Strong Superconducting Proximity Effects in PbS Semiconductor Nanowires

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ABSTRACT: We report the fabrication of strongly coupled nanohybrid superconducting junctions using PbS semiconductor nanowires and Pb0.5In0.5 superconducting electrodes. The maximum supercurrent in the junction reaches up to ∼15 μA at 0.3 K, which is the highest value ever observed in semiconductor–nanowire-based superconducting junctions. The observation of microwave-induced constant voltage steps confirms the existence of genuine Josephson coupling through the nanowire. Monotonic suppression of the critical current under an external magnetic field is also in good agreement with the narrow junction model. The temperature-dependent stochastic distribution of the switching current exhibits a crossover from phase diffusion to a thermal activation process as the temperature decreases. These strongly coupled nanohybrid superconducting junctions would be advantageous to the development of gate-tunable superconducting quantum information devices.

KEYWORDS: PbS semiconductor nanowire, superconducting proximity effects, Josephson junction, supercurrent, switching current distribution

When a semiconductor nanowire (NW) is contacted with two superconducting electrodes, a supercurrent can flow through the non-superconducting nanowire because of the superconducting proximity effect. The NW-based superconducting junctions, or nanohybrid Josephson junctions, acquire an advantage of a gate-tunable supercurrent flow by controlling the carrier density in the NW, resulting in a supercurrent-based field-effect transistor. When the supercurrent is combined with phase-coherent quantum electronic transport in the NW, semiconductor NWs provide a useful platform to develop quantum electronic devices, such as gate-tunable superconducting quantum interference devices, Cooper pair splitters, and quantum electron pumps, and to explore Majorana Fermions and gate-tunable superconducting qubits. So far, most of the NW-based superconducting junctions have been made of Al superconducting electrodes with a very low transition temperature at TC ~ 1.2 K. The ultralow transition temperature and very small superconducting gap energy of Al, ΔAl ~ 0.15 meV, result in a relatively small critical current IC (≤0.1 μA), which is defined as the maximum supercurrent. Recently, a very short (L ~ 30 nm) channel device of InAs NW contacted with Al electrodes has exhibited a maximum supercurrent up to IC ∼ 0.8 μA. Because the Josephson coupling energy EJ = hIC/2e, where h is Planck constant divided by 2π and e is the elementary charge, must exceed thermal fluctuations for the observation of nonzero supercurrent, a large critical current would be essential for a wide variety of applications of the NW-based superconducting devices. Although other superconducting electrodes with higher transition temperatures, such as V (TC ∼ 5.0 K), Pb (TC ∼ 7.2 K), and Nb (TC ∼ 9.2 K), have been used to increase the critical current of NW-based superconducting junctions, the improvements were not significant in most cases, except for ref 22 (IC ∼ 6 μA).

In this work, we demonstrate very strong superconducting proximity effects between a PbS semiconductor NW and PbIn superconducting electrodes. The maximum supercurrent reaches up to IC ∼ 15 μA at T = 0.3 K, which is, to the best of our knowledge, the highest value for semiconductor-NW-based superconducting junctions. Moreover, the ICRC product, which is a figure of merit for a Josephson junction with a normal-state resistance RC, is larger than the superconducting gap energy of PbIn, resulting in εC/RC/ΔPbIn = 1.05, which is the largest value reported so far for the semiconductor-NW-based superconducting junctions. We also examined the superconducting proximity effects in the presence of the microwave and magnetic fields, which are consistent with the theoretical expectations. Furthermore, our measurements of the...
switching current distribution from the superconducting to resistive branches reveals that the phase diffusion and thermal activation processes are responsible for the stochastic switching behavior, which is dependent on temperature.

RESULTS AND DISCUSSION

The PbS NWs were synthesized using the chemical vapor deposition method in a tube furnace, while the doping of the NWs was modulated by varying the weight ratio of lead chloride and sulfur (see Methods). Figure 1a shows the

scanning electron microscopy (SEM) image of the as-grown PbS NWs. The details of NW growth17 and device fabrication6 have been reported elsewhere. A representative SEM image of the PbS NW device is displayed in Figure 1b, where two neighboring superconducting PbIn electrodes were used to apply the bias current (from I+ to I−) and to measure the voltage difference (between V+ and V−). The diameter w of the PbS NW and the distance L between two superconducting electrodes are found to be 130–200 nm and 180–220 nm, respectively, while the normal-state resistance RN of the device ranges from 20 to 130 Ω (see Table S1). The electrical transport properties of the NW devices were measured using a closed-cycle 4He refrigerator (Cryogenic, Ltd.) down to the base temperature of 0.3 K. For the low-noise measurements, two-stage RC filters (cutoff frequency ~30 kHz) and π filters were connected in series to the measurement leads.18

The PbIn superconducting electrode exhibits a superconducting transition below TC,SC = 7.0 K upon lowering the temperature, while the supercurrent can flow through the NW junction below TC,ff = 3.5 K for device D3, as shown in Figure 1c. The highest temperature for the observation of the supercurrent was found to be T = 5.2 K in PbS NW-based Josephson junctions.8 Figure 1d shows the current−voltage (I−V) characteristic curves of devices D1 and D2, displaying hysteresis depending on the sweep direction of the bias current. The switching from the superconductive to dissipative branches occurs at a critical current IC, while the reversed switching occurs at a return current IR. The hysteresis can be understood in terms of the presence of an effective capacitance19,20 in the junction or the quasiparticle heating effect.21 Here, we note that device D1 exhibits a maximum switching current IC ~ 15 μA, corresponding to a supercurrent density Jc ~ 1.1 × 106 A/cm2. To the best of our knowledge, these are the highest IC and Jc values recorded for the semiconductor-NW-based superconducting junctions,13–16,22 which is rather close to those observed for Au-NW-based junctions.20 For device D2, the εLcRN product is larger than ΔPbIn, as shown in Figure 1e for device D1. The overshoot of dV/dI/V peak positions

Table 1 shows a summary of the junction properties for a quantitative comparison.

<table>
<thead>
<tr>
<th>Sample</th>
<th>IC (μA)</th>
<th>JC (kA/cm²)</th>
<th>Ic,RN (mV)</th>
<th>dIR/R/Δ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb-based InAs NW15</td>
<td>0.61</td>
<td>31</td>
<td>0.35</td>
<td>0.26</td>
</tr>
<tr>
<td>Al-based InAs NW15</td>
<td>0.8</td>
<td>16</td>
<td>0.13</td>
<td>0.98</td>
</tr>
<tr>
<td>Nb-based InN NW22</td>
<td>5.7</td>
<td>50</td>
<td>0.44</td>
<td>0.31</td>
</tr>
<tr>
<td>this work (D1)</td>
<td>15</td>
<td>110</td>
<td>0.29</td>
<td>0.36</td>
</tr>
<tr>
<td>this work (D2)</td>
<td>7.3</td>
<td>55</td>
<td>0.84</td>
<td>1.05</td>
</tr>
</tbody>
</table>

The differential conductance curve as a function of voltage, dI/dV(V), is shown in Figure 1e for device D1 at T = 0.3 K. The overshoot of dI/dV near zero voltage is caused by the supercurrent branch, while the dI/dV peaks are attributed to the existence of multiple Andreev reflections23 (MARs) at the interfaces between the PbIn superconducting electrodes and the PbS semiconductor NW. When an electron coming from the semiconductor is incident upon the highly transparent superconductor, known as the Andreev reflection,1 when the Andreev reflections occur successively at two superconductor–semiconductor interfaces on the opposite side of the junction, the MARs result in conductance enhancements (or dI/dV peaks) occurring at Vc,* = 2Δ/ne, where n is an integer and Δ is the superconducting gap energy. It is evidently shown that the dI/dV peaks occur at Vc,* = 1.62 mV and Vc,* = 0.81 mV, indicating ΔPbIn = 0.81 meV. The temperature dependence of the dI/dV peak positions turns out to be consistent with the Bardeen–Cooper–Schrieffer (BCS) theory of superconductivity1 (see Figure S1). Similar features of the subgap structures can be found for different devices, implying the transparency at the interface to be Tc = 0.86, which is deduced from the excess current (see Figure S2).
obtained from previous superconducting junctions based on InAs semiconductor NWs.\textsuperscript{2,13,14} The large superconducting gap energy of Δ_{PbIn} and formation of highly transparent contact between the PbIn and PbS NW are responsible for the very strong Josephson coupling observed in our experiment. An application of the gate voltage V_G can tune the values of I_{C} and R_N as shown in Figure 1f. Here, the I_{C}(V_G) and R_G(V_G) data were taken at T = 2.5 K and T = 10.0 K, respectively. The increase in R_N with decreasing V_G indicates that the PbS NW has a strong n-type character in the experimental range. The electron density and mobility are obtained to be n = 1.04 × 10^{19} cm^{-3} and \mu = 690 cm^{2}/Vs, respectively. It is also noted that I_{C} decreases with decreasing V_G, which is contrary to R_N.

The progressive change in the I−V curves with increasing temperature is displayed in Figure 2a. It is shown that I_{C} decreases monotonically and the hysteresis becomes reduced at higher temperatures than T = 1.2 K. The temperature dependences of I_{C} and I_R are depicted in Figure 2b, together with the fitting result for the I_{C}(T) curve. We fitted I_{C}(T) to the theoretical expression\textsuperscript{24} cl_{c}R_N = \varepsilon E_{T} \frac{E_{Th}}{3.2f_{Th}} \frac{1}{(1 - b \exp(-aE_{Th}/3.2f_{Th}))}, where E_{Th} = \hbar D/L^2 is the Thouless energy, and a and b are fitting parameters. The value of E_{Th} is estimated to be 0.19 meV for the channel length L = 190 nm and diffusion coefficient\textsuperscript{2} D = 103 cm^{2}/s. Thus, the best fit (the solid-line curve in Figure 2b) is obtained with a = 4.6 and b = 3, which are comparable to the parameters in the long and diffusive junction model (see text).

Figure 2. (a) Temperature dependence of the I−V characteristic curves for device D2. Plots are offset for clarity. (b) Temperature dependence of I_{C} (circles) and I_R (squares). The solid line is a theoretical calculation of I_{C} using the long and diffusive junction model (see text).

Figure 3. (a) Microwave-power dependence of the I−V characteristics under external microwave at frequency f_{mw} = 10.4 GHz. The measurement temperature was T = 1.5 K. Voltage plateaus (or Shapiro steps) are clearly observed at V = V_{j}, with n = 1, 2, 3. (b) I−V characteristics at f_{mw} = 4.4, 7.3, 10.4, and 15.5 GHz. Here, the voltage axis was normalized by hf_{mw}/2e. The inset shows the frequency dependences of the voltage interval, ΔV, data (squares) and the theoretical prediction (solid line) based on the ac Josephson relation. (c) Current width ΔI_{n} of the nth Shapiro step (n = 0, 1) with respect to the square root of the microwave power (P^{1/2}). The solid lines correspond to the theoretical calculations (see text).

Additional evidence for the Josephson coupling in the PbS NWs can be found in the dependence of I_{C} on the magnetic field B perpendicular to the substrate. Figure 4a shows the color plot of the differential resistance dV/dI as a function of B and I, where the supercurrent region is denoted by the dark blue color. We note that I_{C} decreases monotonically with increasing B and vanishes at B ~ 0.16 T. The I_{C}(B) data (symbols) in Figure 4b are quite different from the conventional expectation of a periodic modulation of I_{C} with B, i.e., the Fraunhofer diffraction pattern,\textsuperscript{3} where the I_{C} minima occur at the integer magnetic flux quanta in the junction area. Our observations can be explained using the narrow junction model,\textsuperscript{25} where the applied B field acts as a pair breaker for the Cooper pairs induced in the normal conductor with a junction width much smaller than or comparable to the magnetic length l_{B} = (\Phi_0/\Phi_{0}/B_0)^{1/2}, where \Phi_0 = h/2e is the magnetic-flux quantum and B_0 = B_0/Lw (see the Supporting Information). Here, we obtained l_{B} = 157 nm for device D1 with w = 130 nm, satisfying a narrow-junction condition of w < l_{B}. Similar features have also been observed in other narrow superconducting junctions.\textsuperscript{15,16,22,26} Additionally, the I_{C} hump near zero field is attributed to the magnetic field focusing effect.\textsuperscript{15}

Another peculiar feature of the I_{C}(B) data is shown in Figure 4c for device D3 with w = 200 nm and l_{B} = 190 nm, resulting in w < l_{B}. We note that there exist two different types of I_{C}(B) patterns, both indicated by dV/dI peak structures. The inner structure enclosing the dark blue region resembles a
Fraunhofer-type modulation of $I_C$ with $B$, which is commonly observed in a wide Josephson junction.\textsuperscript{18} It follows the Fraunhofer relation $I_C(B) = I_C(0)\sin[\pi(\Phi_0/\Phi)]/(\Phi/\Phi_0)$, where $\Phi$ is the magnetic flux through the superconducting junction. Then, the first minimum of $I_C$ is expected to be located at the magnetic field $B_1 = \Phi_0/[(L + 2\lambda)w]$, where $\lambda$ is the London penetration depth of the superconducting electrodes.\textsuperscript{18} Because $B_1$ is found to be 19 mT, we obtain $\lambda = 180$ nm for the PbIn electrodes, which is comparable to the value obtained in our previous study.\textsuperscript{6} The abrupt switching of $I_C(B)$ above $B_1$ is attributed to the penetration of magnetic vortices into the PbIn electrodes. The outer $dV/dI$ peak structure, occurring at $I_C$, reveals a monotonic suppression of $I_C$ by $B$, which is consistent with the narrow junction model with $r = 0.53$ (see the Supporting Information). Thus, we conclude that the monotonic suppression and periodic modulation of $I_C$ with $B$ can occur simultaneously in intermediate-width ($w \gtrsim \xi_B$) junctions.

As displayed in Figure 5a, the repetition of the current sweep reveals that there exists a stochastic distribution of $I_C$ switching from the superconducting to the resistive branches (see the Methods for measurement details). It is well-known that the $I_C$ distribution is closely related to the dynamics of the Josephson phase particle in the superconducting junction,\textsuperscript{1} where the $I_C$ switching event corresponds to the escape of the phase particle from the local minima of the washboard potential $U(\phi) = -E_{\text{ref}}\cos(\phi) + (1/I_C\omega)\phi$, where $\phi$ is the phase difference across the junction and $I_C\omega$ and $E_{\text{ref}} = \hbar I_C/2e$ are the fluctuation-free $I_C$ and Josephson coupling energy,\textsuperscript{1} respectively (see Figure 5b), with assuming a sinusoidal current–phase relation. Then, the escape is governed by the thermal activation\textsuperscript{27} (TA), phase diffusion\textsuperscript{26,29} (PD), and macroscopic quantum tunneling\textsuperscript{30} (MQT) processes, depending on relative strength of $E_{\text{ref}}$ over thermal fluctuations.

Figure 5c shows the $I_C$ distribution data measured at various temperatures, indicating that the sharp $I_C$ distribution obtained at high temperature ($T = 1.2$ K) becomes remarkably broadened at lower temperatures. As a result, the standard deviation of the $I_C$ distribution increases monotonically with decreasing temperature down to $T = 0.46$ K, implying that the PD process is the dominant switching current mechanism in this experiment,\textsuperscript{31} where thermally activated phase particles are repeatedly retrapped in the neighboring potential minima because of a strong dissipation during the escape. The fitting lines of the switching probability $P(I_C)$ in the PD model are consistent with the observed $I_C$ distribution data, as shown in Figure 5c. The switching probability is related to the escape rate $\Gamma$ by $P(I_C) = [\Gamma(I_C)/(dI/d\Gamma)] \{1 - \int_{0}^{\Gamma} P(\Gamma') d\Gamma'\}$.\textsuperscript{27} Here, $dI/d\Gamma$ is the sweep rate of the applied current. Figure 5d reveals that the behavior of the current dependence of $\Gamma$ changes below $T = 0.33$ K, indicating that there occurs a crossover from the PD to the TA regimes as the temperature decreases. The MQT behavior, however, was not observed in our experimental range, in contrast to graphene-based superconducting junctions.\textsuperscript{31,32} It may be caused by an enhanced electron temperature caused by an incomplete filtering of the high-frequency noise.

**CONCLUSIONS**

In conclusion, we fabricated and characterized the electronic-transport properties of nanohybrid superconducting junctions made of PbS NWs contacted with PbIn superconducting electrodes. We observed the highest values of the critical current and $I_C R_N$ product ever reported in semiconductor-NW-based superconducting junctions. Very strong superconducting proximity effects in PbS NWs are attributed to the formation of...
highly transparent semiconductor—superconductor contacts with PbIn electrodes, which yield the relatively large superconducting gap energy. Both the microwave and magnetic field dependences of the $I$–$V$ characteristics of the PbS–NW superconducting junctions are in good agreement with theoretical expectations. Measurement and analysis of the stochastic distribution of the switching current reveals the underlying mechanism of escape dynamics of the phase particle in the NW-based superconducting junctions.

METHODS

Synthesis. The PbS nanowires (NWs) were synthesized using the chemical vapor deposition method in a horizontal tube furnace (Lindberg Blue M). Lead chloride (PbCl₂, 99.999%, Alfa Aesar) and Sulfur (S, 99.9999%, Alfa Aesar) powders were placed in the center and outside the heating zone of the furnace, respectively. The growth substrate was prepared using electron-beam evaporation of a 100 nm Ti thin film onto a SiO₂-coated Si wafer. The substrate was placed 5 cm downstream from the center of the heating zone ($T ∼ 550 °C$). The synthesis system was first evacuated to a base pressure of 15 mTorr and then flushed with $N₂$ (99.999%) three times before being filled to atmosphere pressure. The furnace temperature was quickly ramped to 630 °C at 60 °C/min, while 150 sccm $N₂$ flow was maintained. At the peak temperature, the quartz boat containing $S$ was transferred to the heating zone to trigger the growth. The growth duration varied from 30 min to 2 h. After the growth, the furnace was naturally cooled to room temperature over approximately 3 h. The $N₂$ flow was kept during the entire cooling process to remove any $S$ residue. The doping of the PbS NWs was modulated by varying the weight ratio of PbCl₂ and $S$. Additional details of the synthesis, characterization, and doping level control can be found in a previous publication.¹⁷

Device Fabrication and Transport Measurements. For the fabrication of the PbS-NW-based superconducting junctions, the as-grown PbS NWs were mechanically transferred by using a tungsten tip, controlled by three-axis micrometer stages, onto a highly doped $n$-type silicon substrate served as a back-gate electrode. Figure 1b shows the scanning electron microscope (SEM) image of a typical PbS-NW-based Josephson junction with a channel length $L = 190$ nm and the four-probe measurement configuration. For the measurement, we used Keithley 2400 as a current source and Keithley 2182A to measure the voltage difference. We also measured the transport properties of 14 devices in three different refrigerator systems: ²He refrigerator (cryogenic) with a base temperature of $T = 0.3$ K, a 1.5 K cryostat using microwave response, and a 2.5 K cryostat. In particular, the switching current was observed in devices with channel lengths that were less than 250 nm. The detailed parameters of the devices are shown in Table S1. To obtain the switching current distribution, a triangle-wave-shape bias current was applied to the sample with a ramping rate $dI/dt = 240 \, \mu A \, s^{-1}$, while 5000 $I₂$ data were recorded at a threshold voltage $V_{th} = 30 \, \mu V$ and a fixed temperature.

REFERENCES


