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 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 $\exp[-(3\beta T_c^*/T)(1-T/T_c^*)^{3/2}]$, 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.

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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, there should be a finite width in the resistive transition in quasi-one-dimensional (quasi-1D) superconductors with a finite number of transverse channels. An important quantum theory to describe the resistive transition in quasi-1D superconductors was developed by Langer, Ambegaokar, McCumber, and Halperin (LAMH) over 30 years ago.[1] 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 μm in diameter. The agreement between the data and theory seems satisfactory although the specimens are not truly quasi-1D superconductors. Recently, great experimental efforts have been made to fabricate ultrathin superconducting wires of amorphous MoGe, whose diameter can be smaller than 10 nm.[4,5] Although there are still several thousand transverse conduction channels in these ultrathin wires, the resistive transitions appear to agree with a model that includes both thermally activated phase slips close to T_{c0} and quantum phase slips at low temperatures.[5]

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] The number of transverse conduction channels of the bundle[6] is around 200, which is significantly lower than that for the ultrathin wires of amorphous MoGe.[4,5] 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 $\exp[-(3\beta T_c^*/T)(1-T/T_c^*)^{3/2}]$, where the barrier height has the same form as that predicted by the LAMH theory.

In the 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]

$$R_{TA} = \frac{h}{4e^2}\frac{\hbar\Omega}{k_BT}\exp[-\Delta F_o(T)/k_BT], \qquad (1)$$

where the attempt frequency Ω is[1]

$$\Omega = \frac{\sqrt{3}}{2\pi^{3/2}}\frac{L}{\xi}\sqrt{\frac{\Delta F_o(T)}{k_BT}}\frac{1}{\tau_{GL}}. \qquad (2)$$

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 $\Delta F_o(T)$ is

$$\Delta F_o(T) = \frac{8\sqrt{2}}{3}\frac{H_c^2(T)}{8\pi}A\xi, \qquad (3)$$

where $H_c^2(T)/8\pi$ 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 $\xi(T)=\xi(0)(1-T/T_{c0})^{-1/2}$,[7] we then have

$$\frac{\Delta F_o(T)}{k_BT} = \frac{3cT_{c0}}{T}\left(1-\frac{T}{T_{c0}}\right)^{3/2}, \qquad (4)$$

where

$$c = \frac{\Delta F_o(0)}{k_BT_{c0}} = \frac{8\sqrt{2}}{3}\frac{H_c^2(0)}{8\pi k_BT_{c0}}A\xi(0). \qquad (5)$$

Combining the above equations, we finally get

$$R_{TA} = \frac{m}{T^{1.5}}\left(1 - \frac{T}{T_{c0}}\right)^{9/4} \exp\left[-\frac{3cT_{c0}}{T}\left(1 - \frac{T}{T_{c0}}\right)^{3/2}\right], \quad (6)$$

with

$$m = 2.55T_{c0}(3cT_{c0})^{1/2}\frac{L}{\xi(0)}, \quad (7)$$

where m is in units of $k\Omega\ \mathrm{K}^{3/2}$. We would like to mention that the exponent in Eq. (6) is a 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, $H_c^2(0)/8\pi$, 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, $N(0)A = 4/3\pi a_{\text{C-C}}\gamma_o$,[8] $\hbar v_F = 1.5a_{\text{C-C}}\gamma_o$,[9] where γ_o is the hopping integral and $a_{\text{C-C}}$ (0.142 nm) is the bonding length. Using the BCS relations $\xi_{BCS} = \hbar v_F/\pi\Delta(0)$ and $\Delta(0)/k_BT_{c0} = 1.76$, and the above relations, one can readily show that

$$c = 0.68\frac{\xi(0)}{\xi_{BCS}}. \quad (8)$$

If a bundle of single-walled nanotubes or a multiwalled nanotube consists of N_{ch} transverse channels, then

$$c = 0.34N_{ch}\frac{\xi(0)}{\xi_{BCS}}. \quad (9)$$

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 ($t \leqslant 1$), $R_Q = h/2e^2 = 12.9\ \mathrm{k}\Omega$ 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 $R_t = 12.9\ \mathrm{k}\Omega/N_{ch}$ 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 bulk 3D systems, does R_t become zero such that the four-probe resistance can go to zero below the superconducting transition temperature.

Figure 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 $T_{c0} \simeq 0.44$ K and saturates to a value of 74 Ω. From the saturated value of $R_0 = 74\ \Omega$, 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 the tubes should have metallic chiralities and become superconducting, we find the total number (N_m) of the superconducting tubes to be 117, implying that $t \geqslant 0.74$.

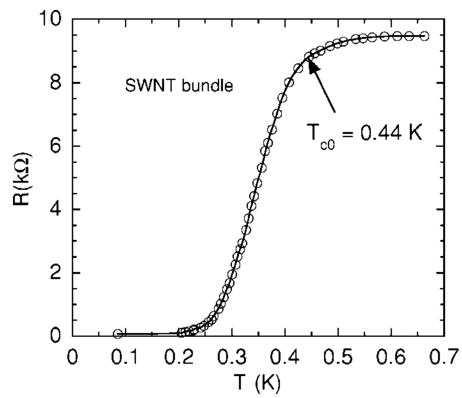

FIG. 1. The temperature dependence of the two-probe resistance for a SWNT bundle that consists of about 350 tubes. The data are extracted from Ref. 6.

The value of N_m can also be 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 transverse channels.[6] According to the BCS theory, the mean-field critical current in the clean limit is given by[7] $I_c^*(0) = en_sA\Delta(0)/\hbar k_F$. The superfluid density n_s is equal to the normal-state carrier density n which is given by $n = 2N_mN(0)E_F = 2N_mN(0)\hbar v_Fk_F = 4N_mk_F/A\pi$. Here we have used the relations for a single metallic tube: $N(0)A = 4/3\pi a_{\text{C-C}}\gamma_o$, $\hbar v_F = 1.5a_{\text{C-C}}\gamma_o$, and $E = \hbar v_F|k|$. Using $\Delta(0)/k_BT_{c0} = 1.76$, we finally get $I_c^* = 7.04k_BT_{c0}N_m/eR_Q$. With $I_c^* = 2.4\ \mu$A (Ref. 6) and $T_{c0} = 0.44$ K, we have $N_m = 116$, in remarkably good agreement with the value deduced above.

In Fig. 2, we fitted the resistance data below $0.88T_{c0}$ by

$$R = R_0 + \alpha \exp\left[-\frac{3\beta T_c^*}{T}\left(1 - \frac{T}{T_c^*}\right)^{3/2}\right]. \quad (10)$$

Here the first term is the sum of the tunneling and contact resistances discussed above, and the second term is the on-

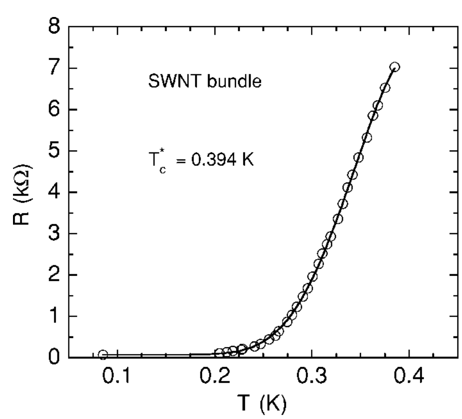

FIG. 2. The temperature dependence of the two-probe resistance for a SWNT bundle below $0.88T_{c0}$. The solid line is the curve best fitted by Eq. (10) with $\beta = 2.99 \pm 0.05$ and $T_c^* = 0.394 \pm 0.002$ K. Reducing or increasing the temperature region for the fit tends to worsen the fit quality. It is striking that the on-tube resistance below $0.88T_{c0}$ decreases exponentially to zero.

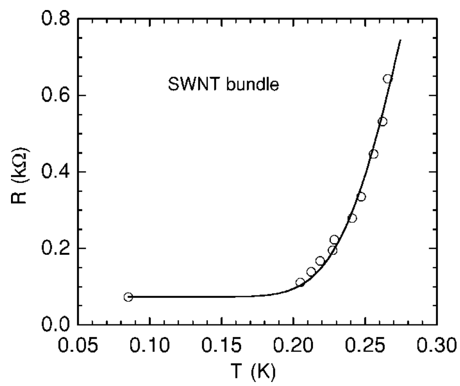

FIG. 3. The temperature dependence of the two-probe resistance for a SWNT bundle below $0.06R_N$. The solid line is the curve fitted to the data below $0.06R_N$ by Eq. (11) predicted by the LAMH theory. The estimated region of validity for the LAMH theory is below $0.07R_N$ for dirty wires where $l \ll \xi_{BCS}$ (Ref. 11). The condition of $l \ll \xi_{BCS}$ is well satisfied in the SWNT bundle (see text).

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 $\beta = 2.99 \pm 0.05$ and $T_c^* = 0.394 \pm 0.002$ K. Reducing or increasing the temperature region for the fit tends to worsen the fit quality. Therefore, the on-tube resistance goes to zero exponentially below $T_c^* = 0.89T_{c0}$. The microscopic origin of this simple exponential dependence up to $0.89T_{c0}$ is not clear, so we consider Eq. (10) only as an empirical formula.

We also tried to fit the resistance below $0.88T_{c0}$ by

$$R = R_0 + \frac{m}{T^{1.5}}\left(1 - \frac{T}{T_{c0}}\right)^{9/4} \exp\left[-\frac{3cT_{c0}}{T}\left(1 - \frac{T}{T_{c0}}\right)^{3/2}\right]. \tag{11}$$

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 applies only to the temperature region where the barrier height is far larger than k_BT so that the current-carrying states involved are truly metastable.[11] The estimated region of validity for the LAMH theory is below $0.07R_N$ for dirty wires where the mean free path $l \ll \xi_{BCS}$.[11] For the SWNT bundle, the condition of $l \ll \xi_{BCS}$ is well satisfied, as seen below. In Fig. 3, we fitted the resistance data below $0.06R_N$ by Eq. (11). Here we have ignored the normal conduction because R_{TA} is a factor of about 20 smaller than R_N. We have also neglected the finite resistance R_{QPS} caused by quantum phase slips.[12] Because this resistance is small and has a much weaker temperature dependence,[12] we cannot differentiate between the contact resistance and R_{QPS} in a narrow temperature region.

One can see that the fitting is very good with the fitting parameters $m = 26.6 \pm 4.7$ kΩ K$^{1.5}$ and $c = 3.08 \pm 0.13$. It is remarkable that the value of c is nearly the same as the value of β (2.99 ± 0.05) deduced above from a simple exponential fit [Eq. (10)]. We will see below that the fitting parameters c and m are in quantitative agreement with the LAMH theory.

From the values of m, c, and T_{c0}, we can evaluate the zero-temperature coherence length $\xi(0)$ using Eq. (7). Substituting $m = 26.6$ kΩ K$^{1.5}$, $c = 3.08$, $T_{c0} = 0.44$ K, and $L = 10\,000$ Å into Eq. (7), we obtain $\xi(0) = 850$ Å. From the measured $R_N(0.25K)/L = 12$ kΩ/μm (Ref. 6) and the relation $R_N/L = R_Q/2N_m l$,[13] we can calculate the mean free path l at 0.25 K. With $N_m = 117$, we get $l = 46$ Å. Substituting $l = 46$ Å and $\xi(0) = 850$ Å into the dirty-limit formula[11] $\xi(0) = 0.85\sqrt{\xi_{BCS} l}$, we have $\xi_{BCS} = 21\,739$ Å. With $\xi_{BCS} = 21\,739$ Å, $\xi(0) = 850$ Å, and $N_{ch} = 2N_m = 334$, 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 ξ_{BCS} and the formula $\xi_{BCS} = 0.18\hbar v_F / k_B T_{c0}$. With $\xi_{BCS} = 21\,739$ Å and $T_{c0} = 0.44$ K, we get $\hbar v_F = 4.6$ eV Å. Then we estimate $\gamma_o = 2.16$ eV from $\hbar v_F = 1.5 a_{\text{C-C}} \gamma_o$. This value is very close to an independent estimate (2.26 eV) from the measured semiconducting gap for a $d = 1.34$ nm SWNT.[14]

The deduced value of $\xi(0) = 850$ 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]

$$I_c = \frac{\Delta(0)}{eR_N}\frac{L}{\xi(0)}. \tag{12}$$

Using $\Delta(0) = 1.76k_BT_{c0}$ and substituting $R_N(0.1\text{ K})/L = 12.5$ kΩ/μm (Ref. 6) and $\xi(0) = 850$ Å into Eq. (12), we find $I_c = 62.4$ nA, which is very close to the measured value (62 nA) at 0.1 K.[6]

We would like to mention that, in deriving all the formulas above, we have used the BCS weak-coupling theory. The validity of the use of the BCS weak-coupling theory might be questionable considering that the system may exhibit Luttinger liquid physics in the normal state.[6] However, the quantitative agreement between the data and theory suggests that the mean-field BCS weak-coupling theory should be valid for this material. This may also imply that the Josephson coupling among the tubes should be strong enough to lead to a mean-field transition.

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 $T_c^* = 0.90T_{c0}$ is simply proportional to $\exp[-(3\beta T_c^*/T)(1 - T/T_c^*)^{3/2}]$, where the barrier height has the same form as that predicted by the LAMH theory.

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