

学位論文要旨

**Spin- and angle-resolved photoemission spectroscopy
studies of Ln(O, F)BiS₂ (Ln = La, Ce, Pr, Nd) superconductors**(スピンおよび角度分解光電子分光による Ln(O, F)BiS₂ (Ln = La, Ce, Pr, Nd) 超伝導体の研究)氏名 **Wu Shilong** **ABSTRACT**

According to the Kramers theorem, spatial inversion symmetry $E(k, \uparrow) = E(-k, \uparrow)$ and time-reversal symmetry $E(k, \uparrow) = E(-k, \downarrow)$ result in $E(k, \uparrow) = E(k, \downarrow)$, guarantee that the electronic states of non-magnetic centrosymmetric materials must be spin degenerate. Namely, a momentum-dependent spin-split state usually comes from global inversion asymmetry of system. However, recent theoretical formalism reconstructed the traditional Rashba and Dresselhaus effects in atomic scale [1]. Microscopically, atomic site that belongs to a non-centrosymmetric point group can carry either a local dipole field or site inversion asymmetric crystal field, inducing local Rashba effect (denoted as R-2) or local Dresselhaus effect (denoted as D-2), respectively. The theoretical works suggested that LaOBiS₂ and the related compounds can be such systems possessing R-2 and/or D-2 due to the breaking of local inversion symmetry in each BiS₂ bilayer and the opposite polar fields caused by ionic bonding between (BiS₂)⁻ bilayer and (La₂O₂)²⁺ layer. Since the projected local spin polarization on each real-space sector of BiS₂ bilayer in LaOBiS₂ crystals holds opposite orientation, so called spin-layer locking effect, has been theoretically predicted in the LaOBiS₂ film at first which could offer advantages for the design of new generation of spin-field effect transistors (SFET) [2]. Theoretical study of either film or bulk LaOBiS₂ further pointed out that spin texture of conduction band at each X point in Brillouin zone must be non-helical originating from D-2 effect whereas the valence band possesses helical spin texture originating from R-2 effect [1]. Moreover, with electron doping by substitution of oxygen with fluorine, LaO_{1-x}F_xBiS₂ is manifested as one of BiS₂-based superconductors with similar properties of cuprate superconductors such as rather high value of $2\Delta/k_B T_C$ and novel Cooper pairing symmetry. The system exhibits the highest superconducting critical temperature (T_C) of 11 K at $x \sim 0.5$ among all the BiS(Se)₂ based superconductors [3], hence serves an excellent platform to combine superconductivity with local Rashba effect that could lead to the unconventional mechanism of Cooper pairing. Note that, heretofore, Rashba superconductors with mixed singlet and triplet pairings have been limited to non-centrosymmetric compounds or surface systems. The evidence of spin-polarized states caused by SOI and the breaking of local inversion symmetry on LaOBiS₂ or family compounds (LaOBiSe₂, etc.), however, has not yet been reported so far. In this thesis, we have investigated the hidden spin polarization by means of spin- and angle-resolved photoelectron spectroscopy (SARPES) and found the evidence of the spin polarized states caused by

interaction (SOI) for the first time. Interestingly, the conversion from Rashba-like to Dresselhaus-like spin texture with varying binding energy has been directly observed in the conduction band. In addition to $\text{LaO}_{0.55}\text{F}_{0.45}\text{BiS}_2$, a series of BiS_2 -based superconductors such as $\text{LnO}_{1-x}\text{F}_x\text{BiS}_2$ compounds (Ln = Ce [4], Pr [5], Nd [6], etc) were discovered after 2012, in which the sandwiched crystal structure possesses the BiS_2 electronically active layers and the alternative LnO buffer-layers, along with providing us a pathway to introducing charge, lattice mismatch and crystal electric field to the interfacial structure by element-substitution of buffer layers such that the novel features (superconductivity, Rashba effect, etc.) emerge in the systems. Although the mechanism of superconductivity is not yet clear, it seems reasonable to attribute the emerging of superconductivity to the electron-phonon interaction. Nevertheless, using ab initio calculation of $\text{LaO}_{0.5}\text{F}_{0.5}\text{BiS}_2$, the electron-phonon coupling was estimated to be $\lambda < 0.5$ [7], which is too low to explain the actual superconducting transition temperature in this system. Moreover, the revealed hidden Rashba/Dresselhaus spin polarization in our study may suggest an unconventional picture of spin-mediated Cooper pairing. In spite of its clear interest, the possibilities of having novel features in such new Rashba superconductors, for instance, both the enhancement of T_C because of Rashba/Dresselhaus effect and the potential applications of spintronics, have not been sufficiently investigated to date. Here, in my thesis we aimed to accurately determine the Fermiology of $\text{LnO}_{1-x}\text{F}_x\text{BiS}_2$ (Ln = Ce, Pr) superconductors by performing higher resolution SARPES analysis. We show that circular-like constant energy contour of electron pocket in $\text{PrO}_{0.87}\text{F}_{0.13}\text{BiS}_2$ possesses nonhelical spin texture whereas helical spin texture was observed in higher F-doping samples $\text{PrO}_{0.77}\text{F}_{0.23}\text{BiS}_2$. The tunable spin texture of the Fermi Surface may induce the unconventional superconductivity. Besides, we confirmed the Ce-S-Ce electron hop channel in $\text{CeO}_{0.73}\text{F}_{0.27}\text{BiS}_2$ by tracking doped electrons along two flow paths. Our findings serve as an excellent platform for fabricating the dual-gate spin field effect transistor (Spin-FET) and fault-tolerant qubits [8].

References

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