Coexistence of superconductivity and ferromagnetism in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$

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Through the measurement of X-ray diffraction, electrical transport, magnetic susceptibility, and heat capacity, we have studied the effect of Ce doping in the newly discovered SrFBiS$_2$ system. It is found that Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ exhibits ferromagnetic ordering of Ce moments at 7.5 K, followed by a superconducting transition with critical temperature at 2.8 K. The negative magnetoresistance and a clear specific heat jump below 7.5 K indicate that the moments of Ce ion order ferromagnetically.

The most important result of the study is that Ce doping not only induces superconductivity at 2.8 K, but also forms ferromagnetic ordering below 7.5 K. Our works provide a new sight for studying the interplay between superconductivity and ferromagnetism in the BiS$_2$ based family.

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The fascinating relationship between superconductivity (SC) and magnetic ordering has been a central issue in condensed matter physics community for several decades. It has been generally believed that within the context of the Bardeen-Cooper-Schrieffer (BCS) theory, the condensation electrons cannot be both magnetically ordered and superconducting$^{[1]}$. In other words, superconductivity and magnetism are two antagonistic phenomena. Even though it is thought that the superconducting pairing in cuprates, heavy fermions and Fe-based superconductors are mediated by antiferromagnetic spin fluctuations$^{[2–3]}$, SC can be generally induced after suppressing the magnetic ordering by chemical doping or pressure$^{[4–5]}$. However, the evidence for the coexistence of superconductivity with FM ordering has been found in a few compounds (UGe$_2$, URhGe, EuFe$_2$As$_{2−x}$P$_x$)$^{[6–9]}$.

Recently, superconductivity with a superconducting transition temperature ($T_c$) of 8.6 K in a novel BiS$_2$ based superconductor Bi$_3$O$_4$S$_3$ has been discovered$^{[10]}$. Following this report, several other BiS$_2$-based superconductors, LnO$_{1−x}$F$_x$BiS$_2$(Ln=La, Ce, Pr, Nd)$^{[11–15]}$ with the highest $T_c$ of 10 K have then been reported and studied. Similar to the cuprates and iron-based superconductors, the BiS$_2$-based compounds possess a layered crystal structure consisting of superconducting BiS$_2$ layers intercalated with various block layers, e.g., Bi$_3$O$_4$(SO$_4$)$_{1−x}$ or [Ln$_2$O$_2$]$^{2−}$. Obviously, the common BiS$_2$ layer is believed to be a basic structure for searching a new superconducting family, where superconductivity can be induced by chemical doping into the block layers. Actually, through replacement of LaO layer by SrF block layer, a new BiS$_2$ based layered superconductor Sr$_1−x$La$_x$FBiS$_2$, which is iso-structural to LaOBiS$_2$, have been synthesized and studied$^{[16–18]}$. The parent compound SrFBiS$_2$ shows a semiconducting-like behaviors, and doping by La into Sr site can induce superconductivity with $T_c$ of 2.8 K.

Up to now, main studies about LnOBiS$_2$-based system have focused on the electronic structure$^{[19]}$, superconducting transition temperature$^{[20]}$ and pairing symmetry$^{[21–23]}$. Even though experimental results$^{[24–20]}$ as well as theoretical models$^{[21, 22]}$ seem to support a conventional s-wave superconductor, the study of exotic superconducting properties is still lacking so far. A typical example is the coexistence of superconductivity and ferromagnetism at low temperature as recently proposed for CeO$_{1−x}$F$_x$BiS$_2$ samples$^{[14–27]}$. But the interaction between SC and magnetic ordering of rare earth elements, as well as the detailed physical properties have not been investigated. Moreover, SrFBiS$_2$, as an important and promising BiS$_2$-based superconductor family, displays the different transport properties compared with the iso-structural LaOBiS$_2$$^{[16–18]}$. In the paper, we report a successful synthesis of Ce-doped Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ superconductor. Through the measurement of resistivity, magnetization and specific heat, we found that superconductivity can be induced around 3 K by Ce doping, and simultaneously the dilute Ce lattices order ferromagnetically below 7.5 K, with only 50% Ce contents in the (Sr, Ce)F layer.

The polycrystalline sample of Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ was synthesized by two-step solid state reaction method. The detailed synthesis methods can be found in previous literature$^{[16]}$. Crystal structure characterization was performed by powder X-ray diffraction (XRD) at room temperature using a D/Max-rA diffractometer with Cu K$_\alpha$ radiation and a graphite monochromator. The XRD data were collected in a step-scan mode for $10^\circ \leq 2\theta \leq$
Lattice parameters were obtained by Rietveld refinements. The electrical resistivity was measured with a standard four-terminal method covering temperature range from 0.4 to 300 K in a commercial Quantum Design PPMS-9 system with a $^3$He refrigeration insert. The measurements of magnetic susceptibility and specific heat were also performed in this system. D.C. magnetic properties were measured on a Quantum Design Magnetic Property Measurement System (MPMS-7).

Figure 1 shows the powder XRD patterns of the Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ sample at room temperature, as well as the result of the Rietveld structural refinement. Overall, the main diffraction peaks of this sample can be well indexed based on a tetragonal cell structure with the P4/nmm space group. In addition to principal phase, extra minor peaks arising from impurity phase of Bi$_2$S$_3$ with Pnma symmetry can also be observed, and its content is estimated to be about 10% by Rietveld refinement. The refined lattice parameters are extracted to be $a = 4.0695 \text{Å}$ and $c = 13.3208 \text{Å}$, which are shorten by 0.32% and 3.4% respectively, compared with that of parent compound SrFBiS$_2$, reflecting the electron doping. As a result, the cell volume shrinks by 4.1% for Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$. This result implies that the Ce ion was indeed doped into the lattice.

Fig. 2(a) shows temperature dependence of the electrical resistivity ($\rho$) under zero field and 9 T for Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ sample. The zero field resistivity increases monotonously with decreasing temperature but its value drops by several orders of magnitude compared to the un-doped sample[16]. Meanwhile, it also shows thermally activated behavior with decreasing temperature from 300 K. Using the thermal activation formula $\rho(T) = \rho_0 \exp(E_a/k_B T)$ to fit the $\rho(T)$ at the temperature range from 150 K to 300 K, we obtain the thermal activation energy ($E_a$) of about 11.8 meV, which is far smaller than that of the undoped SrFBiS$_2$ sample (38.2 meV)[16, 17], suggesting the decrease of gap size due to electron doping. As temperature further cooled down, a sharp superconducting transition with $T_c$ of 2.8 K, developing from the semiconducting-like normal state, is clearly observed. This feature is commonly found in Bi$_2$S$_3$-based superconductors[11–15]. As the magnetic field ($H$) increases to 9 T, superconductivity is suppressed completely. Instead, the resistivity remains rapidly increase with temperature down to 0.6 K and recovers the semiconducting-like behaviors in the normal state. On closer examination, as shown in inset of Fig. 2(a), the negative magnetoresistivity under $B = 9$ T can be obviously observed below 7.5 K, at which temperature Ce moments order ferromagnetically (see below), implying the reduction of spin-scattering of Ce moments. On the contrary, no significant magnetoresistivity can be found in parent compound SrFBiS$_2$[17] and Sr$_{1-x}$La$_x$FBiS$_2$[16, 18], or the small positive magnetoresistivity is reported in Bi$_2$O$_4$S$_3$ system above $T_c$[20].

In order to reveal the ferromagnetic behaviors in the
FIG. 3. (Color online) (a) Temperature dependence of magnetic susceptibility for Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$. (b) Isothermal magnetization of Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ sample at several different temperatures. The inset of (a) shows the ZFC (open symbols) and FC (solid symbols) susceptibilities under $H = 20$ Oe.

present sample, the magnetic field dependence of resistivity under several different temperatures is plotted in Fig. 2(b). The resistivity increases rapidly below $\sim 1$ T due to the suppression of superconductivity, and then decreases monotonously with increasing the magnetic field to 9 T at 2 K which is below $T_c$. At 3 K, the magnetoresistance is almost negligible below 1 T, but the negative magnetoresistance is also observed and reaches -4% under $B = 9$ T. While at 6 K, only tiny negative magnetoresistance can be detected. Further increasing temperature to 10 K, the resistivity remains constant with magnetic field and no magnetoresistance is found. The feature is tentatively attributed to the FM ordering of Ce$^{3+}$ moments (to be shown below).

Figure 3(a) shows the temperature dependence of field-cooled (FC) dc magnetic susceptibility for Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ under $H = 1$ kOe. It is easy to observe that magnetic susceptibility of the sample exhibits a Curie-Weiss law behavior well above 80 K, which is associated with the moment of magnetic rare earth element Ce. By fitting the Curie-Weiss law, $\chi = C/(T - \theta)$, where $C$ is the Curie-Weiss constant, and $\theta$ denotes the paramagnetic Curie temperature, we obtained the $C = 0.73$ emu-K/mol-Ce and $\theta = -19.54$ K. The effective magnetic moment $\mu_{eff}$ is thus calculated to be $2.42 \mu_B$ per formula unit, which is close to the theoretical value of $2.53 \mu_B$ for a free Ce$^{3+}$ ion. To further investigate the coexistence of superconductivity and ferromagnetism in this sample, the magnetic susceptibility under 20 Oe with ZFC and FC mode below 10 K is shown in inset of Fig. 3(a). A rapid increase in magnetic susceptibility and an obvious divergence between ZFC and FC data are observed below 7.5 K, manifesting the long-range FM-ordered of the Ce 4$f$ electrons. Further cooled down temperature, an obvious drop around 2.8 K in both the ZFC and FC data is observed because of the superconducting transition. These results imply that FM ordering coexists with superconductivity in the present system. The isothermal magnetization hysteresis loops in Fig. 3(b) support this conclusion as well. Even at 2 K, the clear hysteresis loop indicates the dominating ferromagnetic signal, which is ascribed to the proximity of superconducting transition and ferromagnetic ordering. The size of loop gradually shrinks with increasing temperature, and then disappears at 10 K. Noted that the loop is not reported in other Bi$_2$-based superconductors, such as CeO$_{1-x}$F$_x$BiS$_2$ system[27].

FIG. 4. (Color online) Temperature dependence of resistivity for Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ samples under several different magnetic field.

Figure 4 shows the enlarged low-temperature resistivity for Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ samples under various magnetic fields below 6 K. With the application of magnetic fields, the superconducting transition becomes broadened and $T_c$ decreases towards lower temperature. Superconductivity can be completely suppressed above 0.6 K by a magnetic field as low as 1 T, and its resistivity displays semiconducting-like feature. Further increasing the mag-
magnetic field to 9 T, the negative magnetoresistance is observed in the normal state, consistent with the magnetoresistivity data as shown in Fig. 2. The similar result is also observed in EuFe$_2$As$_{2−x}$P$_x$ superconductor\cite{8}, which is reported to be a rare ferromagnetic superconductor in iron-based compounds. We note that, in other Bi$_2$ superconductors i.e., Sr$_{0.5}$La$_{0.5}$FBiS$_2$, its magnetoresistivity is absent\cite{10}. The inset of Fig.4 displays the temperature dependence of the upper critical field $\mu_0H_{c2}(T)$, determined by using 90% normal state resistivity criterion. The $\mu_0H_{c2} − T$ diagram shows nearly linear in the measured temperature range. According to Ginzburg-Landau theory, the upper critical field $H_{c2}$ evolves with temperature following the formula:

$$H_{c2}(T) = H_{c2}(0)(1 − t^2)/(1 + t^2),$$  

(1)

where $t$ is the renormalized temperature $T/T_c$. The upper critical field $H_{c2}$ is then estimated to be 1.03 T, very close to that of Sr$_{0.5}$La$_{0.5}$FBiS$_2$ (1.04 T). Noted that both of Ce and La doping in SrFBiS$_2$ seem to have similar $T_c$, while Ce moments can order ferromagnetically in the SrF layer. These findings imply that FM orderings may have less destruction effect to SC in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$. The feature is in contrast to the case of EuFe$_2$As$_{2−x}$P$_x$, which has far smaller $T_c$ and $H_c(0)$ than that of BaFe$_2$As$_{2−x}$P$_x$ because of FM ordering of the Eu moments.

![FIG. 5. (Color online) The specific heat of Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ samples under zero field below 15 K. The upper left panel shows the magnetic specific-heat anomaly around 7.5 K under magnetic fields. $C/T$ vs. $T^2$ diagram is plotted in the lower right panel of this figure. The red line is a fit of the expression $C/T = \gamma + \beta T^2$ to the data.](image)

The specific heat measurement of Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ sample was performed in the temperature range from 2 K to 15 K. As plotted in Fig. 5, a clear $\lambda$ shape kink observed at $T_{FM} = 7.5$ K strongly demonstrates the second order phase transition. Associated with the magnetic susceptibility and resistivity data, the peak is ascribed to the ferromagnetic ordering of Ce moments. With increasing magnetic field, the anomaly is less shifted but becomes more broadened, confirming that the moment of Ce orders ferromagnetically below $T_{FM}$, as shown in inset of Fig.5. However, the specific heat jump caused by superconducting transition is not seen below $T_c$. In Bi$_2$-based superconductors, absence of specific-heat jump at the superconducting transition is observed in some cases. It has been reported that both CeO$_{0.5}$F$_{0.5}$BiS$_2$ and YbO$_{0.5}$F$_{0.5}$BiS$_2$ with magnetic rare earth elements do not show any anomaly at around $T_c$ from specific heat data\cite{30}, while the clear jump is always observed in Sr$_{0.5}$La$_{0.5}$FBiS$_2$, LaO$_{1−x}$F$_x$OBiS$_2$ and La$_{1−x}$M$_x$OBiS$_2$ system\cite{10, 31} where those compounds are paramagnetic in normal state because of non-magnetic Sr or La element. These results suggest that the anomaly around $T_c$ may be overwhelmed by the enhanced specific heat signal because of the contribution of FM ordering.

To further analyze the specific heat data, we fit the data in the normal state using the $C/T = \gamma + \beta T^2$ formula, where $\gamma$ and $\beta T^2$ account for the electronic and lattice contributions, respectively. In the lower inset of Fig. 5, a linear fit to $C/T$ versus $T^2$ plot is seen from 8 to 15 K, and yields values of $\gamma = 32.2$ mJ/mole K$^2$ and the lattice coefficient $\beta = 1.46$ mJ/mole K$^2$, respectively. The Debye temperature is then estimated to be 188 K. The value falls in between with those of CeO$_{0.5}$F$_{0.5}$BiS$_2$ (224 K) and YbO$_{0.5}$F$_{0.5}$BiS$_2$ (186 K)\cite{21}. Noted that CeO$_{0.5}$F$_{0.5}$BiS$_2$ is the coexistence of superconductivity and ferromagnetism, and YbO$_{0.5}$F$_{0.5}$BiS$_2$ exhibits antiferromagnetic order and coexists with superconductivity below 5.4 K\cite{30}. Considering 50% Ce doped into lattice, the $\gamma$ value should be 64.4 mJ/K$^2$/mole-Ce in the present compound. The value is very close to that of CeO$_{0.5}$F$_{0.5}$BiS$_2$ (58.1 mJ/mole K$^2$)\cite{30}, but is far larger than those of Sr$_{0.5}$La$_{0.5}$FBiS$_2$ (1.42 mJ/mole K$^2$)\cite{10, 31} and La$_{1−x}$M$_x$OBiS$_2$ ($M=\text{Tl, Zr, Th}$) (0.58−2.21 mJ/mole K$^2$). The substantially enhanced $\gamma$ may be mainly originating from electronic correlation effect of Ce-$4f$ electrons.

So far, a growing evidence for the coexistence of superconductivity with ferromagnetic ordering has been reported. The vast majority of these systems show superconductivity before the ferromagnetic ordering, and will lead to re-entrant superconductivity overlapping with a magnetically ordered phase, such as ErRh$_4$B$_4$\cite{32}, ErNi$_2$B$_2$C\cite{33}, EuFe$_2$As$_{2−x}$P$_x$\cite{8}. Actually, in these systems, two separate sets of electrons may be responsible for magnetic ordering and superconductivity, respectively. The moments of rare-earth elements devote mainly to the ferromagnetic ordering, and the 3d electrons of transition metal play a key role in causing superconductivity. Generally, superconductivity is in competition with magnetic ordering due to the interaction of 3d electron and 4f electrons. While in our case, the ferromagnetic transition temperature is substantially higher than $T_c$. On the other hand, Ce doping provides carri-
ers inducing superconductivity and meanwhile forms ferromagnetic ordering, implying that the interaction between Ce 4f electron and conduction electrons may be anisotropic. Thus, there may be an intrinsic coexistence of FM and SC, both of them are ascribed to the Ce doping. Compared with the previous report[23], the remarkable feature here is that the FM ordering can be established in the diluted Ce lattice, with 50% Ce in the SrF layer. The reason may be mainly attributed to the RKKY interaction mediated by carriers. More experiments (neutron and NMR) and theoretical explanations may provide a new sight and inspiration for this issue in BiS$_2$-based superconductors.

In summary, by partially substituting Ce for Sr in the newly discovered SrFBiS$_2$ system, we discovered a new BiS$_2$-based superconductor Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ with $T_c$ as high as 2.8 K. Meanwhile, the measurement of magnetic susceptibility and heat capacity has suggested that Ce$^{3+}$ moments order ferromagnetically below 7.5 K, and co-exist with superconductivity in the Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ system. In other words, Ce doping can provide carriers to the superconducting BiS$_2$ layers and meanwhile form FM ordering in the blocking (Sr, Ce)F layers. It remains a puzzle to understand that superconductivity occurs in the BiS$_2$ layers and simultaneously the diluted Ce lattice orders ferromagnetically in the (Sr, Ce)F layer using only 50% Ce doping, because superconductivity requires to establish the interlayer coupling across the ferromagnetic layers. Thus, it would certainly be interesting to explore the interplay of superconductivity and ferromagnetism in the BiS$_2$-based superconductors.

Note: At the completion of this work, we became aware of the recent paper[34], which also observed SC in Sr$_{1-x}$Ln$_x$FBiS$_2$ (Ln = Ce, Pr, Nd, Sm).

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