Electron transport in a bilayer graphene/layered superconductor NbSe$_2$ junction: effect of work function difference

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Abstract. We have experimentally studied electron transport in a bilayer graphene (BLG)/layered superconductor NbSe$_2$ junction encapsulated with hexagonal boron nitride. The junction exhibits nonlinear current-voltage characteristics which strongly depend on the gate voltage around the charge neutrality point (CNP) of the BLG. Besides, we observe that the gate voltage dependence of electron transport in the BLG portion close to the junction interface is different from that of the BLG portion apart from the interface, indicating that the spatial variation of the Dirac point in the charge transfer region due to the difference in work function between superconductor and graphene needs to be considered in the analysis of the superconducting proximity effect.

1. Introduction

At a superconductor/normal metal interface, an incident electron ($-e$) from the normal metal enters the superconductor forming a Cooper pair ($-2e$) and a hole ($+e$) is reflected back to the normal metal due to the charge conservation. This process is called the Andreev reflection.$^1$ Usually, because both the incident electron and the reflected hole belong to the conduction band of the normal metal, the reflected hole goes back along the same path of the incident electron (intraband retro-reflection). On the other hand, when graphene is used as the normal metal, the reflected hole is in the valence band for the Fermi level close to the Dirac point (DP), leading the interband specular Andreev reflection. Theory$^2$ tells that when the Fermi level of graphene is swept across the DP, the transitions from retro- to specular and from specular to retro-reflections cause an anomalous behavior in the current-voltage ($I$-$V$) characteristics. This can be used as an experimental proof of the existence of the specular reflection.

In experiments, the specular Andreev reflection in (single layer) graphene has not been confirmed for a long time,$^3,4,5,6,7,8,9$ presumably due to the spatial fluctuation of the carrier density (electron/hole puddles) in graphene caused by the charged impurities,$^{10}$ which allows the retro- and specular reflections to coexist. Actually, the magnitude of the DP fluctuation in graphene placed on SiO$_2$ is around 50 meV, which further exceeds the superconducting gap energy of conventional superconductors. Besides, due to the difference in work function between
graphene and the superconductor, charge carriers (electrons or holes) are doped to graphene in the vicinity of the interface (charge transfer region).\cite{11, 12, 13} The charge transfer region is reported to extend over $\sim 500$ nm with the energy difference of 60 meV for a Ni/graphene interface.\cite{13} The effect of the work function difference and the charge transfer region were not taken into account in the original theory of Andreev reflections in graphene.\cite{2} Recently, Efetov et al.\cite{14} reported observation of the anomalous $I-V$ characteristics due to the transition between specular and retro-reflections. In their device, they successfully decreased the number of charged impurities by encapsulating the junction with hexagonal boron nitride (hBN), and further reduced the spatial fluctuation of the DP by using bilayer graphene (BLG) instead of single layer graphene. Here, BLG is a zero-gap semiconductor with larger density of states around the DP due to the parabolic bands. It is noted that in their analysis, the experimental result is compared with the theoretical one which does not take into account the effect of the work function difference and the charge transfer region, and it is not clear how the work function difference affects the electron transport in the graphene/superconductor junctions.

In this study, to elucidate the effect of the work function difference between graphene and superconductor, we investigate the electron transport in a superconductor/BLG device which has a device structure similar to that reported in Ref. \cite{14}.

2. Experiment

We fabricated an NbSe$_2$/BLG junction encapsulated with hBN using the mechanical exfoliation of layered materials and van der Waals dry transfer technique\cite{15} in a glove box filled with Ar to prevent the oxidization of NbSe$_2$. Here, NbSe$_2$ is a layered superconductor with the critical temperature of 7.0 K. The device structure is shown in Fig. 1. The BLG is rectangular-shaped with several side arms for the four terminal measurement. A flake of NbSe$_2$ with thickness of $\sim 35$ nm is placed on Cr/Au electrodes and the BLG. We measured voltage $V_1$ across the junction (with the BLG portion “A” with length $d_1 = 2.51$ $\mu$m, see Fig. 1(a)), and voltage $V_2$ across the side arms in the BLG portion “B” (with length $d_2 = 2.20$ $\mu$m) simultaneously in the four terminal configuration, as a function of the bias voltage (applied between leads I+ and I− in Fig. 1) and the gate voltage $V_g$ which was applied to the highly doped Si substrate.

![Figure 1.](image-url)
Figure 2. Voltage $V_1$ dependence of the differential conductance of the junction, $dI/dV_1$, for several gate voltages in the superconducting state at 4.2 K (a) and in the normal state at 10 K (b). (c) The differential resistances $dV_1/dI$ and $dV_2/dI$ at 10 K as a function of the gate voltage.

3. Results and discussion

Figure 2 shows the differential conductance of the junction, $dI/dV_1$, as a function of $V_1$ for several gate voltages with NbSe$_2$ in the superconducting state at 4.2 K (Fig. 2(a)) and in the normal state at 10 K (Fig. 2(b)). In Fig. 2(b), the conductance is almost independent of $V_1$, but strongly depends on the gate voltage due to the modulation of the carrier density of BLG. The gate voltage which gives the minimum of $dI/dV_1$ corresponds to the charge neutrality point (CNP) of the BLG. In the superconducting state (Fig. 2(a)), while the value of $dI/dV_1$ at $|V_1| \gtrsim 3$ mV is close to the value of $dI/dV_1$ at the same $V_g$ in Fig. 2(b), a dip appears in $dI/dV_1$ at $|V_1| \lesssim 1$ mV. This is due to the Andreev retro-reflection, corresponding to the superconducting energy gap of NbSe$_2$ ($\Delta \sim 1.1$ meV). Here, we note that the depth of the dip strongly depends on the gate voltage, i.e., the dip becomes shallower as the gate voltage approaches the CNP. The origin of this gate voltage dependence is not clear at this moment. Figure 2(c) shows the differential resistances $dV_1/dI$ and $dV_2/dI$ at 10 K as a function of the gate voltage. The resistance peak appears at $V_g = -1.7$ V and $-2.0$ V for $dV_1/dI$ and $dV_2/dI$, respectively. The difference in the gate voltage dependence indicates the existence of the charge transfer region near the interface. Actually, since the work function of BLG ($\sim 4.5$ eV[16]) is smaller than that of NbSe$_2$ (5.9 eV[17]), holes are doped to BLG, so that the gate voltage
Figure 3. (a) The ratio of $G_{NS}$ in the superconducting state (4 K) to $G_{NS}$ in the normal state (10 K) plotted in the $V_{NS} - V_g$ plane. (b) The bias voltage plotted in the $V_{NS} - V_g$ plane. Red and blue correspond to positive and negative bias voltages. (c) The gate-voltage dependence of the measured voltages, $V_1$ and $V_2$ and the calculated interface voltage, $V_{NS}$, for bias voltage of +5 mV and temperature of 4.2 K.

Corresponding to the resistance peak of region A near the interface is expected to be larger than that of region B apart from the interface, which agrees with the experimental finding in Fig. 2 (c).

Following the analysis in Ref. [14], we derive the voltage of the interface, $V_{NS}$, by subtracting the nominal voltage drop in the BLG portion A from $V_1$ as $V_{NS} = V_1 - (d_1/d_2) V_2$ and calculated the differential conductance of the junction interface, $G_{NS} = dI/dV_{NS}$.

Figure 3(a) shows the ratio of $G_{NS}$ in the superconducting state (4.2 K) to $G_{NS}$ in the normal state (10 K) plotted in the $V_{NS} - V_g$ plane. At $|V_{NS}| \lesssim 1$ mV and large $|V_g|$, suppression of $G_{NS}^{4K}/G_{NS}^{10K}$ is seen, which agrees with the behavior of the Andreev retro-reflection. Besides, anomalous behavior is seen around $V_g = -2.5$ V. Here, we point out that inside the anomaly region ($-2.7 \leq V_g \leq -2.0$ V), the sign of the calculated interface voltage $V_{NS}$ is opposite to that of the bias voltage applied between leads I+ and I−, as shown in Figs. 3(b) and 3(c), which is physically incorrect. This occurs because the gate voltage dependence of the resistance of the BLG portion A is different from that of the BLG portion B due to the carrier doping from the superconductor, and thus, $V_{NS} = V_1 - (d_1/d_2) V_2$ does not hold. Since our device has a large $d_1/d_2$ ratio, this difference is clearly seen as the sign inversion of $V_{NS}$.

In recent theoretical works,[18, 19] the effect of the work junction difference on the differential conductance of BLG/superconductor interface have been discussed. In the model of Ref. [18],
a part of BLG is covered with a superconductor, and the covered region exhibits a potential (corresponding to a DP shift) due to the work function difference. Here, a stepwise change of the potential is assumed, so that the gradual spatial variation of the DP near the interface within the charge transfer region is not taken into account. Another theoretical work by Takane et al. has adopted a similar model but taken into account the spatial variation of the DP in BLG close to the interface.[19] They predicted that even when the Andreev retro-reflection occurs at the interface, the incident electron and/or the reflected hole exhibits diffraction when passing over a naturally formed $p - n$ junction near the interface, leading to the seemingly specular Andreev reflection. The interpretation of our results based on Refs. [18] and [19] is in progress.

4. Conclusions
We have experimentally investigated electron transport in a BLG/superconductor junction. We observe that the gate voltage dependence of the resistance of the BLG portion near the junction interface is different from that of the BLG portion apart from the interface. This is presumably due to the work function difference between BLG and the superconductor and the existence of the charge transfer region. We point out that the analysis of the electron transport in BLG/superconductor junctions needs to take into account the work function difference and the charge transfer region.

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