First-Principles Study for the Anisotropy of Iron-based Superconductors toward Power and Device Applications

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Performing the first-principles calculations, we investigate the anisotropy in the superconducting state of iron-based superconductors to gain an insight into their potential applications. The anisotropy ratio \(\gamma\) of the \(c\)-axis penetration depth to the \(ab\)-plane one is relatively small in \(\text{BaFe}_2\text{As}_2\) and \(\text{LiFeAs}\), i.e., \(\gamma \approx 3\), indicating that the transport applications are promising in these superconductors. On the other hand, in those having perovskite type blocking layers such as \(\text{Sr}_2\text{ScFePO}_3\) we find a very large value, \(\gamma \gtrsim 200\), comparable to that in strongly anisotropic high-\(T_c\) cuprate \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8\). Thus, the intrinsic Josephson junction stacks are expected to be formed along the \(c\)-axis, and novel Josephson effects due to the multi-gap nature are also suggested in these superconductors.

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Since the discovery of iron-based superconductor \(\text{LaFeAsO}_{1-x}\text{F}_x\) with \(T_c = 26\text{K}\) by Kamihara \textit{et al.}\cite{1}, high-\(T_c\) superconductivity has been reported in some of its family materials. All the superconductors in this family have \(\text{FePn}\) layers (\(\text{Pn} = \text{P, As, or Se}\)), in which the \(\text{high-}\text{\(T_c\)} superconductivity emerges, and are classified into five groups according to the non-superconducting blocking layers sandwiched between \(\text{FePn}\) layers as seen in Fig. 1, i.e., 1111 (e.g., \(\text{LaFeAsO}\)), 122 (e.g., \(\text{BaFe}_2\text{As}_2\))\cite{2}, 111 (e.g., \(\text{LiFeAs}\))\cite{3}, 11 (e.g., \(\text{FeSe}\))\cite{4}, and the other type that contains very thick perovskite-based blocking layers (\(\text{Sr}_2\text{ScFePO}_3\))\cite{5}.

The quasi two-dimensionality of the electronic states due to the layer structure of the iron-based superconductors is significant for their applications, since it causes anisotropy in superconducting properties such as the critical current \(J_c\) and the upper and lower critical fields, \(H_c2\) and \(H_c1\). A convenient quantity characterizing the superconducting anisotropy is the anisotropy parameter \(\gamma\) defined as the ratio of the \(c\)-axis penetration depth to the in-plane one. From the values of \(\gamma\), one can extract important information about their potential applications. For example, superconductors with small \(\gamma\) are suitable for power applications using superconducting wires, tapes or cables. On the other hand, those with extremely large \(\gamma\) are expected to work as the intrinsic Josephson junctions\cite{6, 7}, which will be promising as a THz emission device\cite{8, 9}.

It is well known that the penetration depth anisotropy \(\gamma\) directly leads to the anisotropy of \(H_c2, H_c1\) and \(J_c\) in single-band superconductors. In the multi-band systems such as the iron-based superconductors their relations become much more complicated and are still controversial\cite{10}. But, in any case the parameter \(\gamma\) describes the scale of the superconducting anisotropy. Thus, in this paper we perform a systematic first-principles study for \(\gamma\) in the whole groups of iron-based superconductors and elucidate the origin of the general trend in the superconducting anisotropy observed in various measurements such as torque, microwave surface impedance, \(J_c\), etc. \cite{11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21}. Though accurate values of \(\gamma\) have not yet been settled experimentally, i.e., the reported values are still scattered, we find a clear trend in \(\gamma\) among the five groups of the iron-based superconductors. Moreover, since we can predict the anisotropy ratio of a superconductor in which single crystals are not available, our method presented in this paper will provide a powerful tool for the exploration of the superconducting materials suitable for each application.

Our calculations reveal that the anisotropy ratio \(\gamma\) depends sensitively on the nature of the blocking layers and its values are widely distributed as seen in experiments, e.g., \(\gamma \sim 3\) in the superconductors belonging

![FIG. 1: Crystal structures of typical iron-based superconductors.](image-url)
to the 111 and 122 groups and $\gamma_\lambda \gtrsim 200$ in those with perovskite-based blocking layers. These results remind us of the variety in high-$T_c$ cuprates, e.g., $\gamma_\lambda \sim 8$ in YBa$_2$Cu$_3$O$_{6-\delta}$ and $\gamma_\lambda > 100$ in Bi$_2$Sr$_2$CaCu$_2$O$_{6-\delta}$ (Bi-2212) [21]. It is noted that the anisotropy in the 111 and 122 groups is small in spite of the layer structure. The large value of $\gamma_\lambda$ in the group with perovskite-based blocking layers strongly suggests the formation of the intrinsic Josephson junction stacks along the $c$-axis.

In this paper, we evaluate $\gamma_\lambda$ for the typical iron-based superconductors from numerical results for the anisotropy ratio of the normal state resistivity at $T = 0K$, $\gamma_\rho(0)$ obtained by means of the first-principles calculations for the electronic states [22], that is, the superconducting anisotropy originating from the band-structure dependent Fermi velocities is considered. Let us first summarize the method employed in this paper to evaluate the anisotropy parameters, $\gamma_\rho$ and $\gamma_\lambda$. Assuming that the relaxation time of the conduction electrons is isotropic and independent of their velocities, we consider the quantity $\gamma_\rho(0) = (v_F^2)^2/(v_F^2)^{FS}$ to be equal to the ratio of the $c$-axis resistivity to the $a$-axis one. Here, $v_a$ and $v_c$ are the Fermi velocities, respectively parallel and perpendicular to the FePn layers and $\langle \cdots \rangle_{FS}$ denotes the average on the Fermi surface [23]. The Fermi velocities are derived from the derivative of the band energy, $v_i = \partial \epsilon_k / \partial k_i$, where the band energy $\epsilon_k$ is calculated by the first-principles calculations. In our calculations we use the first-principles density functional package VASP [24], which adopts GGA exchange-correlation energy [25] and PAW method [26]. The electron self-consistent loops to obtain the charge density are repeated until the total energy difference becomes smaller than $10^{-6}$ eV. In these loops the spacing between nearest-neighbor $k$-points is taken to be $0.1$–$0.2$ Å$^{-1}$. Once the charge density is determined, the energy bands are again calculated for more $k$-points located with a much finer spacing, $79 \times 79$, in the $xy$-plane (the number of $k$-points along the $z$-axis depends on the compounds), in order to determine the Fermi surfaces and the Fermi velocities.

The average $\langle \cdots \rangle_{FS}$ is calculated by the standard tetrahedron method. In obtaining the anisotropy of the penetration depth at zero temperature, we utilize the simplest relation, $\gamma_\rho(0) = \sqrt{\gamma_\rho(0)}$ [27], which is convenient for systematic and comparative studies on the superconducting anisotropy among various iron-based superconductors, though its validity is limited in the multi-band systems [27]. The compounds that we examine in this paper are FeSe, LaFeAsO, LaFePO, BaFe$_2$As$_2$, LiFeAs, and Sr$_2$ScFePO$_3$. The calculations are done for tetragonal undoped compounds without any magnetic order, except for BaFe$_2$As$_2$. We calculate the electronic structures based on the observed crystalline structures, that is, the crystalline structures are not optimized, since it is known that the optimized structure deviates from the observed one in iron-based superconductors [21].

The numerical results for the anisotropy parameters, $\gamma_\rho$ and $\gamma_\lambda$, in the iron-based superconductors employed in this paper are summarized in Table I. Let us discuss the 1111-system, first. The experimental values in this group are widely distributed, depending on the compounds [11, 13, 15, 16, 17, 20, 21]. For example, $\gamma_\lambda \sim 15$–$20$ in ReFeAsO ($\text{Re} = \text{Nd, Sm}$) [11, 15, 21], while $\gamma_\lambda \sim 3$ in PrFeAsO [20]. Thus, we notice that there are two groups in the 1111-system, i.e., the one with relatively large anisotropy and the other with moderate anisotropy. In our calculations, we obtained $\gamma_\lambda(0) \sim 10.81$ for LaFeAsO, as shown in Table I. This value is close to that in the group with relatively large anisotropy. We also performed the calculations for stoichiometric PrFeAsO and obtained $\gamma_\lambda(0) \sim 8.6$, which is smaller than that in LaFeAsO as expected and also not far from the observed one in PrFeAsO$_{1-y}$. This difference between LaFeAsO and PrFeAsO can be mainly attributed to the difference of the main elements inside the blocking layers. On the

| $a$ [Å]  | $c$ [Å]  | $h_{F_{Pn}}$ [Å] | $\gamma_\rho(0)$ | $\gamma_\lambda(0)$ |
|--------|--------|------------------|------------------|-------------------|
| FeSe   | 3.7738 | 5.5248           | 1.4652           | 18.44             | 4.29             |
| LiFeAs | 3.7914 | 6.3639           | 1.6769           | 9.06              | 3.01             |
| BaFe$_2$As$_2$ | 3.9625 | 13.0168         | 1.3602           | 10.69             | 3.27             |
| LaFePO | 3.9636 | 8.5122           | 1.1308           | 17.34             | 4.16             |
| LaFeAsO| 4.020  | 8.7034           | 1.3238           | 116.8             | 10.81            |
| Sr$_2$ScFePO$_3$ | 4.016 | 15.543          | 1.1984           | $6.19 \times 10^5$ | 248              |

FIG. 2: The Fermi surfaces of (b) Sr$_2$ScFePO$_3$, (c) antiferromagnetic BaFe$_2$As$_2$, (d) LaFePO, (e) FeSe, (f) BaFe$_2$As$_2$, (g) LaFeAsO, and (h) LiFeAs. The contrast on the surfaces indicates the magnitude of the Fermi velocity. The inset (a) shows typical symmetry points.
other hand, it is also noted that the calculated value in non-arsonic 1111-compound LaFePO ($\gamma_\lambda(0) = 4.16$) is much smaller than that of LaFeAsO, though the crystal structures are equivalent in both compounds. The Fermi surface shapes are different in these two compounds as seen in Figure 2(d) and (g), that is, the Fermi surfaces in LaFeAsO are almost cylindrical, while those in LaFePO are not, indicating that the two-dimensionality is weak in LaFePO. From these results, one understands that the two-dimensionality differs among the 1111 compounds even if the blocking layers are equivalent. This remarkable feature comes from the fact that the band is weak in LaFePO, indicating that the two-dimensionality is weak in this compound.

We also calculated the anisotropy parameter

$$\lambda(0) = \frac{\rho_{\parallel}}{\rho_{\perp}}$$

in the AFM phase as seen in Fig.2(c). These results clearly demonstrate the advantage of the present evaluation technique. In Table I we also list the anisotropy parameters of LiFeAs in the 111-system. The $\gamma_\lambda$ value is comparable to BaFe$_2$As$_2$, that is, the 111-system is also suitable for transport applications. This low anisotropy basically reflects the winding features of the Fermi surface like 122-systems as seen in Fig.2(h).

In the 111-compounds FeSe, which has the simplest crystal structure composed of a stack of only FePn layers, the Fermi surfaces calculated in this compound shows clear curvature along the $z$-direction as seen in Fig.2(e), indicating that the two-dimensionality is weak in this system. In fact, we obtained a relatively small value, $\gamma_\lambda(0) = 4.29$, in the superconducting state. However, we note that this value is larger than those in the 122 and 111 systems and rather close to that in LaFePO. From this result one understands that metallic ions in the blocking layers enhance the three dimensionality in the 122 and 111 systems. We emphasize that only the first-principles calculations can systematically derive such delicate material-dependent difference in the anisotropy.

Now, we discuss the anisotropy in the final group, i.e., the newly discovered iron-based superconductors with perovskite-type blocking layers, which are now under intensive exploration. The superconductors in this group do not show any magnetic order at all, while the superconducting transition temperature is rather high. In this paper we focus on a typical compound, Sr$_2$ScFePO$_3$, which shows the highest $T_c$ at ambient pressure among non-arsonic iron-based superconductors. Since the blocking layers in this compound are very thick as shown in Fig.1, strong anisotropy is expected in the normal state resistivity. In fact, a huge value $\gamma_\rho \sim 10^6$ is obtained, which is several hundred times larger than that in the other groups, i.e., 1111-, 122-, and 11- compounds. From this normal state value it follows, $\gamma_\lambda(0) \sim 250$, for the anisotropy in the London penetration depth of Sr$_2$ScFePO$_3$. Note that the value is comparable to that in Bi-2212 in high-$T_c$ cuprates. From this fact one may infer that the c-axis transport in this system is brought about by the electron tunneling between neighboring FePn layers, as in Bi-2212. In the superconducting state the Cooper-pair tunneling will be also expected to occur between the superconducting FePn layers as in Bi-2212, that is, a single crystal of Sr$_2$ScFePO$_3$ may be regarded as a stack of nano-scale Josephson junctions schematically shown in Fig.3 which is called the intrinsic Josephson junctions. In the intrinsic Josephson junctions the $I-V$ characteristics under no external magnetic field show a remarkable feature, i.e., the multiple branch structure composed of many $I-V$ curves, the number of which corresponds to that of stacked intrinsic SIS junctions as shown in Fig.3. Hence, we naturally expect that the multiple-branch structure appears in the c-axis

![FIG. 3: A schematic illustration of the intrinsic Josephson junction stacks in Sr$_2$ScFePO$_3$. An alternate stack of FePn (superconducting) and blocking layers (insulating) are regarded as a series of S-I-S Josephson junctions. $\varphi_i$ stands for the phase of the superconducting order-parameter of the $i$th electron energy band. The arrows represent possible tunneling channels.](image-url)
I − V characteristics in the superconducting state in a single crystal of Sr$_2$ScFePO$_3$. As for the damping nature in the Josephson effects, this system should be underdamped one, because the paring symmetry is expected to be mainly full gapped s-wave one. It is also predicted that a new oscillation mode exists in the phase differences in addition to the Josephson plasma [34]. Moreover, the Josephson vortex has an internal structure [35]. These new features originate from the multi-tunneling channels due to the multi-gap nature in these superconductors as shown in Fig.3. The existence of the new phase oscillation modes will enrich the physics in the intrinsic Josephson junctions expected in the new compounds.

We also mention that the superconducting Sr$_2$ScFePO$_3$ is a stoichiometric compound, that is, the superconductivity appears without doping. Then, it will be easier to make a stack of homogeneous intrinsic Josephson junctions in a large-scale, which is contrasted with the Bi-2212 intrinsic Josephson junctions, in which the doping for getting the high-$T_c$ superconductivity brings about disorders.

Finally, we summarize our numerical results for $\gamma_\lambda$ vs. distance between adjacent iron-planes in Fig.4. From this figure one finds a rough trend between the distance and the anisotropy, which indicates that the anisotropy increases with increasing the distance. However, our results also suggests the existence of the other factors that affect the penetration depth anisotropy. As discussed in this paper, the anisotropy is sensitive to the distance between the pnictogen ions and the Fe-plane, and also the elements forming the blocking layers. From these results we claim that the method presented in this paper, using the first-principles calculations, is a powerful tool for evaluating the superconducting anisotropy.

In summary, we investigated systematically the anisotropy of the London penetration depth for various iron-based superconductors on the basis of the first-principles calculations for their electronic states. It was shown that the anisotropy parameters, $\gamma_\rho$ and $\gamma_\lambda$, are strongly dependent on the structure of the blocking layers and pnictogen ions situated next to Fe layers. Our numerical results well explain the variety of the $\gamma_\lambda$ values observed in these superconductors. The 122- and the 111-compounds show weak anisotropy, being enough for transport power applications, while those with perovskite-type blocking layers have a very large value of $\gamma_\lambda$, which is comparable to that in Bi-2212. We predict that the intrinsic Josephson junction stacks are realized in the superconducting state of these compounds.

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