Hidden robust presence of a hole Fermi surface in a heavily electron-doped iron-based superconductor LaFe_2As_2

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We investigate the electronic structure of a recently discovered, heavily electron-doped iron-based superconductor LaFe_2As_2 . Although first-principles calculation shows apparent absence of hole Fermi surfaces around the Γ point, we reveal, by hypothetically removing the La d orbital contribution, that a hole Fermi surface around the Γ point is essentially present. In the collapsed phase of LaFe_2As_2 , which is nonsuperconducting, the hole Fermi surface is found to be absent, and the difference from the uncollapsed superconducting phase can be naturally understood within the spin-fluctuation-mediated pairing scenario.

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I. INTRODUCTION

The Fermi surface (FS) configuration in the iron-based superconductors [1] has been an issue of great importance from the early stage of their study. This is because the FS consists of small electron and hole pockets (or cylinders in the three-dimensional sense), so it is sensitive to carrier doping, elemental substitution [2,3], and/or pressurization, and also because many theories suggest that the number and/or asymmetry of the electron and hole FSs play a key role in controlling the superconducting transition temperature (T_c) and/or the pairing symmetry [4,5]. For instance, within the spin-fluctuation-mediated pairing scenario, Cooper pair scattering occurs between electron and hole FSs, so that too much carrier doping is expected to result in a disappearance of hole or electron FSs, and hence suppression or absence of superconductivity. T_c dome obtained in a case when, e.g., electrons are introduced in BaFe_2As_2 by partially substituting Fe with Co [6,7] is considered to be a manifestation of such a FS variance [8].

From this viewpoint, cases with heavy electron doping have attracted much attention. In $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$, a large amount of electrons are doped, and in fact, the angle-resolved photoemission spectroscopy (ARPES) experiments show that the hole bands sink below the Fermi level [9–11]. Still, T_c is high, apparently implying that the hole bands are not playing an important role in the occurrence of superconductivity. A similar situation (with even a higher T_c) is observed in FeSe thin films grown on a substrate [12–14]. Partially motivated by these studies, the importance of the hole bands sinking below the Fermi level (the “incipient band”) has been pointed out in

various studies [4,15–23]. On the other hand, another ARPES experiment on $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ found that a hole band actually intersects the Fermi level [24]. Another series of material that is of great interest are the hydrogen-doped 1111 compounds such as $\text{LaFeAsO}_{1-x}\text{H}_x$ [25–30]. In these materials, high T_c superconductivity survives up to large amounts of electron doping close to 50%. Although such heavy electron doping is expected to wipe out the hole FS, some theoretical studies have pointed out that one of the hole bands (the d_{xy} band) actually exhibits a non-rigid-band shift upon electron (hydrogen) doping, so that the hole band can in fact give rise to a FS and play an important role [25,31].

Given this controversial situation regarding heavily electron-doped iron-based superconductors, a recent discovery of a new superconductor LaFe_2As_2 , exhibiting $T_c = 12.1$ K, opens up a renewed avenue in this hotly debated field [32]. In fact, considering BaFe_2As_2 as the mother (starting) compound, a complete substitution of $\text{Ba}(+2)$ with $\text{La}(+3)$ corresponds to a very heavy electron doping of 50%. A band structure calculation of LaFe_2As_2 has shown that the hole FS around the Γ point is indeed missing [32], which may be taken as natural considering the large amount of electron doping. This study also found that one electron FS interestingly possesses an unusual “jungle-gym-like” shape, whose role played in superconductivity, if any, is difficult to understand. It has also been found that the crystal structure transforms from a collapsed tetragonal structure to an uncollapsed tetragonal structure by annealing samples [32]. The superconductivity is found to emerge only in the uncollapsed phase, and no superconductivity has been observed in the collapsed phase. As discussed for other iron-based superconductors [4,33], it is expected that the difference in the electronic structure of collapsed and uncollapsed LaFe_2As_2 may hold the key to understanding the origin of superconductivity.

Here, we theoretically investigate the electronic structure of the uncollapsed and collapsed phases of LaFe_2As_2 to reveal its relevance to the origin of superconductivity. In the uncollapsed phase, we surprisingly find that the d_{xy} cylindrical

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FIG. 1. (a) Electronic band structure and density of states of uncollapsed LaFe₂As₂. (b)–(f) Five FSs of LaFe₂As₂.

hole FS around the Γ point, in the usual sense of the term used for the iron-based superconductors, can be considered as essentially present despite the heavy electron doping. Interestingly, both the survival of the d_{xy} hole FS and its apparent absence can be traced back to the hybridization between the Fe 3d and La 5d orbitals. In the collapsed phase, on the other hand, the d_{xy} hole FS is lost due to the large Fe-As-Fe bond angle. The correspondence between the presence or absence of the hole FS and the T_c can be naturally understood within the spin-fluctuation-mediated pairing scenario.

II. METHOD

First-principles calculations are performed by means of the full-potential linearized augmented plane wave method as implemented in WIEN2K [34]. We use the Perdew–Burke–Ernzerhof (PBE) exchange-correlation functional [35], and take $RK_{\text{max}} = 7$ and a $16 \times 16 \times 7$ k mesh for self-consistent calculation and a $22 \times 22 \times 22$ k mesh for calculation of the density of states. We start with the electronic structure of the uncollapsed phase, whose lattice constants and the internal coordinates of the Fe atoms are given in Ref. [32].

III. RESULTS AND DISCUSSION

A. Band structure of the collapsed phase

The calculated band structure, the density of states, and the FS of the uncollapsed phase are shown in Fig. 1. In Fig. 1(a), it can be seen that the Fe d orbitals are dominant for the construction of the conduction bands around the Fermi level. As found in Ref. [32], the three hole and two electron FSs are obtained as shown in Figs. 1(b)–1(d) and 1(e)–1(f), respectively. The hole Fermi pockets and the cylindrical electron FS shown in Figs. 1(a)–1(e) are obtained, which can be seen in other iron-based superconductors. On the other hand, the

FIG. 2. (a) Band structure and (b) the FS of uncollapsed LaFe₂As₂ obtained from the 15-orbital tight binding model. (c) and (d) are the band structure and the FS of paramagnetic BaFe₂As₂, respectively, obtained in a similar way.

electron FS shown in Fig. 1(f) does not seem to correspond to any of the FSs seen in usual iron-based superconductors.

To understand the origin of this strange FS, we construct a 15-orbital tight binding model for LaFe₂As₂ exploiting the maximally localized Wannier functions of d orbital projections centered at the Fe or La sites (5 d orbitals $\times$ 3 atoms = 15 orbitals) using the WANNIER90 package [36] and WIEN2WANNIER code [37]. The band structure and the FS of the tight binding model are shown in Figs. 2(a) and 2(b), respectively. In these figures, the orbital components of the La $d_{x^2-y^2}$ and Fe d orbitals are expressed as colored circles. Other La d orbitals have only small weight within the energy window presented here. For comparison, as shown in Figs. 2(c) and 2(d), we also perform similar calculation for BaFe₂As₂ (assuming a paramagnetic state) adopting the experimentally determined lattice structure [38].

First glance at the density of states of LaFe₂As₂ [Fig. 1(a)] might give an impression that the La orbitals do not strongly affect the band structure around the Fermi level. However, the La $d_{x^2-y^2}$ orbital weight is seen to be spread from -2 to 6 eV measured from the Fermi level, namely, the La $d_{x^2-y^2}$ band crosses the Fermi level. In fact, if we look at the orbital weight in the band structure and the FS, we find that the La $d_{x^2-y^2}$ orbital does make a large contribution [see the green colored circles in Figs. 2(a) and 2(b)]. The jungle-gym-like shape of the FS therefore can be understood as originating from the

FIG. 3. (a) FS constructed from the 14-orbital tight binding model of uncollapsed LaFe₂As₂, where the La $d_{x^2-y^2}$ orbital contribution is hypothetically removed. (b) Band structure of the 15-orbital models of uncollapsed LaFe₂As₂ (left) and BaFe₂As₂ (center), and the 10-orbital model of uncollapsed LaFe₂As₂ (right), where all the La d orbitals are hypothetically removed from the 15-orbital model. The Fermi energy of the hypothetical models of LaFe₂As₂ is fixed at that of the original LaFe₂As₂.

hybridization between the Fe $3d$ and La $d_{x^2-y^2}$ orbitals. This is why the strange shape is hardly seen in other iron-based superconductors. In fact, in BaFe₂As₂, nearly the entire Ba $d_{x^2-y^2}$ orbital weight lies above the Fermi level [Fig. 2(c)], so the Ba $d_{x^2-y^2}$ orbital makes a small contribution on the FS. We note that the presence of a La d orbital component around the Fermi level has also been discussed in LaFe₂P₂, which belongs to the same space group [39–41].

B. Hidden hole Fermi surface

Since the Fe $3d$ orbitals are expected to play a main role in the occurrence of superconductivity, it is interesting to look at the “genuine Fe $3d$ appearance” of the FS by hypothetically removing the La $d_{x^2-y^2}$ orbital contribution. For this purpose, we constructed a 14-orbital model, in which the hopping integrals among La $d_{x^2-y^2}$ and all the orbitals are neglected in the 15-orbital model. We adopt the same Fermi level as that of the 15-orbital model in order to directly compare the FSs with and without considering the La $d_{x^2-y^2}$ orbital contribution. The obtained FS is shown in Fig. 3(a). The hole FSs originating from the Fe $d_{xz/yz}$ orbitals are barely affected; they remain to be small pockets present only around the Z point, consistent with the heavy electron doping. However, most interestingly, there appears a cylindrical hole FS around the Γ -Z line, originating from the Fe d_{xy} orbital, whose volume is similar to that in BaFe₂As₂. We will later come back to the origin of this apparent contradiction with the heavy electron doping. If we turn to the FSs around the X point, there appear two cylindrical electron sheets, similar to those seen in BaFe₂As₂. Coming back once again to the original FS in Fig. 2 from the above viewpoint, we can trace portions with strong Fe d orbital weight to find that there essentially exist a d_{xy} hole FS and a pair of electron FSs usually seen in 122 iron-based superconductors. The bottom line here is that there actually exist electron and hole cylindrical FSs originating from the d_{xy}

orbital, which can be favorable for spin-fluctuation-mediated superconductivity because this mechanism is based on the presence of electron and hole FSs originating from the same orbital.

The robust survival of the d_{xy} hole FS against heavy electron doping by elemental substitution in the blocking layer apparently resembles the case of the hydrogen-doped 1111 systems [25,27,29,30] and is in contrast to the case of Co-doped BaFe₂As₂, where the elemental substitution takes place within the conducting layer, and only a rigid band shift of the Fermi level occurs as theoretically expected and experimentally observed [8,42]. For the hydrogen-doped 1111, it was theoretically shown in Ref. [31] that the substitution of oxygen with hydrogen leads to an increase of the positive charge in the blocking layer, which lowers the As $4p$ level (moves away from Fe $3d$) in the conducting layer. This in turn reduces the indirect electron hopping between nearest neighbor Fe $3d_{xy}$ orbitals via As $4p$, which raises the d_{xy} hole band energy. One might expect a similar mechanism to work when Ba is substituted with La in BaFe₂As₂ because the positive charge in the blocking layer increases also in this case.

To see whether this is indeed the case, we construct 21-orbital models for both La and Ba cases, where not only Fe d and La d but also As p orbitals are explicitly considered. It is found that the on-site energies of both the As p orbitals and the Fe d orbitals are lowered by substituting Ba with La (for details, see the Appendix). In particular, the energy reduction of Fe d orbitals and that of As p_z , through which the indirect Fe-Fe hopping mainly occurs, is nearly the same about 0.1 eV, which implies that the robustness of the d_{xy} FS is not due to the same mechanism as in hydrogen-doped 1111 systems.

Since the answer to the puzzle must lie in the difference between La and Ba, we go back to the 15-orbital model of LaFe₂As₂, and now hypothetically remove all the La d orbitals to end up with a $15 - 5 = 10$ orbital model, whose band structure is shown in Fig. 3(b) (right). Interestingly, we find that the position of the Fe d_{xy} and $d_{xz/yz}$ hole bands at the Γ point is reversed by totally removing the La d orbitals, namely, the energy level of the $d_{xz/yz}$ hole bands becomes higher. This means that the energy of the Fe $d_{xz/yz}$ hole bands are suppressed due to the mixing of the La d orbitals. What is surprising here is that the role played by the weakly hybridized La d orbitals other than $d_{x^2-y^2}$ (note that there is small weight of other La d orbitals near the Fermi level) is important because removing just the La $d_{x^2-y^2}$ orbital does not give rise to $d_{xz/yz}$ hole FSs around the Γ point as we have seen in the 14-orbital model [Fig. 3(a)]. If we compare LaFe₂As₂ and BaFe₂As₂ in Fig. 3(b), the energy level of the $d_{xz/yz}$ hole bands of the latter is higher than that of the former. This is because the on-site energy of the Ba d orbitals is higher than that of La d (see Table I in the Appendix). Actually, similar analysis for KFe₂As₂ shows that the energy of the $d_{xz/yz}$ band becomes even higher with respect to d_{xy} (not shown), because the energy level of the K d orbitals is higher than that of Ba d . Our conclusion here is that the d_{xy} FS remains upon heavy electron doping in LaFe₂As₂ because the doped electrons selectively enter the $d_{xz/yz}$ bands, whose energy is significantly lowered by the La $5d$ hybridization [43].

FIG. 4. (a) Band structure and (b) the FS obtained from a 15-orbital tight binding model of collapsed LaFe_2As_2 .

C. Comparison of the collapsed and uncollapsed phases

Next, we will discuss the differences between the band structures of the uncollapsed and collapsed phases. Using the crystal structure of the collapsed phase described in Ref. [32], we calculate the band structure and FS as shown in Fig. 4. In the collapsed phase, the hole FS originating from the Fe d orbitals almost disappears. In particular, the disappearance of the d_{xy} hole FS, as compared to that in the uncollapsed phase, is due to the large Fe-As-Fe bond angle, namely, that of the uncollapsed phase is 110.8° , while that of the collapsed phase is 118.1° . For such a large bond angle, the d_{xy} bands sink deeply below the Fermi level in the iron-based superconductors [44]. From the stand point of spin-fluctuation-mediated pairing, the absence of $d_{xy}/y_z/x_z$ hole bands is unfavorable for superconductivity, so the absence of superconductivity in the collapsed phase can be naturally understood from this viewpoint [45]. We also note that the band structure of collapsed LaFe_2As_2 is

TABLE I. Wannier spread Ω of the 21-orbital model of uncollapsed LaFe_2As_2 .

		Ω ($\text{\AA}^2$)
Fe	d_{z^2}	1.01
	d_{xz}/yz	1.20
	d_{xy}	1.26
	$d_{x^2-y^2}$	0.97
As	$p_{x/y}$	3.39
	p_z	3.45
	d_{z^2}	4.43
La	d_{xz}/yz	5.31
	d_{xy}	5.72
	$d_{x^2-y^2}$	4.06

similar to that of LaFe_2P_2 , where superconductivity also does not emerge [39,40,46].

Finally, we comment on the superconducting gap structure of LaFe_2As_2 expected from the viewpoint of the orbital component on the FS. One may expect that the electron-electron interaction between the Fe $3d$ and La $5d$ orbitals are weak because they are spatially apart. Therefore, when the Fe $3d$ orbitals play a main role in the occurrence of superconductivity, one can expect that the superconducting gap is small (or vanishing) around the portion of the FS where the La d orbital character is strongly dominating. This may be one reason why T_c is not as high as other 122 or 1111 compounds. Experimental probes are therefore expected to detect some kind of nodal feature in the gap structure.

IV. CONCLUSION

In summary, we calculated the electronic structure of uncollapsed and collapsed LaFe_2As_2 . In both cases, the unique FS is a consequence of the hybridization between the Fe $3d$ and La $5d$ orbitals. The main difference between the uncollapsed and collapsed phases is the presence of the d_{xy} hole FS in the former, which is clearly revealed by hypothetically removing the La $d_{x^2-y^2}$ orbital contribution. The robustness of the d_{xy} FS against heavy electron doping is due to the decrease of the on-site energy of the La/Ba d orbitals when Ba is replaced with La. On the other hand, the Fe-As-Fe bond angle is too large in the collapsed phase, resulting in the disappearance of the d_{xy} hole FS. The correspondence between the absence or presence of the d_{xy} hole FS and absence or presence of T_c can be naturally understood within the spin-fluctuation-mediated pairing scenario. Assuming this scenario, there may be room for further increasing T_c by optimizing the volume of the d_{xy} FS and/or removing the La $5d$ orbital contribution, by, say, partially substituting the La and/or As atoms.

TABLE II. For uncollapsed LaFe_2As_2 , the absolute values of the largest nearest neighbor hopping integrals t from Fe to As, La to As, and Fe to La in each orbital. The nearest neighbor hoppings from La $d_{x^2-y^2}$ to Fe d orbitals are also listed.

	From	To	$ t $ (eV)
Fe	d_{z^2}	As p_z	0.48
	d_{xz}/yz	$p_{x/y}$	0.60
	d_{xy}	p_z	0.63
	$d_{x^2-y^2}$	$p_{x/y}$	0.73
La	d_{z^2}	As p_z	0.48
	d_{xz}/yz	$p_{x/y}$	0.69
	d_{xy}	p_z	0.60
	$d_{x^2-y^2}$	$p_{x/y}$	0.40
La	d_{z^2}	Fe d_{xz}/yz	0.27
	d_{xz}/yz	d_{z^2}	0.29
	d_{xy}	d_{xz}/yz	0.05
	$d_{x^2-y^2}$	d_{z^2}	0.20
		d_{xz}/yz	0.06
		d_{xy}	0.08
		$d_{x^2-y^2}$	0.04

TABLE III. Value of the on-site energy (eV) measured from the Fermi energy for the 21-orbital models of uncollapsed LaFe_2As_2 and BaFe_2As_2 . $\Delta_{\text{La-Ba}}$ is the energy difference between LaFe_2As_2 and BaFe_2As_2 .

		La	Ba	$\Delta_{\text{La-Ba}}$
Fe	d_{z^2}	-0.87	-0.69	-0.12
	$d_{xz/yz}$	-0.55	-0.43	-0.12
	d_{xy}	-0.61	-0.49	-0.12
	$d_{x^2-y^2}$	-0.87	-0.78	-0.09
As	$p_{x/y}$	-1.99	-1.71	-0.28
	p_z	-1.73	-1.59	-0.14
	p_z	-1.73	-1.59	-0.14
La/Ba	d_{z^2}	1.97	2.96	-0.99
	$d_{xz/yz}$	2.69	3.49	-0.80
	d_{xy}	2.59	3.33	-0.73
	$d_{x^2-y^2}$	1.83	2.74	-0.92

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APPENDIX: ADDITIONAL INFORMATION ON THE La-Fe HYBRIDIZATION

We present information regarding the mechanism of La-Fe hybridization, which is the origin of the unique Fermi surface as revealed in the main text, and in particular, the role of the As p orbitals played there. Namely, electrons can move from Fe $3d$ to La $5d$ orbitals through the direct hoppings from Fe to La, but another possible path is via the indirect hoppings from Fe to La through As. In fact, the latter may play an important role because the nearest neighbor distance between La and Fe is larger than that between Fe and As; the nearest neighbor distances are 2.39 Å for Fe-As, 3.20 Å for La-As, and 3.53 Å for Fe-La. In the main text, we have mainly discussed the band structure within the 15-orbital model constructed from the La d and Fe d Wannier orbitals, where the As p orbitals are implicitly included in these Wannier orbitals. Since this model does not suffice to directly understand the role of the As p orbitals, here we have further investigated the role of the As p orbitals by constructing a 21-orbital model, where we explicitly consider not only La and Fe d but also As p orbitals. Parameters obtained for the 21-orbital model are listed in Tables I–III. (Table III shows that the robustness of the d_{xy} Fermi surface is not due to the same mechanism as in hydrogen-doped 1111 systems. See the main text for details.)

We first show the spread of the Wannier orbitals in the 21-orbital model in Table I. It is found that the average of the Wannier spreads (variances) is about 1.1 Å² for Fe d , 3.4 Å² for As p , and 5.0 Å² for La d orbitals. This already

FIG. 5. Band structure and orbital component of the three 21-orbital models of uncollapsed LaFe_2As_2 ; (left) the 21-orbital model constructed from the Fe d , La d , and As p orbitals, (center) the model without considering all the direct hoppings between La and As, and (right) the model without considering all the direct hoppings between La and Fe.

suggests that the large Wannier spread of the La d orbitals likely contributes to the hybridization between La d and Fe d , considering the La-Fe distance of 3.53 Å. This is indeed seen in Table II, where the hoppings from La to Fe or La to As are found to be appreciable.

To clearly understand the effect of the indirect and direct hoppings on the band structure, we further construct two different 21-orbital models, where we neglect the hopping integrals between La and As, or between La and Fe. In Fig. 5, we compare the band structure of the three 21-orbital models. As discussed in the main text, the band splitting of the Fe d_{xy} orbital around -0.2 eV originates from the La-Fe hybridization. When we switch off the hoppings between La and As, the band splitting still appears because of the direct hoppings. In addition, even if we do not consider La-Fe direct hoppings, the band splitting between the La and Fe d orbitals exists because of the mixture through As p orbitals. The magnitude of the band splitting without considering La-Fe and that without the La-As hoppings are almost the same, namely, the indirect and direct hoppings have almost the same contribution on the unique Fermi surface. Thus we conclude that the hybridization between La d and Fe d orbitals originates from both the direct hoppings between La and Fe d orbitals and the indirect hoppings through the As p orbitals.

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that the Fermi surface configuration of LaFe₂As₂ is essentially similar to those of conventional iron-based superconductors, our band structure calculation can be considered as implicitly taking into account the magnetic effect in the employed structural parameters. In fact, recently, we came to know that I. I. Mazin, M. Shimizu, N. Takemori, and H. O. Jeschke in [arXiv:1905.06190](https://arxiv.org/abs/1905.06190) show that the stripe-type antiferromagnetic order is required to reproduce the structural parameters of LaFe₂As₂ within DFT calculation.

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