Coexistence of superconductivity and ferromagnetism in Sr$_{0.5}$Ce$_{0.5}$FBiS$_{2-x}$Se$_x$ ($x = 0.5$ and $1.0$), a non-U material with $T_c < T_{FM}$

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We have carried out detailed magnetic and transport studies of the new Sr$_{0.5}$Ce$_{0.5}$FBiS$_{2-x}$Se$_x$ ($0.0 \leq x \leq 1.0$) superconductors derived by doping Se in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$. Se–doping produces several effects: it suppresses semiconducting–like behavior observed in the undoped Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$, the ferromagnetic ordering temperature, $T_{FM}$ decreases considerably from 7.5 K (in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$) to 3.5 K and the superconducting transition temperature, $T_c$ gets enhanced slightly to 2.9–3.3 K. Thus in these Se–doped materials, $T_{FM}$ is marginally higher than $T_c$. Magnetization studies provide evidence of bulk superconductivity in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$, Se$_x$ at $x \geq 0.5$ in contrast to the undoped Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ ($x = 0$) where magnetization measurements indicate a small superconducting volume fraction. Quite remarkably, as compared with the effective paramagnetic Ce–moment (~2.2 $\mu_B$), the ferromagnetically ordered Ce–moment in the superconducting state is rather small (~0.1 $\mu_B$) suggesting itinerant ferromagnetism. To the best of our knowledge, Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$, Se$_x$ ($x = 0.5$ and 1.0) are distinctive Ce–based bulk superconducting itinerant ferromagnetic materials with $T_c < T_{FM}$. Furthermore, a novel feature of these materials is that they exhibit a dual and quite unusual hysteresis loop corresponding to both the ferromagnetism and the coexisting bulk superconductivity.

Traditionally, superconductivity and long–range ferromagnetism had been considered mutually exclusive (BCS pair–breaking and Meissner effect). For example, in several ternary materials RM$_6$X$_8$ (X = S, Se) and RRh$_4$B$_4$ $^{3,4}$, studied in the early eighties, superconductivity was observed coexisting with long range antiferromagnetic order. But uniform ferromagnetism suppressed superconductivity at low temperatures $^{4–6}$. The situation has changed strikingly over the last several years with the discovery of a number of materials that do exhibit coexistence of superconductivity and long-range ferromagnetism. In materials such as ErNi$_2$, RuSr$_2$GdCu$_2$O$_{8}$, the ferromagnetic ordered moment of U is drastically reduced, 0.03 $\mu_B$ and ferromagnetic superconductors such as UCoGe, URhGe and UGe$_2$ have been proposed to be compatible with itinerant ferromagnetism. For $p$-wave pairing, one needs to go beyond electron–phonon interaction (pairing mechanism in conventional superconductivity) with Cooper-pairing mediated via spin fluctuations. The material UCoGe is of particular interest from the viewpoint of the present work. In this material the paramagnetic effective moment of U is ~1.7 $\mu_B$ whereas the ferromagnetic ordered moment of U is drastically reduced, 0.03 $\mu_B$. The materials under investigation (the title compounds) in this work, exhibit coexisting superconductivity and itinerant ferromagnetic properties, as we shall see below, similar to those of UCoGe.

The parent material Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$, of the title compounds Sr$_{0.5}$Ce$_{0.5}$FBiS$_{2-x}$Se$_x$, belongs to a small class of AFBiS$_2$ ($A = $ Sr and Eu) $^{15,16}$ of a larger family of BiS$_2$ layered tetragonal materials LnOBiS$_2$ $^{17,18}$ ($P4/nmm$) recently shown...
to exhibit superconductivity. SrFBS is derived by replacing Ln–O layers by Sr–F layers. Its structure essentially consists of alternate stacking of conducting BiS₂ layers and blocking (insulating) layer LnO/SrF in the electron carriers are doped into the conducting BiS₂ layers employing the commonly used doping strategy, namely, replacing O partially by F, for instance LaOₓFₓₐBiS₂ exhibits superconductivity at T_c = ~2.8 K. In AFBiS₂ electron doping and eventual superconductivity is achieved/enhanced byLn (La and Ce) doping at A sites. Structurally, these materials are quite similar to high-T_c cuprates and iron nictides and superconductivity is quite robust as evident from numerous studies on various site substitutions and pressure via partial or complete substitution of La by a smaller rare-earth (Ln = Ce, Pr, Nd, Sm and Yb)³³,³⁴–³⁶. YbOₓFₓₐBiS₂ has the highest T_c = 5.4 K among LnOₓFₓₐBiS₂ and, interestingly, it undergoes an anti-ferromagnetic transition (T转变为 ~2.7 K) also³³,³⁴. Se substitution has been realized in LaOₓFₓₐBiS₂ and EuOₓFₓₐBiS₂, where an enhancement of T_c is observed with maximum T_c of 3.8 K for LaOₓFₓₐBiS₂ composition (x = 1.0). T_c decreases on further Se substitution. For other rare earths (Ce and Nd), however, the effect of Se on T_c is different. In Ce(O/F)BiS₂ enhancement in T_c is only marginal (2.4 to 2.6 K)³⁶. Se substitution induces bulk superconductivity in La and Ce materials. In Nd(O/F)BiS₂ and Bi₁₋ₓFₓBiS₂, Se doping has been shown to depress T_c³³,³⁶. Se substitution in AFBiS₂ has not been tried so far. Under applied pressure, T_c is enhanced in LnOₓFₓₐBiS₂ and A₁₋ₓLnₓBiS₂ (Ln = La, Ce, Pr and Nd; A = Sr and Eu) up to a maximum of 10 K³⁶–³⁸. The Bi-S₂ materials are BCS-like and probably have s-wave pairing symmetry. But there is yet no consensus on the origin of superconductivity in these materials³³,³⁴.

Very recently, ferromagnetism and superconductivity have been reported to coexist in CeO₁₋ₓFₓBiS₂ and Sr₁₋ₓCeₓFBiS₂, with T_c ~ 2.5–4 K and T_FM ~ 4–8 K³⁰,³²–³⁴. As these materials have layered structure, magnetism originates in the Ce–O (or Sr/Ce–F) layers and conduction occurs in BiS₂ layers. In Sr₁₋ₓCeₓFBiS₂, the parent materials for our Se–added materials Sr₁₋ₓCeₓFₓₐBiS₂–xSe, Ce–substitution provides conduction electrons as well as gives rise to long range magnetic order³⁷. Ferromagnetic order takes place at a higher temperature (7.5 K) and superconductivity sets in at a lower temperature (2.8 K) in an already ferromagnetically ordered lattice. We report here the effect of substitution of larger isovalent Se ion at the S site on the magnetic and superconducting properties of Sr₁₋ₓCeₓFBiS₂–xSe–doping leads to a modest enhancement of T_c (up to 3.3 K) and a significant depression of T_FM (down to 3.5 K). Thus Se–doping moves T_c and T_FM in opposite directions, bringing them in closer proximity in temperature. We believe the ferromagnetism in our materials is itinerant just as it is in UCoGe³⁵, namely, high Ce-paramagnetic moment (~2.2 μB) and small saturation Ce-magnetic moment (0.1 μB). To the best of our knowledge, the materials Sr₁₋ₓCeₓFₓₐBiS₂–xSe, x = 0.5, 1.0 are unique Ce-containing materials exhibiting coexisting bulk superconductivity and itinerant ferromagnetism. Thus our observation of the coexistence of superconductivity and itinerant ferromagnetism in Sr₀.₅Ce₀.₅FBiS₂–xSeₙ is a timely discovery, in that it puts U– and Ce on equal footing in this respect also.

Results and Discussion

PXRD characterization. PXRD patterns of all the Sr₁₋ₓCeₓFₓₐBiS₂–xSe, (x = 0.0, 0.3, 0.5 and 1.0) compositions are shown in Fig. 1. All the peaks could be easily indexed on the basis of a SrFBS₂ type tetragonal unit cell (SG: P4/mmm). Minor peaks corresponding to the impurity of Bi₁₋ₓS (S) and Bi₁₋ₓSeₙ (Se) were also observed for composition with x > 0. The estimated impurity phase of Bi₁₋ₓS was ~4% observed in x = 0.3 composition whereas the amount of Bi₁₋ₓSeₙ was ~6% and ~14% in x = 0.5 and x = 1.0 composition respectively. It is evident from X-ray studies that the impurities increase with the increase of Se content. The samples with x > 1.0 were obtained as multiphase products. This indicates a Se solubility limit of x ~ 1.0. Lattice parameters a and c show an expected increase upon Se doping (a = 4.0886(2) Å, c = 13.4143(8) Å for x = 0.5 and a = 4.1057(1) Å, c = 13.4756(8) Å for x = 1.0) resulting in the monotonous unit cell expansion (inset in Fig. 1). Compositional analysis on x = 0.5 and 1.0 samples gives a stoichiometry close to the nominal value for both the compositions (Figure S1 in supplementary material (SM)). For x = 1.0 sample, the Se:S ratio was slightly less than 1, possibly due to the formation of small amount of the impurity phase Bi₁₋ₓSeₙ, which is non-magnetic and insulating under ambient pressure³⁹. It does not interfere with superconducting and magnetic properties of the materials under investigation.

![Figure 1. Powder X-ray diffraction of Sr₀.₅Ce₀.₅FBiS₂–xSe, (x = 0.0, 0.3, 0.5 and 1.0). Symbols (*) and (#) indicate impurity phases Bi₁₋ₓSeₙ and Bi₁₋ₓS respectively. Inset shows the variation of cell volume with Se content.](image-url)
Resistivity. Resistivity of the materials as a function of temperature is shown in Fig. 2. In the normal state, resistivity of Sr0.5Ce0.5FBiS2-xSex with x = 0 and 0.3 exhibit semiconducting–like temperature dependence, namely, it increases with the decrease of temperature just before the onset of superconducting transition at 2.4 K and 2.7 K respectively as shown in Fig. 2(a). Note that the resistivity values for x = 0 and 0.3 were divided by a factor of 20 and 4 respectively for the purpose of clarity. In the higher Se–doped materials, x = 0.5 and x = 1.0, this semiconducting behavior is progressively subdued and metallic conductivity is observed in the normal state. Superconductivity sets in at Tc = 2.9 and 3.3 K in materials with x = 0.5 and 1.0 respectively. Our estimate of Tc onset is based on a 90% criterion as shown in Fig. 2(b). Se–doping clearly enhances Tc by ~1 K (inset of Fig. 2(b)). In the material with x = 1.0, a sharp superconducting transition is observed with a transition width ΔT = 0.2 K. Similar small enhancement in Tc with Se substitution was previously observed in LnO1-xFxBiS2 (Ln = La and Ce)38,56,57. This enhancement in Tc is attributed to the in-plane chemical pressure induced by the Se substitution at S sites as elucidated by Mizuguchi et al.58. The plot of upper critical field, Bc2 (T) as a function of temperature is given in the inset of Fig. 2(a). We estimated Bc2 below 2 K using a standard single-band Werthamer–Helfand–Hohenberg (WHH) formula with the Maki parameter59 α = 0. Upper critical field, Bc2(0) at T = 0 is estimated to be 2.6 T for x = 0.5 and 3.3 T for x = 1.0. These Bc2 values are atleast twice higher than those reported for the Se–free samples Sr0.5Ce0.5FBiS2.26,27. Enhancement of Tc and Bc2 in the Se–doped samples clearly indicates that Se atoms have entered the lattice. Enhancement of Bc2 implies reduction of the coherence length or stronger impurity scattering due to Se doping in Sr0.5Ce0.5FBiS2.

Magnetic susceptibility in low field of 10 Oe. Figure 3(a) shows dc susceptibility of Sr0.5Ce0.5FBiS2-xSex (x = 0.5 and 1.0), in both the field-cooled (FC) and the zero field-cooled (ZFC) conditions in an applied field of 10 Oe. Clear diamagnetic signal, of magnitude close to the theoretical value, for both the x = 0.5 and 1.0 compositions is observed in ZFC condition (Fig. 3a) establishing the superconducting state. Poor Meissner response in both cases is possibly due to flux pinning. A superconducting volume fraction of >95% is estimated for both x = 0.5 and 1.0 compositions. In several studies60–65 on a variety of materials, such large diamagnetic superconducting signals have been observed and have been considered suggesting bulk superconductivity therein. Inset of Fig. 3(a) shows dc susceptibility of the Se free sample Sr0.5Ce0.5FBiS2 (x = 0.0) which shows a ferromagnetic behavior with Curie temperature ~7.5 K, similar to that reported earlier by Li et al.57. A weak drop in the ZFC susceptibility below 3 K is due to the superconducting transition that was also observed in our resistivity measurements. Such a weak diamagnetic signal rules out bulk superconductivity in parent sample x = 0.0 and is consistent with weak superconductivity. Figure 3(b) shows both the real and the imaginary parts of the ac susceptibility. A large superconducting screening indicates bulk superconductivity. Moreover a larger imaginary part of the

Figure 2. Variable temperature resistivity curves for Sr0.5Ce0.5FBiS2-xSex; x = 0, 0.3, 0.5 and 1.0. The data for x = 0 and 0.3 have been divided by 20 and 4 respectively (a) in temperature range 2–300 K and (b) in low temperature range. Inset of (a) shows the upper critical field (Bc2) versus temperature (T) curve for the x = 0.5 and 1.0 compositions (open circles) along with the WHH fit (solid lines). Inset of (b) shows the variation of Tc onset and Tc (ρ = zero) as a function of Se-doping.
signal indicates a considerable energy loss due to movement of vortices. Such a behavior cannot be explained if superconductivity is present only in thin surface layers. As deduced from these measurements, superconducting transition temperature increases from $T_{c \text{onset}} = 2.65\, \text{K}$ for $x = 0.5$ to $T_{c \text{onset}} = 3.20\, \text{K}$ for $x = 1$ which corroborates well with the resistivity data described above. It must be pointed out that in the earlier measurements on Se–free Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ materials diamagnetic signal was not observed and the occurrence of superconductivity was inferred from the resistivity measurements only. Inset of Fig. 3b shows the low temperature ac susceptibility (real part) data for the compositions with $x = 0.3$ in comparison with $x = 0.5$ and 1.0 samples. It shows a ferromagnetic behavior similar to $x = 0.0$ with a reduced Curie temperature $T_{FM} = 4.1\, \text{K}$. No diamagnetic signal was observed indicating that similar to the parent compound $x = 0.3$ is also a weak superconductor. A clear diamagnetic signal is observed only for $x = 0.5$ and 1.0 samples.

Further, in Fig. 3(a), a weak magnetic anomaly is discernible at 3.5 K for the sample $x = 0.5$ which corresponds to a ferromagnetic transition as evidenced in our high field measurements, (see below), for both the samples $x = 0.5$ and $x = 1.0$. This anomaly is not observed clearly for the sample $x = 1.0$. $T_c$ and $T_{FM}$ were ascertained from the derivative plots of susceptibility (see Figure S2 in SM). It is evident from the susceptibility studies that Se substitution depresses ferromagnetic ordering and enhances $T_c$ and $T_{FM}$ in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$$_x$.

**High field DC magnetization measurements.** Magnetic susceptibility $\chi(T)$, measured in an applied field of 10 kOe, and its inverse in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$$_x$Se$_x$ ($x = 0.5$ and 1.0) is presented in the Figure S3 in SM. By fitting the data above 50 K to the Curie–Weiss law $\chi(T) = \chi_c + C/(T - \theta)$, the paramagnetic effective magnetic moments obtained for the two samples are: $\mu_{eff} = 2.22\, \mu_B$ for $x = 0.5$ and 2.29$\, \mu_B$ for $x = 1.0$ (see Figure S3 in SM). These values are close to the theoretical value 2.54$\, \mu_B$ for free Ce$^{3+}$ ions. Thus Ce–ions are in trivalent (or nearly trivalent state) state.

We display in Fig. 4 the results of our magnetization measurements, at a few selected temperatures 5 K, 3.5 K and 2 K, in Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$Se$_{0.5}$ and Sr$_{0.5}$Ce$_{0.5}$FBiSSe. At 5 K, magnetization $M$ varies linearly with applied magnetic field, suggesting a paramagnetic state (no magnetic order). At 3.5 K, $M$ is no longer linear in $H$ in the low
field region and shows a sign of a ferromagnetic behavior. Ferromagnetic state is clearly observed at a lower temperature 2 K and, remarkably, at this temperature in both the samples, we observe a ferromagnetic hysteresis loop and a superimposed superconducting hysteresis loop, demonstrating unambiguously the coexistence of ferromagnetism and bulk superconductivity. A dual loop, displaying the two ordered states, superconductivity and ferromagnetism, with such clarity, is a novel feature of this material. In UCoGe, ferromagnetic hysteresis is observed in the ferromagnetic state ($T_c < T < T_{FM}$) but no superconducting hysteresis loop as such was observed\(^{66}\). Inset of Fig. 4a shows the isothermal magnetization (at 2 K) for $x = 0.0$ where only a ferromagnetic hysteresis loop is observed. In the inset of Fig. 4(b) and (d) the diamagnetic response is clearly seen in the virgin low field region (from which $H_{c1}$ is easily estimated to be $\sim 44$ Oe and 40 Oe for $x = 0.5$ and 1.0 respectively. It is important to point out that in the selenium-free compound Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ ($T_c \sim 2.6$ K & $T_{FM} \sim 7.5$ K)\(^{27,44}\) and in a similar material Ce(O, F)BiS$_2$ ($T_c \sim 2.5–4$ K & $T_{FM} \sim 6.5–7.5$ K)\(^{20,52,53,67}\) no superconducting hysteresis loop was observed. This is consistent with our own results on Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ (inset of Fig. 4a). Thus Se–doping has created crucial changes in the superconducting and magnetic properties of the parent material Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$. Observation of superconducting loop is a good indication of bulk superconductivity.

Dual hysteresis loop has been observed very recently in [(Li$_{1-x}$Fe$_x$)OH][(Fe$_{1-y}$Li$_y$)Se]\(^{68}\). However, there is a fundamental difference in this material and our samples, namely, in this case, $T_c$ is easily estimated to be $\sim 44$ Oe and 40 Oe for $x = 0.5$ and 1.0 respectively. It is important to point out that in the selenium-free compound Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ ($T_c \sim 2.6$ K & $T_{FM} \sim 7.5$ K)\(^ {27,44}\) and in a similar material Ce(O, F)BiS$_2$ ($T_c \sim 2.5–4$ K & $T_{FM} \sim 6.5–7.5$ K)\(^ {20,52,53,67}\) no superconducting hysteresis loop was observed. This is consistent with our own results on Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$ (inset of Fig. 4a). Thus Se–doping has created crucial changes in the superconducting and magnetic properties of the parent material Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$. Observation of superconducting loop is a good indication of bulk superconductivity.

The spontaneous magnetization $M_s$ is estimated by linear extrapolation of the high–field data to $H = 0$ (Fig. 4(a) and (c)). From the estimated $M_s$, we obtain at $T = 2$ K, the spontaneous Ce–moment $\mu_0 = 0.09 \mu_B$ for the sample $x = 0.5$ and 0.11 $\mu_B$ for the sample $x = 1.0$. These values are quite small as compared with what is expected for free Ce$^{3+}$ ion. We may note here that in Ce(O, F)BiS$_2$, a reduced moment $M_s = 0.52 \mu_B$/Ce was reported\(^{34}\) which, possibly, suggests that in this case Ce–ions may be in the crystal–field split doublet state (localized moment). In our case, we observe a drastically reduced, but non–zero, Ce–moment.

Figure 4. Hysteresis loops at different temperatures for Sr$_{0.5}$Ce$_{0.5}$FBiS$_1.5$Se$_{0.5}$ and Sr$_{0.5}$Ce$_{0.5}$FBiSe in $H \leq 10$ kOe (a) and (c) and $H \leq 1.5$ kOe (b) and (d). The superconducting loop is superimposed on the ferromagnetic loop at 2 K. Inset in Fig. 4(a) shows the ferromagnetic hysteresis loop for $x = 0$ composition. Insets in Fig. 4(b) and (d) show initial diamagnetic signal with arrows indicating lower critical field, $H_{c1}$.
The transition of the high Ce–paramagnetic effective moment $\mu_{\text{eff}} \approx 2.2$ $\mu_B$ to a small ordered moment $\mu_0 \approx 0.1$ $\mu_B$ in the superconducting state is an important observation as the drastic loss of Ce–moment signals a delocalization of the 4f electrons concurrent with the appearance of superconductivity. Thus, 4f–electrons may also be involved in superconductivity in these materials. A high ratio $\mu_{\text{eff}}/\mu_0$ ($\approx 22$) implies an itinerant ferromagnetic state$^{13,70}$ in both materials Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$Se$_x$, $x = 0.5$ and $x = 1.0$. Further, as Ce–atoms are responsible both for ferromagnetism and coexisting bulk superconductivity we think the two phenomena can coexist uniformly. These materials fill the glaring void, namely, so far no Ce–based material has been hitherto known exhibiting superconductivity within the itinerant ferromagnetic state.

**Specific heat.** Figure 5 shows the temperature dependence of specific heat of Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$Se$_x$ ($x = 0.5$) in the low temperature range 2–16 K. Inset shows C/T data before (blue circle) and after subtraction (black circle) of a Schottky contribution which was approximated by the dashed line. A broad peak, not in the low temperature range 2–16 K. Inset shows C/T data before (blue circle) and after subtraction (black circle) of a Schottky contribution which is represented by the dashed line.

![Figure 5. Temperature dependence of Schottky corrected specific heat C/T vs. T for x = 0.5 sample at H = 0. Red line is the linear fit to the equation C/T = γ + βT$^2$. Inset shows C/T data before (blue circle) and after subtraction (black circle) of a Schottky contribution which was approximated by the dashed line.](image)

After submission of this manuscript we came across a report on the coexistence of superconductivity and ferromagnetism in CsEuFe$_4$As$_4$ compound$^{73}$. A dual hysteresis loop has been observed in this compound also. These materials fill the glaring void, namely, so far no Ce–based material has been hitherto known exhibiting superconductivity within the itinerant ferromagnetic state. A dual hysteresis loop has been observed in this compound also.

**Concluding Remarks.** We have observed superconductivity ($T_c \approx 3.0$ K) and itinerant ferromagnetism ($T_{\text{FM}} \approx 3.5$ K) coexisting in the new materials Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$Se$_x$, $x \geq 0.5$. Thus in these materials, superconductivity occurs much closer to the border of ferromagnetism than in UCoGe. A novel feature of these materials, as compared with the other ferromagnetic superconductors reported so far, is a dual hysteresis loop corresponding to both the coexisting bulk ferromagnetism and superconductivity. Thus Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$Se$_x$ is an important and timely addition to the exciting Ce–based materials exhibiting coexisting superconductivity and magnetism. The materials Sr$_{0.5}$Ce$_{0.5}$FBiS$_2$Se$_x$ are potential candidates for the unconventional p–wave superconductivity$^{74}$ and deserve to be further pursued in this regard. We are making efforts to grow single crystals of these materials. In single crystals (if we succeed to grow) or else in polycrystalline materials, we would carry out studies such as NMR, MuSR, neutron diffraction
and Andreev reflection to throw further light on the coexistence of superconductivity and ferromagnetism and nature of the superconducting state (p-wave or s-wave) in these materials.

Methods

Polycrystalline compounds of the series \( \text{Sr}_{1-x}\text{Ce}_x\text{FBiS}_2-x\text{Se} \) (\( x = 0.0, 0.3, 0.5 \) and 1.0) were prepared by the usual solid state synthesis procedure as reported elsewhere \(^{23, 35}\). \( \text{SrF}_2\), Bi and Se powder, pre-reacted \( \text{Ce}_2\text{S}_3 \) and \( \text{Bi}_2\text{S}_3 \) powder were thoroughly mixed, pelletized and sealed in quartz tube under vacuum. The tubes were then heated twice at 800°C for 24 hours with an intermediate grinding. The end products were black/dark grayish in color. Phase purity of all the compositions was checked by powder X-ray diffraction (PXRD) technique using Cu Kα radiation source. Temperature dependent resistivity, magnetization and specific heat measurements were performed using a 14 T PPMS (Quantum Design). The specific heat was measured using a relaxation technique.

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Figure 6. Ferromagnetism and superconductivity phase diagram of Sr0.5Ce0.5FBiS2-xSex (0 ≤ x ≤ 1).
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Author Contributions
G.S.T. carried out the synthesis and preliminary characterization (XRD and EDX) with help of Z.H., G.F. and K.N. carried out the resistivity, magnetic and specific heat measurements. All authors reviewed the manuscript. A.K.G., L.C.G. and G.F. supervised the work.

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