Consistency of Superconducting Correlations with One-Dimensional Electron Interactions in Carbon Nanotubes

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We show that a model of interacting electrons in one dimension is able to explain the order of magnitude as well as the temperature dependence of the critical supercurrents recently measured in nanotube samples placed between superconducting contacts. We use bosonization methods to deal with the long-range Coulomb interaction, ending up with a picture in which the critical current does not follow the temperature dependence of the gap in the contacts. Our results also reveal the presence of a short-range attractive interaction in the nanotubes, which accounts for a significant enhancement of the critical supercurrents.

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Since their discovery, carbon nanotubes have offered a great potential for novel electronic properties and technological applications. The theoretical prediction that there should be semiconducting as well as metallic nanotubes [1] has been checked experimentally [2]. Also remarkable has been the experimental observation of unconventional transport properties [3], that seem to be compatible with the expected Luttinger liquid behavior of one-dimensional electron systems [4]. Different approaches have predicted the appearance of phases with broken symmetry in the carbon nanotubes at very low energies [5–8]. Anyhow, the estimates are in general that there should be enough margin to observe the characteristic scaling behavior of the Luttinger liquid over a wide range of temperatures.

A different class of experiments has been aimed to test the superconducting properties of the carbon nanotubes [9,10]. One of the most striking results has been the observation of supercurrents along carbon nanotubes placed between superconducting contacts [9]. In a sample made of a single-walled nanotube, for instance, critical supercurrents have been measured that are about 40 times higher than expected from the value of the gap in the contacts [9]. They also show a very flat dependence with temperature, until the critical value of the superconducting contacts is approached. In that respect, there is a marked difference from the behavior of another sample made of a rope of nanotubes, where the critical supercurrent seems to follow the BCS gap in a certain range of low temperature [9].

A model of the electron interaction in the carbon nanotubes should give a quantitative account of all these different observations of superconducting correlations. We show in this Letter that the mentioned features of the critical supercurrent can be understood in the framework of a one-dimensional theory of interacting electrons. We will see that the experimental data are consistent with a definite form of the one-dimensional interaction, as it is actually nontrivial to reproduce both the shape and the order of magnitude of the supercurrents in the single-walled nanotube and in the rope of nanotubes. In particular, the experimental values of the supercurrent point to a sensible renormalization of the strength of the long-range Coulomb interaction, especially in the sample made of a rope of nanotubes, in agreement with earlier theoretical predictions [11,12].

A metallic single-walled nanotube has several one-dimensional subbands, with two pairs of linear branches crossing at Fermi points k_F and $-k_F$. We deal in this Letter with an effective description of the nanotubes for energies below the scale E_c at which all the gapped subbands decouple in the computation of low-energy properties, so that the relevant modes left belong to the linear branches close to the Fermi level. We can estimate this energy E_c as a few tenths of eV, for a typical single-walled nanotube with about 10 subbands.

The low-energy excitations can be encoded into four boson fields, each boson corresponding to a linear branch in the same fashion as in the Luttinger model [4]. The Hamiltonian of the effective theory can be written in terms of the respective density operators $\rho_{ia\sigma}$, labeled by the Fermi point $a = 1, 2$ and by the chirality $i = L, R$,

$$H = \frac{1}{2} v_F \int_{-k_c}^{k_c} dk \sum_{ia\sigma} :\rho_{ia\sigma}(k)\rho_{ia\sigma}(-k): + \frac{1}{2} \int_{-k_c}^{k_c} dk \sum_{ia\sigma} \rho_{ia\sigma}(k) V(k) \sum_{jb\sigma'} \rho_{jb\sigma'}(-k) . \quad (1)$$

In the above expression, k_c is related to E_c through the Fermi velocity v_F, $k_c = E_c/v_F$.

Our assumption regarding the interaction will be the presence of the long-range Coulomb interaction $V(k) \approx e^2/(4\pi^2) \log|(k_c + k)/k|$ [13], which remains unscreened in one spatial dimension [14], plus an additional short-range effective attraction coming from the coupling to the elastic modes of the nanotube. In this framework, we are neglecting backscattering and umklapp processes that mix different chiralities and Fermi points, relying on the fact that those interactions have smaller relative

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strength ($\sim 0.1/n$, in terms of the number n of subbands [7,15]) and they remain small down to extremely low energies [7].

The correlators in the model governed by (1) can be computed by changing variables to the total charge density operators $\rho_i(k) = (1/\sqrt{N})\sum_{a\sigma} \rho_{ia\sigma}(k)$, $i = L, R$, where N stands in general for the number of channels $\{(a, \sigma)\}$, so that $N = 4$ in the case of a single-walled nanotube.

A typical propagator of Cooper pairs, for instance, becomes

$$G(x,t) \equiv \langle \Psi_{L1\uparrow}(x,t)\Psi_{R2\downarrow}(x,t)\Psi^+_{L1\uparrow}(0,0)\Psi^+_{R2\downarrow}(0,0)\rangle = C(x,t)F(x,t)\,, \quad (2)$$

where F is the part that does not depend on the interaction and C corresponds to the propagation of the total charge. At zero temperature, for instance, we have

$$C(x,t) = \exp\biggl(-\frac{2}{N}\int_0^{k_c} dk\,\frac{1}{\mu(k)k} \times [1 - \cos(kx)\cos(\tilde{v}_F kt)]\biggr), \quad (3)$$

where $\mu(k) = 1/\sqrt{1 + 2NV(k)/v_F}$ and $\tilde{v}_F = v_F/\mu(k)$. The other factor has the simple dependence

$$F(x,t) = 1/|k_c^2(x - v_F t)(x + v_F t)|^{(N-1)/N}. \quad (4)$$

The critical supercurrent I can be estimated under the assumption that (i) the normal-superconductor junctions are perfectly transmitting [16], or (ii) the single-particle scattering is relevant at the interfaces [17]. The latter is more realistic for the experiments that we are considering. The distance L between the superconducting contacts is large enough that I can be expressed as a function of L and the temperature T as

$$I_L(T) = ev_F k_c \int_0^{1/T} d\tau\, G(L, -i\tau)\,. \quad (5)$$

In the above equation, G stands for the appropriate expression at finite temperature. The analytic continuation to imaginary time, however, cannot be taken directly in expressions such as (3), and for computational purposes it is more convenient to introduce the temperature dependence through the Matsubara formalism

$$C(x, -i\tau) = \exp\biggl[-\frac{2}{N}\int_0^{k_c} dk\,\frac{2T}{v_F}\sum_{m=-\infty}^{m=+\infty}\frac{1 - \cos(kx)\cos(2\pi mT\tau)}{(2\pi mT/\tilde{v}_F)^2 + k^2}\biggr]. \quad (6)$$

We can use Eq. (5) to test whether the behavior of the critical currents measured in Ref. [9] can be reproduced within the present framework. The comparison should be fairly direct for the sample that is made at one end of a single nanotube (called ST_1 in Ref. [9]). According to the above discussion, we consider a momentum-dependent parameter $\mu(k) = 1/\sqrt{1 + N[e^2/(2\pi^2 v_F)\log|(k_c + k)/k| - g/(\pi v_F)]}$, taking in this case a number of channels $N = 4$ in the above equations.

We have checked first that the critical current $I \approx 0.1\ \mu$A of the ST_1 sample at $T \approx 0$ K, found anomalously high in the BCS framework, can be explained with the present model. The results represented in Fig. 1 show the magnitude of $I_L(0)$ for different values of the interaction, the distance being measured in units of k_c^{-1}. The actual values of the supercurrent are obtained by multiplying the magnitudes in Fig. 1 by the prefactor in Eq. (5). The Fermi velocity can be obtained from the hopping amplitude $t \approx 2.1$ eV and the nearest-neighbor distance $a \approx 1.4$ Å, by using the expression $v_F = 3ta/2$. A reasonable estimate of the cutoff k_c for the single-walled nanotube is $k_c \approx 0.5\ \text{nm}^{-1}$, which gives $ev_F k_c \approx 30\ \mu$A.

We observe that the coupling corresponding to the bare parameters of a graphite layer, $2e^2/(\pi^2 v_F) \approx 8.0$, does not lead to sensible results. The total length of the sample ST_1 is $\approx$300 nm, and we should expect to get the correct order of magnitude of the critical current at $L \sim 50/k_c$. The most appropriate value for the Coulomb interaction seems to be $2e^2/(\pi^2 v_F) \approx 1.0$. The order of magnitude $I \approx 0.1\ \mu$A is then reached, most precisely if one takes into account a coupling for the short-range attractive interaction $g/(\pi v_F) \sim 0.2$. The sensible reduction in the value of $2e^2/(\pi^2 v_F)$ can be understood by the presence of nearby charges and the renormalization of the interaction

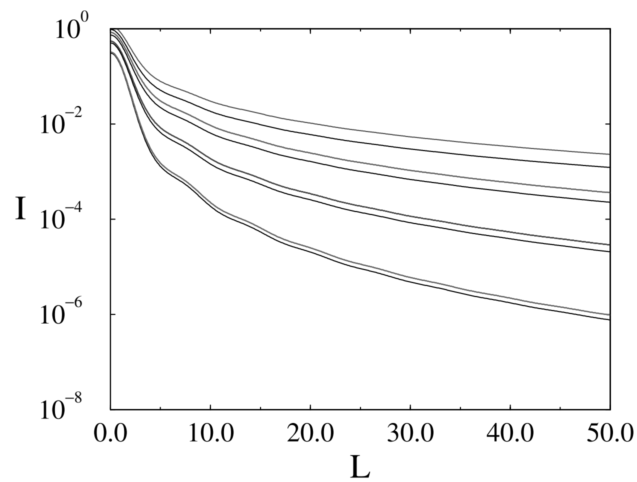

FIG. 1. Plots of the critical current (in units of $ev_F k_c$) versus distance, at $T = 0$, for different strengths of the Coulomb interaction. From top to bottom, the solid curves correspond to $2e^2/(\pi^2 v_F) = 1.0, 2.0, 4.0, 8.0$. The dotted lines correspond in each case to the correction by effect of the additional short-range interaction, with $4g/(\pi v_F) = 0.8$.

in the narrow rope into which the nanotube merges, as we discuss afterwards.

Moreover, the dependence of the critical current on T for the mentioned interaction strength reproduces the shape that has been observed in the measurements of the sample ST_1. In the theoretical model, the temperature T is given in units of the unique energy scale E_c. This means that the critical temperature $T_c \approx 0.4$ K of the contacts for the sample ST_1 corresponds to the dimensionless value $T_c/E_c \approx 2 \times 10^{-4}$. We have represented in Fig. 2 the results for the critical current $I_L(T)$ at $L = 50/k_c$, with and without the effect of the short-range attractive interaction. It is remarkable the smooth behavior of the critical current in the low-temperature regime below T_c. The slight increase observed near zero temperature is related to the renormalization of the transmission at the interfaces [18]. Near T_c, the suppression of superconductivity in the contacts should be incorporated to produce a sharp decrease, leading then to the full agreement with the experimental results of Ref. [9].

Moving now to the sample made of a rope of nanotubes (called $R0_3$ in Ref. [9]), we have to take into account two main differences with respect to the preceding discussion. First, the energy scale E_c up to which the rope can be seen as a purely one-dimensional system is smaller, compared to the cutoff introduced for the sample ST_1. Given that the diameter of $R0_3$ can be approximately 15 times larger than that of the single-walled nanotube of ST_1, we may assume that the energy cutoffs in the two samples differ in the inverse proportion by a factor of 15. This means that a given temperature of the sample $R0_3$, when measured in units of the corresponding E_c, looks comparatively higher than the same temperature in the sample ST_1. Thus, the critical temperature $T_c \approx 1.1$ K of the contacts used for the sample $R0_3$ gives a dimensionless value $T_c/E_c \approx 9 \times 10^{-3}$, which is more than 1 order of magnitude higher than the ratio for the sample ST_1.

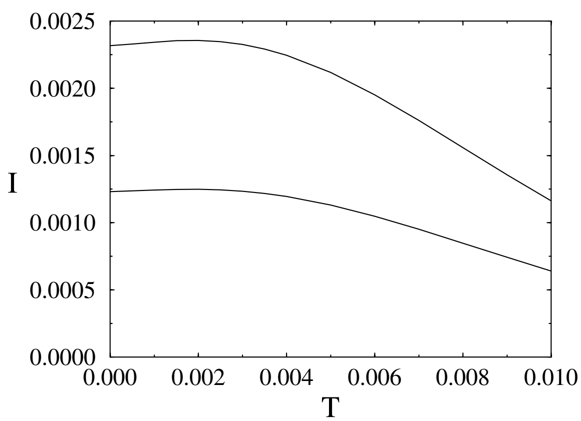

FIG. 2. Plots of the critical current (in units of $ev_F k_c$) versus T/E_c, for $2e^2/(\pi^2 v_F) = 1.0$ and a coupling of the short-range attractive interaction $4g/(\pi v_F) = 0$ (lower curve) and 0.8 (upper curve).

The second important difference between the rope of nanotubes $R0_3$ and the sample ST_1 is the interaction among the large number of nanotubes ($\approx$200) in the former [19], which leads to a significant renormalization of the strength of the Coulomb interaction. This can be understood in the bosonization approach developed above, if we consider that each metallic nanotube in the rope contributes with four units to the number N, while there is only one channel for the interaction of the total charge density [20]. The picture is more involved considering the whole number of nanotubes in the rope, but the overall physical effect can be taken into account by assuming a convenient renormalization of the bare coupling. It turns out, for instance, that the correlations in an aggregate of 100 metallic nanotubes with a bare coupling $2e^2/(\pi^2 v_F) = 1.0$ can be reproduced in a system of decoupled nanotubes in which the interaction has been renormalized down to 0.2, as observed in Fig. 3. On the other hand, the attractive coupling g is not affected by this kind of renormalization, as it refers to an interaction with the elastic modes that takes place within each nanotube.

Given that the total length of the sample $R0_3$ is $\approx$1.7 μm, we have estimated the supercurrent by the decay of I_L through a distance $L = 50/k_c \approx 1.5$ μm. The evaluation of the prefactor in front of Eq. (5) is now more delicate, compared to that for the sample ST_1. On the one hand, we have to bear in mind that the value of k_c decreases according to the increase in the diameter of the sample. On the other hand, there are more metallic nanotubes in the sample $R0_3$, in a number that may be estimated as $1/3$ of the total number, which gives $\approx$60 metallic nanotubes. Balancing both points, it is appropriate to take now a prefactor in Eq. (5) that is four times the value for the single-walled nanotube.

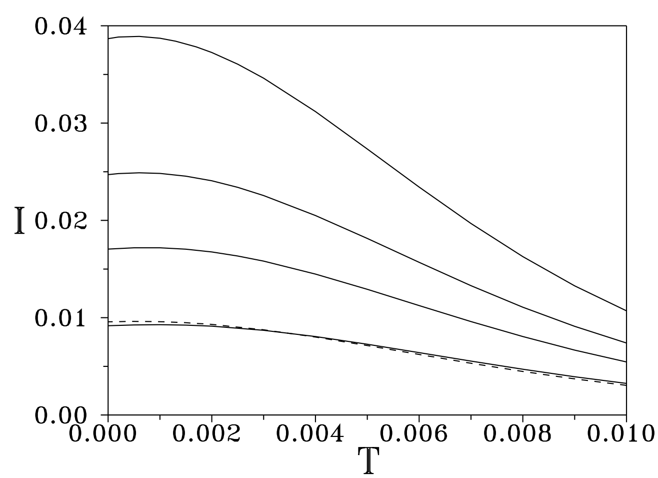

FIG. 3. Plots of the critical current (in units of $ev_F k_c$) versus T/E_c for a renormalized interaction $2e^2/(\pi^2 v_F) = 0.2$ corrected within each tube by the additional short-range attraction with $4g/(\pi v_F) = 1.0$, 0.75, 0.5, and 0 (solid lines from top to bottom), and for an aggregate of 100 metallic nanotubes interacting with strength $2e^2/(\pi^2 v_F) = 1.0$ (dashed line).

We show in Fig. 3 the plots of $I_L(T)$, within the above picture of renormalization of the Coulomb interaction and including the short-range attractive interaction in each nanotube. The critical current in the sample $R0_3$ at $T \approx 0$ K is $I \approx 2.5\ \mu$A. We observe that the correct order of magnitude can be obtained from our results by considering a renormalization of the coupling $2e^2/(\pi^2 v_F)$ down to a value $\approx$0.2, together with the effect of a weak short-range attractive interaction with coupling $g/(\pi v_F) \sim 0.15$.

The value of the attractive coupling g is similar to that required for the sample ST_1, and it actually matches what is expected from the coupling to the elastic modes of the nanotube. The short-range effective attraction can be estimated on theoretical grounds from the modulation of the hopping $t' = \partial t/\partial a \approx 4.2$ eV Å^{-1}, the speed of sound $v_s \approx 2.1 \times 10^4$ ms^{-1}, and the mass M of the atoms. This gives $g/v_F \sim t'^2 a^3/(M v_s^2 v_F) \sim 0.2$, which is of the same order of magnitude needed in our fits.

As a final check of the consistency of our approach, we observe that the curves in Fig. 3 reproduce the dependence on temperature measured experimentally in the sample $R0_3$, with the characteristic inflection point and the very slow decay around the critical temperature at $T \sim 10^{-2} E_c$ [9].

To summarize, we have seen that a model of interacting electrons in one dimension is able to explain the order of magnitude as well as the temperature dependence of the critical currents in both the ST_1 and the $R0_3$ samples of Ref. [9]. Our description is free of the shortcomings arising from the conventional picture of the proximity effect, which relates the value of the critical supercurrent to the gap Δ and the normal resistance R through the expression $I = \pi\Delta/(eR)$. Our approach focuses on the strong correlations in the one-dimensional electron system, explaining in this way why the experimental data of the supercurrent do not follow in general the temperature dependence of the gap in the superconducting contacts.

Our discussion also stresses the relevance of the coupling to the elastic modes of the nanotube, which reveals itself through the presence of a short-range attractive electron interaction. This is also supported by recent experiments on the intrinsic superconductivity of ropes of nanotubes [21]. In a sample such as $R0_3$, it can be already observed that the supercurrent measured experimentally does not vanish near T_c, which is at odds with the conventional picture of the proximity effect but in accordance with the results of our model. This enhancement of the superconducting correlations should deserve further study, in order to understand the experimental conditions under which the effect of the short-range attraction may dominate over the Coulomb repulsion.

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