Nonhelical spin texture in the normal states of the centrosymmetric superconductor $\beta$-PdBi$_2$

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A nonhelical spin texture was observed in the centrosymmetric superconductor $\beta$-PdBi$_2$ using an imaging-type spin- and angle-resolved spectroscopy. The observed surface and bulk states near the Fermi energy are found to be spin polarized with nonhelical textures. First-principles calculations and effective models show that the inter- and intralayer hoppings account for the complex spin texture and thus play a significant role in the spatial distribution of the layer-locked spin states. Our work not only provides insights into the local spin states in inversion-symmetric systems but also paves the way to the identification of the nature of superconducting pairing in the presence of a complex spin texture in $\beta$-PdBi$_2$ and the related centrosymmetric superconductors.

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The realization of controllably creating and manipulating the spin polarization in the absence of an external magnetic field is a key subject in the research of spintronics [1]. The traditional methods to stabilize spin-polarized states in nonmagnetic systems are mostly associated with global inversion-asymmetric structures [2,3] to break Kramers degeneracy. Recently, it has been suggested that atomic site asymmetry may induce local spin-polarized states while global inversion symmetry is preserved [4]. This understanding leads to the identification of the hidden spin states in centrosymmetric systems [5–16]. While the total electronic states are spin degenerate, states with different spin polarizations can be spatially localized on different atomic sites. The research on the local spin states has attracted attention as it not only extends the candidate spintronic systems to the centrosymmetric systems, but has also shown the promising possibility to control spin states by tuning local inversion asymmetry with atomic operations [17].

The local spin states have been observed and studied in several transitional metal dichalcogenide compounds [5–10,13], cuprates [12,14], iron-based superconductors [15], and some other compounds [11]. However, previous reports are mostly focused on the experimental fact and methods to observe spin polarization in nonmagnetic centrosymmetric materials. There have not been many discussions on the origin and spatial distribution of the detailed local spin texture, which is probably due to the lack of an efficient probing method of local spin states.

$\beta$-PdBi$_2$ is a superconductor with the centrosymmetric space group $I4/mmm$. It has drawn much attention as a candidate of topological superconductors, which have the potential to be used as the building block of quantum computers [10,18–26]. Recent studies suggest that the superconducting pairing in $\beta$-PdB$_2$ may be influenced by the local inversion asymmetry [22,26,27]. Thus, it is crucial to study the influence of the local inversion asymmetry on the electronic states in $\beta$-PdBi$_2$, which can be reflected by the local spin-polarized states.

In this Rapid Communication, we studied the spin texture in the normal state of $\beta$-PdBi$_2$ in the vicinity of the Fermi energy ($E_F$) by utilizing a high-efficiency image-type spin- and angle-resolved photoemission spectroscopy (SARPES). Large spin polarization was observed not only in the surface, but also in the bulk states. Moreover, some of the bulk states and even surface states can hold a nonhelical spin texture, with reversed chiralities along the $\Gamma$-$M$ and $\Gamma$-$X$ directions. First-principles calculations and effective models show that the local inversion asymmetry induces local spin states. Mediated by inter- and intralayer hoppings and the surface Rashba effect, the local spin states form a complex spin-polarized Fermi surface with nonhelical spin textures spatially localized on different atom layers. Our findings have not only revealed the spatial distribution of spin states in $\beta$-PdBi$_2$ and the crucial role of inter- and intralayer hoppings in the local spin physics, but also provide information for the clarification of the superconducting pairing mechanism in $\beta$-PdBi$_2$ and related centrosymmetric superconductors.

Spin-integrated and spin-resolved angle-resolved photoemission spectroscopy (ARPES) measurements were
FIG. 1. Spin-polarized electronic states in β-PdBi₂. (a) Momentum distribution curve (MDC) along the $\bar{M}$-$\Gamma$-$\bar{M}$ direction obtained by integrating the intensity of $\sim$0.05 to 0 eV energy regions (upper panel) and the spin-integrated ARPES image (lower panel). (b) Calculated electronic structure on the (001) surface. The bulk and surface states are marked as $\alpha$, $\beta$, $\gamma$, $\delta$, S1, S2, and S2', and indicated by shadowed areas and black lines, respectively. (c) The MDC and spin-integrated ARPES image along the $\bar{X}$-$\bar{\Gamma}$-$\bar{X}$ direction. (d) The spin-resolved MDCs along the $\bar{M}$-$\Gamma$-$\bar{M}$ direction with the in-plane spin component perpendicular to the momentum. (e) In-plane spin polarization along the $\bar{M}$-$\Gamma$-$\bar{M}$ direction with the spin component perpendicular (parallel) to the momentum is shown in the up (down) panel. (f) Indication of the experimental spin polarizations near $E_F$. Blue and red arrows are used to indicate the clockwise and anticlockwise spin chiralities, respectively. (g) The spin-resolved MDCs along the $X$-$\bar{\Gamma}$-$X$ direction with the in-plane spin component perpendicular to the momentum. (h) In-plane spin polarization along the $\bar{X}$-$\bar{\Gamma}$-$\bar{X}$ direction with the component perpendicular (parallel) to the momentum is shown in the upper (lower) panel.

performed with a home-made spin polarimeter consisting of a Scienta R3000 analyzer fitted with a multichannel very low electron energy (MCVLEED) spin detector [28] using a He I $\alpha$ light source. The home-made MCVLEED spin detector enables a parallel detecting window of $\sim$11° × 0.3 eV, with an effective Sherman function $\sim$0.25 and a detecting efficiency over $10^5$ times higher than a commercial Mott polarimeter. The energy and angular resolution were set to $\sim$35 meV and $\sim$0.255°, respectively. This home-made spin polarimeter is more readily able to measure the in-plane than the out-of-plane spin component. Since the in-plane inversion symmetry is always maintained in β-PdBi₂, the spin texture induced by spin-orbital coupling (SOC) is expected to be aligned in plane. Single-crystal β-PdBi₂ samples were prepared by a melting method [29]. The superconducting transition temperature $T_C$ is $\sim$5.3 K as shown in Fig. S2(a) in the Supplemental Material [29]. The samples were cleaved in situ under an ultrahigh vacuum of better than $1 \times 10^{-10}$ Torr and were held at $\sim$6 K during the experiments.

The experimental ARPES images along the $\bar{M}$-$\Gamma$-$\bar{M}$ and $\bar{X}$-$\bar{\Gamma}$-$\bar{X}$ directions are shown in Figs. 1(a) and 1(c). The bulk and surface states near $E_F$ are distinguished by comparing with the calculated band structure shown in Fig. 1(b) [29], and marked as $\alpha$, $\beta$, $\gamma$, $\delta$, S1, S2, and S3 in Figs. 1(a) and 1(c), respectively. The observed electronic dispersions are in agreement with previous experiments [19] with a hole pocket $\alpha$ near the $\Gamma$ point and electron pockets $\gamma$ and $\delta$ near the $\bar{M}$ and $\bar{X}$ points, respectively.

Now we focus on the spin texture in the vicinity of $E_F$. The spin-resolved MDCs and the SARPES images along the $\bar{M}$-$\Gamma$-$\bar{M}$ direction with the in-plane spin component perpendicular and parallel to the momentum are shown in Figs. 1(d) and 1(e), respectively. Though the crystal structure of β-PdBi₂ is centrosymmetric, obvious spin polarization is observed not only in the surface states but even in the bulk states with the in-plane spin component perpendicular to the momentum. The bulk states $\alpha$ and $\beta$ are spin polarized along the same direction opposite to that of the bulk $\gamma$ states and the surface states S1 and S2, which cannot be explained solely by the surface Rashba effect.

Then we turn to the in-plane spin texture along the $\bar{X}$-$\bar{\Gamma}$-$\bar{X}$ direction as shown in Figs. 1(g) and 1(h). No obvious spin polarization is observed in the bulk band $\delta$ while the rest of the bulk and surface states are spin polarized with the spin component perpendicular to the momentum. It is noted that the bulk band $\beta$ and surface band S2 hold spin polarizations opposite to those along the $\Gamma$-$\bar{M}$ direction.

Figure 1(f) concludes the observed spin texture. Blue and red arrows indicate the clockwise and anticlockwise spin chiralities, respectively. Most of the surface and bulk bands near $E_F$ are spin polarized except for the $\delta$ band. It is noted that not only the bulk state $\beta$ but even the surface state S2, which is usually considered to have a helical spin texture due to the electrostatic potential gradient on the surface, host a nonhelical spin texture. Moreover, the four bulk bands $\alpha$, $\beta$, $\gamma$, and $\delta$ hold spin textures different from each other near $E_F$, which suggests a complicated origin of the observed spin polarizations.

To study the local spin polarization in centrosymmetric systems, we built a tight-binding model inspired by previous reports [30,31]. The structure of β-PdBi₂ is shown in Fig. 2(a).
The Pd-site point group ($D_{6h}$) is centrosymmetric while the Bi-site point group ($C_{4v}$) is not. Thus, only the SOC-induced local spin splitting on the Bi sites needs to be considered. Moreover, since the in-plane inversion symmetry is always maintained in β-PdBi$_2$, we only consider the inversion asymmetry out of plane and divide the Bi sites within the unit cell into four Bi layers indicated by red dotted squares in Fig. 2(a). The two-dimensional Rashba Hamiltonian of a Bi layer is given as

$$H_R = -\frac{\hbar^2}{2m} \nabla^2 + \frac{\hbar^2 E_{in}}{4m^2 c^2} (\sigma_x k_x - \sigma_y k_y) = -\frac{\hbar^2}{2m} \nabla^2 + \alpha_R (\sigma_x k_x - \sigma_y k_y),$$

where $m$, $c$, $\sigma_x$, $\sigma_y$, and $E_{in}$ are the mass of electron, speed of light, Pauli matrices, and the local dipole field directed out of the plane, respectively. The energy splitting and the wave functions are given as

$$E_k = E_k \pm \alpha_R |k|,$$

$$\psi_{\pm} = \frac{1}{\sqrt{2}} \left( \begin{pmatrix} \pm e^{i\phi} \nabla \theta \\ -k \end{pmatrix} \right) = \frac{1}{\sqrt{2}} \left( \begin{pmatrix} \pm e^{i\phi} \\ -k \end{pmatrix} \right),$$

where $E_k$ is the on-site energy. The $\psi_{\pm}$ states hold in-plane helical spin polarization with opposite chiralities.

Then we introduce the Hamiltonian for the whole unit cell of β-PdBi$_2$ as shown in Fig. 2(b),

$$H_{uc} = \begin{pmatrix} H_{R+} & T_{inter} & 0 & T_{intra} \\ T_{inter} & H_{R-} & T_{intra} & 0 \\ 0 & T_{intra} & H_{R+} & T_{inter} \\ T_{intra} & 0 & T_{inter} & H_{R-} \end{pmatrix},$$

$$H_{R\perp} = \begin{pmatrix} E_k & \pm \alpha_R (k_x - i k_y) \\ \pm \alpha_R (k_x - i k_y) & E_k \end{pmatrix},$$

$$T_{intra} = \begin{pmatrix} t_{intra} & 0 \\ 0 & t_{intra} \end{pmatrix},$$

$$T_{inter} = \begin{pmatrix} t_{inter} & 0 \\ 0 & t_{inter} \end{pmatrix},$$

where $H_{R\perp}$, $T_{intra}$, and $T_{inter}$ correspond to the Bi layers with opposite local dipole field, and the hopping terms between adjacent Bi layers via the Pd layers and via the van der Waals gap, respectively. When the hopping term is isotropic, $t_{intra} = t_{inter}$, by solving Eq. (2), it can be proved that no extra spin splitting would be induced, and the spin textures of the band with $E_k + |\alpha_R|k|$ in each layer are indicated in Fig. 2(c). When the hopping terms are not equal, for example, when $t_{inter} \gg t_{intra}$, the influence of $t_{intra}$ can be ignored, and the fourfold degenerate eigenvalues and the wave functions are given by solving Eq. (2),

$$E_k^\pm = E_k \pm \sqrt{\alpha_R^2 k^2 + t_{inter}^2},$$

$$\psi_+^\pm = (\sqrt{1-D_{k^2}^2} \psi_+^{(1)} + D_{k^2} \psi_+^{(2)}), (D_{k^2} \psi_+^{(1)} + \sqrt{1-D_{k^2}^2} \psi_+^{(2)}),$$

$$\psi_-^\pm = (-D_{k^2} \psi_+^{(1)} + \sqrt{1-D_{k^2}^2} \psi_+^{(2)}), (\sqrt{1-D_{k^2}^2} \psi_+^{(1)} - D_{k^2} \psi_+^{(2)}),$$

$$D_{k^2} = t_{inter} \left[ (\sqrt{\alpha_R^2 k^2 + t_{intra}^2 + \alpha_R |k|} \right]^{-1/2}. (3)$$

When the hopping between the van der Waals layers is also weak, $t_{inter} \ll |\alpha_R|k|$, the spin polarizations in the four Bi layers remain the two-dimensional Rashba type as indicated in Fig. 2(c). However, when the hopping between van der Waals layers is strong, $t_{inter} \gg |\alpha_R|k|$, the spin polarization in each layer will be reversed as indicated in Fig. 2(d). The situation $t_{intra} \gg t_{inter}$ can also lead to a similar solution. The unequal local hopping term tends to redistribute or even reverse the local spin states. Yet, there are some differences between the situations $t_{intra} \gg t_{inter}$ and $t_{intra} \gg t_{inter}$. For one thing, when $t_{intra} \gg t_{inter}$, the hopping term tends to hybridize the electron states within the single PdBi$_2$ layer while when $t_{inter} \gg t_{intra}$, electron states on the two sides of the van der Waals gaps are hybridized. For another, the large $t_{intra}$ term also comes with a small local dipole field $E_{in}$, which results in a small local spin polarization.

To identify the relative distribution of $t_{intra}$ and $t_{inter}$ near $E_F$, first-principles calculations on the PdBi$_2$ slabs were carried out. Figure 3(a) is the calculated electronic structure along the $X^{-1}M$ direction. The contributions of Bi and Pd elements are indicated by purple and green, respectively. It is noted that the electronic states near $E_F$ indicated by the red dotted square in Fig. 3(a) are mostly composed by Bi. Moreover, for Bi layers, $t_{inter} (t_{intra})$ tend to hybridize the electronic states in the adjacent Bi (Pd and Bi) layers, which tends to lead to a 100% (50%) Bi contribution in the electron states. Thus, the distribution of $(t_{inter} - t_{intra})$ near $E_F$ can be estimated by the distribution of the Bi contribution near $E_F$. The distribution of $(\text{Bi}\% - 100\% + 50\%)$ near $E_F$ is shown in Fig. 3(b). The purple and green regions suggest the regions with $t_{inter} > t_{intra}$ and
t_{\text{inter}} < t_{\text{intra}}$, respectively. The electronic states marked with white are the states with a similar strength of $t_{\text{inter}}$ and $t_{\text{intra}}$. The calculated spin textures in the first six Bi layers with the in-plane spin component perpendicular to the momentum are shown in Figs. 3(c)–3(h). The calculated spin texture of the first Bi layer is consistent with previous studies [10], which confirms the validity of our calculations. The summation of the calculated results in the first two Bi layers is in accordance with our experimental results, which can be understood from the $\sim 7$ Å mean free path of 21 eV photoelectrons.

With the calculated layer-locked spin polarization and the model explained in Eq. (2), we can illustrate the spin-polarized states in $\beta$-PdBi$_2$ one by one. The $\omega$ band near $E_F$ is almost purely from Bi as shown in Fig. 3(a), which indicates small $t_{\text{intra}}$. Moreover, considerable spin polarization in the $\alpha$ band appears from the second Bi layer where the van der Waals gap appears as shown in Figs. 3(c)–3(h). These suggest a dominant role of $t_{\text{inter}}$ in the local spin polarization of the $\alpha$ bands which is consistent with Fig. 3(b). For the $\beta$ bands, as shown in Fig. 3(a), Pd also contributes to the electronic states and the influence of $t_{\text{intra}}$ needs to be considered. Along the $\Gamma$-$M$ direction, the first three Bi layers possess similar spin polarization shown in Figs. 3(c)–3(e), which suggests that the surface Rashba effect plays an important role here. The rest of the layers and the textures along the $\Gamma$-$X$ direction hold the local Rashba-type behavior as shown in Figs. 3(c)–3(h), which is due to the similar strength of $t_{\text{inter}}$ and $t_{\text{intra}}$ as shown in Fig. 3(b). The dominant roles of the surface Rashba effect along the $\Gamma$-$M$ direction and the local Rashba effect with a similar strength of $t_{\text{inter}}$ and $t_{\text{intra}}$ along the $\Gamma$-$X$ direction are responsible for the observed nonhelical spin texture of the $\beta$ band. For the $\gamma$ band, the calculations show a local Rashba-type behavior along the $\Gamma$-$X$ direction but a reversed spin polarization along the $\Gamma$-$M$ direction as shown in Figs. 3(c)–3(h), which is related to the large hopping with Pd near $E_F$ along the $\Gamma$-$M$ direction as shown in Fig. 3(b) and thus $t_{\text{intra}}$ plays an important role. It is noted that, for the $\gamma$ band along the $\Gamma$-$M$ direction, there is energy splitting between the spin-polarized states contributed by the first two layers and the spin-polarized electronic states in the second layer are mostly unoccupied as shown in Figs. 3(c) and 3(d).

Thus, the experimentally observed spin-polarized states along the $\Gamma$-$M$ direction are mostly from the first Bi layer, while along the $\Gamma$-$X$ direction, the observation comes from the first two Bi layers, which host opposite spin polarizations and the summation is along the direction antiparallel to that along the $\Gamma$-$M$ direction. For the $\delta$ band, no obvious spin polarization is observed in the experimental and calculated results, which is related to the large hopping between Bi and Pd as shown in Fig. 3(b) that leads to a small local dipole field $E_{\text{in}}$.

For the surface states, the calculation is in agreement with the experimental results that the chirality of spin polarization of S2 near $E_F$ is opposite along the $\Gamma$-$M$ and $\Gamma$-$X$ directions as shown in Figs. 1(f), 3(c) and 3(d). From the calculation, it is noted that the surface states near $E_F$ along the $\Gamma$-$M$ direction, S1 and S2, hold the same spin polarization in the first six Bi layers, while the surface state along the $\Gamma$-$X$ direction, S2’, possesses a layer-dependent spin polarization showing the contribution from bulk states and the effect of hopping terms, which suggests that the observed S2’ is the surface resonant state. From our calculations, it is originated from the hybridization of S2, $\beta$, and $\gamma$ bands, which is also consistent with the calculated spatial distribution in previous reports [23]. The formation of the surface resonant state along the $\Gamma$-$X$ direction leads to the nonhelical spin texture of S2 in our experiments.

In conclusion, we have observed a nonhelical spin texture near $E_F$ in the normal state of the centrosymmetric superconductor $\beta$-PdBi$_2$ with an image-type SARPES. Not only the surface states but also the bulk states are spin polarized with the direction of the polarization helical and nonhelical on the Fermi surface. With an effective model and first-principles calculations, we reveal that the observed spin-polarized bulk states are originated from an atomic dipole field as well as the surface Rashba effect, and are mediated by the anisotropic distributed inter- and intralayer hoppings in the momentum space, which leads to spatially localized spin polarizations on different atomic sites. Moreover, the formation of the surface resonant state S2’ along the $\Gamma$-$X$ direction leads to the nonhelical spin-polarized surface state. Our work not only reveals the complex spin texture in $\beta$-PdBi$_2$ and illustrates the important role of interlayer and intralayer hoppings in the spatial distri-
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bution of the local spin states, but also provides a foundation for further discussions on the superconducting pairing in β-PdBi2 and related centrosymmetric superconductors.

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