Topological Crystalline Insulator Pb$_{1-x}$Sn$_x$Se Nanowires with \{100\} Facets

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Topological insulators (TIs) are a recently discovered class of unusual quantum state of matter characterized by a gapless metallic surface state and insulating bulk gap.\cite{1} The electron spin on the surface of TIs is locked perpendicular to the momentum. Under the protection of time-reversal symmetry, the surface state of TIs is immune to any time reversal perturbations such as non-magnetic impurities and crystal defects.\cite{2} The distinct properties of TIs make them promising materials for the potential applications of novel spintronics and low-dissipation quantum computation.\cite{3,4} In addition, it is strongly expected to realize unique quantum mechanical effect on TIs such as quantum anomalous hall effect\cite{5} and Majorana fermions.\cite{3}

Recently, topological crystalline insulators (TCIs) have been proposed as a new topological state in which crystal symmetry replaces the roles of time-reversal symmetry in guaranteeing the topological protection.\cite{6-9} Different from conventional TIs, gapless metallic surface states resides on only highly-symmetry crystal surfaces of TICs such as crystal planes \{110\} and \{111\}.\cite{6,7,9} Soon after the theoretical predication of SnTe\cite{6} and its related alloys Pb$_x$Sn$_{1-x}$(Se/Te)\cite{10,11} as TCI, they have triggered considerable research interest of theorists and experimentalists from all over the world. Angle-resolved photoemission spectra (ARPES) has confirmed the existence of Dirac cones on surface (001) of SnTe\cite{6} and its related alloys Pb$_x$Sn$_{1-x}$(Se/Te).\cite{10,11} More importantly, the electronic structure of SnTe or Pb$_x$Sn$_{1-x}$(Te, Se) is found to be continuously tunable by exerting external perturbation such as electrical field\cite{13} and elastic strain.\cite{6} These intriguing properties of TCIs may allow for applications of tunable electronic and spintronic devices.\cite{7}

Low-dimensional TIs nanostructures are the ideal system to investigate the surface topological nature since they effectively suppress the carrier transport from the bulk due to their large surface-to-volume ratio.\cite{14-17} Significantly, low-dimensional TIs nanostructures are crucial for realizing novel quantum mechanics phenomena and applications as nanoscale electronic and spintronic devices.\cite{14,15,17,18} From the point of this view, it is highly important to develop the growth technique for high quality single crystalline TIs nanostructures. Great success has been achieved on the growth of TIs nanostructures by vapor deposition method such as Bi$_2$Se$_3$ nanoribbons\cite{14} few-layer Bi$_2$(Se/Te)$_3$ nanoplates\cite{19} and Ag$_2$Te nanowires\cite{20}. As for TCIs, controlling the morphologies of nanostructures with distinct surfaces is of great importance for revealing the unique surface topological nature because topological surface states are strongly dependent on highly symmetric crystal surfaces.\cite{11,21} Our group and S. X. Zhang et al. have demonstrated controllable synthesis of highly single crystalline SnTe nanostructures with distinct surfaces, such as nanowires terminated by only \{100\} surfaces or \{100\} and \{111\} surfaces\cite{22} and microcrystals covered by only \{100\} or \{111\} surfaces by using the similar vapor deposition method.\cite{25} However, few works report the low-dimensional nanostructures of another important TCI Pb$_{1-x}$Sn$_x$(Se/Te), which may be due to the more complicated growth conditions compared with the simple stoichiometric SnTe.

Pb$_{1-x}$Sn$_x$(Se/Te) crystallizes in the rock-salt structure and has a direct electronic bandgap ($E_g \leq 0.29$ eV). Moreover, Pb$_{1-x}$Sn$_x$(Se/Te) possesses some unique physical properties in contrast to SnTe. In the alloy Pb$_{1-x}$Sn$_x$(Se/Te), the system undergoes a band inversion when Sn content $x$ exceeds a critical value $x_c$.\cite{22} The band inversion also occurs when temperature is lowered below gap-inversion temperature $T_{inv}$ in the samples $x > x_c$. It is worth noting that recent theoretical calculation and ARPES experiments have pointed out that Pb$_{1-x}$Sn$_x$(Se/Te) changes from a trivial insulator to a TCI accompanying with the band inversion.\cite{10,12} That means tunable electronic and spintronic device may be realized based on Pb$_{1-x}$Sn$_x$(Se/Te) by manipulating composition or temperature parameters. The newly discovered topological nature makes it more meaningful to conduct the synthesis of one dimensional Pb$_{1-x}$Sn$_x$(Se/Te) nanostructures.

Here, we report the first synthesis of cylinder Pb$_{1-x}$Sn$_x$Se nanowires using vapor phase deposition method on silicon (Si) with Sn nanoparticles as the catalyst. Further, high quality rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowires with distinct \{100\} surfaces are obtained when changing Si substrate into mica. Significantly, two-dimensional Pb$_{1-x}$Sn$_x$Se nanowires with top and bottom \{100\} surfaces are formed due to

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the van der Waals epitaxial growth of Pb$_{1-x}$Sn$_x$Se nanowires along lateral direction on mica surface. Substrate surface electronic structures are found to be the critical factor that affects the vapor phase deposition process and final shapes of Pb$_{1-x}$Sn$_x$Se nanowires. Lastly we fabricate four terminal devices of Pb$_{1-x}$Sn$_x$Se nanowires. The electrical transport properties at ultralow temperature of rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowires grown on mica are shown to be dominated by topological surface transport.

Pb$_{1-x}$Sn$_x$Se nanowires were grown by vapor phase deposition method in a horizontal tube furnace as shown in Figure 1a. A mixture of Pb and SnSe powders was ground and then placed in the center of quartz tube as precursors. The source and substrate temperatures were set to 650 and 420 °C, respectively. Si with exposed surface of (100) and layered mica were used for the growth substrates. More experimental details are described in the Experimental Section. Figure 1b is the schematic diagram of cubic crystal structure of Pb$_{1-x}$Sn$_x$Se. Straight and smooth Pb$_{1-x}$Sn$_x$Se nanowires with diameter of ≈100–200 nm and a typical length of ≈50 µm are obtained on Si substrate (Figure 1c). Figure 1d shows a representative scanning electron microscopy (SEM) image of a Pb$_{1-x}$Sn$_x$Se nanowires at the top of which a nanorod is clearly seen. The diameter of the nanorod is a little larger than that of nanowire. Energy-dispersive X-ray spectroscopy (EDX) analysis in SEM (SEM-EDX) of several nanowires proves the as-grown Pb$_{1-x}$Sn$_x$Se nanowires are consisted of Pb, Sn and Se with Sn content (x) = 0.30. Inset in Figure 1d is the 25°-tilted-view SEM image of the Pb$_{1-x}$Sn$_x$Se nanowire cross-section, indicating the cylindrical shape of Pb$_{1-x}$Sn$_x$Se nanowire. Magnified SEM images of Pb$_{1-x}$Sn$_x$Se nanowire body and tip are displayed in Figure 1e and f, respectively. SEM-EDX mapping shown in Figure 1g–i demonstrates Pb, Sn and Se are uniformly distributed in the entire nanowire body without detectable phase separation. Quantitative analysis of SEM-EDX shows the elemental ratio of Pb, Sn and Se on nanowire body is about 0.68:0.32:1.00. The Pb/Sn/Se alloy nanorod on the top of nanowire also shows homogeneous distribution of Pb, Sn and Se. However, the tip of Pb/Sn/Se alloy nanorod displays more Sn as indicated by a white dashed rectangle in Figure 1k. Thus Sn nanoparticles are suggested to initiate the growth of Pb$_{1-x}$Sn$_x$Se nanowires in vapor-liquid-solid (VLS) deposition process. This can be further proved by the SEM-EDX mapping of another Pb$_{1-x}$Sn$_x$Se nanowire (Figure S2, Supporting Information) where obvious signature of Sn element are observed. The SEM-EDX quantitative analysis of Pb/Sn/Se alloy nanorod reveals the atomic ratio of Pb, Sn and Se is about 0.69:0.42:0.86 which slightly deviates from the stoichiometric ratio of Pb$_{1-x}$Sn$_x$Se.

We now turn our attention to the products on mica substrate. Substantial differences can be found on the morphologies of Pb$_{1-x}$Sn$_x$Se nanowires grown on mica and Si. The experimental parameters for the synthesis of Pb$_{1-x}$Sn$_x$Se nanowires on mica are kept exactly as that on Si. Figure 2a presents the SEM image of Pb$_{1-x}$Sn$_x$Se nanowires grown on mica surface. Different from Si surface where a rough buffer layer was formed before the growth of Pb$_{1-x}$Sn$_x$Se nanowires, Pb$_{1-x}$Sn$_x$Se nanowires were directly grown on mica substrate. The SEM image of a single nanowire in Figure 2b clearly
shows Pb$_{1-x}$Sn$_x$Se nanowires obtained on mica surface are of regular shape. The magnified SEM image of Pb$_{1-x}$Sn$_x$Se nanowire body (right bottom inset of Figure 2b) implies smooth surface and distinct facets. The following transmission electron microscopy (TEM) analysis points out the side facets are highly symmetric {100} equivalent surfaces. The side-view SEM image of another Pb$_{1-x}$Sn$_x$Se nanowire (left top inset of Figure 2b) demonstrates the cross-section of Pb$_{1-x}$Sn$_x$Se nanowire is in square shape. Statistics show shape of Pb$_{1-x}$Sn$_x$Se nanowire grown on mica surface is rectangular prismatic morphology with {100} surfaces. Rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowires synthesized on mica will favor the study of topological surface states on highly symmetric {100} facets. The width of rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowire distributes in $\approx 200$–$400$ nm with a typical length of $\approx 10$ µm. SEM-EDX of a nanowire body proves rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowires is composed of Pb, Sn and Se with atomic ratio of $\approx 0.52:0.48:1.00$. Elemental mapping of a nanowire body (Figure 2c–f) confirms uniform distribution of Pb, Sn and Se without clear segregation of either pure PbSe or SnSe, while signals from Sn elements can be clearly observed at the tip of the top nanoparticle (Figure 2h–j), indicating the fact that Sn plays the role of catalyst for inducing the growth of Pb$_{1-x}$Sn$_x$Se nanowire, which is consistent with the results in cylinder Pb$_{1-x}$Sn$_x$Se nanowires grown on Si substrate as described above. Interestingly, the top nanoparticle shows sharp edges and larger size than Sn catalyst and nanowire body (Figure 2g). We will discuss this in detail in the next growth mechanism part.

The TEM was applied to identify the fine crystal structure and chemical composition of Pb$_{1-x}$Sn$_x$Se nanowires on both Si and mica substrate. Figure 3a shows one cylinder Pb$_{1-x}$Sn$_x$Se nanowire synthesized on Si substrate. High-resolution TEM (HRTEM) obtained from edge of cylinder Pb$_{1-x}$Sn$_x$Se nanowire clearly illustrates the well-defined single crystalline Pb$_{1-x}$Sn$_x$Se nanowire with growth direction of $<100>$. Figure 3c is the TEM image of a rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowire obtained from mica substrate. Distinct lattice fringes shown in Figure 2d verifies the higher single crystalline of rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowire than nanowires on Si substrate. Perfect cubic symmetry of
SAED pattern as indicated by the inset in Figure 3d proves rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowire crystallizes along the direction of [100]. On the basis of the cubic SAED pattern and square cross-section (Figure 2b) of Pb$_{1-x}$Sn$_x$Se nanowire, the four perpendicular side facets of rectangular prismatic Pb$_{1-x}$Sn$_x$Se nanowire grown on mica surface can be identified as highly symmetric {100} surfaces. EDX in TEM (TEM-EDX) as shown in Figure 3e, which further confirms that Pb$_{1-x}$Sn$_x$Se nanowires grown on both Si and mica are constituted by Pb, Sn and Se. The peaks of C and Cu originate from copper grip. Quantitative TEM-EDX analysis suggests elemental ratios of Pb, Sn and Se are ≈0.69:0.31:1 and ≈0.51:0.49:1, respectively. The ratio of Pb to Se in Pb$_{1-x}$Sn$_x$Se nanowires grown on both Si and mica are ≈0.69:0.31 and ≈0.51:0.49:1, respectively. Raman scattering measurements (Figure 3f) further confirm the higher quality of Pb$_{1-x}$Sn$_x$Se nanowires grown on mica. Since Pb$_{1-x}$Sn$_x$Se and PbSe shares the same crystal structure, theoretical analysis of Raman scattering carried out for PbSe remain valid for Pb$_{1-x}$Sn$_x$Se alloy. As shown in Figure 3f, the dominant peak of ~140 cm$^{-1}$ should originate from the non-degenerate longitudinal optical (LO) branch. The weak peak located at ~250 cm$^{-1}$ is associated with two-phonon scattering (2LO). Huge differences of the two nanowires in the peak intensity at 140 cm$^{-1}$ could be ascribed to much lower structural quality of nanowires grown on Si substrates.

The evidently morphology differences of Pb$_{1-x}$Sn$_x$Se nanowires grown on Si and mica suggest the electronic structure of the substrate surface critically affects the vapor deposition growth of Pb$_{1-x}$Sn$_x$Se nanowires. Based on the above experimental data, the growth models of Pb$_{1-x}$Sn$_x$Se nanowires are proposed as shown in Figure 4. Figure 4a displays the initial growth stage of cylinder Pb$_{1-x}$Sn$_x$Se nanowires on Si substrate with growth duration of 10 mins. A buffer layer covered with a high density of nanoislands is observed on the Si surface. This phenomenon can be clearly elucidated by the schematic illustration of structure evolution process of cylinder Pb$_{1-x}$Sn$_x$Se nanowires shown in Figure 4b. The surface of three-dimensionally bonded Si has a large number of dangling bonds. The large lattice mismatch and strong electrostatic interaction between Si substrate and adatoms hinders the coherent heteroepitaxy growth. The lattice mismatch is estimated to be 12.3% between Pb$_{0.69}$Sn$_{0.31}$Se nanowires and (100) surface of Si substrate. In this case, a buffer layer is firstly formed to relax the strained energy in the interface between Si substrate and overlayer. Then Sn nanocatalyst reduced from SnSe compound is deposited on the surface of buffer layer. The growth model of layer-plus-island at the initial growth stage obviously results from strong vertical adatom-substrate attractive interaction and lateral adatom-adjacent attractive interaction as well. The formation of Sn catalyst can be reasonably correlated to its lower melting point (232 °C for Sn and 327 °C for Pb), which similarly occurred in the vapor deposition growth of PbSe nanowires by using...
other metals with low-melting-point such as In, Ga, and Bi.\[^{28}\] In the growth process, SnSe compound first decomposes into Sn atoms and Se atoms. Then Sn vapor condenses to form a nanoscale liquid droplet for Pb\(_{1-x}\)Sn\(_x\)Se nanowires nucleation and growth. And then Pb\(_{1-x}\)Sn\(_x\)Se nanowire is induced by continuously feeding the Pb, Sn and Se vapor with Sn nanocatalyst. However, Pb\(_{1-x}\)Sn\(_x\)Se nanowire is not strictly straight at the beginning of nanowire growth. One thing should be noted that, even a buffer layer is firstly formed on the surface of Si, the surface of buffer layer is not atomic-scale smooth. A pre-growth process is required to eliminate interface strain. Thus a short and curved nanorod is firstly assembled on the top of buffer layer. In addition, the rough surface of the buffer layer with dangling bonds increases the energy barrier for the growing of top Pb\(_{1-x}\)Sn\(_x\)Se nanorod along lateral direction on buffer layer surface. Hence, the growth of Pb\(_{1-x}\)Sn\(_x\)Se nanorod is confined into vertical direction, and its shape and size are defined by spherical Sn nanocatalyst, which results in cylindrical morphology.\[^{29}\] After that Pb\(_{1-x}\)Sn\(_x\)Se nanowire grows out from top Pb\(_{1-x}\)Sn\(_x\)Se nanorod (Figure 4c,d). A continuous growth of Pb\(_{1-x}\)Sn\(_x\)Se nanowires in vertical direction could be ascribed to the high growth active along <100> directions.

From the above discussion, we see the vapor deposition growth process is strongly influenced by the surface properties of Si and buffer layer. And the shape and size of Pb\(_{1-x}\)Sn\(_x\)Se nanowire are controlled by Sn catalyst. However, things will be different if we change the Si substrate into mica as shown in Figure 4e. At the initial growth stage with growth time of 10 min, Pb\(_{1-x}\)Sn\(_x\)Se nanoparticles are straightly assembled on the surface of mica without the formation of a buffer layer. This is because mica substrate used here are absent of surface dangling bonds. The chemically inert surface effectively circumvents the requirements of lattice matching between substrate and overlayer. In addition, as presented in Figure 2g, the top Pb\(_{1-x}\)Sn\(_x\)Se nanoparticles show regular shape with sharp edges. Due to the weak van der Waals interaction between adsorbed atoms and substrate\[^{30}\] energy barrier for the growing of Pb\(_{1-x}\)Sn\(_x\)Se nucleus along mica surface is dramatically reduced.\[^{31}\] Thus Pb\(_{1-x}\)Sn\(_x\)Se nucleus grows up according to their most active growth planes without restriction from Sn catalyst as shown in Figure 4f. The isotropic growth leads to a regular Pb\(_{1-x}\)Sn\(_x\)Se nanoparticle with sharp edges. After the growth of Pb\(_{1-x}\)Sn\(_x\)Se nanoparticle, Pb\(_{1-x}\)Sn\(_x\)Se finds it most active growth direction and grows out from top Pb\(_{1-x}\)Sn\(_x\)Se nanoparticle along <100> direction with distinct [100] equivalent surfaces (Figure 4g,i). The formation of dominated [100] equivalent facets of Pb\(_{1-x}\)Sn\(_x\)Se nanowire would be due to their lowest surface energy compared with other crystal planes. The rectangular prismatic Pb\(_{1-x}\)Sn\(_x\)Se nanowires surrounded by highly symmetric [100] surfaces will allow the study of topological states study on {100} surfaces. It is worth noting, for the growth on Si, even Sn content x and nanowires diameter increase to \(\approx 0.44\) and \(\approx 500\) nm respectively by decreasing the substrate temperature into \(\approx 380^\circ\)C, Pb\(_{1-x}\)Sn\(_x\)Se nanowires still exhibit cylindrical morphology (Figure S3, Supporting Information). While for the growth on mica, Pb\(_{1-x}\)Sn\(_x\)Se nanowires remain rectangular prismatic shape with Sn content \(\approx 0.50\) and nanowires width \(\approx 200-400\) nm although the substrate temperature decrease to \(\approx 380\) °C (Figure S4, Supporting Information), further confirming the importance of substrate surface electronic structure on the growth of Pb\(_{1-x}\)Sn\(_x\)Se nanowires. Interestingly, weak van der Waals interaction between mica substrate and adatoms enables the additional growth of Pb\(_{1-x}\)Sn\(_x\)Se nanowires along lateral direction. As shown in Figure 4h and Supporting Information Figure S5, 2D Pb\(_{1-x}\)Sn\(_x\)Se nanoparticles are formed due to the van der Waals epitaxy (vdWE) of Pb\(_{1-x}\)Sn\(_x\)Se nanowire along the lateral direction, which would be related to vapor-solid (VS) deposition process (Figure 4j). The enlarged top and bottom [100] surface of 2D Pb\(_{1-x}\)Sn\(_x\)Se nanoplate maximizes topological surface states on these surfaces.

Lastly we demonstrated that four-terminal devices of rectangular prismatic Pb\(_{1-x}\)Sn\(_x\)Se nanowires grown on mica exhibited topological surface electrical transport properties.
suggested their great potentials for the study of topological surface states. Pb$_1$-Sn$_x$Se nanowires grown on Si has also been executed electrical transport measurements for comparison study. In order to clearly describe the electrical transport behavior, devices fabricated based on Pb$_1$-Sn$_x$Se nanowires on Si and mica are denoted as device-1 and device-2 respectively. Details of device fabrication process are provided in the Supporting Information. SEM-EDX displays nanowires used in device-1 and device-2 are Pb$_{0.56}$Sn$_{0.44}$Se and Pb$_{0.59}$Sn$_{0.41}$Se, respectively. The resistance of device-1 increases with the decrease of temperature from 300 to ~225 K due to the decreasing intrinsic thermal excited carriers (Figure 5a). With further decrease of temperature from ~225 to ~150 K, resistance of the Pb$_{0.56}$Sn$_{0.44}$Se nanowire decreases, indicating the dominant electron-phonon interaction in this temperature range. And then resistance of the Pb$_{0.56}$Sn$_{0.44}$Se nanowire again increases with the decrease of temperature (~150 to ~44 K). Both the increasing band gap and decreasing thermal exciting carriers could reduce the conductivity of the Pb$_{0.56}$Sn$_{0.44}$Se nanowire. The carrier freeze-out effect may also cause this fast rising channel resistance. Interestingly, the channel resistance of Pb$_{0.56}$Sn$_{0.44}$Se nanowire slightly decreases at lower temperature (~44 to 2 K). Similar behavior also occurred in bulk Pb$_{0.56}$Sn$_{0.44}$Se where extrinsic-hole concentration approaches to a constant while electron concentration dramatically decreases. Electrical transport of Pb$_{0.56}$Sn$_{0.44}$Se nanowire at low temperature may also mainly be attributed to hole carriers. This implies Pb$_{0.56}$Sn$_{0.44}$Se nanowires are highly impure with hole doping. As for device-2 (Figure 5b), it initially shows a semiconductor-like behavior between 210 and 300 K. The channel resistance gradually increases with the decrease of temperature, which is likely due to the decreasing intrinsic thermal excited carriers with the decrease of temperature. And then channel resistance decreases when the temperature reduces from 210 to 100 K, indicating the metallic behavior and dominated electron-phonon scattering of Pb$_{0.56}$Sn$_{0.50}$Se nanowire in this range. After that device-2 again shows semiconductor-like behavior between 50 and 100 K. This could be explained by two synergistic mechanisms. The first one of these is the increasing band gap reduces the number of thermally excited carriers at low temperature. The second one is the decreasing thermal energy decreases the number of carriers as the temperature goes down. The dramatically increase of channel resistance at low temperature (<50 K) is ascribed to carrier freeze-out effect. When the temperature approaches to 2 k, the resistance hardly rises, which is likely due to the dominant topological surface transport. Here, thermal activation energy $E_a$ cannot be extracted simply by the relation $R = R_0 e^{E_a/kT}$ due to multiple-channel conduction (surface and bulk carriers), where $k_B$ is the Boltzmann constant.

In summary, we have synthesized highly single crystalline Pb$_1$-Sn$_x$Se nanowires using a simple vapor phase deposition method with the utilization of Sn as the catalyst. Two distinct morphologies, cylinder Pb$_1$-Sn$_x$Se nanowires and rectangular prismatic Pb$_1$-Sn$_x$Se nanowires with [100] exposed surfaces, are obtained by modulating the surface electrical properties of growth substrates. Significantly, 2D Pb$_1$-Sn$_x$Se nanoplates with dominant [100] surfaces are also obtained due to the vdW growth of Pb$_1$-Sn$_x$Se nanowires along lateral direction. Electrical transport properties indicate rectangular prismatic Pb$_1$-Sn$_x$Se nanowires are suitable for the further study of topological surface states. Additionally, our work may pave the way for the development of a general synthetic strategy that designs the growth of compound nanostructures by controlling substrate surface properties.

**Experimental Section**

**Synthesis of Pb$_1$-Sn$_x$Se Nanowires**: Pb$_1$-Sn$_x$Se nanowires were synthesized using horizontal vacuum tube furnace with single temperature zone. Pb and SnSe powder (99.99%, alfa aesar) were mixed, ground and loaded in the center of quartz tube. Si and mica substrates were placed in the downstream area. The quartz tube was evacuated and flushed few times with high purity Ar gas. During the experiment, Ar gas was fed with a constant flow rate 20 sccm by maintaining tube pressure of 65 pa. The furnace temperature was adjusted to 650 °C for the source and 420 °C for the substrate. The whole reaction process was maintained for 30 min. And then the furnace was allowed to cool naturally down to room temperature.

**Characterization**: Morphologies were characterized by field emission scanning electron microscopy (FESEM) S4800 (Tokyo, Japan) at 10 KV. Transmission electron microscopy (TEM) and high-resolution transmission electron microscope (HRTEM) were done on FEI Tecnai F20 at an accelerating voltage of 200 kV. Selected area electron diffraction (SAED) attached to the TEM were used to characterize the crystal structure. Electron energy dispersive X-ray spectroscopy (EDX) attached to the SEM and TEM were used to characterize the composition. Raman (Renishaw InVia, 532 nm excitation laser) was further used to characterize the quality of Pb$_1$-Sn$_x$Se nanowires.

**Device Fabrication**: Pb$_1$-Sn$_x$Se nanowires were mechanically separated in acetone by 30 s sonication and transferred onto a
cleaned Si substrate which has SiO₂ dielectric layer of 300 nm. Then FEI Nanolab 600i SEM/FIB dual beam system was used to fabricate the Pt-electrodes onto an individual PbₓSnₓSe nanowires.

Transport Measurements: Temperature-dependent channel resistance of PbₓSnₓSe nanowires was carried out in Quantum design PPMS-9 instrument. The resistance was measured using a standard four-terminal device to eliminate the contact resistance.

Supporting Information

Supporting information is available from the Wiley Online Library or from the author.

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