Resistance and current-voltage characteristics of individual superconducting NbSe$_2$ nanowires

Zhixian Zhou, Rongying Jin, Gyula Eres, David Mandrus, Victor Barzykin, Pedro Schlottmann, Yew-San Hor, Zhili Xiao, and John F. Mitchell

1Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
2Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996, USA
3Department of Physics, Florida State University, Tallahassee, Florida 32306, USA
4Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA
5Department of Physics, Northern Illinois University, DeKalb, Illinois 60115, USA

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Resistance and current-voltage characteristics of individual superconducting NbSe$_2$ nanowires are investigated. In the current-voltage curves, a stairlike structure is observed, indicating the possible formation of phase-slip centers. A close examination of the current-voltage characteristic in a selected high quality NbSe$_2$ nanowire with a diameter of 75 nm reveals that the characteristic voltages in the stairlike structure follow the BCS-like temperature dependence of superconducting gaps vanishing at $T_C$. While the phase-slip center mechanism remains to be a plausible explanation of the observed features, an alternative model involving multigap Josephson tunneling is proposed to account for the BCS-like temperature dependence. From the BCS fits, two distinct superconducting gaps are extracted. Moreover, the critical current of the 75 nm nanowire at low temperatures as well as near $T_C$ can also be described by the Ambegaokar-Baratoff relation for multigap Josephson junctions. Our data suggest the possible observation of multiband superconductivity in NbSe$_2$ and are in good agreement with the predictions of recent band structure and Fermi surface calculations on NbSe$_2$.

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I. INTRODUCTION

The existence of distinct superconducting energy gaps on different sheets of the Fermi surface (FS) gives rise to an unusual phenomenon termed as multiband superconductivity (MBSC). The observation of this phenomenon in MgB$_2$, a phonon-mediated $s$-wave superconductor with a high transition temperature, $T_C$=39 K, has spurred considerable research interest in the physics of MBSC. To gain a deeper understanding of the physics of MBSC, it is important to study multiband superconductors other than MgB$_2$. NbSe$_2$ is another system that is host to MBSC. NbSe$_2$ as a classic type-II superconductor has been studied intensively over the past four decades. A variety of interesting phenomena have been discovered in this system, including the coexistence of seemingly competitive charge density wave (CDW) order and superconductivity and the “peak effect.” An early study on the superconducting gap of NbSe$_2$ using far-infrared transmission spectroscopy has shown that the temperature dependence of the gap energy is in good agreement with the BCS theory over the limited temperature range of measurement.

Johannes et al. have demonstrated in their recent density function calculations that the FS sheets of NbSe$_2$ can be divided into two groups: the small Se-derived pancake surface around the $\Gamma$ point and the large bonding and antibonding Nb-derived FS sheets. In addition, they predicted that these two sets of FS sheets with different gap sizes can be probed by tunneling measurements. Experimentally, however, the low temperature scanning tunneling spectroscopy measurements performed on NbSe$_2$ single crystals only indicated a distribution of energy gaps between 0.6 and 1.3 meV. Angle resolved photoemission spectroscopy measurements revealed a sizable difference in the magnitude of the superconducting gap on two sets of FS sheets at 5.3 K only. The superconducting NbSe$_2$ nanowires (NWs) recently synthesized by Hor et al. provide a new platform for studying the MGSC in NbSe$_2$, and the observation of grain boundaries (GBs) in some of the NWs by transmission electron microscopy (TEM) inspired us to study the MGSC in NbSe$_2$ through GB tunneling. These NWs are also good candidates for studying the current-induced breakdown of superconductivity, which has been a long-standing problem of recurrent interest. As the bias current is raised above a local minimum critical current of a NW, a phase-slip center (PSC) develops, creating a finite voltage. The successive nucleation of PSCs leads to the appearance of a stairlike structure in the $I$-$V$ characteristics.

In this paper we report electrical transport studies of individual NbSe$_2$ NWs. A stairlike structure has been observed in the current-voltage ($I$-$V$) characteristic, indicating the possible formation of PSCs under a bias current. The temperature dependences of the characteristic voltages in the $I$-$V$ characteristic of a selected NbSe$_2$ NW with a diameter of 75 nm follow the BCS-like temperature dependence of superconducting gaps. Accordingly, an alternative mechanism involving the multigap Josephson tunneling is proposed to explain the observed BCS-like temperature dependences, which cannot be understood in the context of phase slips. The extrapolated superconducting gaps are in quantitative agreement with the theory. We have also found that the critical current of the 75 nm diameter NbSe$_2$ NW is consistent with the theoretical predictions of Ambegaokar and Baratoff.

II. EXPERIMENTAL DETAILS

NbSe$_2$ NWs were synthesized from NbSe$_3$ NWs and Nb powder. The as-grown NWs have the crystal structure of...
$2H$-NbSe$_2$ as determined by x-ray diffraction. Although nearly perfect electron diffraction patterns were observed in selected areas, grain boundaries were indeed found inside some NWs through high-resolution TEM imaging. In order to perform electrical transport measurements on individual NbSe$_2$ NWs, they were first ultrasonically dispersed in isopropyl alcohol, and then deposited onto Si/SiO$_2$ substrates with predefined alignment marks. Subsequently, custom metal electrodes were defined by electron beam lithography followed by electron-gun assisted deposition of a 10 nm Ti adhesion layer and 200 nm of Au. Fast thermal annealing at 400 °C was carried out in vacuum ($10^{-7}$ torr) to improve the electrical contacts. Both four-probe (4P) and two-probe devices were fabricated. Electrical transport measurements were carried out in a Quantum Design physical properties measurement system.

III. RESULTS AND DISCUSSIONS

We have fabricated and characterized over 40 devices, most of which showed signatures of impurity phases such as the two CDW transitions of NbSe$_3$. High quality devices are obtained only in optimally converted NWs. In this paper, we focus on one 4P device with high sample quality. For comparison, data on a slightly underconverted NbSe$_2$ NW (but without the signatures of NbSe$_3$) and two other high quality NbSe$_2$ NWs of substantially larger diameters are also presented and briefly discussed. Figure 1 shows the low bias ($I=200$ nA $\ll I_C$) resistance $R$ as a function of temperature $T$ for the 4P device consisting of a 75 nm diameter superconducting NbSe$_2$ NW as depicted in the lower right inset of Fig. 1(b). The general aspects of the $R(T)$ data resemble those of NbSe$_2$ bulk crystals, except that the $T_C$ in this NW is suppressed from the typical bulk value of 7.2 K to 6.6 K. The slight decrease of $T_C$ may be attributed to the polycrystalline nature of the wire.17,18 In the normal state, a hump is seen in $R(T)$ [Fig. 1(b)], corresponding to the CDW transition of NbSe$_2$ with $T_{CDW}=33$ K determined by plotting $dR/T$ as a function of $T$, indicating high sample quality.16,19,20

Figure 2(a) shows the voltage-current ($V$-$I$) characteristics of the NW for $T<T_C$, applying a gradually increasing dc current. The voltage drop is immeasurably low at low bias currents until the critical current $I_C$ is reached, where the voltage jumps abruptly from zero to a finite value before entering a dissipative region with a linear $V$-$I$ behavior at higher bias currents. Nearly identical results were obtained with reversed polarity, while the $V$-$I$ characteristic was always measured with increasing $I$. At low temperatures, a stairlike structure is clearly visible within the big voltage jump, which in superconducting NWs is often attributed to the formation of PSCs.11–13

Since the self-field generated by the current does not exceed the lower critical field in the wire (or at the surface), we rule out the possibility of filamentary vortex flow, which may also generate steps in $V$-$I$ curves.21–24 In Fig. 2(b), we plot the $T$ dependence of the critical current $I_C$ (defined as the maximum supercurrent). For 0.85$T_C<T<T_C$, $I_C$ increases linearly with decreasing $T$, significantly deviating from the Ginzburg-Landau theory, which predicts $I_C \sim (1-T/T_C)^{3/2}$.
near $T_C$. This discrepancy casts reasonable doubt on “pair breaking” (at a critical velocity of the Cooper pairs) as a possible current-limiting mechanism in our NW.25–27

Falk et al. also observed stairlike structures in their NbSe$_2$ NWs of comparable or larger diameters but with significantly lower $T_C$ (2–2.3 K), where the voltage steps were attributed to the formation of PSCs possibly due to some type of internal structures arising from the variation of the order parameter over the cross section of the superconductor.28 Besides the difference in $T_C$, two other import dissimilarities between our 75 nm NbSe$_2$ NW and their NbSe$_2$ NWs are also noted. First, the voltage drop in our sample is zero at low bias in contrast to the finite $V$ drop in their samples, indicating that thermally activated phase slips (TAPSs) near $T_C$ (Refs. 29–31) and quantum phase slips (QPSs)25–27 which may have contributed to the finite resistances in their samples have a much weaker effect in our NW. Second, the critical current density in our NW is also substantially higher. These differences signify higher sample quality of our selected NW. Although the QPSs and TAPSs are negligible in our sample, the stairlike structure observed in our high quality NW is consistent with the formation of PSCs in thin superconducting filaments such as whisker crystals, thin-film microbridges, and NWs as described by a model proposed by Skocpol, Beasley, and Tinkham (SBT).11–13 Based on the SBT model, every step in the V-I characteristic is associated with the formation of a new PSC with finite resistance. The spatial extent of the PSC is given by the quasiparticle diffusion length $\Lambda=V_{PSC}/2R_N(I-I_s)$, where $V_{PSC}$ is the voltage contributed by a PSC, $L$ is the length of the sample, and $R_N$ is the normal state resistance. Fitting the $L$, $V_{PSC}$, $R_N$, and $(I-I_s)$ values obtained from Figs. 1 and 2 to the formula, the quasiparticle diffusion length of the 75 nm NbSe$_2$ NW is estimated to be $\Lambda \sim 1 \mu$m, comparable to $L$.

For comparison, we have also investigated slightly underconverted NbSe$_2$ NWs (with Se impurities) and high quality NbSe$_2$ NWs with large diameters. Figure 3(a) shows the low bias ($I=10$ nA) $R(T)$ data for a representative slightly underconverted NbSe$_2$ NW device with $d \sim 40$ nm, where not only the onset of $T_C$ is significantly reduced but also the superconducting transition is much broader than that in the high quality sample as depicted in Fig. 1. In addition, the $R(T)$ exhibits two distinct regimes below $T_C$, similar to the findings of Tian et al. in their Sn NWs, where the $R(T)$ data were interpreted as a superposition of TAPS near $T_C$ and QPS far below $T_C$.35 Although the relatively small diameter ($\sim 40$ nm) of the underconverted NbSe$_2$ NW may contribute to the TAPS and QPS, similar phenomena such as significantly reduced $T_C$ and a resistance tail have also been observed in our underconverted NbSe$_2$ NWs with diameters larger than 100 nm. Therefore, as reported by Falk et al., internal structures within the NWs are likely to be the primary cause of the TAPS and QPS in our underconverted NbSe$_2$ NWs. Figure 3(b) shows the V-I characteristic and
dV/dl of the same sample as in Fig. 3(a). At very low I, the voltage drop across the NW is small. As \( I \) increases above a switching current \( I_{SW} \), the slope \( (dV/dl) \) swiftly increases followed by multiple small voltage steps, which are clearly revealed as peaks in the \( dV/dl \) versus \( I \) plot. These small voltage steps are also consistent with the PSC scenario. Figures 3(c) and 3(d) show the \( R(T) \) and \( V-I \) data on two larger diameter NbSe\(_2\) NWs, respectively. Sharp superconducting transitions (\( \Delta T \approx 0.1 \) K) have been observed at temperatures very close to the bulk \( T_C \) of NbSe\(_2\) (7.0 and 6.9 K for the 250 and 300 nm NWs, respectively). In the \( V-I \) characteristic, an abrupt voltage jump from zero to a finite value appears in both samples at \( I_C \). However, we were unable to resolve any internal structures in the large voltage jump at low temperatures, and smaller bias current steps in the transition region only yield increased fluctuations. The absence of voltage steps in the larger diameter samples at low \( T \) may be attributed to the heating effect at high power levels.\(^{11}\)

Based on the low temperature critical currents and the NW diameters determined by scanning electron microscopy (SEM), the critical current densities of the NbSe\(_2\) NWs are estimated, yielding comparable values for the three high quality NWs \( (1.5 \times 10^6 \text{ A cm}^{-2} \text{ for the 75 nm}, 1.4 \times 10^6 \text{ A cm}^{-2} \text{ for the 250 nm}, \text{ and } 7.8 \times 10^5 \text{ A cm}^{-2} \text{ for the 300 nm samples}) \) and a much lower critical current density for the slightly underconverted NW \( (1.6 \times 10^5 \text{ A cm}^{-2}) \), where the \( I_C \) in the underconverted NW is defined as the current at which the first jump in the \( V-I \) occurs. The critical current densities of our NbSe\(_2\) NWs are consistent with the current-induced phase-slip scenario, in which the nucleation of PSC occurs at the same critical current density for NWs with similar material characteristics.\(^{13}\) The substantially lower critical current density of the underconverted NW can be attributed to its smaller diffusion constant \( D \).\(^{11}\) Even in the high quality samples, the critical current density is still substantially below the critical depairing current density, possibly due to the voltage driven nonequilibrium of the quasiparticles.\(^{36}\) Since \( \Lambda \approx \sqrt{D} \), the quasiparticle diffusion length of the underconverted NbSe\(_2\) NW is also smaller, which may contribute to the larger number of voltage steps observed in its \( V-I \) characteristic. Within the framework of the SBT model, the differential resistance of the linear \( V-I \) region following each voltage jump \( dV/dl \approx 2\Delta \rho /A \), where \( \rho \) and \( A \) are the resistivity and cross section of the sample, respectively.\(^{11}\) Therefore, the differential resistance associated with each PSC is expected to be smaller than the total normal state resistance. However, the differential resistances associated with the two voltage steps in our 75 nm NW are both significantly larger than the total normal resistance of the sample [see Fig. 2(a)]. We do not understand this discrepancy, but speculate that it may be caused by the interactions between the PSCs and the electrodes.

To shed more light on the origin of the stairlike structure (which may be connected to the current-limiting mechanism), a close examination of the \( I-V \) characteristic in the finite voltage region is necessary. In Fig. 4(a), we replotted the data shown in Fig. 2(a) in the form of current versus voltage \( (I-V) \). The sudden increase of the voltage at the critical current and the subsequent entrance to the linear \( I-V \) region is also reminiscent of Josephson tunnel junctions, where the transition to the linear part of the \( I-V \) occurs at a voltage corresponding to twice the lower energy gaps \( (\Delta_1) \), the sum of the two different gaps \( (\Delta_1+\Delta_2) \), and twice the high energy gap \( (2\Delta_2) \). (b) Temperature variations of \( 2\Delta_2 \), and \( \Delta_1+\Delta_2 \) Solid lines are the fits to the BCS theory.
four tunneling processes are expected: $\Delta_1 \rightarrow \Delta_1$, $\Delta_1 \rightarrow \Delta_2$, $\Delta_2 \rightarrow \Delta_1$, and $\Delta_2 \rightarrow \Delta_1$, where $\Delta_1$ and $\Delta_2$ are the small and large gaps, respectively. As a result, three current jumps are expected to occur sequentially at $2\Delta_1/e$, $(\Delta_1+\Delta_2)/e$, and $2\Delta_2/e$ in the quasiparticle tunneling region as the voltage increases. Since the FS sheets associated with $\Delta_2$ carry most of the DOS, $\Delta_2 \rightarrow \Delta_2$ is the dominant tunneling process. While the minijump at $V_1$ disappears quickly above 0.5$T_C$, the minijump at $V_2$ survives until near $T_C$. $eV_2(T)$ follows a BCS behavior with $T_C=6.6$ K and $eV_2(0) \approx 1.2$ meV, suggesting that the minijump at $V_2$ originates from the quasiparticle tunneling between the Se band and the Nb band across the junction: $eV_2=\Delta_1+\Delta_2$. Consequently, the size of the small gap can be determined: $\Delta_1(0) \approx 0.2$ meV. This value agrees with the energy of the minijump at $V_1$ (2$\Delta_1$ = 0.4 meV) for $T=2$ K. Since both $\Delta_1(T)$ and $[\Delta_1(T)+\Delta_2(T)]$ follow the BCS theory and diminish at a nearly identical $T_C$, $\Delta_1(T)$ should also be BCS-like. In general, the multigap equation yields a non-BCS $T$ dependence. However, for DOS$[\Delta_1]/$DOS$[\Delta_2] \approx 0.05$ as the ratio of DOS and 0.2 as the gap ratio, we found from the multigap equation that both gaps indeed have the BCS $T$ dependence.

Additional support for the tunneling between two two-band superconductors can be found in the comparison between the experimentally determined critical current and the theoretical calculations. The critical current of Josephson junctions was theoretically calculated by Ambegaokar and Baratoff. They established for a symmetrical superconductor-insulator-superconductor (SIS) Josephson junction: $I_C R_N = (\pi/2e) \Delta(T) \tanh[\Delta(T)/2k_B T]$, where $R_N$ is the normal state resistance of the junction, $\Delta$ is the energy gap of the superconductor, $e$ is the elementary charge, and $k_B$ is Boltzmann’s constant. For asymmetric junctions ($\Delta_1 \neq \Delta_2$), $I_C(0) R_N = \pi \Delta_1(0) \Delta_2(0)/2e[\Delta_1(0)+\Delta_2(0)]$ at $T=0$ K. At $T=0$, the critical current for each tunneling process is determined by $R_N$ and $\Delta(0)$, where $R_N$ is calculated from the slope ($dV/dI$) of the corresponding critical jump. The critical current of the dominant tunneling process ($\Delta_2 \rightarrow \Delta_2$) is $56 \mu$A; the critical currents for the other processes are approximately 5.9 $\mu$A ($\Delta_1 \rightarrow \Delta_1$) and 5.8 $\mu$A ($\Delta_1 \rightarrow \Delta_2$ and $\Delta_2 \rightarrow \Delta_1$ together). Assuming that these four processes are independent and parallel, the total calculated zero temperature critical current (67.7 $\mu$A) is consistent with the experimental critical current (67 $\mu$A, when it is extrapolated to zero temperature). The linear temperature dependence of the critical current near $T_C$ is also in qualitative agreement with the predictions of Ambegaokar and Baratoff. However, the temperature dependence of the critical current in the medium temperature range below $T_C$ deviates from the Ambegaokar and Baratoff relation, which requires further investigations. While the exact origin of the Josephson tunnel junction is unclear, we speculate that the dc Josephson effect may occur at the boundary between two nanoscale crystalline NbSe$_2$ grains. It was found that the critical current of polycrystalline YBa$_2$Cu$_{3−\delta}$O$_{7−\delta}$ can be described by assuming formation of grain boundary Josephson junctions. As mentioned earlier, grain boundaries may exist in our NWs.

Although we cannot completely rule out the possibility that the quantitative agreement between our data on a selected NbSe$_2$ NW and the multigap tunneling scenario is merely a coincidence, we believe that this scenario is worth being presented as an alternative explanation of the data on the 75 nm NbSe$_2$ NW and merits further investigations. Finally, we point out that multiple SIS-type grain boundary junctions may exist in an individual NbSe$_2$ NW and the critical current (maximum supercurrent) of each junction is inversely proportional to the $R_N$ of the junction. Therefore, it is the junction with the highest $R_N$ that determines the critical current as well as the quasiparticle tunneling characteristics of the NW. Our 75 nm high quality NbSe$_2$ NW device has relatively high extrapolated zero temperature critical current density ($1.5 \times 10^6$ A cm$^{-2}$) and very weak magnetic field dependence (a magnetic field of $B$=0.3 applied perpendicular to the wire axis only slightly suppresses $I_C$), which is not unusual for natural grain boundary junctions. The very weak magnetic field modulation of the critical current in our devices can also be attributed to the small sizes of the junctions. Moreover, the magnetic flux penetrates the entire grains across the barrier, leading to significantly reduced magnetic field difference between the barrier and inside the grains. Unfortunately, the high quality NbSe$_2$ NW was destroyed by electrostatic dissipation before detailed measurements of the magnetic field dependent $I$-$V$ characteristic could be carried out.

IV. CONCLUSIONS

In conclusion, we have investigated the resistance and $V$-$I$ characteristics of individual NbSe$_2$ NWs. Overall, the $V$-$I$ characteristics and critical current density of the NWs can be described by the PSC model. Evidence of TAPS and QPS mechanisms has also been observed in slightly underconverted NbSe$_2$ NWs. A close examination of the temperature dependences of the data on a selected 75 nm NW leads to a possible alternative explanation of the stairlike structure in the $V$-$I$ characteristic and the critical current. Based on the alternative multigap tunneling junction model, two distinct superconducting gaps are extracted from the $I$-$V$ characteristic of the 75 nm sample, and both gaps follow the BCS-like temperature dependence vanishing at a common $T_C$. The extrapolated zero temperature values for the low and high energy gaps (approximately 0.20 and 1.0 meV, respectively) are consistent with previous studies. Although the grain boundary tunnel junction scenario provides a possible explanation of our data obtained from a high quality NbSe$_2$ NW to support the recent theoretical predictions of two distinct energy gaps in NbSe$_2$, further experimental studies are needed to fully understand the nature of the junctions in NbSe$_2$ NWs.
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*Author to whom correspondence should be addressed. Present address: Department of Physics and Astronomy, Wayne State University, Detroit, Michigan 48201. zhous@ornl.gov or zxzhou@wayne.edu


