Gate-tunable superconducting quantum interference devices of PbS nanowires

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We report on the fabrication and electrical transport properties of gate-tunable superconducting quantum interference devices (SQUIDs), made from a semiconducting PbS nanowire contacted with PbIn superconducting electrodes. With a magnetic field applied perpendicular to the plane of the nanohybrid SQUID, periodic oscillations of the critical current due to the flux quantization in SQUID are observed up to $T = 4.0$ K. A nonsinusoidal current–phase relationship is obtained as a function of temperature and gate voltage, which is consistent with a short and diffusive junction model. © 2016 The Japan Society of Applied Physics

An nanohybrid superconducting junctions,1,2 made from a single-crystalline nanostructure contacted with conventional superconductors, provide a useful tool for exploring the combined effects of quantum electrical transport in the nanostructure and superconducting phase coherence. Supercurrent transistors, in which the superconducting coupling through the nanostructure is gate-tunable, have been developed using semiconductor nanowires (NWs),1,3 carbon nanotubes,4,5 and graphene.5,6 Controlled Cooper pair splitters for nonlocal entanglement have also been realized using the nanostructures.6,7 Furthermore, recent studies on the gate-tunable macroscopic quantum tunneling8,9 and Majorana bound states10,11 in the nanohybrid superconducting junctions would pave the way for developing nanohybrid superconducting qubits.12,13

A superconducting quantum interference device (SQUID),14 formed with two superconducting junctions connected in parallel, is used as a very sensitive magnetometer and a key building block for a superconducting flux qubit15 as well. So far, nanohybrid SQUIDs have been made from various nanostructures such as NWs,16 carbon nanotubes,17 and graphene.18 Very low operation temperatures below $T = 2.5$ K,19 however, hinder their wide applications in superconducting electronics and quantum information devices. In addition, the gate-tunable current–phase relation (CPR) has not yet been studied in the NW-based superconducting junctions.20 CPR in the superconducting weak links is expected to be nonsinusoidal,21 while tunneling-type superconducting junctions exhibit a sinusoidal one, $I_c = I_c \sin \phi$, where $I_c$ is the supercurrent, $I_c$ is the critical current, and $\phi$ is the phase difference between two superconducting electrodes.22 Since the CPR determines the shape of an anharmonic potential well for the Josephson phase particle,23 the CPR in the NW-based superconducting junctions would be important for developing gate-tunable superconducting qubits.

In this Letter, we report on the gate-tunable operation of nanohybrid SQUIDs, made from a PbS NW and a PbIn superconductor. Employing PbIn alloy as superconducting electrodes,24,25 enables us to achieve a higher operation temperature of the NW SQUIDs above the liquid-helium temperature, which is the highest reported to date. Modulation of $I_c$ as a function of magnetic flux through the SQUID loop was achieved by varying the temperature and gate voltage, resulting in a nonsinusoidal CPR at lower temperatures. Our observations are consistent with a short and diffusive junction model and suggest that the skewness of the CPR can be controlled by the application of gate voltage.

PbS NWs were synthesized via chemical vapor deposition in a tube furnace, as described elsewhere26 (see also the online supplementary data at http://stacks.iop.org/APEX/9/023102/mmedia). NW-based SQUIDs with different loop areas were fabricated by electron-beam lithography using PbS NWs (see Fig. S1 in the online supplementary data at http://stacks.iop.org/APEX/9/023102/mmedia), as the details are explained in Supplementary Data. Figure 1(a) shows a scanning electron microscopy (SEM) image of a typical NW SQUID, which consists of two superconducting junctions along the NW and a supercurrent loop. Electrical transport properties of the NW SQUID are measured by a four-point measurement in a closed-cycle helium cryostat and a $^3$He refrigerator (Cryogenic) down to the base temperatures of 2.6 and 0.3 K, respectively.

The PbIn electrodes become fully superconducting below $T_s = 6.7$ K (see Fig. S1 in the online supplementary data at http://stacks.iop.org/APEX/9/023102/mmedia), and the supercurrent through the PbS NW SQUID is observable up to $T = 5.2$ K, as shown in the inset of Fig. 1(b). The maximum operation temperature in this work is two times higher than that of InAs NW SQUID,19 which is attributed to a very strong Josephson coupling between the PbS NW and the superconducting PbIn electrodes.27 When the magnetic field is applied perpendicular to the NW SQUID loop, gradual changes in the current–voltage (I–V) characteristic curves are observed with increasing magnetic flux $\Phi$, as shown in Fig. 1(b). It is clearly shown that $I_c$ is maximum at $\Phi = 0$ and absent at $\Phi = h/2e$, where $h = h/2e$ is the magnetic flux quantum, $h$ is Planck’s constant, and $e$ is the elementary charge. For $\Phi > h/2e$, $I_c$ increases to reach its maximum value at $\Phi = \Phi_0$ and then exhibits an oscillatory behavior with a period of $\Phi_0$ as a signature of the SQUID. The periodic modulation of $I_c$ is displayed in a color plot of differential resistance, $dV/dI$, in Fig. 1(c). Here, we used the effective area of the SQUID loop to be $A = 3.39 \mu m^2$ [the yellow dashed line in Fig. 1(a)], which is calculated from the magnetic field periodicity of $H_0 = 6.1$ Oe. The difference between the effective area and the geometrical inner area of the loop is due to the London penetration depth $\lambda$ of PbIn electrodes,24 which is estimated to be $\lambda = 0.43 \mu m$ for sample D1 and $0.48 \mu m$ for sample D2.

The periodic modulation of $I_c(\Phi)$ can be explained by the sinusoidal CPR in the NW Josephson junction and flux quantization in the SQUID loop.22 Under the assumption of negligible self-inductance of the loop, $I_c$ is given by...
where $I_{c1}$ and $I_{c2}$ are the critical currents of the weak links.\(^{19}\)

At $T = 2.8$ K, $I_{c1} = 298$ nA and $I_{c2} = 295$ nA are obtained by fitting Eq. (1) to experimental data [see the white line in Fig. 1(c)]. It is inferred that two weak links in the NW SQUID are almost identical with $I_{c2}/I_{c1} = 0.99$. The self-inductance is calculated to be $L_S \approx 3$ pH from the SQUID geometry, corresponding to the screening parameter $eta_S = 2\pi L_S I_0/\Phi_0 = 3 \times 10^{-3} \ll 1$. Here, $I_0 = (I_{c1} + I_{c2})/2$ is the average critical current. We note that the periodic $I_c(\Phi)$ relation is maintained up to $T = 4.0$ K, as shown in Fig. 1(d), which confirms the sinusoidal CPR in the NW weak links at higher temperatures.

The NW SQUID, as a flux-to-voltage transducer, can provide an output voltage related to $\Phi$. When biased with constant $I$, the periodic modulation of $V(\Phi)$ is as shown in Fig. 2(a). The sensitivity of the SQUID becomes maximum, $|\partial V/\partial \Phi| \sim 39 \mu V/\Phi_0$, near $I_c$ [see Fig. 2(b)], which is similar to the previous result in the InAs NW SQUID.\(^{19}\) Another plot of differential resistance, $dV/dI$, with $\Phi$ exhibits a similar behavior at a low bias current. At a high bias current above $I = 0.8 \mu A$, however, $\pi$-phase-shifted oscillations occur, as shown in Fig. 2(c). This $\pi$-junction behavior in the weak link has been attributed to the existence of a ferromagnetic layer,\(^{28}\) a nonequilibrium electron distribution in the normal region,\(^{29}\) a quasiparticle-pair interference effect,\(^{30}\) or a quantum dot\(^{17}\) between two superconducting electrodes.

The numerical differentiation of the $I$–$V$ curves, however, reveals that the $\pi$-phase shift observed in this work is a direct result of the flux-dependent $dV/dI$ vs $I$ curves, as shown in Fig. 2(d). At a near-zero bias current, $dV/dI$ becomes maximal at half-integral flux quantum and minimal at the integral one. At a higher bias current above $I = 0.8 \mu A$, the opposite behavior is obtained to induce the $\pi$-phase shift. In the intermediate bias region, $h/4e$ oscillations instead of $h/2e$ ones are observed. A metallic dc SQUID made from Al–Au–Al junctions exhibits a similar phenomenon.\(^{31}\)

The color plot of $dV/dI$, obtained from device D2 at a much lower $T$, is shown in Figs. 3(a)–3(d). It is noted that there are several differences between Figs. 3(a)–3(d) and Fig. 1(c). Firstly, the supercurrent “off” state ($I_c = 0$) is not seen in D2.

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**Fig. 1.** (a) False-colored SEM image of a typical NW SQUID with PbS NW (yellow) and PbIn superconducting electrodes (blue). The yellow dashed line indicates an effective area of the supercurrent loop. Bias current flows from I+ to I−, while voltage is measured between V+ and V−. (b) Current–voltage curves at $T = 2.8$ K with different magnetic fluxes through the NW SQUID loop (sample D1). Inset: temperature dependence of the critical current. The line is a guide to the eye. (c) Color plot of differential resistance, $dV/dI$, as a function of magnetic flux and dc bias current. The black-colored region is the supercurrent regime and the white line indicates a sinusoidal current-phase relationship described by Eq. (1). (d) Temperature dependence of $I_c(\Phi)$ relation. The line is a fit using Eq. (1). The measurements were carried out with the gate voltage $V_g$ of $0$ V.

**Fig. 2.** (a) Modulation of output voltage as a function of $\Phi$ at $T = 2.8$ K and $V_g = 0$ V. The bias current $I$ increases from 0.1 to 1.0 $\mu A$ in 0.1 $\mu A$ steps from bottom to top. (b) Flux-to-voltage transfer function with different $I$ values. Error bars are indicated. (c) Flux-dependent $dV/dI$ measurement with $I = 0, 0.4, 0.5, 0.6, 0.7, 0.8, \text{and } 1.0 \mu A$ from bottom to top. The lock-in bias current is $I_c = 200$ nA. (d) $dV/dI$ vs $I$ curves with different $\Phi$ values, which were obtained from the numerical differentiation of $I$–$V$ curves in Fig. 1(b).
but the periodic modulation of $I_c(\Phi)$ between the maximum ($I_{c,\text{max}}$) and minimum ($I_{c,\text{min}}$) values of $I_c$. Since the screening effect is negligible ($\beta_l \ll 1$), the absence of the $I_c$-off state can be caused by the asymmetric weak links,\textsuperscript{14)} where $I_{c1}$ and $I_{c2}$ are estimated as $I_{c1} = (I_{c,\text{max}} + I_{c,\text{min}})/2$ and $I_{c2} = (I_{c,\text{max}} - I_{c,\text{min}})/2$. This results in $I_{c1} = 510 \text{nA}$, $I_{c2} = 150 \text{nA}$, and $I_{c2}/I_{c1} = 0.29$ for D2 at $T = 0.3 \text{ K}$. The asymmetric $I_c$’s are also responsible for the shift of $I_c(\Phi)$ curves in opposite polarity.\textsuperscript{14)} Secondly, the skewed $I_c(\Phi)$ curves are obtained instead of the sinusoidal ones. The skewness can be defined by $(2\varphi_{\text{max}}/\pi - 1)$, where $\varphi_{\text{max}}$ is the position of $I_{c,\text{max}}$.\textsuperscript{32)} Figure 3(e) shows that the skewness decreases monotonically with temperature, evolving into a sinusoidal $I_c(\Phi)$ curve at a higher temperature, as observed in D1. The skewness can be caused by the asymmetric $I_c$’s in two weak links in the SQUID. The ratio $I_{c2}/I_{c1}$, however, decreases with temperature to enhance the $I_c$ asymmetry, resulting in $I_{c2}/I_{c1} = 0.17$ at $T = 2.7 \text{ K}$ (see Fig. S2 in the online supplementary data at http://stacks.iop.org/APEX/9/023102/mmedia), which is contrary to the temperature dependence of the skewness.

A more plausible explanation can be found in the CPR of the NW-based superconducting weak link. It is well known that the CPR in the superconducting weak links is nonsinusoidal at low temperatures and transforms into a sinusoidal one near $T_c$.\textsuperscript{33)} For a short and diffusive weak link, the CPR is given by

$$I_c(\varphi) = \frac{4\pi k_B T}{e R_N} \sum_{\omega > 0} \frac{\Delta \cos(\varphi/2)}{\delta} \arctan \frac{\Delta \sin(\varphi/2)}{\delta},$$

where $R_N$ is the normal-state resistance of the junction, $\delta = \sqrt{\Delta^2 \cos^2 \varphi + (\hbar \omega)^2}$, $\hbar \omega = \pi k_B T(2n + 1)$ is the Matsubara energy, $\Delta$ is the superconducting energy gap, $\varphi$ is the phase difference between two superconducting electrodes, and $n$ is an integer.\textsuperscript{34)} After applying Eq. (2) for $I_{c1}$ and $I_{c2}$ in combination with flux quantization in the SQUID loop, the calculation results are obtained and depicted in Fig. 3(f) as solid lines, which are in good agreement with the $I_c(\Phi)$ data. $\Delta(T = 0) = 1.04 \text{ meV}$ was used as a fitting parameter, which is consistent with the experimental value (see Fig. S3 in the online supplementary data at http://stacks.iop.org/APEX/9/023102/mmedia). Since the elastic mean free path and the Thouless energy of the PbS NW are obtained to be $l_e = 26 \text{ nm}$ and $E_{\text{Th}} = \hbar D/L^2 = 112 \text{ µeV}$, respectively, where $D = 103 \text{ cm}^2/\text{s}$ is the diffusion coefficient and $L = 250 \text{ nm}$ is the length of the superconducting weak link for D1, the PbS NW weak link is in short ($E_{\text{Th}}/\Delta \sim 0.11$) and diffusive ($l_e \ll L$) junction regimes.

Figures 4(a)–4(d) show the color plots of $dV/dI$ as a function of magnetic flux and bias current at different gate voltages, $V_g$. Note that the $V_g$-dependent change in $I_c(\Phi)$ curves is quite similar to the temperature-dependent one shown...
in Figs. 3(a)–3(d). As the gate voltage decreases, $I_c$ also decreases and the skewness becomes smaller, as shown in Fig. 4(e). We used Eq. (2) for fitting the short and diffusive junction model to the $V_F$-dependent $I_c(\Phi)$ data, where the effective temperature $T_{\text{eff}}$ was used as a fitting parameter. The fitting results [solid lines in Fig. 4(f)] are in good agreement with the $I_c(\Phi)$ data (symbols), while yielding $T_{\text{eff}}$ in a reasonable range [see Fig. 4(e)].

The application of negative $V_F$, $I_c$ is suppressed and the distorted CPR becomes sinusoidal. These features are quite similar to the effects seen at increased temperature, as shown in Fig. 3, and thus $T_{\text{eff}}$ increases at a negative $V_F$. To the best of our knowledge, this work is the first to demonstrate the gate-voltage dependence of the CPR in the NW-based SQUIDs. Since the CPR determines the shape of an anharmonic potential well for the Josephson phase particle, our observed gate-tunable CPR would be important for developing nanohybrid superconducting qubits.

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1. Y.-J. Doh, J. A. van Dam, A. L. Roest, E. P. A. M. Bakkers, L. P. Kouwenhoven, and S. De Franceschi, Science 309, 272 (2005).
2. S. De Franceschi, L. P. Kouwenhoven, C. Schönenberger, and W. Wernsdorfer, Nat. Nanotechnol. 5, 703 (2010).
3. J. Xiang, A. Vidan, M. Tinkham, R. M. Westervelt, and C. M. Lieber, Nat. Nanotechnol. 4, 208 (2009).
4. J. P. Cleuziou, W. Wernsdorfer, V. Bouchiat, T. Onarcuha, and M. Monthioux, Nat. Nanotechnol. 1, 53 (2006).
5. C. Girit, V. Bouchiat, O. Naaman, Y. Zhang, M. F. Crommie, A. Zettl, and I. Siddiqi, Nano Lett. 9, 198 (2009).
6. P. Spathis, S. Biswas, S. Roddaro, F. Giazotto, and F. Beltram, Nanotechnology 22, 105201 (2011).
7. G.-H. Lee, S. Kim, S.-H. Jhi, and H.-J. Lee, Nat. Commun. 6, 6181 (2015).
8. A. A. Golubov, M. Y. Kupriyanov, and E. Il’ichev, Rev. Mod. Phys. 76, 411 (2004).
9. M. Tinkham, Introduction to Superconductivity. (McGraw-Hill, New York, 1996).
10. J. A. van Dam, Y. V. Nazarov, E. P. A. M. Bakkers, S. De Franceschi, and L. P. Kouwenhoven, Nature 442, 667 (2006).
11. B.-K. Kim, H.-S. Kim, Y. Yang, X. Peng, M.-H. Bae, D. Yu, and Y.-J. Doh, in preparation.
12. V. V. Ryazanov, A. V. Oboznov, A. Y. Rusanov, A. V. Veretennikov, A. A. Golubov, and J. Aarts, Phys. Rev. Lett. 86, 2427 (2001).
13. J. J. A. Baselmans, A. F. Morpurgo, B. J. van Wees, and T. M. Klapwijk, Nature 397, 43 (1999).
14. J. Kim, Y. J. Doh, and H.-J. Lee, Phys. Rev. B 52, 15095 (1995).
15. J. Wei, P. Cadden-Zimansky, and V. Chandrasekhar, Appl. Phys. Lett. 92, 102502 (2008).
16. C. Chialvo, I. C. Moraru, D. J. V. Harlingen, and N. Mason, arXiv:1005.2630.
17. K. K. Likharev, Rev. Mod. Phys. 51, 101 (1979).