Orbital-driven two-dome superconducting phases in iron-based superconductors

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Recent several experiments revealed that novel bipartite magnetic/superconducting phases widely exist in iron pnictides and chalcogenides. Nevertheless, the origin of the two-dome superconducting phases in iron-based compounds still remains unclear. Here we theoretically investigated the electronic structures, magnetic and superconducting properties of three representative iron-based systems, i.e. LaFeAsO₁₋ₓHₓ, LaFeAs₁₋ₓPₓO and KFe₂As₂. We found that in addition to the degenerate in-plane anisotropic \( xx/yz \) orbitals, the quasi-degenerate in-plane isotropic orbitals drive these systems entering into the second parent phase. Moreover, the second superconducting phase is contributed by the isotropic orbitals rather than the anisotropic ones in the first superconducting phase, indicating an orbital-selective pairing state. These results imply an orbital-driven mechanism and shed light on the understanding of the two-dome magnetic/superconducting phases in iron-based compounds.

Iron-based superconductors display rich phase diagrams with spin-density wave (SDW) [1 2], orbital order [3 4], nematicity [5–7] and superconductivity [8–10]. These competing or coexisting phases arise from the interplay among charge, spin, orbital and lattice degrees of freedom of iron-based compounds. In the early stage it had already realized that the multi-orbital characters of band structures and orbital order in many different families of iron pnictides and chalcogenides [11–19], and the orbital selective Mott physics was also suggested in some iron-based compounds [16 18], however, our understand to the roles of orbital degree of freedom seem to be no more than that. The key roles of orbital degree of freedom on superconducting (SC) pairing interactions, pairing symmetry, and phase diagrams in these different iron-based superconductors seem to be far from understood or revealed.

The parent compound LaFeAsO possesses a stripe antiferromagnetic (SAFM) ground state associated with a weak \( xx \) orbital order in the low-temperature phase, and exhibits an orthorhombic to tetragonal structural phase transition upon doping or increasing temperature. Once substituting O with F, such an SAFM is gradually suppressed and an SC dome emerges in LaFeAsO₁₋ₓFₓ [8]. While for hydrogen doping, recent experiments demonstrated that the phase diagram shows two SC domes accompanied with bipartite magnetic parent phases in LaFeAsO₁₋ₓHₓ, as seen the AF1-SC1-SC2-AF2 phase diagram in Refs. [20 21]. The neutron diffraction experiment showed that the second magnetic parent phase lies around \( x=0.5 \) in LaFeAsO₁₋ₓHₓ. This novel parent phase also shows SAFM order except that Fe spin has a large magnetic moment of \( \sim 1.21 \mu_B \), in comparison with \( \sim 0.63 \mu_B \) in LaFeAsO. Moreover, it possesses an unusual orthorhombic lattice distortion accompanied by the instability of As atoms which are away from the center of the Fe square lattice in the low-temperature phase. These unusual phenomena demonstrate that the orbital physics in LaFeAsO₁₋ₓHₓ could be different from LaFeAsO. One naturally arises a question whether the two SC domes in LaFeAsO₁₋ₓHₓ is particular? Detailed analysis to recent experimental data shows that some other iron-based compounds also have two distinct AFM/SC phases, demonstrating that such two SC domes may be a general phenomenon in various iron-based SC families.

More recently, several experiments revealed that LaFeAs₁₋ₓPₓO exists a different AF1-SC1-AF2-SC2 phase diagram [22 24]. Also in K₁₋ₓFe₂₋ₓSe₂, a second SC phase re-emerges under high pressure [25]. In KFe₂As₂, a second SC phase is also found accompanied with a collapsed tetragonal phase under high pressure [26]. In addition, two SC domes are also observed in the K-doped FeSe thin films grown on SiC substrate [27]. And in the intercalated FeSe [28], the high pressure also could lead to two SC domes, similar to K₁₋ₓFe₂₋ₓSe₂. On the other hand, the pressure can induce a new AFM parent phase in pure FeSe [24]. The more recent nuclear magnetic resonance (NMR) experiment showed that there also exist two SC domes and a second orthorhombic phase in LaFeAsO₁₋ₓPₓO [29]. More and more bipartite AFM/SC phases have been found in various iron-based compounds, however, the nature of the second AFM/SC phase still remains unclear.

In this Letter, we aim to clarify the origin of the bipartite AFM/SC phases in LaFeAsO₁₋ₓHₓ, LaFeAs₁₋ₓPₓO, and KFe₂As₂ compounds. For this purpose, we first...
study the electronic, magnetic and SC properties of LaFeAsO$_1$-$x$H$_x$ using the first-principles method, projected Wannier functions, the random phase approximation (RPA) and mean-field approximation based on five-orbital Hubbard models. We find that quasi-degenerate $xy/xz/yz$ orbitals drive the emergence of the second magnetic parent phase, and the in-plane isotropic $xy$-orbital dominates an orbital-selective SC pairing state in LaFeAsO$_1$-$x$H$_x$. While in LaFeAs$_1$-$x$P$_x$O$_2$ one of the quasi-degenerate orbitals, i.e. in-plane isotropic $3z^2-r^2$ orbital, is responsible for the second magnetic parent phase. In KFe$_2$As$_2$, a quasi-degenerate $xz/yz/xy$ orbital instability leads to a structural phase transition from a tetragonal phase into a collapsed tetragonal (CT) phase under pressure. Therefore we propose that the quasi-degenerate orbitals play key roles in the emergence of the second AFM phase and the active isotropic orbital controls the second SC phase, leading to the two SC domes in the phase diagrams of these iron pnictides and chalco-
tides.

Fig. 1 schematically displays the phase diagram of LaFeAsO$_1$-$x$H$_x$ according to the recent experiments [20, 21, 31] and our numerical calculations and analysis. We find that the degenerate $xz/yz$ orbitals dominate the AF1 phase at $x$=0 and the SC1 phase, while the quasi-
degenerate $xy/xz/yz$ orbitals dominate the AF2 phase at $x$=0.5 and the SC2 phase, respectively. In unveil-
ing the different orbital physics in LaFeAsO$_1$-$x$H$_x$, we first study the electronic structures of two parent phases LaFeAsO$_1$-$x$H$_x$ at $x$=0 and 0.5, respectively. With the help of the projected Wannier function technique, the five-orbital tight-binding model $H_0$ is constructed. The orbital-resolved band structures are displayed in Fig. 2 (a) and (b). It is clearly showed that besides the $xz/yz$ orbitals, the $xy$ orbital also contributes considerable weight near Fermi level ($E_F$) at $x$=0.5, different from that at $x$=0. For comparison, the crystal field splittings, i.e. on-site energies of Fe-3$d$ orbitals in two compounds can be seen in Supplemental Material [32] and Ref. [33].

The most obvious multi-orbital feature of LaFeAsO$_1$-$x$H$_x$ at $x$=0.5 is that the three $t_{2g}$ orbitals, $xz/yz$ and $xy$, have nearly identical on-site energies, thus are almost degenerate. We call these three orbitals quasi-degeneracy, since the bandwidths of these three orbitals in LaFeAsO$_{0.5}$H$_{0.5}$ are clearly distinguished with $W_{xz/yz} > W_{xy}$. The anion-height instability, which was recently suggested to be responsible for the AF2 phase [31], is in fact a natural consequence of the orbital quasi-degeneracy; according to the Jahn-Teller effect, these quasi-degenerate orbitals are unstable, resulting in the lattice distortion at low temperatures. On the other hand, due to different orbital components and weights in the two parent phases with $x$=0 and 0.5, a single rigid-band tight-binding model could not describe the entire $T$-$x$ phase diagram. Thus to depict the low-energy physics in LaFeAsO$_1$-$x$H$_x$, we have to adopt two sets of different tight-binding parameters at $x$=0 for AF1 and SC1 phases, and at $x$=0.5 for AF2 and SC2 phases, respectively.

In order to explore the role of Fermi surface nesting and the possible magnetic instability in LaFeAsO$_1$-$x$H$_x$ at $x$=0.5, we study the dynamical spin susceptibility within RPA. The bare spin susceptibility is given by the formula [35, 37]

$$\chi_0(q, i\omega) = -\frac{1}{N} \sum_{\vec{k}, \mu \nu} \frac{a_{\mu}^{+}(\vec{k})a_{\nu}^{+}(\vec{k})a_{\mu}^{+}(\vec{k} + \vec{q})a_{\nu}(\vec{k} + \vec{q})}{\omega + \varepsilon_{\mu}(\vec{k} + \vec{q}) - \varepsilon_{\nu}(\vec{k}) + i\eta} \times [f(\varepsilon_{\mu}(\vec{k} + \vec{q})) - f(\varepsilon_{\nu}(\vec{k}))]$$

(1)

The RPA susceptibility is expressed as

$$\chi^{RPA}(q, i\omega) = \chi_0(q, i\omega)[\pm \Gamma(x)\chi_0(q, i\omega)]^{-1},$$

(2)

FIG. 1. (Color online) Schematic phase diagram of LaFeAsO$_1$-$x$H$_x$. Different orbital configurations are displayed in the first and second AFM/SC phases.

FIG. 2. (Color online) Orbital-resolved band structures of the five-orbital tight-binding model for (a) LaFeAsO, (b) LaFeAsO$_{0.5}$H$_{0.5}$, (c) LaFeAsO$_{0.5}$P$_{0.5}$O and (d) LaFePO.
where $\chi_0$ is the bare susceptibility defined in Eq. (1) and the nonzero components of the matrices $\Gamma_c$ and $\Gamma_s$ are given as $(\Gamma_c)_{aa,aa} = U$, $(\Gamma_c)_{aa,bb} = 2U' - J_H$, $(\Gamma_c)_{ab,ab} = -U' + 2J_H$, $(\Gamma_s)_{aa,bb} = J_H$ and $(\Gamma_s)_{aa,aa} = U$, $(\Gamma_s)_{ab,ab} = J_H$, $(\Gamma_s)_{ab,ba} = U'$, $(\Gamma_s)_{ba,ab} = J_H$ with orbitals $a \neq b$. It is found that the peaks of the dynamical spin susceptibility appear at $Q \sim (\pi, \pi/3)$, rather than $(\pi, 0)$, implying that the nesting scenario fails in LaFeAsO$_5$H$_{0.5}$. This is due to the absence of the hole pocket at $\Gamma$ point like $K_{1/2}$Fe$_2$Se$_2$ [38], in contrast to that in LaFeAsO [32, 39, 40].

To further understand the AF2 parent phase, we also investigate the magnetic ground state properties of LaFeAsO$_5$H$_{0.5}$ by the first-principles electronic structures calculations. We find that a stable magnetic ground state is SAFM, in agreement with the neutron diffraction experiment [21]. Based on the total energy differences among different magnetic configurations and fitting by the $J_{1a}$-$J_{1b}$-$J_2$ Heisenberg model, we estimate the spin exchange parameters: $J_{1a} = 7.8$ meV/$s^2$, $J_{1b} = 4.7$ meV/$s^2$, and $J_2 = 31.4$ meV/$s^2$ in LaFeAsO$_5$H$_{0.5}$. The next nearest-neighbor (N.N.N.) coupling $J_2$ is much larger than the N.N. spin exchange parameter $J_2$, which explains the strong anisotropy between $J_1$ and $J_2$ [41]. Therefore, the orbital order scenario involved with three quasi-degenerate orbitals in LaFeAsO$_{1-x}$H$_x$ ($x=0.5$) is significantly different from the weak orbital order in LaFeAsO.

![FIG. 3. (Color online) Orbital occupancies and magnetic moments dependence on Coulomb interaction $U$ within the five-orbital Hubbard model for (a) LaFeAsO and (b) LaFeAsO$_5$H$_{0.5}$ with parameter $J_H=0.1U$.](image)

From Fig. 3 (b), one notices that in LaFeAsO$_5$H$_{0.5}$, when $U > U_c \sim 1.6$ eV, $n_{xy}=n_{xy}\uparrow + n_{xy}\downarrow \sim 1$ and $m_{xy}=m_{xy}\uparrow - m_{xy}\downarrow \sim 1$, indicating $n_{xy}\uparrow \sim 1$ and $n_{xy}\downarrow \sim 0$, i.e. the spin-up channel of $xy$ orbital is almost fulfilled. This demonstrates that an orbital-selective Mott phase (OSMP) occurs with the insulating $xy$ orbital and the metallic $xz$ and $yz$ orbitals, as seen the sketch of the OSMP [32]. The insulating $xy$ orbital exhibits strong ordering with respect to $xz$/$yz$ orbitals, similar to manganites and other transition-metal oxides. This OSMP accompanied by a strong orbital order contributes to the second AFM parent phase at $x=0.5$.

To look into the emergence of the second SC phase...
SC2, we investigate the dependence of orbital order parameters on doping x within the five-orbital Hubbard model of LaFeAsO$_{1-x}$H$_x$ at 0.5 [32]. These two orbital ordering parameters show a competing behavior: the $xz$-orbital weight increases while the $xy$-orbital weight decreases when x varies from 0.5 to 0.35, favoring a paramagnetic metallic phase and the emergence of SC2 around $x<0.5$ end. Out of expectation, when $x>0.5$, the $xy$-orbital weight becomes more large, consistent with the experimental observations [21, 31]. These results explain why the SC2 phase appears at the left end of AF2 phase in the phase diagram of LaFeAsO$_{1-x}$H$_x$.

In analyzing the SC pairing properties of the SC2 phase, we calculate the pairing vertex arising from the exchange of spin and orbital fluctuations using RPA. The singlet orbital vertex function $\Gamma_{\alpha\beta\mu\nu}$ is given by

$$\Gamma_{\alpha\beta\mu\nu}(k, k', \omega) = \frac{3}{2} U^s \chi_s^{\alpha\mu}(k - k', \omega) U^s + \frac{1}{2} U^c \chi_c^{\alpha\mu}(k - k', \omega) U^c + \frac{1}{2} U^c \chi_c^{\alpha\mu}(k - k', \omega) U^c + \frac{1}{2} U^s \chi_s^{\alpha\mu}(k - k', \omega) U^s. \quad (4)$$

The dominant orbital pairing vertices for the AF2 and SC2 phases, which is consistent with the experimental observations [23]. The orbital-resolved band structures are shown in Fig. 2 (c) and (d) for LaFeAs$_{1-x}$P$_x$O at $x=0$ and 0.5, respectively. From Fig. 2 (d) it is clearly found that in the AF2 phase, in addition to the degenerate $xz/yz$ orbitals, there also exist quasi-degenerate $3z^2-r^2/x^2+y^2$ orbitals at $x=0.5$, which could be seen from the crystal field splittings, i.e. the on-site energy differences, of Fe-3d orbitals [32].

Finally, we also perform similar analysis for KFe$_2$As$_2$ with $n=5.5$ under pressure, and find that the quasi-degenerate character of orbitals occurs when the SC1 or SC2 phase disappears, as seen from the schematic phase diagram in Fig. 3 according to the experimental result in Ref. [26]. For comparison, the on-site energies of Fe-3d orbitals are given in Supplemental Material [32]. The instability of the quasi-degenerate $xz/yz/xy$ orbitals at about 10 GPa drives a structural phase transition from a tetragonal phase to a CT phase under high pressure up to 15 GPa. The dominant $xy$-orbital character is presented at 10 GPa, but absent at 20 GPa, which is also verified by a recent DFT and dynamical mean-field-theory (DMFT) calculations [32]. In addition, we predict that another quasi-degenerate $xz/yz/xy$ orbital instability emerges above 30 GPa.

Through the analysis of the above three representative systems, we conclude that the active quasi-degenerate orbitals drive the emergence of the second AFM or SC phase [32]. Meanwhile, the orbital physics of the fully degenerate $xz/yz$ orbitals is the dominant scenario in the AF1/SC1 phase [32]. Actually, the doping/substitution (chemical internal pressure) or hydrostatic pressure pushes the in-plane anisotropic $xz/yz$ orbitals with C2 symmetry away from $E_F$, but it activates the in-plane isotropic $xy$, $x^2-y^2$ or $3z^2-r^2$ orbital with C4 symmetry in the planar square Fe lattice. Therefore, it is readily understood that the isotropic orbitals emerge around $E_F$. The in-plane isotropic orbital would...
FIG. 6. (Color online) Schematic orbital configurations in the phase diagram of KFe$_2$As$_2$ under pressure.

suppress ($\pi$, 0) spin fluctuations, but enhance the ($\pi$, $\pi$) spin fluctuations and orbital fluctuations. Consequently, the orbital fluctuations in the second AF/SC phase play a dominant role, which is supported by a quite recent NMR experiment [44].

In summary, we have shown that in addition to conventional magnetic and superconducting phases in LaFeAsO$_{1-x}$F$_x$, LaFeAs$_{1-x}$P$_x$O and KFe$_2$As$_2$, the quasi-degenerate orbitals drive the emergence of the second AFM/SC phases, lead to universal two-dome SC phase in iron pnictides. Due to the orbital modulation, there exist two distinct types of SC phases, one is in-plane anisotropic orbital dominant SC1 phase with low $T_c$, the other is in-plane isotropic orbital dominant SC2 phase with high $T_c$. Actually, our scenario could be generally extended to the similar two-dome SC phases observed experimentally in iron chalcogenides, such as the FeSe-based compounds under pressure, as well as the heavy fermion superconductors.

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