Quantitative test of a quantum theory for the resistive transition in a superconducting single-walled carbon nanotube bundle

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The phenomenon of superconductivity depends on the coherence of the phase of the superconducting order parameter. The resistive transition in quasi-one-dimensional (quasi-1D) superconductors is broad because of a large phase fluctuation. We show that the resistive transition of a superconducting single-walled carbon nanotube bundle is in quantitative agreement with the Langer-Ambegaokar-McCumber-Halperin (LAMH) theory. We also demonstrate that the resistive transition below \( T^*_c = 0.89T_{c0} \) is simply proportional to \( \exp(\frac{3.9T_c}{T_c^*}(1 - \frac{T}{T_c^*})^{3/2}) \), where the barrier height has the same form as that predicted by the LAMH theory and \( T_{c0} \) is the mean field superconducting transition temperature.

The phenomenon of superconductivity depends on the coherence of the phase of the superconducting order parameter. The phase coherence of the superconducting order parameter leads to the zero-resistance state. For three-dimensional (3D) bulk systems, the transition to the zero-resistance state occurs right below the mean-field superconducting transition temperature \( T_{c0} \) such that the resistive transition is very sharp and the transition width is negligibly small. In contrast, the resistive transition in quasi-one-dimensional (quasi-1D) superconductors is broad because of a large superconducting fluctuation. A quantum theory to describe the resistive transition in quasi-1D superconductors was developed by Langer, Ambegaokar, McCumber and Halperin (LAMH) [1] over 30 years ago. The theory is based on thermally activated phase slips (TAPS), which cause the resistance to decrease to zero exponentially. Experiments to test this theory were done by Lukens et al. [2] and Newbower et al. [3] on tin whiskers. These are single-crystal, cylindrical specimens, typically ~0.5 \( \mu \)m in diameter. The agreement between the data and theory is satisfactory although the specimens are not truly quasi-1D superconductors. Recently, great experimental efforts have been made to fabricate altrathin superconducting wires of amorphous MoGe, whose diameter can be smaller than 10 nm [4,5]. The resistive transitions of the altrathin wires appear to be in good agreement with the LAMH theory [5] although granularity and inhomogeneity may occur in these altrathin wires.

Now a question arises: Can we find a truly quasi-1D superconductor to quantitatively test the LAMH theory? Fortunately, it has been shown that the electronic structure of individual single-walled carbon nanotubes (SWNTs) has 1D nature. The carbon nanotubes can be metallic or semiconducting, depending on their chiralities. The metallic individual carbon nanotubes should be ideal 1D superconductors if there are significant pairing interactions that overcome direct Coulombic interaction between conduction electrons. Bundling these ideal 1D superconductors will lead to the formation of a quasi-1D superconducting wire with a much smaller superconducting fluctuation. Indeed, quasi-1D superconductivity below 0.5 K has been observed in a bundle of single-walled carbon nanotubes, which consists of about 350 tubes with mixed chiralities [6]. Here, we show that the resistive transition of this nanotube bundle is in quantitative agreement with the LAMH theory. We also demonstrate that the resistive transition below \( T^*_c = 0.89T_{c0} \) is simply proportional to \( \exp(\frac{3.9T_c}{T_c^*}(1 - \frac{T}{T_c^*})^{3/2}) \), where the barrier height has the same form as that predicted by the LAMH theory.

In a theory developed by Langer, Ambegaokar, McCumber and Halperin [1], phase slips occur via thermal activation, leading to a finite width for the resistive transition. The resistance due to the TAPS is given by [7]

\[
R_{TA} = \frac{h}{4e^2 k_B T} \hbar\Omega \exp[-\Delta F_0(T)/k_B T],
\]

where the attempt frequency \( \Omega \) is [1]

\[
\Omega = \frac{\sqrt{3}}{2\pi^{3/2}} \left( \frac{L}{\xi} \right)^{1/3} \frac{\Delta F_0(T)}{k_B T} \frac{1}{\tau_{GL}}.
\]

Here \( L \) is the length of the wire, \( \xi(T) \) is the coherence length, and \( \hbar/\tau_{GL} = (8/\pi)k_B(T_{c0} - T) \). The barrier energy \( \Delta F_0(T) \) is

\[
\Delta F_0(T) = \frac{8\sqrt{2}}{3} \frac{H^2_e(T)}{8\pi} A \xi,
\]

where \( H^2_e(T)/8\pi \) is the condensation energy, \( A \) is the cross-section area of the wire, and the critical field near \( T_{c0} \) is given by \( H_c(T) = 1.73H_c(0)(1 - T/T_{c0}) \) within the BCS theory [7]. Using \( \xi(T) = \xi(0)(1 - T/T_{c0})^{-1/2} \) (Ref. [7]), we then have
\[
\frac{\Delta F_\sigma(T)}{k_B T} = \frac{3cT_{c0}}{T}(1 - \frac{T}{T_{c0}})^{3/2},
\]

where
\[
c = \frac{\Delta F_\sigma(0)}{k_B T_{c0}} = \frac{8\sqrt{2}}{3} \frac{H^2(0)}{8\pi k_BT_{c0}} A\xi(0).
\]

Combining the above equations, we finally get
\[
R_{TA} = \frac{m}{T_{c0}}(1 - \frac{T}{T_{c0}})^{9/4} \exp\left[-\frac{3cT_{c0}}{T}(1 - \frac{T}{T_{c0}})^{3/2}\right],
\]

with
\[
m = 2.55T_{c0}(3cT_{c0})^{1/2} \frac{L}{\xi(0)}.
\]

where \(m\) is in the unit of kΩK\(^{3/2}\). We would like to mention that the exponent in Eq. 6 is factor of 3 larger than that in a similar formula deduced in Ref. [5]. It is possible that the authors in Ref. [5] missed the prefactor of 1.73 in \(H_c(T)\).

The condensation energy at zero temperature \(H^2(0)/8\pi\) is equal to \(N(0)\Delta^2(0)/2\) within the BCS theory, where \(N(0)\) is the density of states near the Fermi level and \(\Delta(0)\) is the superconducting gap at zero temperature. For a single metallic SWNT with two transverse channels, \(N(0)A = 4/3\pi a_{C-C}\gamma_0\) (Ref. [8]), \(h\nu_F = 1.5\pi a_{C-C}\gamma_0\) (Ref. [9]), where \(\gamma_0\) is the hopping integral and \(a_{C-C} = 0.142\) nm) is the bonding length. Using the BCS relations: \(\xi_{BCS} = h\nu_F/\pi\Delta(0)\) and \(\Delta(0)/k_BT_{c0} = 1.76\), and the above relations, one can readily show that
\[
c = 0.68 \frac{\xi(0)}{\xi_{BCS}}.
\]

If a bundle of single-walled nanotubes or a multi-walled nanotube consists of \(N_{ch}\) transverse channels, then
\[
c = 0.34N_{ch} \frac{\xi(0)}{\xi_{BCS}}.
\]

For two-probe or four-probe measurements on carbon nanotubes with finite transverse channels, the total resistance is \(R(T) = R_0 + R_{tube}\), where \(R_{tube}\) is the on-tube resistance and \(R_0 = R_t = R_Q/tN_{ch}\) (tunneling resistance) for four-probe measurements, or \(R_0 = R_Q/tN_{ch} + R_c\) for two probe measurements [10]. Here \(t\) is the transmission coefficient \((t \leq 1)\), \(R_Q = h/2e^2 = 12.9\) kΩ is the resistance quantum, and \(R_c\) is the contact resistance. Both \(R_c\) and \(R_t\) should be temperature independent. For ideal contacts, \(R_c = 0\) and \(t = 1\), so \(R_t = 12.9\) kΩ\(/N_{ch}\) for a bundle comprising \(N_{ch}\) transverse channels. For quasi-1D systems, \(N_{ch}\) is always finite such that \(R(T)\) never goes to zero even if the on-tube resistance is zero. Only if \(N_{ch}\) goes to infinity, as in the bulk 3D systems, \(R_t\) becomes zero such that four probe resistance can go to zero below the superconducting transition temperature.

Fig. 1 shows the two-probe resistance data for a SWNT bundle that consists of about 350 tubes [6]. One can see that the resistance starts to drop below about 0.5 K, decreases more rapidly below \(T_{c0} \approx 0.44\) K and saturates to a value of 74 Ω. From the saturated value of \(R_0 = 74\) Ω, and the relation: \(R_0 = R_Q/tN_{ch} + R_c\), one can easily find that more than 174 transverse channels are connected to the electrodes and participate in electrical transport. This implies that more than 87 metallic-chirality superconducting SWNTs take part in electrical transport. Considering the fact that one third of tubes should have metallic chiralities and become superconducting, we find the total number \((N_m)\) of the superconducting tubes to be 117, implying that \(t \geq 0.74\). The value of \(N_m\) can be also deduced from the measured current \(I_c^*\) at which the last resistance jump occurs. The \(I_c^*\) corresponds to the critical current for a superconducting wire without disorder and with the same number of the transverse channels [6]. For metallic chirality superconducting carbon nanotubes, one can easily deduce that [11] \(I_c^* = 7.04k_BT_{c0}N_m/eR_Q\) with \(I_c^* = 2.4\) μA (Ref. [6]) and \(T_{c0} = 0.44\) K, we have \(N_m = 116\), in remarkably good agreement with the value deduced above.

![Fig. 1. The temperature dependence of the two-probe resistance for a SWNT bundle that consists of about 350 tubes. The data are extracted from Ref. [6].](image)

In Fig. 2, we fit the resistance data below 0.88\(T_{c0}\) by
\[
R = 74 + \alpha \exp\left[-\frac{3\beta I^*_c}{T}(1 - \frac{T}{T_c})^{3/2}\right].
\]

Here the first term is the sum of the tunneling and contact resistances discussed above, and the second term is the on-tube resistance which has a similar exponential dependence on \(T\) as Eq. 6 but with a temperature independent prefactor. We can see that the fit is excellent with the fitting parameter \(\beta = 2.99\pm0.05\) and \(T_c^* = 0.394\pm0.002\) K. Reducing or increasing the temperature region for the fit tends to worsen the fit quality. There-
fore, the on-tube resistance goes to zero exponentially below $T_c^* = 0.89 T_{o,0}$. The microscopic origin of this simple exponential dependence up to $0.89 T_{o,0}$ is not clear, so we consider Eq. 10 only as an empirical formula.

We also try to fit the resistance below $0.88 T_{o,0}$ by

$$R = 74 + \frac{m}{T} \left( 1 - \frac{T}{T_{o,0}} \right) \frac{3}{4} \exp \left[ - \frac{3 c T_{o,0}}{T} \left( 1 - \frac{T}{T_{o,0}} \right)^{3/2} \right].$$

(11)

Here the second term is the on-tube resistance which is the same as Eq. 6 predicted by the LAMH theory. We find that the fit is not good and the fitting parameters have no quantitative agreement with the LAMH theory. This is because the LAMH theory is only applied to the temperature region where the barrier height is far greater than $k_B T$ so that current carrying states involved are truly metastable [12]. The estimated region of validity for the LAMH theory is below $0.07 R_N$ for dirty wires where $l << \xi_{BCS}$ (Ref. [12]). The condition of $l << \xi_{BCS}$ is well satisfied in the SWNT bundle (see text).

From the values of $m$, $c$, and $T_{o,0}$, we can evaluate the zero-temperature coherence length $\xi(0)$ using Eq. 7. Substituting $m = 26.6 \text{k} \Omega \text{K}^{1.5}$, $c = 3.08$, $T_{o,0} = 0.44 \text{K}$, and $L = 10000 \text{Å}$ into Eq. 7, we obtain $\xi(0) = 850 \text{Å}$. From the measured $R_N(0.25 K) / L = 12 \text{k} \Omega / \mu \text{m}$ (Ref. [6]) and the relation $R_N / L = R_Q / 2 N_m l$ (Ref. [13]), we can calculate the mean free path $l$ at 0.25 K. With $N_m = 117$, we get $l = 46 \text{Å}$. Substituting $l = 46 \text{Å}$ and $\xi(0) = 850 \text{Å}$ into the dirty-limit formula [12]: $\xi(0) = 0.85 \sqrt{\xi_{BCS}^*}$, we have $\xi_{BCS} = 21739 \text{Å}$. With $\xi_{BCS} = 21739 \text{Å}$, $\xi(0) = 850 \text{Å}$, and $N_{ch} = 2 N_m = 334$, we calculate $c = 3.11$ from Eq. 9. It is remarkable that the calculated value of $c$ from Eq. 9 is in quantitative agreement with the value (3.08) deduced from the fitting.

We can also estimate the Fermi velocity $v_F$ from the deduced value of $\xi_{BCS}$ and the formula $\xi_{BCS} = 0.18 h v_F / k_B T_{o,0}$. With $\xi_{BCS} = 21739 \text{Å}$ and $T_{o,0} = 0.44 \text{K}$, we get $h v_F = 4.6 \text{eV}$. Then we estimate $\gamma_o = 2.16 \text{eV}$ from $h v_F = 1.5 a_C - c \gamma_o$. This value is very close to an independent estimate (2.26 eV) from the measured semiconducting gap for a $d = 1.34 \text{nm}$ SWNT [14].

The deduced value of $\xi(0) = 850 \text{Å}$ is also in excellent agreement with the measured critical current $I_c$ for this SWNT bundle. For a diffusive superconducting wire, the critical current $I_c$ is given by [15]

$$I_c = \frac{\Delta(0)}{e R_N \xi(0)}.$$

(12)

Using $\Delta(0) = 1.76 k_B T_{o,0}$ and substituting $R_N(0.1 K) / L = 12.5 \text{k} \Omega / \mu \text{m}$ (Ref. [6]) and $\xi(0) = 850 \text{Å}$ into Eq. 12, we find $I_c = 62.4 \text{nA}$, which is very close to the measured value (62 nA) at 0.1 K (Ref. [6]).
In summary, we have shown that the observed resistive transition of a superconducting carbon nanotube bundle is in quantitative agreement with the LAMH theory. We have also demonstrated that the resistive transition below $T^*_c = 0.90 T_c$ is simply proportional to $\exp(-\frac{3\sqrt{T^*_c}}{T} (1 - \frac{T}{T^*_c})^{3/2})$, where the barrier height has the same form as that predicted by the LAMH theory.

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