Supplementary Information

Intrinsic non-stoichiometry and anomalous transport properties of layered oxysulfide LaOPbBiS$_3$

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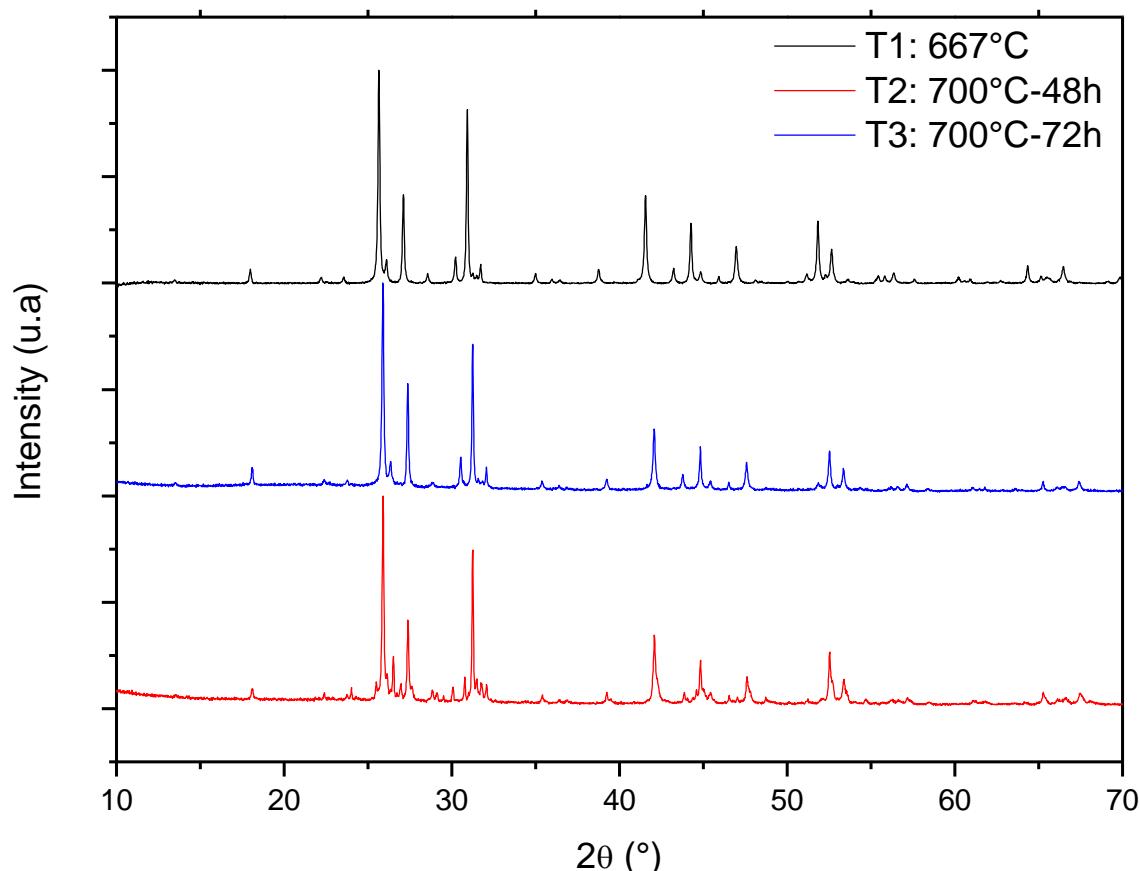
Figure S1: influence of successive thermal treatments on the amount of secondary phases of pristine LaOPbBiS$_3$.

Pb/Bi = 1/1
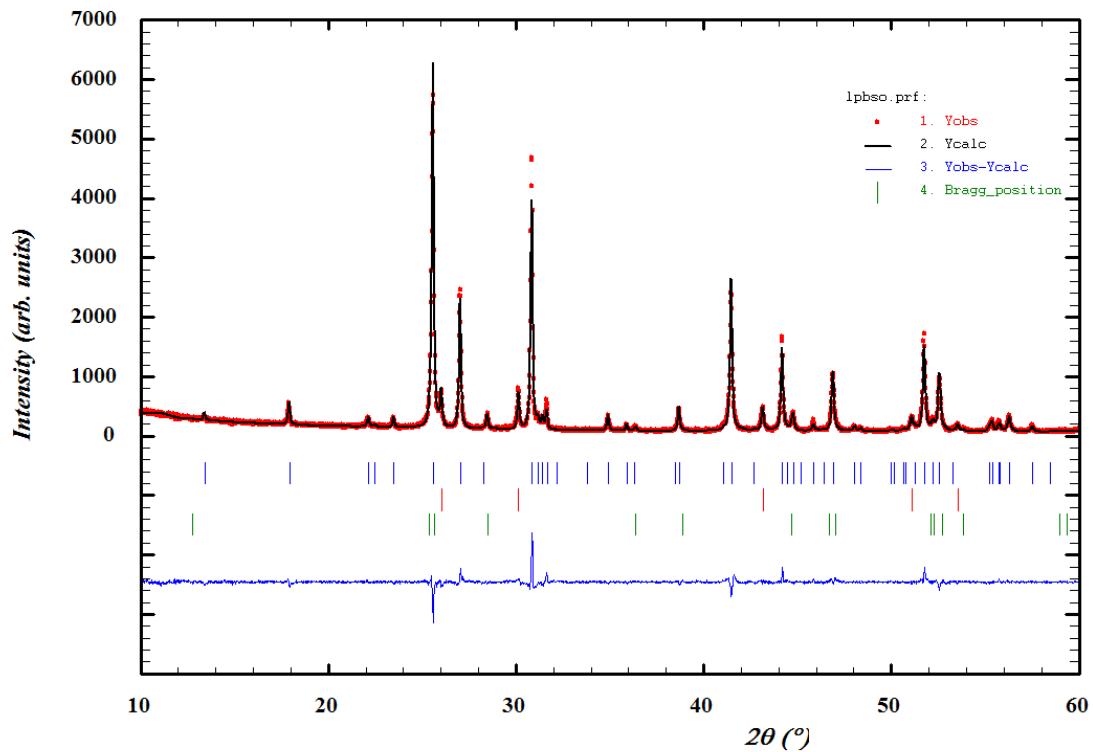
Figure S2a: Rietveld refinement of LaOPbBiS$_3$. Blue Ticks correspond to LaOPbBiS$_3$ phase, red ticks to PbS, and green ticks to La$_2$O$_2$S.

Pb/Bi = 0.99/1.01
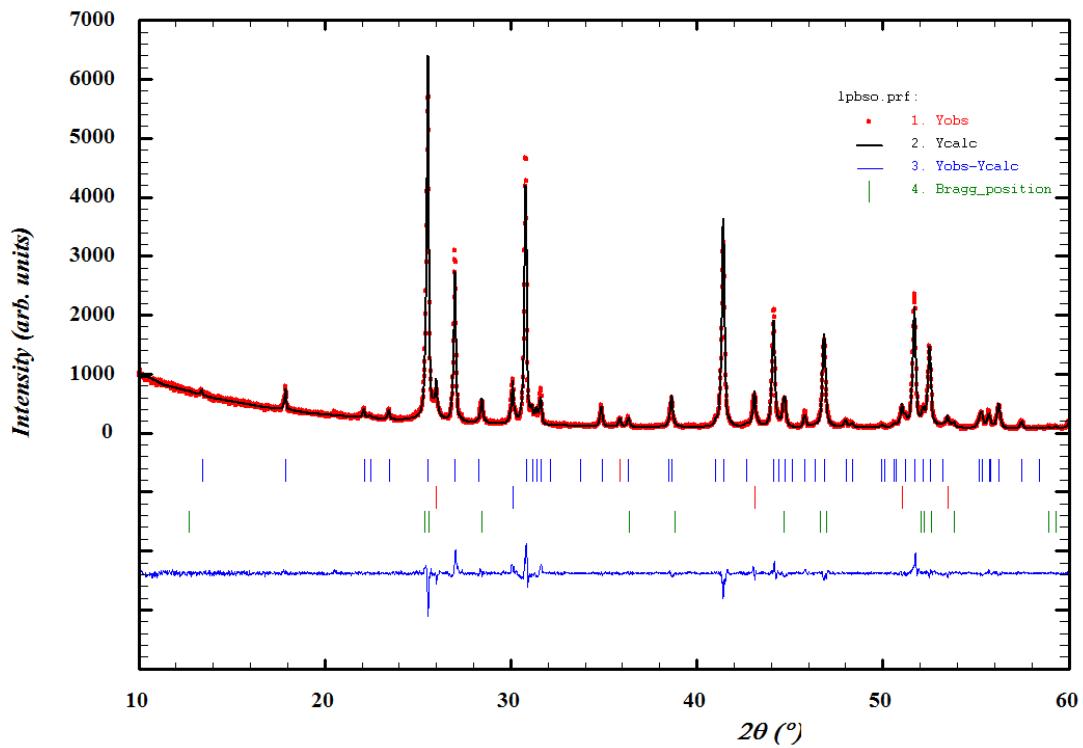
Figure S2b: Rietveld refinement of LaOPb$_{0.99}$Bi$_{1.01}$S$_3$.

Pb/Bi = 0.95/1.05
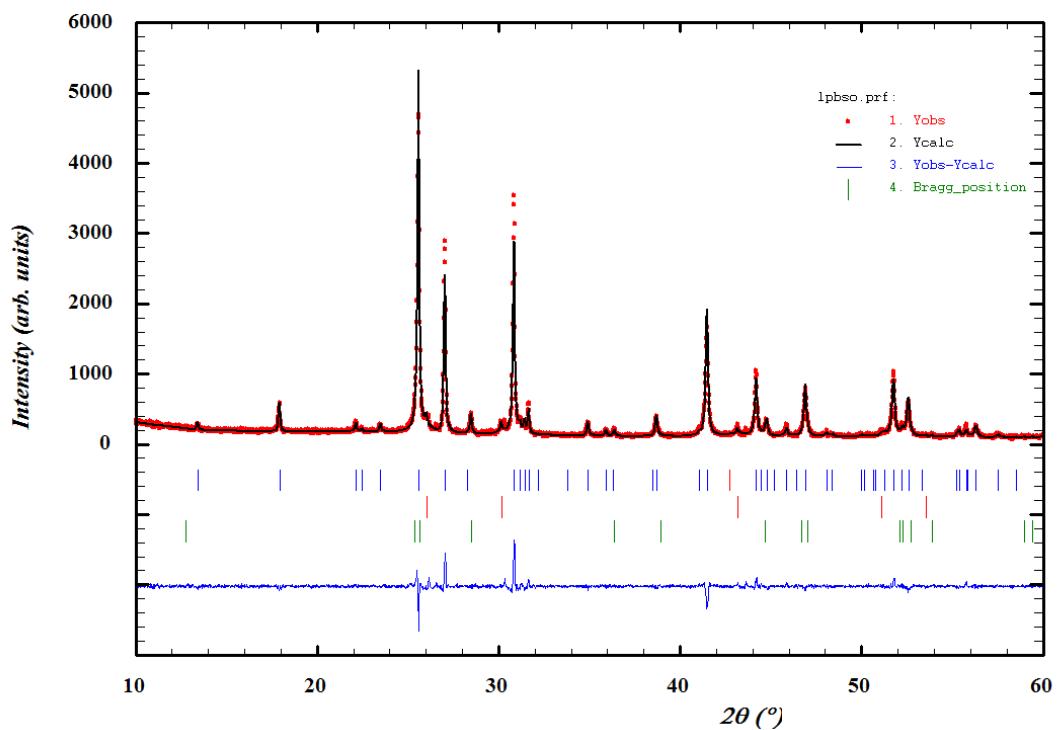
Figure S2c: Rietveld refinement of LaOPb$_{0.95}$Bi$_{1.05}$S$_3$.

Pb/Bi = 0.9/1.1
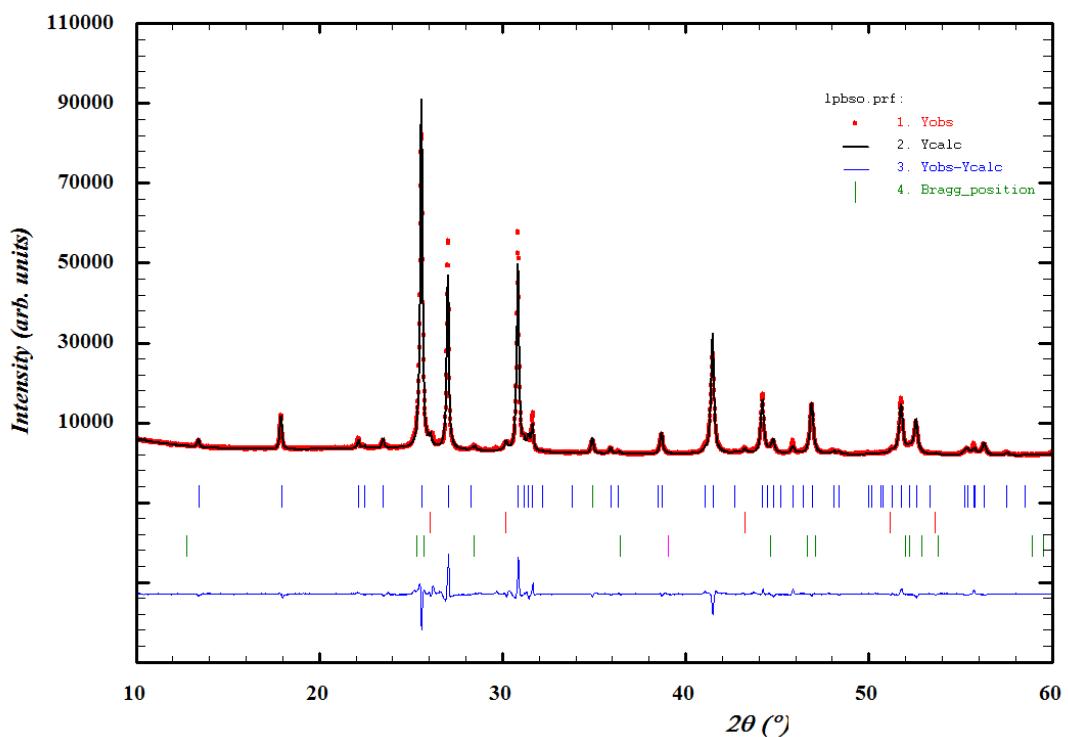

Figure S2d: Rietveld refinement of LaOPb$_{0.9}$Bi$_{1.1}$S$_3$.
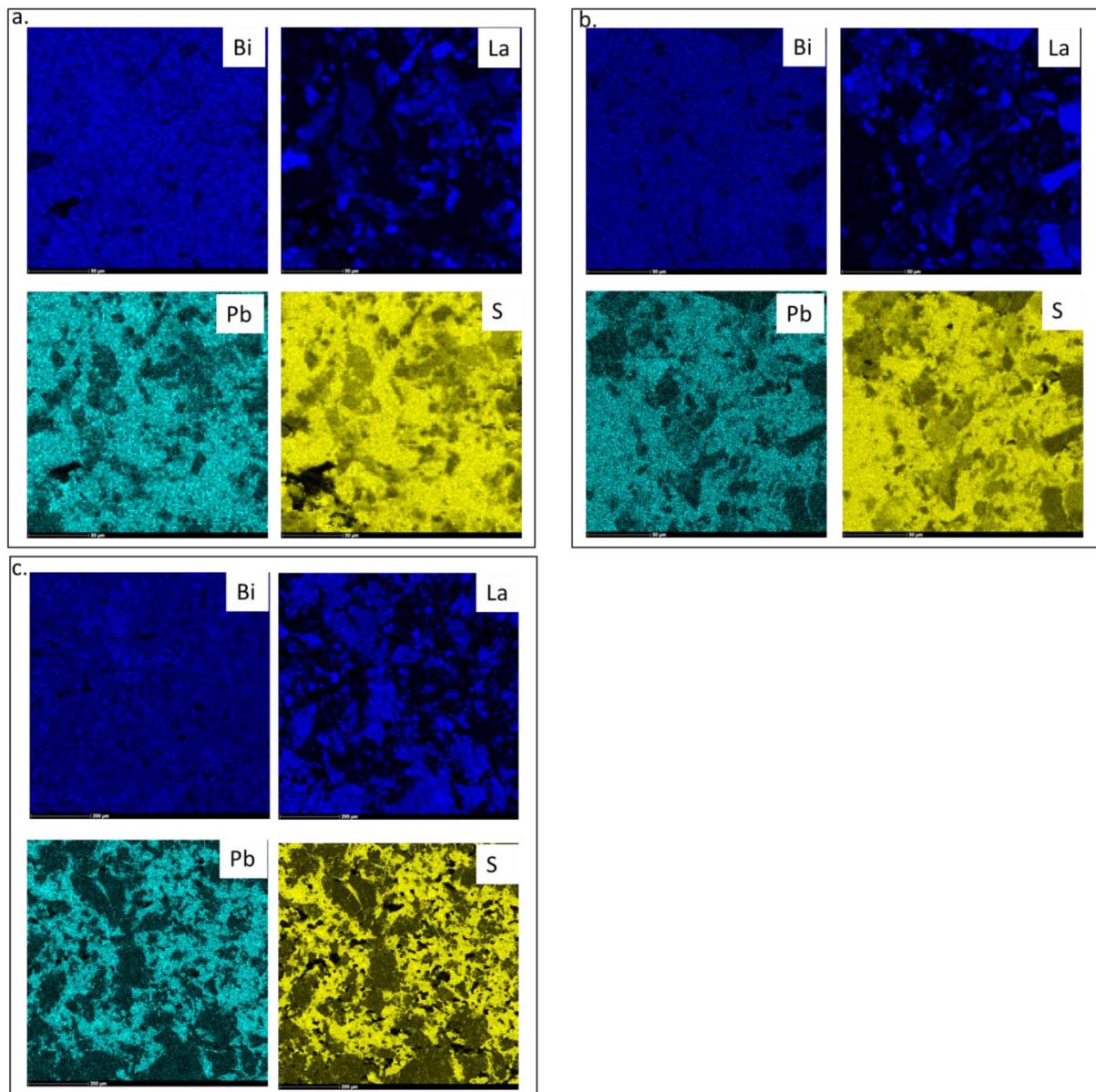
S3: EDX Mapping of LaOPb$_{1-y}$Bi$_{1+y}$S$_3$ synthesized with several proportions of Pb/Bi: 1.05/0.95 (a), 1/1 (b), 0.99/1.01 (c)
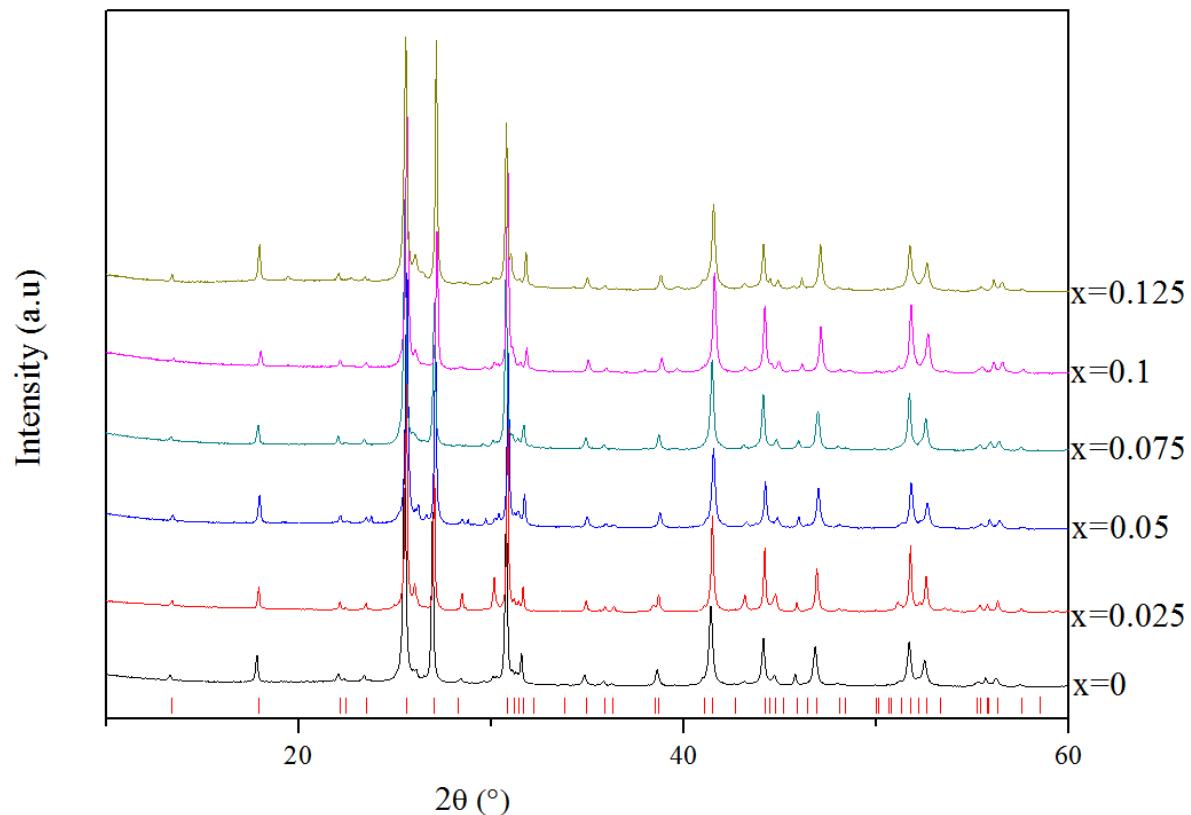
S4: XRD pattern of fluorine-doped samples with nominal composition $LaO_{1-x}F_xPb_{0.9}Bi_{1.1}S_3$ (x=0 to 0.125).
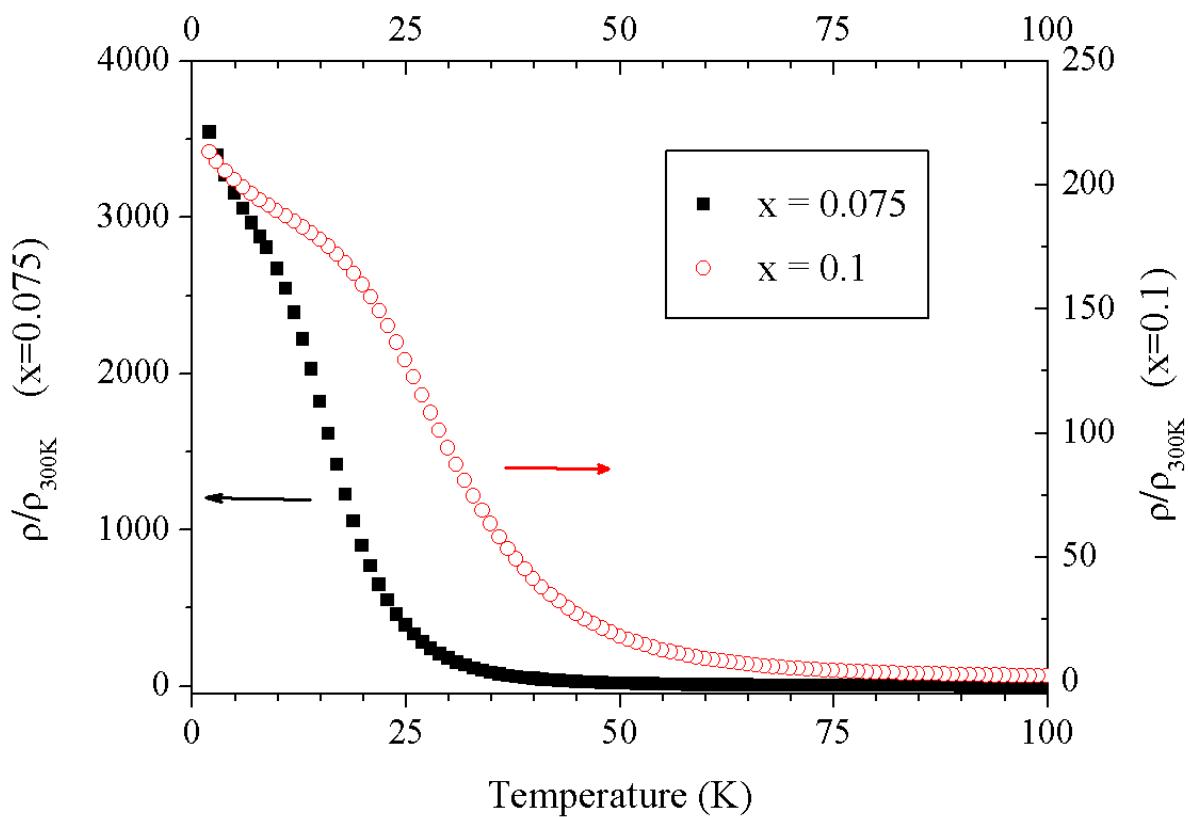
S5: Temperature dependence of the normalized resistivity for fluorine doped $LaO_{1-x}F_xPbBiS_3$ with x=0.075 and x=0.1.
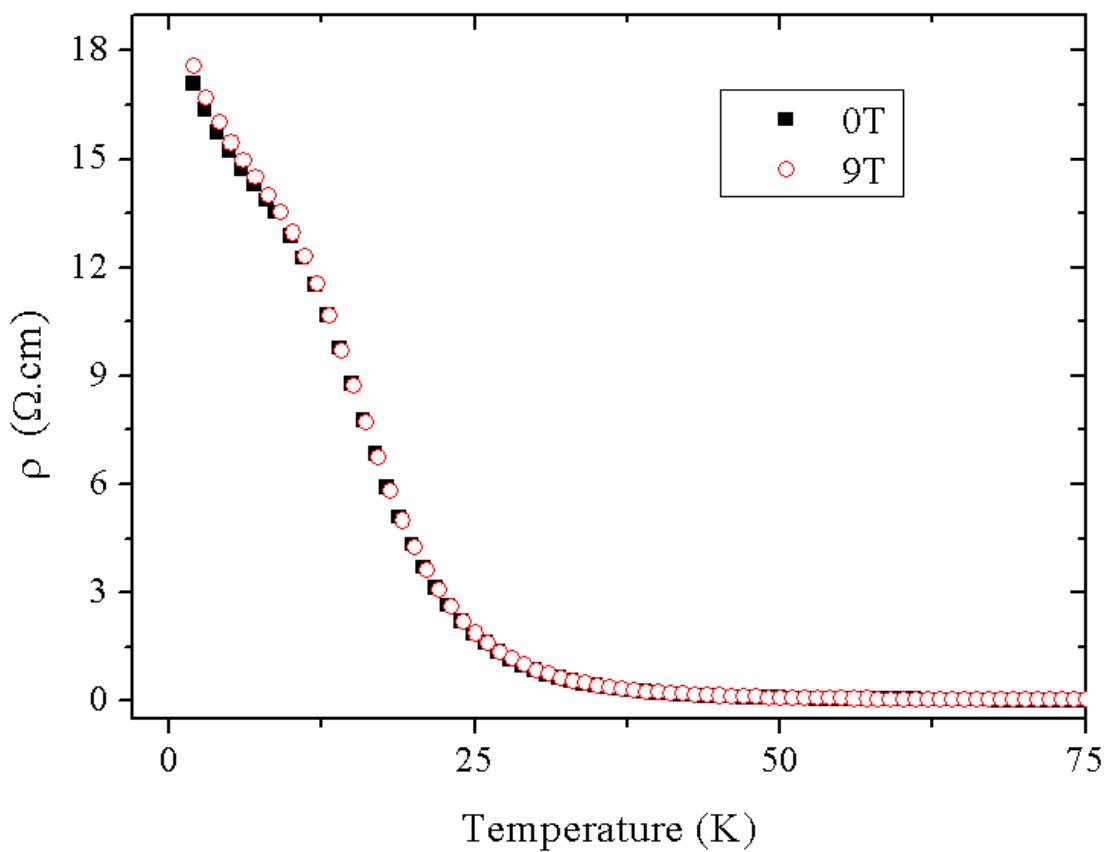
S6: Temperature dependence of the electrical resistivity of LaO$_{0.925}$F$_{0.075}$PbBiS$_3$ under 0T and 9T.
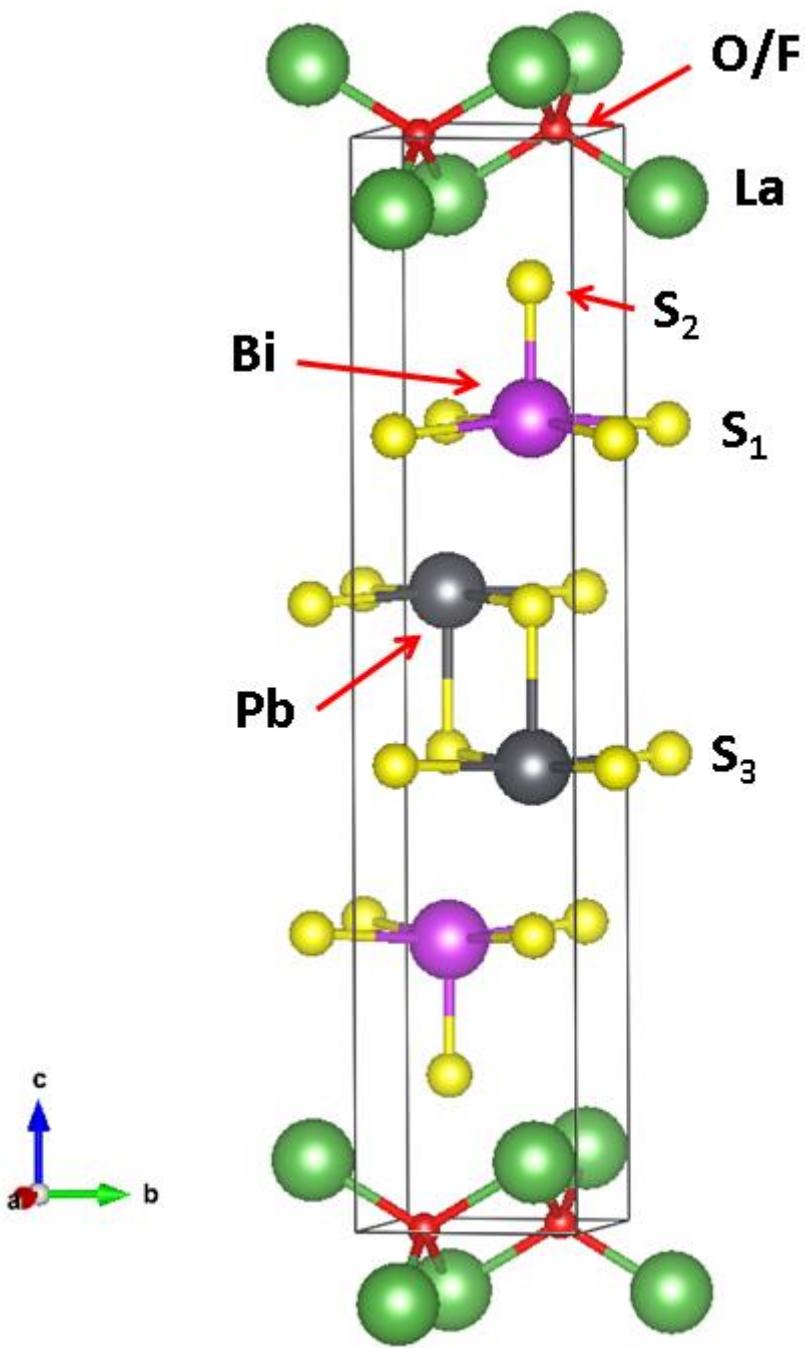
S7: atom position in the unit cell of $LaO_{1-x}F_xPbBiOS_3$ (with a total segregation of Pb and Bi for simplicity). Drawn using VESTA. [1]
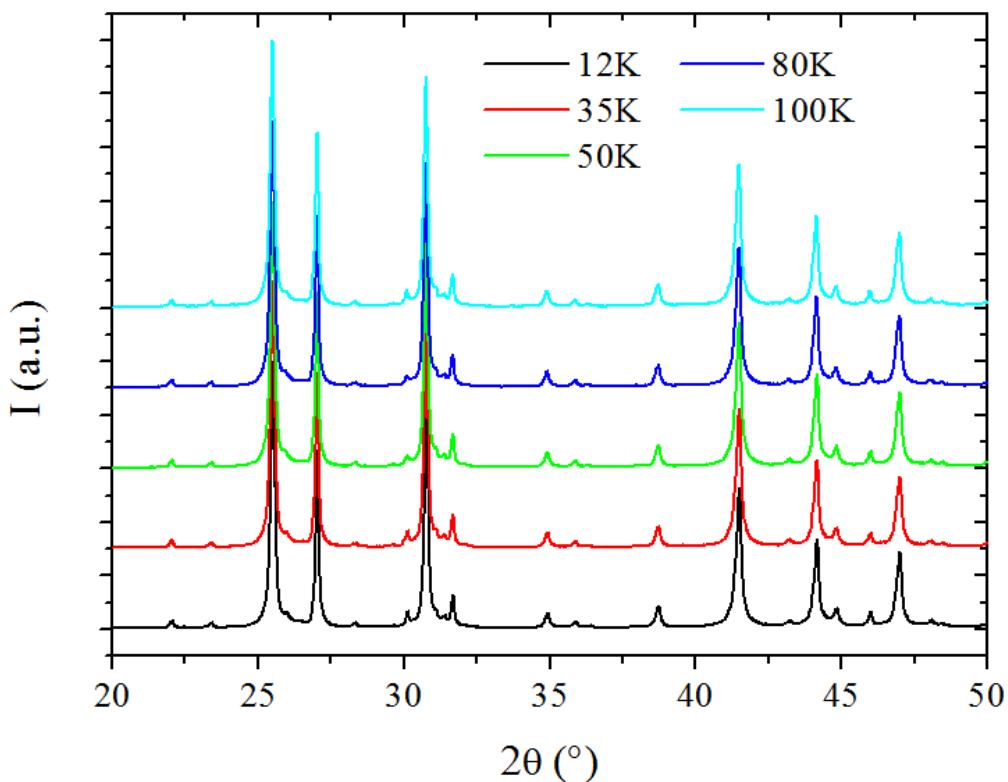
S8: XRD patterns of $LaO_{0.925}F_{0.075}Pb_{0.9}Bi_{1.1}S_3$ at several representative temperatures.
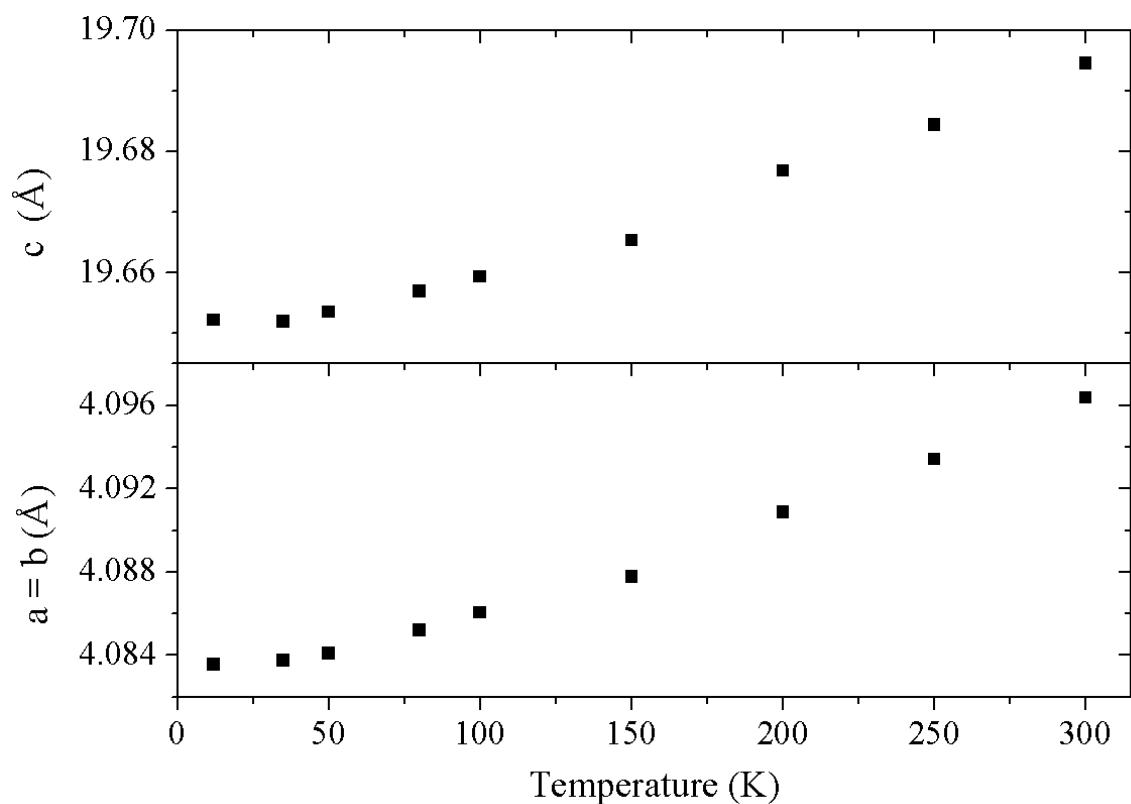
Figure S9: Temperature dependence of the lattice parameters of $LaO_{0.925}F_{0.075}Pb_{0.9}Bi_{1.1}S_3$.
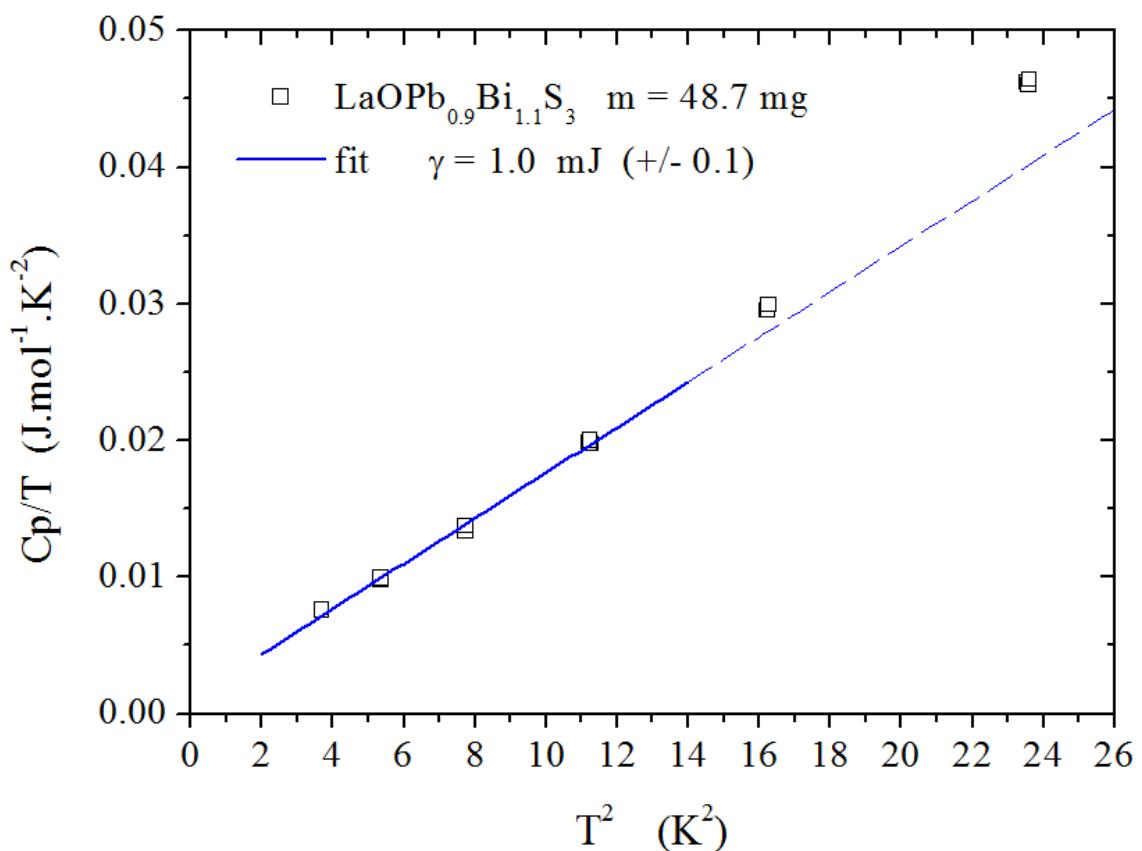
Figure S10: Low temperature fit of the specific heat of $LaOPb_{0.9}Bi_{1.1}S_3$ using a conventional Cp/T vs T^2 plot.

REFERENCES

1. K. Momma and F. Izumi, *J. Appl. Crystallogr.*, 2011, **44**, 1272.