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Planar graphene Josephson coupling via van der Waals superconducting contacts

Jongyun Lee^a, Minsoo Kim^{a,*}, Kenji Watanabe^b, Takashi Taniguchi^b, Gil-Ho Lee^a, and Hu-Jong Lee^a

^a Department of physics, Pohang University of Science and Technology, Pohang 37673, Korea

^b National Institute for Materials Science, Tsukuba 305-0044, Japan

KEYWORDS: graphene, proximity Josephson junction, Van der Waals materials, Van der Waals stacking, dry transfer

Abstract

We report on the fabrication and transport characteristics of van der Waals (vdW)-contacted planar Josephson junctions. In a device, two pieces of cleaved 2H-NbSe₂ superconducting flakes and a monolayer graphene sheet serve as the superconducting electrodes and the normal conducting spacer, respectively. A stack of NbSe₂-graphene-hexagonal boron nitride (hBN) heterostructure with clean and flat interfaces was prepared by a dry transfer technique. The outermost hBN layer protected the NbSe₂-graphene-NbSe₂ Josephson junction from chemical contamination during the fabrication processes. The Josephson coupling was confirmed by a periodic modulation of the junction critical current I_c in a perpendicular magnetic field. The temperature dependence of I_c showed long and diffusive Josephson coupling characteristics. The temperature dependence of the superconducting gap, obtained from the multiple Andreev reflection features, followed the Bardeen-Cooper-Schrieffer (BCS) prediction.

* Present address: School of Physics & Astronomy, University of Manchester, Manchester M13 9PL, United Kingdom

† E-mail: hjlee@postech.ac.kr

1. Introduction

Since the realization of graphene layers, a variety of two-dimensional (2D) materials have been produced by the mechanical exfoliation technique [1], including metals [2, 3], insulators [4, 5], semiconductors [6], topological insulators [7, 8], superconductors [1, 9-11], etc. Fundamental properties of various cleavable 2D materials including graphene have been investigated extensively to date, together with exploring their electronic applications [12, 13]. Recently developed dry transfer technique has allowed to stack such cleaved 2D materials into van der Waals (vdW) heterostructures with atomically flat and clean interfaces [14, 15], which often reveal novel physical properties [16, 17]. Proximity Josephson coupling is a good example: when a normal conductor (N) is sandwiched between two closely spaced superconductors (S), a supercurrent can flow through the N layer without dissipation, forming an SNS proximity Josephson junction.

Such a SNS junction requires electrically transparent interfaces between the S and N layers. However, conventional ways of depositing superconducting materials with electron-beam/thermal evaporation or plasma sputtering, with highly energetic evaporants, may seriously damage the normal conducting insert and the S/N interfaces. Such deterioration can be minimized by dry transferring superconducting materials onto the normal conducting insert as demonstrated in previous studies [18, 19]. Recently realization of vertical Josephson junctions has been demonstrated by vdW stacking of two dry transferred superconducting layers onto each other. Here, the angle mismatch of the two stacked 2H-NbSe₂ flakes leads to formation of a weak-link with tunneling-like junction behavior [20]. On the other hand, vertically stacked NbSe₂-graphene-NbSe₂ vdW junctions has shown strong proximity Josephson coupling [21]. NbSe₂ is one of cleavable superconducting materials, maintaining superconductivity down to a few atomic layers [9-11, 22]. Other form of vertical Josephson junctions were fabricated by electron beam deposition of aluminum superconducting electrodes on both sides of graphene [23]. Although the short and ballistic strong proximity Josephson coupling was demonstrated in the vertical junctions, characteristics of the graphene layer were not gate tunable due to screening of the gating field by the superconducting electrodes.

In this study, we report on the fabrication and the measurements of vdW-contacted planar proximity Josephson junctions by employing NbSe₂ flakes as superconducting electrodes and a graphene layer as a normal-conducting weak-link spacer. Dry transfer technique [14]

enabled to form superconducting interfaces which were sufficiently transparent as to exhibit Josephson coupling. The planar junction geometry allowed electrostatic tuning of Josephson coupling by modulating carrier density of the graphene weak link. To form an NbSe₂–graphene–NbSe₂ planar Josephson junction, we dry-transferred [14] graphene–hexagonal-boron-nitride (hBN) bilayer onto two pieces of NbSe₂ flakes which were closely pre-arranged. The outermost hBN layer protected the Josephson junction from the chemicals and ambient moistures during the fabrication process. Thus-prepared junction exhibited the Josephson current up to temperature of $T \sim 1$ K. We investigated the nature of Josephson coupling by examining the temperature and magnetic-field dependences of the junction critical current I_c . The temperature dependence of I_c showed that the junction was in a long diffusive regime [24]. Gate dependence of the Josephson coupling and Fraunhofer interference under magnetic field were also investigated. This study demonstrates the potentials of vdW heterostructured superconducting hybrid quantum devices [25-27].

2. Device fabrication and basic characteristics

Figures 1(a-f) illustrates device fabrication processes. The hBN flakes were mechanically exfoliated and identified on a SiO₂ substrate coated with a thin polypropylene carbonate (PPC) layer. After peeling off a PPC layer from a SiO₂ substrate, it was transferred onto a Gel-pak layer. Using dry transfer method [14], a graphene layer was picked up by an hBN layer. Consequently, a graphene–hBN bilayer stack was transferred onto two separate pieces of NbSe₂ flakes, which were closely pre-arranged with parallel edges as shown in Fig. 1(a). The NbSe₂ flakes were cover-protected by a graphene–hBN bilayer within an hour of intermission after the exfoliation of NbSe₂, which was to minimize the surface degradation of NbSe₂ [20]. The Gel-pak stamp released the entire vdW structure at 100 °C by melting PPC layer [Fig. 1(b)], followed by vacuum annealing to remove PPC residues [Fig. 1(c)]. Scanning electron microscope (SEM) image of Fig. 1(g) was taken at this stage of fabrication. The hBN layer was partially etched using CF₄ plasma to make electrode contact holes [Fig. 1(d)]. Ti/Au (5/100 nm in thickness) bilayer contact electrodes was deposited in sequence right after the *in-situ* etching of degraded NbSe₂ surface by Ar ion milling without breaking the vacuum [Fig. 1(e)]. During fabrication processes, air could have been trapped in the cavity between two NbSe₂ flakes and graphene. A small hole of 500 nm in diameter (black dotted circle in Fig. 1(h)) was made by using CF₄ plasma etching and Ar ion milling [Fig. 1(f)]. It was to

pump out the air trapped in the cavity and thus to minimize the hysteresis with respect to backgating. Figure 1(h) shows an optical image of the device A together with the measurement configuration. Graphene is denoted by a white dashed line. All of the data in this report, except for the Fig. 3(b), were taken from the device A. The key in this fabrication process was that the graphene and NbSe₂ were not exposed to chemicals during the fabrication processes. The graphene layer was suspended from the substrate by the thickness of the NbSe₂ electrodes, which helped maintain its cleanness. Figure 1(i) shows gate-voltage (V_G) dependence of the normal-state junction resistance (R_N). R_N is asymmetric with respect to the charge neutral point (CNP). The work function of NbSe₂, ~5.9 eV [28], is larger than that of graphene, ~ 4.5 eV [29, 30], so that the graphene layer in contact with NbSe₂ electrodes was hole-doped. A slight increase of R_N in the electron-doped regime ($V_G > 0$) is attributed to the formation of less transparent p-n-p junction by the work function mismatch between graphene and NbSe₂. V_G dependence of the junction conductance in out-of-plane magnetic fields is plotted in Fig. 1(j). In high magnetic fields, the junction shows clear quantized plateaus of $G = \nu e^2/h$ with filling factor of $\nu = 4 \left(n + \frac{1}{2} \right) = 2, 6, 10$ (red horizontal lines; n = the Landau-level index). These quantized half-integer quantum-Hall conductance plateaus confirm that the graphene layer used in the measurements was a monolayer [2, 3]. The plateaus are clearer at negative V_G than positive side (not shown) due to the hole-doping of the graphene at the contacts with NbSe₂.

3. Results and discussion

Current-voltage (I - V) characteristic curves of Fig. 2(a) were taken at base temperature of 15 mK. The I - V characteristics show sharp transitions from a superconducting to resistive state at I_c . In addition, V_G changes both I_c and R_N . There is a negligible hysteresis in I_c and retrapping current (I_r), indicating that the junction is in an overdamped state [31]. Figure 2(b) shows the differential resistance as a function of V_G and the bias current. The upper and lower boundaries between the dark-blue (zero dV/dI) and light-blue regions (finite dV/dI) represent I_c and I_r , respectively. The overlapped red curve represents V_G dependence of the normal-state conductance. I_c shows an asymmetric gate dependence across the CNP, similar to the feature appeared in normal-state junction conductance. Although the graphene layer between the NbSe₂ flakes was electron-doped by the positive V_G , the graphene region

in contact with the NbSe₂ remained to be hole-doped, which led to the formation of a p-n-p junction with weakened Josephson coupling.

The temperature dependence of I_c for various V_G obtained from the device A is plotted in Fig. 3(a). Data are fitted with the long-junction behavior, the temperature dependence of which is written as,

$$eI_c R_N = \alpha a E_{Th} \left[1 - b \exp\left(-\frac{a E_{Th}}{3.2 k_B T}\right) \right], \quad (1)$$

where $E_{Th} = \hbar D/L^2$ is the Thouless energy, $\hbar$ is the Planck constant divided by 2π , and a and b are phenomenological fitting parameters [24]. Here, $D = v_F l_{mfp}/2$ is the carrier diffusion constant in the graphene layer, where v_F is the Fermi velocity and l_{mfp} is the mean free path. α is introduced to reflect reduction of the junction current due to low contact transparency, Fermi-velocity mismatch, and Fermi-level pinning. Using the data of Fig. 1(i), E_{Th} and l_{mfp} are estimated to be 50 μ eV and 30 nm, respectively, for $V_G = -50$ V. As $l_{mfp} \sim 30$ nm is smaller than the channel length of the device A, $L=450$ nm, the junction is in a diffusive limit. In a long junction limit of $E_{Th}/\Delta_{NbSe_2} \rightarrow 0$, the parameters are predicted to be $a = 10.8$ and $b = 1.30$ [24]. Here, Δ_{NbSe_2} is the superconducting gap of NbSe₂. The best fitting with the data gives $a = 1.7 \sim 2.7$, $b = 1.4 \sim 1.7$ and $\alpha = 0.10 \sim 0.14$ with $E_{Th}/\Delta_{NbSe_2} = 0.057$. The device A falls into the intermediate regime between the short ($E_{Th}/\Delta_{NbSe_2} > 1$) and the long ($E_{Th}/\Delta_{NbSe_2} < 0.01$) junction limits. The fitting parameter a was indeed smaller than 10.8 as expected in the intermediate regime between the long and short junction limits [24, 32]. The temperature dependence of I_c for the device B of $L=150$ nm is shown in Fig. 3(b). $I_c - T$ curves shows a convex upward behavior at low temperatures, which is reminiscent of a short-junction behavior [21, 23, 33]. However, the I_c data at higher temperatures are required to claim the short Josephson coupling characteristics.

The periodic modulation of I_c in varying magnetic fields, so-called the Fraunhofer diffraction pattern, shown in Fig. 4 is a firm evidence of the Josephson junction [31]. Color-coded differential resistance maps are plotted as a function of magnetic field and bias current for various V_G [Fig. 4]. Boundaries between the zero-resistance and resistive regions represent I_c and I_r . I_c shows local minima when integer multiples of magnetic flux quanta penetrate the junction area. I_c clearly oscillates in magnetic-field periods of $B_0 =$

$\Phi_0/[(L + 2\lambda)W] \sim 2.0$ G, where $\Phi_0 = h/2e$ is the magnetic-flux quantum, W is the junction width, and λ is the London penetration depth of the superconducting NbSe₂ flakes [31, 34]. The period B_0 is similar at different V_G as indicated by red vertical dashed lines. When current distribution is uniform along the width of the junction, the magnetic field dependence of I_c follows the relation of $I_c(B) = I_0|\sin(\pi\Phi/\Phi_0)/(\pi\Phi/\Phi_0)|$, where $\Phi = B[(L + 2\lambda)W]$ is the magnetic flux penetrating the junction area [31, 34]. The solid lines are the fitting curves to the experimental results. The magnetic field period of the oscillation $B_0 \sim 2.01$ G yields $L + 2\lambda \sim 4.1$ μm . While the London penetration depth λ of the bulk NbSe₂ is known to be 130 nm [35], it should be replaced by an expression, $\lambda_{2D} \sim 2\lambda^2/d$, for a 2D superconductor [36], which turns out to be 1.5 μm for the NbSe₂ electrodes with the thickness of $d=23$ nm as used in this study. The corresponding field penetration range becomes $L + 2\lambda_{2D} \sim 3.5$ μm , which is in reasonable agreement with the value determined from the data, 4.1 μm .

Lastly, we discuss the bias-voltage dependence of the differential resistance shown in Fig. 5(a), exhibiting the multiple Andreev reflection (MAR) at low temperatures, which is a distinctive feature of proximity-type Josephson junctions [37, 38]. Multiple Andreev reflection exhibits differential resistance dips indicated by a red dotted line in Fig. 5(a) at the voltage of $V_{\text{MAR}} = 2\Delta_{\text{NbSe}_2}/ne$ [37, 38], with the integer number corresponding to $n = 2$. As temperature increases, V_{MAR} decreases due to the reduction of superconducting gap energy and vanishes above critical temperature of NbSe₂ as shown in Fig. 5(b). The device A consists of two NbSe₂ flakes of slightly different critical temperatures, $T_{c,u} \sim 5.3$ K for the upper flake and $T_{c,l} \sim 5.6$ K for the lower one as indicated in the inset of Fig. 2(a). To analyze the temperature dependence of V_{MAR} for a Josephson junction consisting of two superconductors of different T_c , we introduce the combined superconducting gap of $\Delta_0(T) = 2\Delta_u\Delta_l/[\Delta_u + \Delta_l]$, where Δ_u and Δ_l are the superconducting gaps of upper and lower NbSe₂ flakes, respectively [23, 39]. Here, we assumed the temperature dependence of Δ_u and Δ_l following the Bardeen–Cooper–Schrieffer (BCS) theory and the zero-temperature superconducting gap $\Delta_{u/l}(T = 0) = \gamma k_B T_{c,u/l}$ with a proportionality factor γ [40]. The solid pink line in Fig. 5(b) represents the best fit with V_{MAR} for the value of $\gamma = 1.72$, which agrees well with the BCS theory of 1.76 [40]. This gives $\Delta_0(T = 0) \sim 0.81$ meV.

4. Conclusions

In conclusion, we successfully fabricated vdW-stacked planar NbSe₂–graphene–NbSe₂ Josephson junctions by using the dry transfer technique [14]. The outermost hBN layer protected the entire device structure from the chemical contamination during the fabrication processes. This made it possible to fabricate Josephson junctions based on the vdW superconductor, NbSe₂, even being exposed to air. The temperature dependence of I_c shows a typical long-diffusive Josephson-junction behavior [24]. The Fraunhofer interference in out-of-plane magnetic fields confirms Josephson coupling with a uniform current distribution along the width of the junction. Multiple Andreev reflection signatures further confirm proximity-type of the Josephson coupling. This study reports on the first realization of vdW-contacted planar Josephson junctions by dry transfer technique, which would pave a road to a potential application of vdW-stacked heterostructures for superconducting hybrid quantum devices.

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Figures and captions

Figure 1. Device fabrication process and normal-state properties of graphene. (a-f) Schematics of the fabrication processes. (g) SEM image of a multilayered heterostructure of Fig. 1(c). The thickness of NbSe₂ (23 nm) and hBN (30 nm) flakes were measured by atomic force microscope. (h) Optical microscope image of the fabricated device and measurement configurations. After loading a device in a sealed can, the trench cavity was pumped out through a hole, denoted in the black dotted circle, to remove any contaminant trapped inside the trench cavity. (i) Gate-voltage dependence of the normal-state junction resistance obtained from the device A at $T=15$ K. (j) The gate-voltage dependence of the conductance in out-of-plane magnetic fields, ranging from 0 to 16 T.

Figure 2. Basic Josephson junction properties. (a) Current-voltage (I - V) characteristic curves of the device A for different gate voltages, V_G . Inset: resistance vs temperature (R - T) curves of the upper and lower NbSe₂ flakes, showing the critical temperature of the upper NbSe₂ flake $T_{c,u} \sim 5.3$ K and the lower one $T_{c,l} \sim 5.6$ K. (b) Differential-resistance map of the device A as a function of the gate voltage V_G and the bias current I . Red curve shows the gate-voltage dependence of the normal-state junction conductance.

Figure 3. Temperature dependence of the junction critical current I_c obtained for (a) the device A and (b) the device B at various gate voltages V_G . (a) Solid curves are the best fits with the long-diffusive junction behavior of I_c with corresponding parameters listed in the table. (b) Upper inset: Optical microscope image of the fabricated device B. Grey dashed line indicates the graphene boundary. Lower inset: Current-voltage (I - V) characteristic curves of the device B for different gate voltages, V_G . The critical current I_c in the main panel is determined by the bias currents giving rise to maxima of the differential resistance dV/dI of the I - V curves.

Figure 4. Magnetic field dependence of the critical current; Fraunhofer interference pattern. Differential resistance map of the device A as a function of magnetic field and bias current for various gate voltages, V_G . Red vertical lines indicate the period of I_c oscillation. Magenta solid lines represent theoretical fitting of the Fraunhofer pattern with a uniform current distribution along the junction width.

Figure 5. Temperature dependence of the multiple Andreev reflection (MAR) signature. (a) Bias-voltage dependence of the differential resistance obtained from the device A, measured at temperatures ranging from 1.6 to 6.9 K. The red dotted line is a guide to the eye, corresponding to the local minima of the differential resistance. (b) Temperature dependence of the differential resistance minima (symbol) and the Bardeen-Cooper-Schrieffer (BCS) fit (solid line).

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