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Published in:
Physica Status Solidi B-Basic Solid State Physics

DOI:
[10.1002/pssb.201700181](https://doi.org/10.1002/pssb.201700181)

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2017

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Chen, Q., Liang, L., Ali El Yumin, A., Lu, J., Zheliuk, O., & Ye, J. (2017). High Quality Superconductor–Normal Metal Junction Made on the Surface of MoS₂ Flakes. *Physica Status Solidi B-Basic Solid State Physics*, 254(11), [1700181]. <https://doi.org/10.1002/pssb.201700181>

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High Quality Superconductor–Normal Metal Junction Made on the Surface of MoS₂ Flakes

Qihong Chen, Lei Liang, Abdurrahman Ali El Yumin, Jianming Lu, Oleksandr Zheliuk, and Jianting Ye*

A superconductor–normal metal (SN) junction is fabricated on the surface of a few-layer MoS₂ flake. Superconductivity is induced by ionic liquid gating, and an *h*-BN flake is used to locally separate ionic liquid from the surface of MoS₂. The *h*-BN covered channel remains semiconducting, therefore an SN junction forms at the interface between the exposed and *h*-BN covered MoS₂ channel. Transport properties at the SN junction are studied at low temperatures and under magnetic fields. The probability of observing Andreev reflection is discussed based on the BTK model.

1. Introduction

Electrical transport of superconductor-normal conductor (SN) junctions has shown a variety of interesting mesoscopic phenomena.^[1–3] Given a particular superconductor, the normal conductor part of the junction can be a metal,^[4,5] semimetal,^[6–9] or degenerate semiconductor.^[10–12] At the interface of an SN junction, the current is converted from a dissipative flow carried by normal electrons to a non-dissipative supercurrent carried by Cooper pairs. This process is known as the Andreev reflection.^[13] In a traditional SN junction, the superconductor and normal conductor are constructed by different materials. Therefore, contact resistance between two different materials is unavoidable for electrical measurement due to difference in work functions. Practically, contaminations, residues from the fabrication process, etc. are regarded as imperfections on interfaces, which act as extrinsic factors of the ideal Andreev reflection process modeled at clean interfaces.

To study the intrinsic transport properties of the Andreev process, we fabricated a SN junction on the surface of a single few-layer molybdenum disulfide (MoS₂) crystal. Superconductivity is induced by the well-known ionic liquid gating

technique.^[14–17] By partially covering the surface of MoS₂ using a thin hexagonal boron nitride (*h*-BN) flake, part of the MoS₂ is isolated from ionic liquid and remains semiconducting. An atomically sharp and perfectly clean SN junction forms at the interface of the exposed and *h*-BN covered part of the flake. Since the SN junction is formed in the same material, we expect to obtain high electrical transparency. Nevertheless, an intrinsic energy barrier forms at the interface due to the different doping levels at the S and N sides. At low temperatures, an abrupt upturn of the

temperature dependence of resistance and non-monotonic magnetoresistance are observed. Our results can be well explained by theoretical calculations of the Andreev reflection probability at the SN interface with a finite barrier.

2. Materials and Methods

To fabricate SN junctions, few-layer MoS₂ flakes were first prepared on silicon wafers by mechanical exfoliating bulk single crystals of 2H polytype (SPI supplies), following the well-known scotch tape method.^[18] Then we used the dry transfer technique^[19] to transfer selected flakes onto a thin (~30 nm) and uniform *h*-BN flake, which serves as the direct contacting substrate. Another carefully chosen *h*-BN flake with a quasi-rectangular shape was laid across the flake to mask the MoS₂ surface from contacting the ionic liquid. Electrodes composed of Ti/Au (5/60 nm) were patterned with the standard e-beam lithography method.

After patterning electrodes, the whole sample is immersed in a small droplet of the ionic liquid: N,N-diethy-N-(2-methoxyethyl)-N-methylammonium bis-(trifluoromethylsulfonyl)imide (DEME-TFSI), which is widely used in ionic gating.^[14,17,20–22] All ionic gating procedures were conducted at $T = 220$ K for inducing carriers electrostatically. By retaining the gate voltage and cooling down to below the glass transition temperature of the ionic liquid, dynamic gating was transformed into static doping after freezing the ion movements. At low temperatures, the induced carriers were maintained even if the gate bias was grounded. After measurement, the gating effect can be thermally released by warming the sample up to $T = 220$ K, at which ion movement reactivates.

For inducing carriers, bias on ionic liquid and SiO₂ back gate were set by a DC source meter (Keithley 2450). The electrical transport was measured by AC locking techniques using standard lock-in amplifiers (Stanford Research SR830).

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DOI: 10.1002/pssb.201700181

3. Results and Discussion

The device structure and gating configuration are schematically shown in **Figure 1(a)** and (b), respectively. **Figure 1(c)** shows the optical image of a device fabricated on a 3 nm-thick MoS₂ flake. For a flake containing N layers, the corresponding thickness measured experimentally is $d = a + (N - 1) \times b$, where a is the height of the bottom layer (including the space between the bottom layer and the substrate) and b is the actual thickness of individual layers. In MoS₂, the typical value of a ranges from 0.8 to 1 nm depending on the adhesion between the flake and underlying substrates, while b is around 0.62 nm. Therefore, the MoS₂ flake used in this study is composed of four monolayers. The width of top h -BN flake is $\sim 2 \mu\text{m}$. To consistently describe different electronic states formed on different parts of the channel, afterwards, we refer the “exposed channel” to the surface of MoS₂ that is exposed to ionic liquid, measured by contacts 2 and 3 [**Figure 1(c)**], the “ h -BN covered channel” to the surface of the MoS₂ that is covered by the top h -BN flake, measured by contacts 4 and 5

[**Figure 1(c)**]. The exposed MoS₂ channel is in contact with the ionic liquid, where superconductivity can be easily induced by ionic gating. Whereas, the h -BN covered channel remains almost unaffected from ionic gating without direct contact with the ionic liquid. The conductivity of this channel can be efficiently tuned by the back gate through dielectrics composed of 300 nm SiO₂ and a 30 nm h -BN flake. The SN junction forms at the interface between the exposed and h -BN covered MoS₂ channel.

The transfer curve of the exposed channel by ionic gating is shown in **Figure 1(d)**. The device can be switched on and off with bipolar transistor operation, showing on/off ratio $> 10^5$. To form the SN junction, we rapidly cool the device down to $< 180 \text{ K}$ to freeze the doping state with $V_{\text{LG}} = 4 \text{ V}$. At low temperatures, the ionic gate voltage can be retracted without losing the gating effect.

Figure 1(e) plots the temperature dependence of the exposed channel resistance, showing overall metallic behavior before a sharp resistance drop corresponding to the superconducting transition. The transition temperature T_c is $\sim 4.3 \text{ K}$, defined as 50% of the normal state resistance. Zero resistance state is reached at $T < 4 \text{ K}$. As shown in **Figure 1(f)**, the superconductivity is gradually suppressed by applying out-of-plane magnetic fields B . Using the same standard of 50% of the normal resistance, the upper critical field B_{c2} is extracted as shown in the inset of **Figure 1(f)**. According to the two-dimensional (2D) Ginzburg–Landau model,^[23] the superconducting coherence length is related to the critical field by: $B_{c2}^{\perp}(T) = \frac{\Phi_0}{2\pi\xi_{\text{GL}}(0)^2} \left(1 - \frac{T}{T_c}\right)$, where the parameter $\Phi_0 = \frac{h}{2e} = 2.07 \times 10^{-15} \text{ Wb}$ is the flux quantum and $\xi_{\text{GL}}(0)$ is the Ginzburg–Landau coherence length at zero temperature. By fitting the experimental data, we obtained $B_{c2}^{\perp}(0) = 0.68 \text{ T}$ for $T_c = 4.26 \text{ K}$. Thus we estimate the Ginzburg–Landau coherence length $\xi_{\text{GL}}(0) = 22 \text{ nm}$. Here, ξ_{GL} is much larger than the thickness of the flake hence the superconductivity can be regarded as two-dimensional. Theoretical calculations show that with 2D carrier density $n_{2\text{D}}$ induced by ionic gating in the order of $\sim 10^{14} \text{ cm}^{-2}$, the density of accumulated carriers decays exponentially for individual layers from the top to bottom due to strong Thomas–Fermi screening effect.^[24] The calculated $n_{2\text{D}}$ of the second layer is reduced by almost 90% compared to that of the first one. Therefore, the topmost layer is electronically isolated from the rest layers below and acts like a freestanding monolayer. As a result, superconductivity only exists in the topmost layer,^[17,21] hence the SN junction also forms at the top surface.

For the h -BN covered channel, back gate can efficiently tune the conductivity as a normal field effect transistor (FET). The measured four-probe resistance as a function of back gate voltage is shown in **Figure 2(a)**. For all different temperatures, R decreases with increasing V_{BG} , as expected for an n -type semiconductor. At $V_{\text{BG}} > 0 \text{ V}$, sample shows

Figure 1. Schematic illustration of (a) the device structure and (b) the ionic gating configuration. (c) Optical image of a device fabricated with four-layer MoS₂. MoS₂ and the top h -BN are outlined by blue and purple dashed lines, respectively. Scale bar: $5 \mu\text{m}$. (d) Transfer curve by ionic gating for the exposed channel, at $T = 220 \text{ K}$ and $V_{\text{DS}} = 0.1 \text{ V}$. (e) Temperature dependence of the resistance of the exposed channel, with fixed $V_{\text{LG}} = 4 \text{ V}$. (f) The resistance of the exposed channel as a function of temperature at $V_{\text{BG}} = 100 \text{ V}$, under different perpendicular magnetic fields: 0, 0.05 T, 0.1 to 1.5 T in 0.1 T steps, 1.7, 2, 2.5, 3 and 5 T. Inset: Temperature dependence of the upper critical field B_{c2} . Solid line is the best linear fitting.

Figure 2. (a) The resistance of the *h*-BN covered channel as a function of V_{BG} at different temperatures from 2 to 170 K. Inset: Comparison of the back gate dependence of the resistance of the *h*-BN covered (black) and exposed (red) channel. (b) The temperature dependence of the resistance (RT curves) of the *h*-BN covered channel under different out-of-plane magnetic fields from 0 to 1.2 T, with $V_{BG} = 100$ V.

metallic behavior, where resistance decreases significantly as temperature goes down. The FET mobility can be calculated by $\mu_{FET} = \frac{1}{C_g} \frac{d\sigma}{dV_{BG}}$, where σ is the conductivity and C_g is the gate capacitance per unit area. With the decrease of temperature, the extracted mobility increases from 100(170 K) to 1000 cm² Vs⁻¹ (2 K). The observed high mobility suggests the high quality channel formed in the *h*-BN covered region. In contrast to the effective tuning of the *h*-BN covered channel, the exposed superconducting channel is almost back gate independent, as shown in the inset of Figure 2(a), because ionic gating induces carrier density in the range of 10¹⁴ cm⁻², which is far beyond the tuning capability of SiO₂ back gate.

Figure 2(b) shows the temperature dependence of the resistance of the *h*-BN covered channel at different magnetic fields with fixed back gate voltage $V_{BG} = 100$ V. At zero field, a sharp drop of resistance associated with the superconducting transition of exposed channel is observed with an onset temperature of 5 K. The transition indicates that Cooper pairs from the superconducting channel diffuse into the *h*-BN covered normal channel due to the proximity effect; therefore the resistance decreases when the normal channel becomes partially superconducting. The diffusion length of the Cooper pair is related to the superconducting coherence length and mean free path of the normal channel. From $\mu = \frac{e\tau}{m^*}$, where $m^* = 0.6m_e$ is the electron effective mass, we can calculate the scattering time $\tau = 340$ fs at $T = 2$ K. The carrier density n_{2D} at $V_{BG} = 100$ V in the *h*-BN covered channel can be calculated using the back gate voltage and capacitance $n_{2D} = \frac{C_g}{e}(V_{BG} - V_{th})$, here $V_{th} \approx -20$ V is the threshold voltage for back gate, $C_g = 10.5$ nF cm⁻² is the capacitance per unit area considering two dielectric layers of 300 nm SiO₂ and 30 nm *h*-BN. Therefore, $n_{2D} \approx 7.9 \times 10^{12}$ cm⁻². The Fermi velocity can be determined by $v_F = \frac{\hbar k_F}{m^*}$, where $k_F = \sqrt{\frac{4\pi n_{2D}}{g_s g_v}}$, $g_s = 2$, and $g_v = 6$ are the spin and valley degeneracy for few-layer MoS₂, respectively. As a result, the electron mean free path in the *h*-BN covered region is $l = v_F \tau \approx 19$ nm, which is comparable to the superconducting coherence length (~ 22 nm). Both the mean free path and coherence length are much smaller than the width of the *h*-BN covered region (~ 2 μm). Therefore, the penetration depth of the Cooper pairs is very small from the SN interface.

This explains why proximity effect only induces a small resistance drop (~ 5 K) in the normal channel, instead of a zero resistance superconducting state. With further decrease of temperature (< 4 K), an abrupt upturn of the resistance appears as an unusual behavior beyond simple expectation that pairing becomes stronger at lower temperature where lower resistance is expected.

Similar unusual behavior is observed in the magnetoresistance (MR) of the *h*-BN covered channel [measured by contacts 4 and 5 in Figure 1(c)]. As shown in Figure 3, at $T = 2$ K with the increase of the magnetic field, the resistance first decreases (negative MR) and then increases (positive MR). The negative MR becomes less pronounced with the increase of temperature. At $T > 3$ K, the negative MR disappears and only a positive MR dip is observed in small field range. This positive MR dip is clearly associated with the suppression of superconductivity, as it disappears above the superconducting transition temperature. When the temperature further increases above T_c , the MR shows a normal parabolic B -field dependence. It should be noted that the measured resistance [represented by R_N in Figure 1(a)] involves both the resistance of the *h*-BN covered normal channel and the resistance of two SN junctions at the left and right sides of the *h*-BN flake. More specifically, the measured resistance can be written as $R = R_{SN} + R_{MoS_2} + R_{SN}$, where R_{SN} is the resistance at the SN interface and R_{MoS_2} is the channel resistance. As aforementioned, since the width of the normal MoS₂ region is much larger than the superconducting coherence length, these two SN junctions are decoupled and behave similarly due to the symmetric arrangement, thus can be described as one SN junction.

Figure 3. Magnetoresistance at different temperatures from 2 to 10 K, with $V_{BG} = 100$ V. Each curve is shifted by 50 Ω for clarity.

The behavior of the negative MR looks similar to that of weak localization, but it changes to a positive MR above 3 K, and disappears above 5 K. It is therefore unlikely to be explained by the weak localization. In **Figure 4(a)**, the different MR features at low fields are schematically mapped to the temperature dependence of resistance (RT curve). It clearly shows that the unusual MR behavior is correlated with the superconducting transition. Above the superconducting transition, MR shows a normal parabolic shape. Positive MR is observed in the middle of the superconducting transition and negative MR appears when a more robust superconducting state is developed below $T = 4$ K.

Our results can be qualitatively understood by the Andreev reflection process^[13] and the Blonder-Tinkham-Klapwijk (BTK) model,^[25] which describes the crossover from metallic to tunneling behavior of a SN micro-constriction contact. In the BTK model, a dimensionless δ -function with potential barrier Z is used to describe the electrical tunneling at the SN interface. When Z equals to zero, there is no tunneling barrier at the interface corresponding to a perfect SN junction; the electrical transport is then dominated by the Andreev reflection. A very large Z corresponds to the limiting case of classical tunneling barrier, such as in a normal metal-insulator-superconductor (MIS) junction, where the electrical transport is dominated by tunneling of normal electrons. With an intermediate barrier Z , both Andreev reflection and normal reflection occur at the SN interface. In our device, because of the large inequivalent doping levels between ion-gated channel and h -BN covered channel, a Schottky-like barrier is expected to form at the interface. As indicated by the resistance (~ 1 k Ω), the barrier is of intermediate size. Using the BTK model, we calculated the

probability of Andreev reflection at the SN interface with a finite-sized barrier. **Figure 4(b)** shows the probability of the Andreev reflection as a function of the electron energy, which is normalized by Δ (where 2Δ is the BCS superconducting gap). The probability of the Andreev reflection increases with the increase of electron energy before reaching the maximum at Δ . Afterwards, the probability decreases dramatically with the further increase of electron energy.

In general, higher Andreev reflection probability means more Cooper pairs enter the normal region, leading to lower resistance. In our measurement, the electron energy does not change because the bias voltage is fixed, but the effective superconducting gap of the exposed channel shrinks with increasing magnetic fields according to the BCS theory. To simulate our system as a function of magnetic field at low temperature, we calculate the Andreev reflection probability by fixing the electron energy at 0.8Δ as shown in **Figure 4(c)**, where the superconducting gap is normalized by its value at zero field. Consistently, as the gap size gradually decreases from Δ ($B = 0$) to 0 ($B > 0$), the probability of Andreev reflection first increases and then decreases; therefore the channel resistance first decreases and then increases, in good agreement with the MR curve measured at $T = 2$ K (**Figure 3**). It should be noted that the transition from negative to positive MR in **Figure 3** has a more rounded shape compared to the sharp peak shown in **Figure 4(c)**. This is because on one hand, the probability of Andreev reflection increases as the superconducting gap decreases, therefore the resistance at the interface R_{SN} decreases concomitantly. On the other hand, the proximity effect also weakens due to weaker Cooper pairing, which leads to the increase of the channel resistance R_{MoS_2} . The rounded shape of the minimum of MR is a signature of the competition between these two mechanisms.

Figure 4. (a) Schematic mapping of the unusual MR behavior of the h -BN covered channel to the RT curve of the exposed superconducting channel at $V_{\text{BG}} = 100$ V. Light gray region represents the negative MR, in light pink region positive MR is observed, and in the light blue region MR shows a normal parabolic behavior. (b) Calculated probability of the Andreev reflection as a function of the electron energy, with an intermediate barrier strength height. The electron energy is normalized by Δ (where 2Δ is the BCS superconducting gap). (c) Calculated probability of the Andreev reflection as a function of superconducting gap, with the electron energy fixed at 0.8Δ . The superconducting gap is normalized by the gap value at zero field.

4. Conclusions

We studied the transport properties of an ideal SN junction made on the surface of a few-layer MoS_2 flake on an h -BN substrate. Due to the huge difference in the doping levels between the exposed and h -BN covered MoS_2 channel, a Schottky-like barrier formed at the interface. A sharp upturn in the RT curve at low temperature and an unusual negative magnetoresistance were observed at the SN junction. These behaviors could be qualitatively explained by the BTK model. With a finite barrier at the SN interface, the probability of Andreev reflection is a non-monotonic function of electron energy. The probability is maximized when the electron energy equals to Δ (where 2Δ is the BCS superconducting gap size). Negative MR was observed as the superconducting gap decreased with the increase of magnetic fields. As the magnetic field further increased, the Cooper pair breaking dominated the transport, therefore a positive MR dip is observed.

Acknowledgements

We thank J. Harkema for technical support. This work is financially supported by the European Research Council (consolidator grant no. 648855 Ig-QPD) and FOM programme: two-dimensional semiconductor crystals.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

ionic gating, MoS₂, superconductivity, superconductor–normal metal junctions, transition metal dichalcogenides

Received: April 23, 2017

Revised: July 4, 2017

Published online: August 8, 2017

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