spin susceptibility, the dependence of the eigenvalues of the gap equation on $U$ is for $U < U'$ the spin fluctuations dominate over the charge-orbital fluctuations as shown in Figs. 3 (a) and (b).

The several components of the effective pairing interaction $V(q)$ for the spin-singlet state given in eq. (6) are plotted in Fig. 3 (c). Since the largest eigenvalue $\lambda$ is always spin-singlet state in the present study, we show the effective pairing interaction only for the spin-singlet state. Since the case for $U = 1.71$, $U' = 1.4$ and $J = J' = 0.1$, the spin fluctuations dominate over the orbital fluctuations as mentioned above, the structures of $V(q)$ are similar to those of the spin susceptibility.

Substituting $V(q)$ into the gap equation eq. (3), we obtain the gap function $\Delta(k)$ with the eigenvalue $\lambda$. In Fig. 3 (d), the eigenvalues $\lambda$ for various pairing symmetries are plotted as functions of $U$ for fixed values of $U'$, $J$, $J'$. With increasing $U$, $\lambda$ monotonically increases and finally becomes unity at a critical value $U_c$ above which the superconducting state is realized. For $U' = 1.4$ and $J = J' = 0.1$ the largest eigenvalue $\lambda$ is for the $s$-wave symmetry and $U_c = 1.71$. The second largest eigenvalue is for $d_{xy}$-wave symmetry and the eigenvalue for the $d_{xy}$-wave symmetry increases as $J$ increases for $U > U'$.

### C. RPA Results for $U < U'$

In this subsection, we concentrate our attention on the case with $U < U'$. We set the typical parameters as $U = 0.4$, $U' = 1.15$ and $J = J' = 0.1$, where the condition for the superconducting transition $\lambda = 1$ is satisfied as mentioned below (see Fig. 4 (d)).

The several components of the spin susceptibility $\chi_s(q)$ given in eq. (4) are plotted in Fig. 4 (a). In contrast to the case with $U > U'$ (see Fig. 3 (a)), the off-diagonal element $d_{x^2-y^2} - d_{yz}$ is most dominant owing to the inter-orbital direct term $U' > U$.

The several components of the charge-orbital susceptibility $\chi_c(q)$ given in eq. (5) are plotted in Fig. 4 (b). In contrast to the case with the spin susceptibility, the diagonal component of $d_{x^2-y^2}$ becomes most dominant and shows peaks around the $\Gamma$ point. It is noted that for $U < U'$ the charge-orbital fluctuations, which corresponds to the fluctuations near the ferro-orbital ordered
state realized in the large $U'$ regime as mentioned later (see Fig. 8), dominate over the spin fluctuations as shown in Figs. 4 (a) and (b).

The several components of the effective pairing interaction $\hat{V}(\mathbf{q})$ for the spin-singlet state given in eq. (8) are plotted in Fig. 4 (c). Since for $U = 0.4$, $U' = 1.15$ and $J = J' = 0.1$, the charge-orbital fluctuations are larger than the spin fluctuations, the diagonal components of $\hat{V}(\mathbf{q})$ are always negative in $\mathbf{q}$ space.

Substituting $\hat{V}(\mathbf{q})$ into the gap equation eq. (8), we obtain the gap function $\Delta(\mathbf{k})$ with the eigenvalue $\lambda$. In Fig. 4 (d), the eigenvalues $\lambda$ for various pairing symmetries are plotted as functions of $U'$ for fixed values of $U$, $J$ and $J'$. With increasing $U'$, $\lambda$ monotonically increases and finally becomes unity at a critical value $U'_c = 1.15$ above which the superconducting state is realized. Similar to the case of $U > U'$, the largest eigenvalue $\lambda$ is for the $s$-wave symmetry but the superconducting gap structure is significantly different from that for $U > U'$ as shown below.

### D. Gap Functions

First, we discuss the gap functions in the case with $U > U'$. Fig. 5 shows the diagonal components of the gap function $\Delta(\mathbf{k})$ for $U = 1.71$, $U' = 1.4$, $J = J' = 0.1$. Figs. 5 (a)-(d) show the gap functions in the orbital representation and Figs. 5 (e)-(h) show those in the band representation. We note that the energy bands are numbered as descending energy. It is found that the gap function has the $s$-wave symmetry and the most dominant component is the $d_{x^2-y^2}$ diagonal component. We find that the gap functions in the band representation have different signs between the electron pockets and the hole pockets without any nodes on the Fermi surfaces ($s$-wave symmetry in the orbital representation, except for the $d_{xy}$ component, also change those signs in $\mathbf{k}$ space. The absolute values of the gap functions on the Fermi surfaces are almost isotropic but largely depend on the energy bands; those on the electron pockets of the 13th and 14th bands are twice or more larger than those on the hole pockets of the 11th and 12th bands. This is because the $d_{x^2-y^2}$ component, which has dominant contribution in $\chi^s(\mathbf{q})$ as shown in Fig. 5 (a), for the 13th and 14th bands is larger than that for the 11th and 12th bands. We note that the 10th band (hole band) with the largest $d_{x^2-y^2}$ component has the largest absolute value of the gap function, although the Fermi level is just above the 10th band and does not cross it for $x = 0.1$.

Next, we discuss the gap functions in the case with $U < U'$. Fig. 6 shows the diagonal components of the gap function $\Delta(\mathbf{k})$ for $U = 0.4$, $U' = 1.15$, $J = J' = 0.1$. Figs. 6 (a)-(d) shows the gap functions in the orbital representation and Figs. 6 (e)-(h) show those in the band representation. The diagonal components of the gap function in the orbital representation have no sign change in the $\mathbf{k}$ space due to the diagonal components of $\hat{V}(\mathbf{q}) < 0$ as shown in Fig. 4 (c). We call this $s$-wave state as the $s_{++}$ wave state. The gap function in the band representation, however, has sign change between the Fermi surfaces and line nodes on the 14th band Fermi surface. These facts reflect that the sign change of the gap function in the orbital representation between the $d_{x^2-y^2}$ diagonal component and the other orbital diagonal components. The 11th band and 12th band Fermi surface has mainly $d_{yz}$ and $d_{zx}$ orbital character, while, the 13th band Fermi
surface has mainly \( d_{x^2-y^2} \) orbital character. Therefore, the gap function has different sign between the hole pockets and the 13th band electron pocket. The 14th band electron pocket has mainly \( d_{yz} \) and \( d_{xy} \) orbital character away from the Brillouin zone boundary, while \( d_{x^2-y^2} \) orbital character on the 14th band electron pocket is comparable with \( d_{yz} \) and \( d_{xy} \) one near the Brillouin zone boundary. Thus, the gap function on the 14th band electron pockets has plus sign near the zone boundary and minus sign away from the zone boundary. By the simple mean field analysis of the pair transfer term of the interacting part of the Hamiltonian eq (3),

\[
\frac{J'}{2} \sum_{i,\ell \neq \ell', \sigma \neq \bar{\sigma}} \langle d_{i\ell \sigma}^\dagger d_{i\ell' \bar{\sigma}} \rangle \langle d_{i\ell' \bar{\sigma}} d_{i\ell \sigma} \rangle \propto \sum_{\mathbf{k}, \ell \neq \ell'} \Delta_{\ell\ell'}^A(\mathbf{k}) \Delta_{\ell\ell'}^A(\mathbf{k}).
\]

It is shown that the pair transfer \( J' > 0 \) favors the sign change between the diagonal components of the gap function in the orbital representation.

In fact, we also examine the case with \( J' = 0 \) and we find that the \( s_{++} \)-wave state without sign change between the orbitals is realized for \( U < U' \). We show the gap function for \( U = 0.4, U' = 1.18, J = 0.1, J' = 0 \) in Fig. 7.

\[
\text{FIG. 6. (Color online) (a), (b), (c), (d) The diagonal components of the gap function } \Delta(\mathbf{k}) \text{ in the orbital representation and (e), (f), (g), (h) those in the band representation for } U = 0.4, U' = 1.15 \text{ and } J = J' = 0.1eV. \text{ The solid and dashed lines represent the Fermi surfaces and the nodes of the gap function, respectively.}
\]

\[
\text{FIG. 7. (Color online) (a), (b), (c), (d) The diagonal components of the gap function } \Delta(\mathbf{k}) \text{ in the orbital representation and (e), (f), (g), (h) those in the band representation for } U = 0.4, U' = 1.18 \text{ and } J = 0, J' = 0eV. \text{ The solid and dashed lines represent the Fermi surfaces and the nodes of the gap function, respectively.}
\]
It is found that all the diagonal components of the gap function in the orbital representation have the same sign and those in the band representation have no sign change between all Fermi surfaces. Therefore, it is considered that the sign change of the gap function between the the $d_{x^2-y^2}$ diagonal component and the others is due to the pair transfer $J^p$. It is helpful for understanding the difference between the $s_\pm$-wave state and the $s_{++}$-wave state in more detail to consider the gap function in the real space. For $s_\pm$-wave state, the on-site pairing is comparable with the nearest neighbor and/or the next nearest neighbor one. On the other hand, for the $s_{++}$-wave state, the on-site pairing is dominant and the off-site pairings are negligibly small as compared to the on-site pairing.

Here we discuss the reason why the on-site part of the gap function for $s_\pm$-wave state is large (especially in the $d_{x^2-y^2}$ diagonal component) even though the most dominant component of the effective interaction is always repulsive in $q$ space (see Fig. 3(b)), and then the on-site effective interaction is repulsive. When we perform the Fourier transformation of the gap equation eq. (5), the on-site part of the left hand side is proportional to the on-site gap function, while that of the right hand side is given by the product of the on-site effective interaction $(\times (-1))$ and the on-site anomalous Green’s function which is proportional to the $q$ summation of the gap-function times the single-particle spectral weight times the thermal factor. In the case with the $d_{x^2-y^2}$ diagonal component, the on-site gap function is negative as the negative contribution of the gap function in $q$ space is much larger than the positive one as shown in Fig. 3 (a). On the other hand, the on-site anomalous Green’s function becomes positive as the single-particle spectral weight of the $d_{x^2-y^2}$ hole band is very large around the $\Gamma$ point where the gap function is positive as compared to that of the electron band around the $M$-point where the gap function is negative. Then, the gap equation can be satisfied with the large value of the on-site gap function against the repulsive on-site effective interaction.

When the doping $x$ increases, the Fermi level rises apart from the $d_{x^2-y^2}$ hole band, and then the effect of the hole band decreases resulting in the decrease in the on-site gap function as well as the decrease in the superconducting transition temperature (not shown). Such doping dependence of the on-site gap function has recently been observed in the 5-band Hubbard model. On the contrary, in the $s_{++}$-wave state, the on-site pairing is always dominant almost independent of the doping $x$.

E. Phase Diagram

The phase diagram on $U'$-$U$ plane for $J = J' = 0.1$ is shown in Fig. 8, where the magnetic and charge-orbital instability is determined by $\det (1 - \hat{\chi}^{(0)}(q) \hat{S}) = 0$ and $\det (1 - \hat{\chi}^{(0)}(q) \hat{C}) = 0$, respectively and the superconducting instability is determined by $\lambda = 1$ as mentioned before. The ISDW with $q \sim (\pi, \pi)$ appears in the large $U$ region, while, the ferro-orbital order appears for $U < U'$ [see also Figs. 3(a), (b) and Figs 4(a), (b)]. It is noted that on the phase boundary where the charge-orbital instability takes place, the longitudinal orbital susceptibility $(\hat{\chi}_c^{(q)})_{\ell \ell', \ell'}^{\alpha \beta} = 0$, charge susceptibility $(\hat{\chi}_c^{(q)})_{\ell \ell', \ell}^{\alpha \beta}$ and $\hat{\chi}_c^{(q)}_{\ell \ell', \ell}^{\alpha \beta}$ diverge, while, the $s_{++}$-wave pairing is realized near the ISDW due to the spin fluctuations, while, the $s_{++}$-wave pairing is realized near the ferro-orbital ordered phase due to the charge-orbital fluctuations, where we regard the superconducting states as the $s_{++}$-wave states if $d_{yz}$, $d_{zx}$ and $d_{x^2-y^2}$ diagonal components of the gap function have no sign change in $k$ space and as the $s_{\pm}$-wave states if not. The way to determine whether the superconducting state is the $s_{++}$-wave state or the $s_{++}$-wave state is not unique. This is because the $s_{\pm}$-wave and the $s_{++}$-wave state are same symmetry ($A_{1g}$) and the change between $s_{++}$-wave and the $s_{\pm}$-wave state is crossover. In fact, as $U$ increases, the on-site pairing decreases, while, the off-site pairing increases continuously. At $U \sim 1.25$, the nodes appear around the $M$-point for the $d_{x^2-y^2}$ diagonal component and those approaches the $\Gamma$-point as $U$ increases. It is noted that we also obtain the phase diagram on $U'$-$U$ plane for $J = J' = 0.25$ and find that the phase diagram is essentially the same as that for $J = J' = 0.1$ except that the magnetic and the $s_{\pm}$-wave superconducting instabilities are slightly enhanced by the larger value of the Hund’s coupling $J$. 

![Phase Diagram](image_url)
F. Effects of Electron-Phonon Coupling

In this subsection, we discuss the effects of the electron-phonon coupling. By performing the group theoretical analysis for LaFeAsO, it is found that there are 14 kinds of the optical phonon modes at the Γ point: $2A_{1g} + 2B_{1g} + 4E_g + 3A_{2u} + 3E_u$. Here, we concentrate on the $A_{1g}$ mode in which La and As ions oscillate along the c-axis. The $A_{1g}$ phonon dose not break the symmetry of the orbital and the resulting electron-phonon coupling matrix $\hat{g}$ is diagonal in the orbital representation. Within the RPA, the charge-orbital susceptibility $\hat{\chi}^{(4)}(q)$ including the effects of both the electron-electron and the electron-phonon coupling is obtained by replacing $U$ with $U - 2U_{ph}$ and $2U' - J$ with $2(U' - U_{ph}) - J$ in eqs. (3) and (4), where $U_{ph} = 2g^2/\omega_{A_{1g}}\omega_{A_{1g}}$ is the frequency of the $A_{1g}$ phonon and we neglect the orbital- and q-dependence of the electron phonon interaction. It is found that the inter-orbital direct term $U'$ which enhances the orbital fluctuations is harder to be reduced by the electron-phonon coupling than the intra-orbital direct term $U$. As a result, the orbital fluctuations are relatively enhanced by the electron-phonon coupling as compared to the spin fluctuations.

IV. SUMMARY AND DISCUSSION

In summary, we have investigated the pairing symmetry of the two-dimensional 16-band $d$-$p$ model by using the RPA and have obtained the phase diagram including the magnetic and orbital orders and the superconductivity. For $U > U'$, the $s_{\pm}$-wave superconductivity is realized near the ISDW with $q \sim (\pi, \pi)$ phase. On the other hand, for $U < U'$, the $s_{++}$-wave superconductivity appears near the ferro-orbital ordered phase. The $s_{\pm}$-wave pairing is mediated by the spin fluctuations, while the $s_{++}$-wave pairing is mediated by the orbital fluctuations.

For $U > U'$, the gap function for the $s_{\pm}$-wave pairing changes its sign between the hole pockets and the electron pockets and the most dominant contribution of the gap function is the $d_{xy}$ orbital diagonal component. This is qualitatively consistent with the results based on the 5-band Hubbard model. However, the $d_{xy}$ diagonal component of the gap function in our 16-band $d$-$p$ model have much larger value than the other matrix elements in comparison with the results based on the 5-band Hubbard model. This may be because the outer hole Fermi surface which has mainly $d_{yz}$ and $d_{zx}$ orbital character obtained by the $d$-$p$ model is almost circular, but that obtained by the 5-band Hubbard model is diamond shaped. Therefore, the nesting effect which enhances the spin fluctuations becomes weak in our $d$-$p$ model, and the resulting components of $\Delta(k)$ related to $d_{xy}$, $d_{xz}$ orbitals have smaller values.

For $U < U'$, the gap function in the orbital representation for the $s_{++}$-wave pairing dose not change its sign in $k$ space. In other words, the on-site pairing is much larger than the off-site pairing in the real space. This is similar to the conventional phonon-mediated superconductivity. However, the gap functions have different signs between orbitals in contrast to the conventional phonon-mediated superconductivity. We have shown that this sign change of the gap functions between orbitals is due to the effect of the pair transfer interaction $J_{ps}^{a1}$. It is noted that the $s_{++}$-wave state has been observed also in the one-dimensional 2-band Hubbard model in the same parameter region with $U < U'$. It seems that the both $s_{\pm}$-wave and the $s_{++}$-wave states with full superconducting gaps are consistent with various experiments such as, the NMR relaxation rate, the Knight shift, the ARPES, the magnetic penetration depth measurements, although the sign of the gap function has not been directly observed there. However, according to the recent theoretical studies of the nonmagnetic impurity effects, Anderson’s theorem is violated for the $s_{+}$-wave superconductivity in contrast to the experimental results of very weak $T_c$ suppression in Fe site substitution and neutron irradiation. Since it can be considered that the impurity potential by the Fe-site substitution is diagonal and local in the orbital basis according to the first principle calculation, it is expected that the $s_{++}$-wave state observed in the present study is more robust against the nonmagnetic impurity effects than the $s_{\pm}$-wave state.

In addition to the Coulomb interaction, we have also discussed the effects of the coupling $g$ between the electron and the $A_{1g}$ phonon within the RPA. It has been found that the $s_{++}$-wave pairing realized in the unrealistic parameter region with $U < U'$ for $g = 0$ is enhanced due to the effect of $g$ and can be expanded over the realistic parameter region with $U > U'$ for a realistic value of $g$. In the first principle calculations for iron-based superconductors in conjunction with the Migdal-Eliashberg theory, the electron-phonon coupling is found to be too small to obtain high $T_c$ observed in experiments. The effect of the Coulomb interaction, however, has not been discussed there. In the present study, the cooperative effect of the Coulomb interaction and the electron-phonon coupling is crucial for the enhancement of the orbital fluctuations which induce the $s_{++}$-wave superconductivity. Recently, the large isotope effects on the transition temperatures for both the SDW and the superconductivity have been observed. This experimental result implies that not only the Coulomb interaction but also the electron-phonon coupling plays crucial effects on the electronic states for iron-based superconductors.

In early theoretical studies for the copper oxide superconductors, the effect of the Coulomb interaction between the $d$ and $p$ electrons $U_{pd}$ was studied by several authors. According to the RPA study based on the $d$-$p$ model with the single $d_{xy}$ orbital, $U_{pd}$ enhances the charge fluctuations with $q = (0, 0)$ and the s-wave superconductivity is realized due to the effect of charge fluctuations. In addition, the $1/N$-expansion ap-
The strong correlation effect enhances the charge fluctuations together with the $s$-wave superconductivity. Therefore, it is expected that, in the present $d$-$p$ model including not only the on-site Coulomb interaction but also the inter-site Coulomb interaction $U_{pd}$ together with the electron-phonon coupling $g$ are now under way.

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The spin and charge-orbital susceptibilities are the particle-hole channel, while the superconducting susceptibility is the particle-particle channel. Then, there is a rotation of the orbital indices of the effective pairing interaction $V$ in the gap equation eq. (8) with respect to those of the spin and charge-orbital susceptibilities in eqs. (4)-(6). This is more explicitly represented in ref. 31 with a different notation.

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