

Composition-spread Growth and the Robust Topological Surface State of Kondo insulator SmB_6 Thin Films

Jie Yong^{1,2,*}, Yeping Jiang^{1,2,*}, Demet Usanmaz³, Stefano Curtarolo³, Xiaohang Zhang^{1,2}, Linze Li⁴, Xiaoqing Pan⁴, Jongmoon Shin⁵, Ichiro Tachuchi^{1,5}, Richard L. Greene^{1,2}

¹Center for Nanophysics & Advanced Materials, University of Maryland, College Park, Maryland 20742, USA

²Department of physics, University of Maryland, College Park, Maryland 20742, USA

³Department of Mechanical Engineering and Materials Science, Duke University, Durham, NC 27708

⁴Department of Materials Science and Engineering, University of Michigan, Ann Arbor, Michigan 48109, USA

⁵Department of Materials Science & Engineering, University of Maryland, College Park, Maryland 20742, USA

Topological insulators are a class of materials with insulating bulk but protected conducting surfaces due to the combination of spin-orbit interactions and time-reversal symmetry. The surface states are topologically non-trivial and robust against non-magnetic backscattering, leading to interesting physics and potential quantum computing applications^{1,2}. Recently there has been a fast growing interest in samarium hexboride (SmB_6), a Kondo insulator predicted to be the first example of a correlated topological insulator^{3,4}. Here we fabricated smooth thin films of nanocrystalline SmB_6 films. Their transport behavior indeed shows that SmB_6 is a bulk insulator with topological surface states. Upon decreasing the temperature, the resistivity ρ of $\text{Sm}_{0.14}\text{B}_{0.86}$ (SmB_6) films display significant increase below 50 K due to hybridization gap formation, and it shows a saturation behavior below 10 K. The saturated resistance of our textured films is similar to that of the single crystals, suggesting that this conduction is from the

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surface and robust against grain boundary scatterings. Point contact spectroscopy (PCS) of the film using a superconducting tip displays both a Kondo Fano resonance and Andreev reflection, suggesting the existence of both an insulating Kondo lattice and metallic surface states.

^{a)} Electronic mail: jyong@umd.edu ypjiang@umd.edu

*These authors contributed equally to this work

Samarium hexboride has recently been predicted to be the first example of topological Kondo insulator^{3, 4}. Upon decreasing the temperature, the resistivity of SmB₆ increases like an insulator but saturates at temperatures below 5K [ref. 5-14]. Recent transport measurements⁸⁻¹⁵ show that the resistance of this saturation is thickness-independent for both longitudinal and transverse directions⁸⁻¹⁰. Moreover, weak anti-localization and linear magnetoresistance have been observed in single crystal SmB₆ to support the presence of spin-momentum locked surface states¹². It has been shown that doping SmB₆ with magnetic impurities diminishes this saturation, while non-magnetic impurities do not⁹. These facts suggest that the conduction is from the surface state, it is protected by time-reversal symmetry, and is robust against non-magnetic scatterings. Quantum oscillations have been observed by torque magnetometry measurements¹⁶ and the tracking of the Landau levels in the infinite magnetic field limit points to -1/2, which indicates a 2D Dirac electronic state. Surface sensitive measurements such as angle resolved photoemission (ARPES)¹⁷⁻²³, scanning tunneling microscopy²⁴⁻²⁶ (STM) confirm the formation of hybridization gap and the existence of the surface states. One study²³ in particular suggests that the surface states are spin polarized with spin momentum locking which is a signature of the topological states. Preparation of SmB₆ in thin film geometries with smooth surfaces can facilitate device patterning and surface sensitive measurements that will give direct insight on the topological nature of this

material. In particular, smooth thin films would allow fabrication of heterostructures for probing the long-sought Majorana fermions by coupling this material with a conventional superconductor²⁷.

In this paper, we report the growth of SmB_6 films by co-sputtering of SmB_6 and boron targets. Composition and structural analyses show that our films are polycrystalline SmB_6 films with smooth surfaces. Transport and point contact spectroscopy (PCS) measurements confirmed a robust surface conduction at low temperatures with an insulating bulk, suggesting a topological behavior as found in single crystal SmB_6 .

We initially attempted growth of SmB_6 thin films via pulsed laser deposition and sputtering using single SmB_6 targets, but these growths resulted in grossly boron deficient films. In order to achieve the correct stoichiometry, we turned to the combinatorial composition-spread approach²⁸. By covering a large range of continuous average composition on a given spread wafer, this method can be used to ensure that somewhere on the wafer, there is a region with the correct average stoichiometry (see Methods for growth conditions).

The composition variation of the $\text{Sm}_x\text{B}_{1-x}$ spreads is mapped by wavelength dispersive spectroscopy (WDS), with an error bar of about 1%. Fig. 1a plots the composition of $\text{Sm}(x)$ mapped across a typical spread: it runs from about 0.01 directly underneath the B target to about 0.36 underneath the SmB_6 target. The desired composition of SmB_6 ($x = 0.14$) is obtained in the middle of the spread wafer. Fig. 1b shows the X-ray diffraction pattern (integrated in the χ direction) from the part of the spread film whose composition is SmB_6 . A broad ring was seen in

the area detector (see Supplementary I), which indicates the highly polycrystalline nature of the film. The pattern indicates that the film is preferentially oriented in the (001) direction, but peaks from other orientations such as (011) and (021) are also present. Upon scanning XRD across the entire range ($0.01 \leq x \leq 0.34$) we found that the predominant phase in the composition spread is always SmB_6 , regardless of x (See Supplementary II). This is surprising because there are reports of several other intermetallic phases^{29, 30} such as SmB_4 , Sm_2B_5 , etc.

To address this, we performed quantum mechanical parameterization of the Sm-B system to understand the phase formation and stability within the AFLOWLIB framework^{31, 32} using the standard computational parameters as specified in Ref. 33. Fig. 2 shows, in blue line, the minimum formation free energy (E_f) at zero temperature. Based on the calculations, the low temperature thermodynamics would indeed dictate that the first precipitating phase should be SmB_4 , having the minimum formation energy E_f in the entire composition range, since E_f represents the strength of the system to oppose phase separation. However, due to the hyper-thermal plasma process of sputtering, the Sm-B feedstock solution in the vapor phase starts off with an extremely large amount of intrinsic entropy (from translational degrees of freedom of vapor), and as a result, the entropic temperature T_s of a compound (a measure of the entropy to overcome the formation energy) emerges as the key factor which determines the strength of the system to compensate for the entropy of the feedstock^{33, 34}. Given the locations of phases in Figure 2, one sees that, from the point of view of the vapor, the most “accommodating” phase to nucleate is actually SmB_6 with its highest relative T_s compared to the other phases. This follows from the fact that during nucleation, the entropy maximization will limit the flow of latent heat and therefore chooses the phase most capable of accepting and transforming entropy (from

translational to configurational). Thus, the non-equilibrium nature of the co-sputtering process has served as the key enabler which allowed us to achieve the desired SmB_6 phase in the present synthesis method.

To further confirm the existence of SmB_6 , we performed cross sectional transmission electron microscopy (TEM) and Raman spectroscopy on films with x near 0.14. Fig. 1c shows the high resolution cross sectional TEM images of films on a MgO substrate. Nanocrystalline SmB_6 grains with different orientations can be clearly seen. Fig. 1c2 and Fig. 1c3 show the enlarged images (after filtering) of two selected grains with (011) and (111) orientations, respectively. The lattice constant determined from the image is 4.14 Å, which is consistent with that of the reported bulk value. We also performed Atomic Force Microscopy (AFM) on our films and the typical rms roughness is less than 1.0nm for a 20µm by 20µm area.

Raman spectrum of a SmB_6 film on Si is taken at room temperature using a 532 nm wavelength laser. The result is shown in Fig 1d. Three modes T_{2g} , E_g and A_{1g} from SmB_6 (cubic structure with the $\text{Pm}3\text{m}$ symmetry) are seen in addition to the Si substrate peak at 510 cm^{-1} [Ref. 35]. The wavenumbers of these three peaks are consistent with those from the single crystals. They are first order Raman modes which involve the displacement of the boron octahedral. The Sm ion is at a site of inversion symmetry and cannot contribute to first order phonon Raman scattering. The fact that the widths of these three peaks are wider than the Si substrate peak also confirms that these films are polycrystalline.

We have carefully extracted different sections of the spread film with different compositions to carry out resistivity measurements. The sheet resistance R_s (Fig. 3 left axis) and resistivity ρ (Fig. 3 right axis) vs temperature curves are plotted for three different boron concentration films with thickness about 100nm. We use sheet resistance here since at low temperatures the measured resistance arises from the surface state as discussed below. Comparing the three concentrations ($x = 0.10, 0.14$, and 0.22), the room temperature resistivity $\rho(300K)$ increases with increasing boron concentration. At 300K, a typical resistivity for a stoichiometric SmB_6 film is around $300 \mu\Omega \text{ cm}$, which is similar to that of bulk SmB_6 crystals^{1, 2, 13, 16}.

For single SmB_6 crystals, though the proposed topological property is yet to be verified, the bulk insulating and surface conductive nature have been established⁸⁻¹¹. Our films show similar behaviors. As the temperature is decreased, all three films show increase in the resistivity with a particularly rapid rise below 50K. This behavior is consistent with the opening of the hybridization gap between the itinerant 5d band and localized Sm 4f band as found in bulk SmB_6 crystals⁵⁻⁷. Below 10 K, the sheet resistance displays a saturating behavior for the film with $x > 0.10$, but it continues to increase for the $x < 0.10$ sample. The saturation behavior, regarded as a mystery for decades, is now widely accepted as the signature of the emergence of the conducting surface state. We note that our saturated sheet resistance ($R_s \sim 50 \Omega$ at 2 K) is very close to the reported sheet resistance of single crystals at low temperatures⁹. If the low temperature conduction is from the bulk of the film, one would expect a much larger sheet resistance because the films are three orders of magnitude thinner than typical crystals. Also if the conduction is from some trivial surface state, one would expect a very large sheet resistance due to grain boundary scattering because our films are nanocrystalline. The fact that we observe a very

similar sheet resistance strongly supports that this low temperature conduction is from the surface and that this surface conduction is protected against conventional grain-boundary scatterings. This agrees with the predicted nontrivial nature of the surface states in SmB_6 , in which the topologically protected surface states exist only at the interfaces between materials of different topology. Thus, the similar and robust surface conduction in our polycrystalline SmB_6 films compared to single crystals strongly supports the topological aspect of the surface states.

The resistivity ratio between 300 K and 2 K is about 1.7, which is substantially lower than that found in single crystals, where the ratio can be as large as 10^4 [Ref. 5-7]. This can be explained by a much larger surface to bulk conduction ratio for films. It has indeed been shown that when single crystals are thinned down from 200 μm to 37.5 μm , this ratio $\rho_{2\text{K}}/\rho_{300\text{K}}$ decreases from 10^4 to less than 3000. A detailed study¹⁵ has shown that the ratio has roughly a linear dependence on the thickness. Given the ratio value of 1.7, an extrapolation of this linear dependence gives a thickness of 242 nm, which is of the order of our film thickness (~ 100 nm). Of course, this linear relationship might not be valid down to 100 nm, but this rough agreement with the linear thickness dependence demonstrates that our thin films are already in a limit where the surface conduction at lowest temperature is comparable to the bulk conduction at room temperature. Therefore, we attribute the saturation and the small resistance increase in our films to the surface conducting state.

The Kondo nature of SmB_6 can be investigated by point contact spectroscopy (PCS)¹¹. Fig. 4a (the bottom curve) shows a typical PCS curve of our SmB_6 films with a PtIr tip, exhibiting an asymmetric Fano line shape^{36, 37}. When a superconducting tip is used for the contact (the top

curve of Fig. 4a), inside the Fano dip and around the Fermi level a clear double peak feature appears, indicating the emergence of Andreev reflection, which takes place in a metal-superconductor junction. The fitting to the Fano line shape (blue curves) gives a Kondo gap of around 19 meV, which is similar to that observed in single crystals¹¹. The red curve is the best fit to the Fano line shape and the Blonder-Tinkham-Klapwijk model³⁸, which gives $\Delta = 1.35$ meV, $Z = 0.57$ (barrier strength), $\Gamma = 1.0$ meV (broadening term). The gap value is consistent with that of Nb, and above its T_c , the spectrum only shows a Fano line shape with no Andreev enhancement. When the point contact force is varied, the barrier strength (Z) of the junction decreases with junction resistance, while clear signature of the superconducting gap remains (Fig. 4b). The co-occurrence of the Kondo feature and Andreev reflection in the SmB₆-superconductor junction clearly signifies the Kondo nature and the simultaneous existence of metallic states at the Fermi level in our films, which, although polycrystalline, reinforces the theoretical prediction that SmB₆ is a topological Kondo insulator. The observation of Andreev reflection implies that high transparency can be achieved in the superconductor/SmB₆ interface. Synthesis of SmB₆ thin films with true bulk insulation is an important step towards pursuing proximity induced topological superconductivity in SmB₆.

Methods:

Fabrication. SmB₆ and B targets were co-sputtered at dc 50 W and rf 100 W, respectively, in a high-vacuum combinatorial co-sputtering chamber. Boron is brittle and insulating, and its deposition rate is much lower than that of the SmB₆ target. The base pressure of the deposition chamber is typically 2×10^{-8} torr. The substrates used were MgO and 3-inch (001) oriented Si

wafers with a 300 nm amorphous SiO₂ layer. Films on different substrates show consistent behavior. A physical shadow mask is placed over the wafer during the deposition to naturally separate the film into 2 mm x 2 mm squares. The typical argon pressure was 6.5 mTorr during the deposition, and the substrate was heated at 800 °C during the deposition and for 3 hours following the deposition in vacuum. The deposition thickness across the spread varied from 80 – 200 nm.

Characterization and measurements X-ray diffraction (XRD) of the spread is carried out with an area detector (Vantec500) using a Bruker D8 Discover with the CuK α line. Raman microscopy is done in a Horiba Jobin Yvon LabRam ARAMIS spectrometer using a 532 nm wavelength laser. Transport and point contact spectroscopy measurements are done in a Quantum Design Physical Property Measurement System (PPMS). Sheet resistance is measured in four point Van der Pauw configurations. Contacts are made by a wire bonder using Al wires. Sheet resistance is calculated by $R_s = V \pi / I / \ln 2$. Resistivity is calculated by $\rho = R_s t$ (thickness of the film)

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Author contributions: J.Y., I. T. and R. G. conceived the project; J.Y. fabricated samples and did XRD, AFM and Raman studies; J. Y., X. Z, and J. S performed transport studies; L.L and X. P. performed TEM study; Y. J. carried out PCS studies; D. U. and S. C. performed the phase

diagram calculations; J.Y., Y. J., D. U., S. C., I. T., R. G. wrote the manuscript. All authors contributed to discussions and gave comments to the manuscript.

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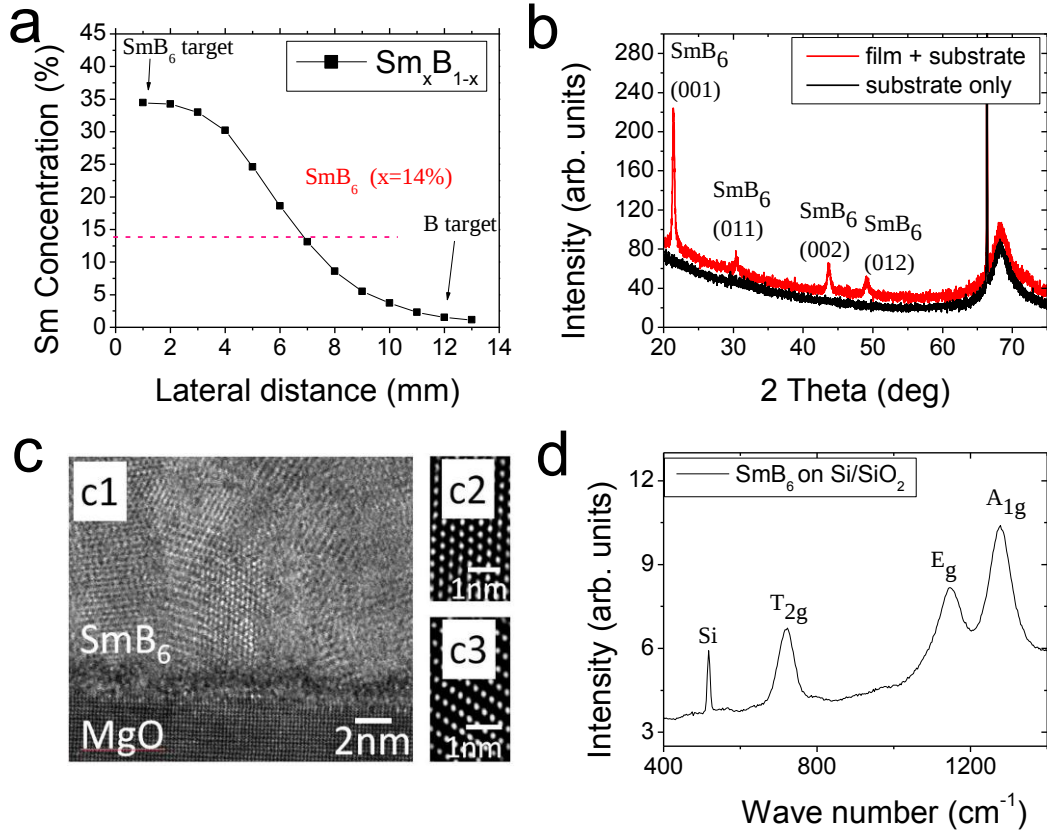


Figure 1: The fabrication and characterization of SmB_6 . a, Samarium concentration x , measured by WDS, as function of lateral distance. b, Integrated X-ray diffraction intensity with aerial detector for the film with the substrate (in red) and with only the substrate (in black). The substrate is 300 nm SiO_2 on Si wafer. c, Cross-sectional TEM image on the interface between SmB_6 film and MgO substrate. (c1) raw image revealing textured structures with amorphous areas (c2) and (c3) are selected areas after filtering showing SmB_6 (011) and (111) planes. d, Raman spectrum of SmB_6 films on Si/SiO₂ substrate. The laser wavelength used is 532nm. Three modes T_{2g} , E_g , A_{1g} from SmB_6 are clearly seen.

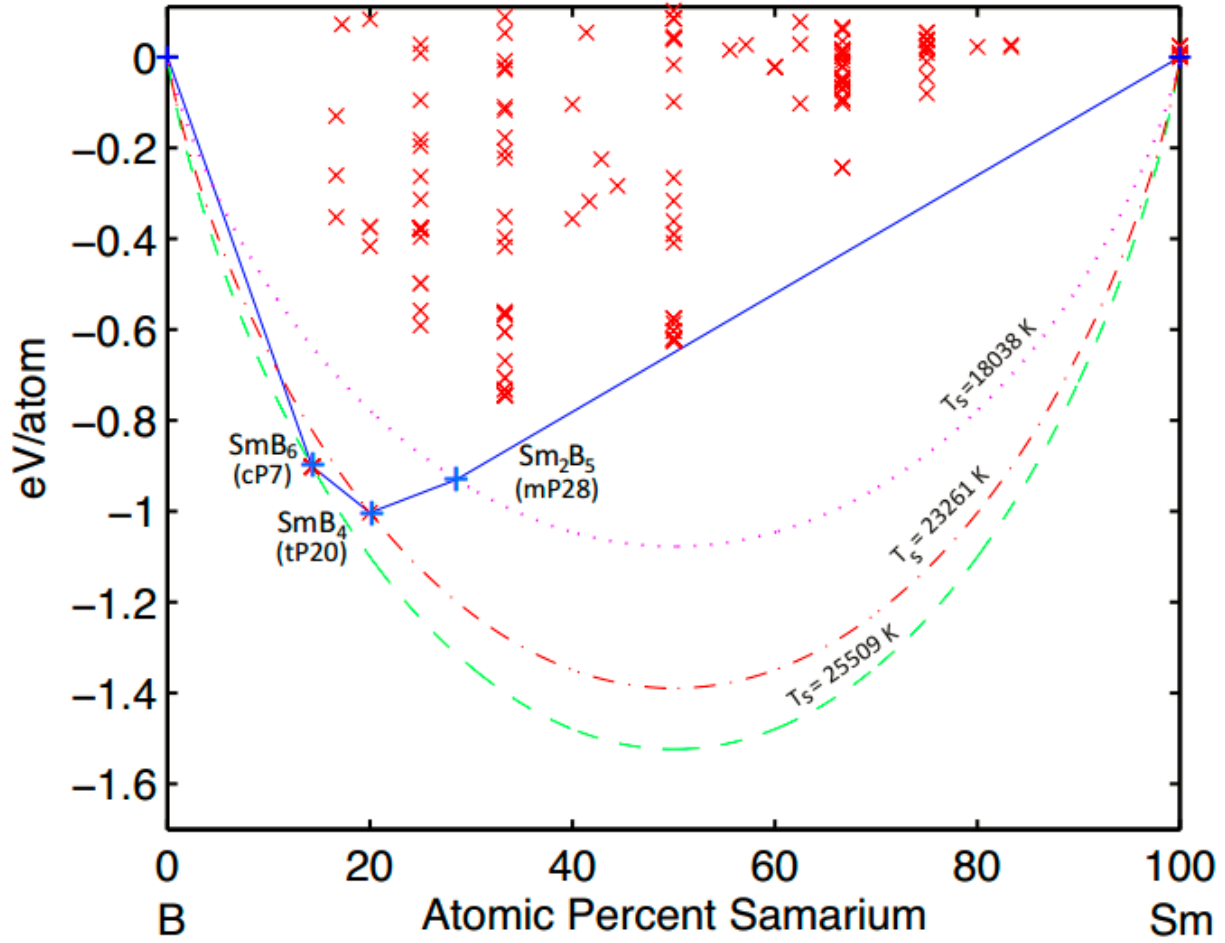


Figure 2: Free energy convex hull for B-Sm at zero temperature (solid blue line). Blue/red crosses indicate stable/unstable phases. Space groups are in parentheses below the stable compound stoichiometries. The three dashed curves indicate the entropic temperature envelopes^{33, 34}. Cooling from the hyper-thermal plasma of sputtering it is actually SmB_6 with the highest T_s which nucleates first.

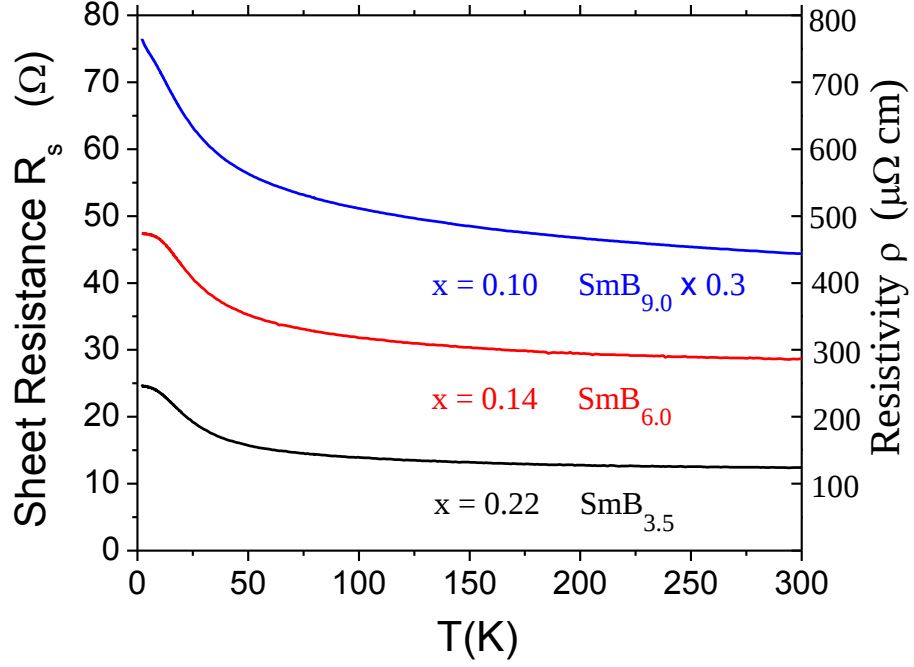


Figure 3: Temperature dependences of sheet resistances (left axis) and resistivities (right axis) for three $\text{Sm}_x\text{B}_{1-x}$ films with $x=0.10, 0.16, 0.22$ respectively. The films are about 100nm thick and resistances are measured using Van Der Pauw method (see Methods).

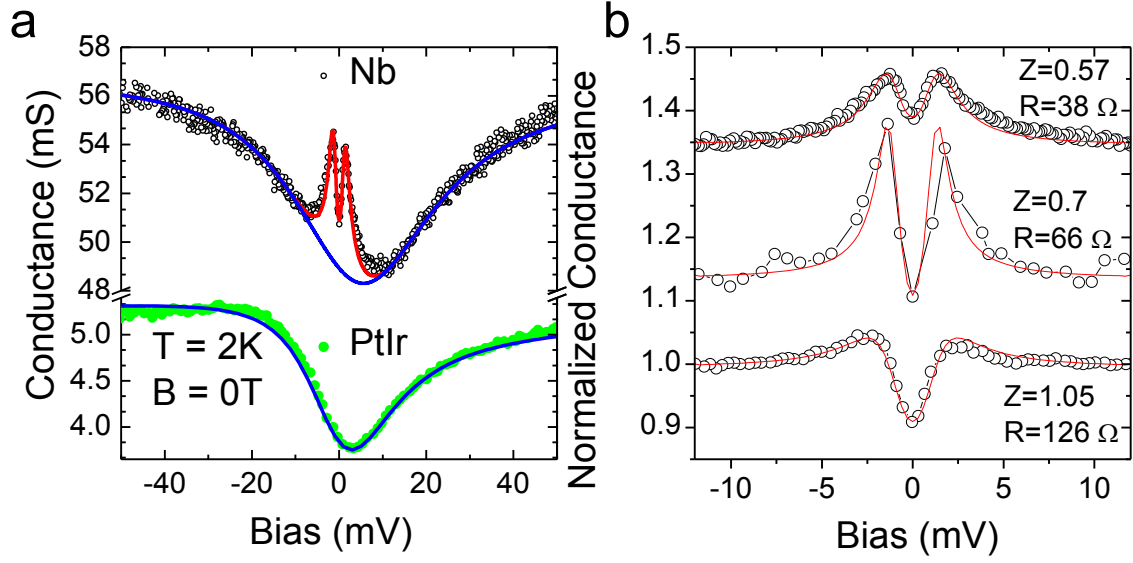


Figure 4: Point Contact Spectroscopy (PCS) on SmB_6 films. (a) PCS on the SmB_6 film with a PtIr tip (green filled squares) and a superconducting Nb tip (black open circles). The blue curves are the best fittings to the Fano line shape. The red curve is the fitting to both Fano line shape and modified BTK model. (b) Andreev reflection data (black open circles) of different Nb/ SmB_6 junction resistances. Fano background is subtracted. The red curves are the fitting to the BTK model. The spectra here were taken at 2 K and in zero magnetic field. Details of BTK fit and data analysis are given in Supplementary III.

Supplementary information

Jie Yong^{1,2,*}, Yeping Jiang^{1,2,*}, Demet Usanmaz³, Stefano Curtarolo³, Xiaohang Zhang^{1,2}, Linze Li⁴, Xiaoqing Pan⁴, Jongmoon Shin⁵, Ichiro Tachuchi^{1,5}, Richard L. Greene^{1,2}

¹Center for Nanophysics & Advanced Materials, University of Maryland, College Park, Maryland 20742, USA

²Department of physics, University of Maryland, College Park, Maryland 20742, USA

³Department of Mechanical Engineering and Materials Science, Duke University, Durham, NC 27708

⁴Department of Materials Science and Engineering, University of Michigan, Anne Arbor, Michigan 48109, USA

⁵Department of Materials Science & Engineering, University of Maryland, College Park, Maryland 20742, USA

I. 2D-XRD using an area detector

Polycrystalline films can be characterized using X-ray detector with an area detector. With a fixed angle between incident X-ray and the sample, the area detector can detect diffractions that come from the grains of different orientations in the sample. Polycrystalline films will show a broad ring (Fig. S1) in the detector while single crystal show a very broad spot (the incident angle must satisfy Bragg's law) which need to be avoided. Integration of the data along χ direction can be carried out to obtain $\theta - 2\theta$ XRD plots (Fig. 1b and Fig. S2)

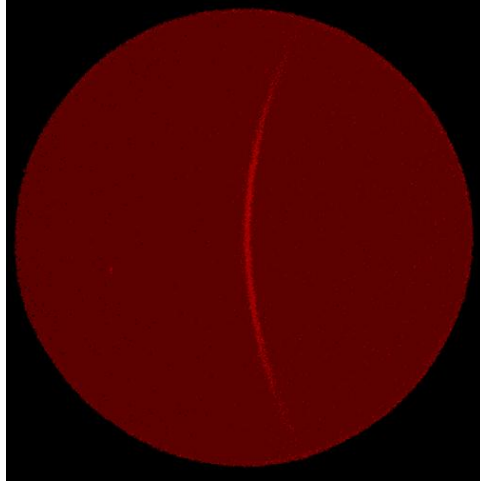


Figure S1: raw image of XRD area detector, the broad ring is from polycrystalline SmB₆ (001) peak.

II. XRD spectra of Sm_xB_{1-x} as a function of x

Figure S1 shows the spectra of Sm_xB_{1-x} for seven different x. Only peaks from SmB₆ phase can be identified for any given x. This indicates SmB₆ is always the dominant phase in our growth setup, regardless of x. The main text explains why this is the case. Figure S3(a) shows the a small shift in SmB₆ (001) peak as x changes. This corresponds to a lattice constant change from 4.16 Å for small x to 4.12 Å for large x. This is because the higher the boron concentration (smaller x), the more boron atoms will fit into the lattice of much larger samarium atoms and the lattice constant increases. It is also interesting to note that the lattice change mostly occurs where samarium concentration is around 20% to 25%.

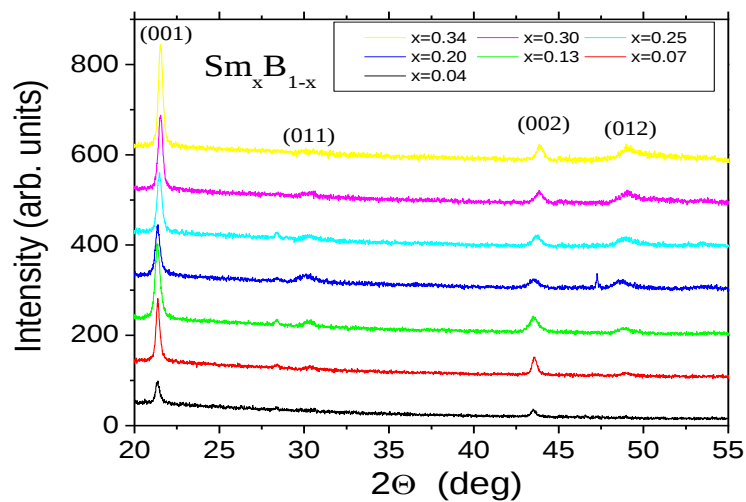


Figure S2: XRD spectra of $\text{Sm}_x\text{B}_{1-x}$ with different composition x . Vertical axis are shifted for clarity purposes.

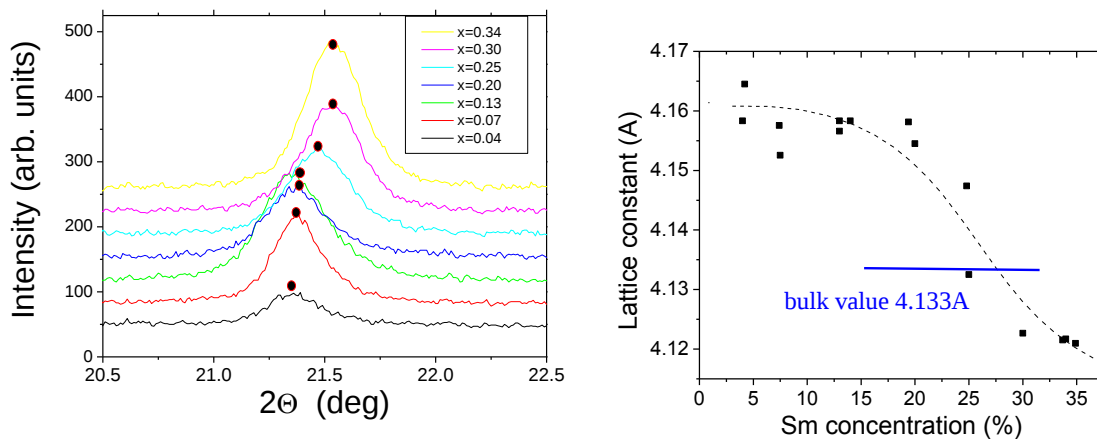


Figure S3: (a) 2-D XRD spectra on $\text{Sm}_x\text{B}_{1-x}$ films with different x showing SmB_6 (001) peak (purple dots) shift. Vertical axis is shifted for clarity purposes. (b) SmB_6 lattice constant, from fit of 2D XRD data, as a function of Samarium concentration in films. The dashed curve is a guide to the eye.

III. Blonder-Tinkham-Klapwijk (BTK) fit

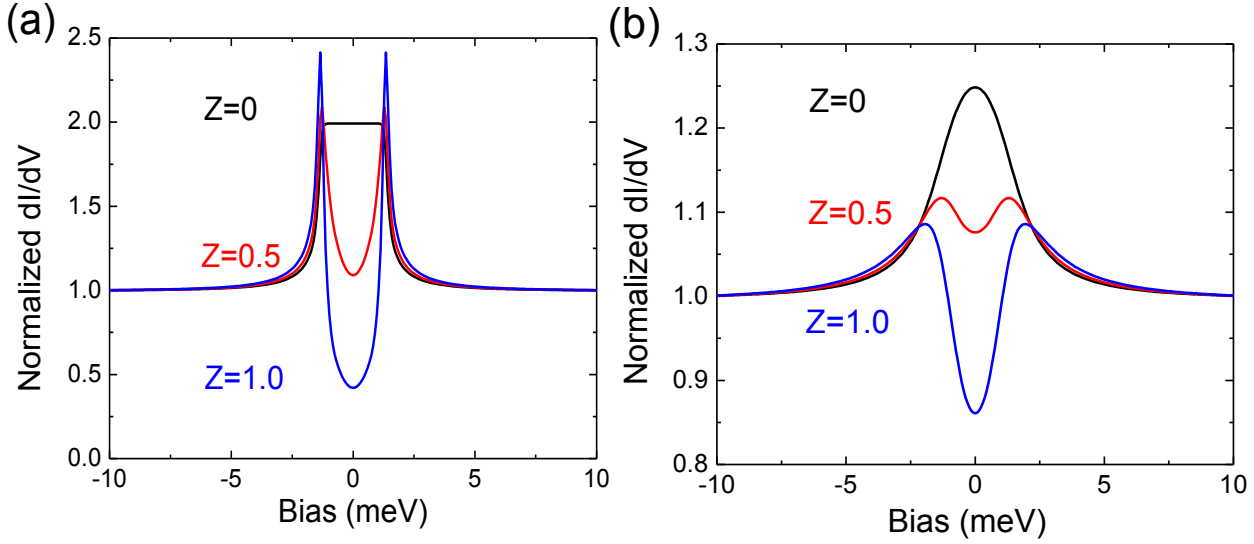


Figure S5: Calculated conductance curves using the modified BTK model. (a) $Z = 0, 0.5, 1.0$ with T , Δ and Γ fixed at 2.0 K, 1.35 meV and 0 meV. (b) $Z = 0, 0.5, 1.0$ with T , Δ and Γ fixed at 2.0 K, 1.35 meV and 1.0 meV.

The conductance G_{NS} (dI/dV) as a function of the bias voltage V of a N/S junction can be described by the BTK theory [ref. S1]:

$$G_{NS}(V) \propto \int_{-\infty}^{+\infty} \frac{\partial}{\partial V} [f(E - eV)][1 + A(E) - B(E)] dE,$$

where $A(E)$ and $B(E)$ are the probability of Andreev reflection and normal reflection, $f(E)$ is the Fermi distribution function. $A(E)$ and $B(E)$ can be calculated as follows,

$$\begin{aligned} A(E) &= a \cdot a^* \\ B(E) &= b \cdot b^* \\ a &= \tilde{u}\tilde{v} / \gamma \\ b &= -(\tilde{u}^2 - \tilde{v}^2)(Z^2 + iZ) / \gamma \\ \gamma &= \tilde{u}^2 + (\tilde{u}^2 - \tilde{v}^2)Z^2 \end{aligned}$$

Z represents the interfacial barrier strength, and the coherence factors $\tilde{u}$ and $\tilde{v}$ are

$$\tilde{u}^2 = \frac{1}{2} \left[1 + \frac{\sqrt{(E + i\Gamma)^2 - \Delta^2}}{E + i\Gamma} \right]$$

$$\tilde{v}^2 = \frac{1}{2} \left[1 - \frac{\sqrt{(E + i\Gamma)^2 - \Delta^2}}{E + i\Gamma} \right],$$

in which an imaginary component Γ is introduced to describe a finite quasiparticle lifetime affected by inelastic scattering near the N/S interface. The inelastic scattering can result in a smearing effect on the conductance spectrum.

The effect of Z and Γ on the conductance spectrum is shown in Figure S5. For an ideal N/S interface with $Z = 0$ and $\Gamma = 0$, the spectrum has a flat top and the enhancement factor is 2 inside the superconducting gap where Andreev reflection takes place. As Z increases, the conductance in the gap is suppressed. With a relatively large Γ , the enhancement at low Z is reduced and the double-peak structure in the case of a finite Z is smeared.

Reference

S1. Blonder, G. E. Tinkham, M. & Klapwijk, T. M. Transition from metallic to tunneling regimes in superconducting microconstrictions: Excess current, charge imbalance, and supercurrent conversion. Phys. Rev. 25, 4515 (1982).