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The influence of dimensionality on superconductivity in carbon nanotubes

S Bellucci[1], **M Cini**[1,2], **P Onorato**[1,3] **and E Perfetto**[4]

[1] INFN, Laboratori Nazionali di Frascati, PO Box 13, 00044 Frascati, Italy
[2] Dipartimento di Fisica, Università di Roma Tor Vergata, Via della Ricerca Scientifica 1 00133, Roma, Italy
[3] Department of Physics 'A. Volta', University of Pavia, Via Bassi 6, I-27100 Pavia, Italy
[4] Consorzio Nazionale Interuniversitario per Le Scienze Fisiche della Materia, Università di Roma Tor Vergata, Via della Ricerca Scientifica 1 00133, Roma, Italy

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Abstract
We investigate the electronic instabilities in carbon nanotubes (CNs), looking for the breakdown of the one-dimensional Luttinger liquid regime due to strong screening of the long-range part of the Coulomb repulsion. We show that such a breakdown is realized in both ultra-small single walled CNs and multi-walled CNs, while a purely electronic mechanism could explain the superconductivity (SC) observed recently in ultra-small (diameter ∼0.4 nm) single-walled CNs ($T_c \sim 15$ K) and entirely end-bonded multi-walled ones ($T_c \sim 12$ K). We show that both the doping and the screening of the long-range part of the electron–electron repulsion, needed to allow the SC phase, are related to the intrinsically 3D nature of the environment where the CNs operate.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recent progresses in nanotechnology has allowed for a detailed study of the transport properties of 1D electron systems. The discovery of carbon nanotubes (CNs) in 1991 [1], as a by-product of carbon fullerene production, opened a new field of research in mesoscopic physics [2] especially because of their potential application to nanoelectronic devices. It is known that two types of CNs, i.e. single-walled carbon nanotubes (SWNTs) and multi-walled carbon nanotubes (MWNTs) (see figure 1), exist and are reported to display different electronic properties depending on their diameter and on the helicity of the carbon rings around the tubule [3]. Because of their sizes, CNs usually behave as ideal one-dimensional (1D) electronic systems and there have been many experiments showing the existence of superconducting (SC) correlations in these devices at low temperatures.

SC behaviour in low-dimensional systems is quite interesting, since 40 years ago Mermin and Wagner [4] proved a famous theorem stating that it is impossible for abrupt phase

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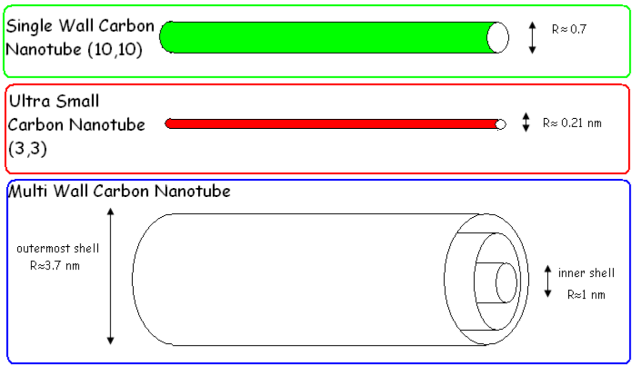
Single Wall Carbon Nanotube (10,10)
R≈0.7
Ultra Small Carbon Nanotube (3,3)
R≈ 0.21 nm
Multi Wall Carbon Nanotube
outermost shell R≈3.7 nm
inner shell R≈1 nm

Figure 1. A SWNT is a rolled up sheet of graphite just nanometres in diameter and up to some micrometres long, while a MWNT is made of several concentrically arranged graphene sheets with a radius of some nanometres.

transitions with long-range order to occur in 1D or 2D systems at finite temperature. Thus CNs are among the best candidates for investigating the possibility of (quasi)1D superconductivity. In general CNs do not show superconducting properties, but some recent experiments found that ultra-small single-walled carbon nanotubes [5] and entirely end-bonded multi-walled ones [6] can superconduct. Clear evidence of superconductivity was also found in CNs suspended between superconducting contacts, showing the so-called proximity effect [7, 8] while genuine superconducting transitions below 1 K have been observed in thick ropes of nanotubes suspended between normal and highly transparent electrodes [9].

Here we mainly discuss the phenomenology concerning the experiments of [5] and [6]. In the first experiment [5], ultra-small-diameter single-walled nanotubes (USCN) have been produced inside the channels of a zeolite matrix (with an inner diameter of ∼0.73 nm). The nanotube diameter $d = 4.2 \pm 0.2$ Å is closer to the value calculated for a $(3, 3)$ CN geometry, although the presence of $(5, 0)$ nanotubes cannot be discounted [10]. These CNs have many unusual properties, such as superconductivity, leading to a transition temperature $T_c \approx 15$ K [5], much higher than that observed in bundles of larger-diameter tubes [11].

In a recent letter [6] it was reported that there is a superconducting phase competing with the Luttinger liquid (LL) phase and even overcoming it in entirely end-bonded MWNTs with a transition temperature T_c as high as 12 K. The lengths of the MWNTs were $L \sim 0.6$ μm, and the high-resolution cross-sectional TEM images showed a MWNT with an outer diameter $2R_o = 7.4$ nm, and inner diameter $2R_i < 2$ nm ($R < 1$ nm). It was also found that the emergence of this superconductivity is highly sensitive to the junction structures of the Au electrode/MWNTs. T_c depends on the number of electrically activated shells; to enhance superconductivity the Au electrodes must be in contact with the tips of all the shells; in contrast, the conventional ‘bulk junction’ contacts only touch the outermost shell of a tube. Below T_c the LL states are suppressed and SC behaviour can appear, while for $T > T_c$ conventional LL behaviour was observed.

In this paper we investigate this phenomenology theoretically by focusing on the central role which the screening of the long-range part of the Coulomb repulsion plays. This screening has to be generally related to the environment (intra- and inter-shell screening in the MWNTs or screening by the zeolite matrix for the USCNs) and has an intrinsic 3D nature. Thus our aim is to show that the interplay between the 1D typical character of the CNs and the 3D nature of the environment allows the SC phase.

In order to pursue our aim we discuss the possibility that superconducting behaviour can arise in these CNs by a purely electronic mechanism, i.e. neglecting the contribution of phonons. We do that by a comparison between two different approaches, one based on the Luttinger model and the other, which emphasizes the role of the lattice and short-range interaction, developed starting from the Hubbard Hamiltonian.

2. Transport in 1D electron systems

Electronic correlations have been predicted to dominate the characteristic features in quasi 1D interacting electron systems, leading to the breakdown of the conventional Fermi liquid picture. In fact Landau quasiparticles are unstable in 1D and the low-energy excitations take the form of plasmons (collective electron–hole pair modes). Thus the 1D character of the system leads to a strong correlation among electrons, inducing the so-called Luttinger liquid (LL) [12]. The LL state has two main features:

(i) the power-law dependence of physical quantities, such as the density of states (DOS), as a function of energy or temperature;
(ii) the spin–charge separation: an additional electron in the LL decays into decoupled spin and charge wavepackets, with different velocities for charge and spin.

Characteristic experimental signatures support the assumed LL behaviour of CNs [13–15], where the temperature dependence of the resistance above a crossover temperature T_c was measured [16]. In fact the power-law dependence of physical observables follows from the behaviour of the DOS as a function of the energy. For example, the tunnelling conductance G in a small-bias experiment [17] follows a power law,

$$G = \mathrm{d}I/\mathrm{d}V \propto T^{\alpha} \tag{1}$$

for $eV_b \ll k_B T$, where V_b is the bias voltage, T is the temperature and k_B is Boltzmann's constant. The critical exponent α assumes different values for an electrode–bulk junction (α_{bulk}) and for an electrode–end junction (α_{end}), as reported for MWNTs in [18].

The theoretical analysis starts from the model of the CN where the electrons have a linear dispersion relation around each of the two Fermi points at $(\pm K_F, 0)$ ($K_F = 4\pi/3a$, and $a = 2.46$ Å is the lattice constant). These branches are highly linear with Fermi velocity $v_F \approx 8 \times 10^5$ m s^{-1}. The linear dispersion relation holds for energy scales $E < D$, with the bandwidth cut-off scale $D \approx \hbar v_F/R$ for tube radius R.

As far as the interaction is concerned we distinguish some processes associated with the Fermi points $\pm K_F$:

(a) the *forward scattering* (g_2 with small transferred momentum $q \sim q_c = 2\pi/L$ which can be assumed as the natural infrared cut-off, depending on the longitudinal length L of the CN),
(b) *backscattering* (g_1 with large transferred momentum, i.e. $q \sim 2K_F$),
(c) an additional *Umklapp* process which is relevant at half-filling and which in our case we neglect, since the sample is assumed to be doped,
(d) an additional *forward scattering* term (f) which measures the difference between intra- and intersublattice interactions; this term is due to the hard core of the Coulomb interaction, i.e. it follows from the unscreened short-range component of the interaction.

f corresponds to [12] $\delta V_p = U_{++} - U{+-}$, where $U_{p,p'}$ is the interaction between electrons belonging to different sublattices (p, p'), and it is strongly suppressed at a distance much larger than $\ell \sim 0.3$ nm [19]. In the same way, the only non-vanishing contribution to g_1 comes from

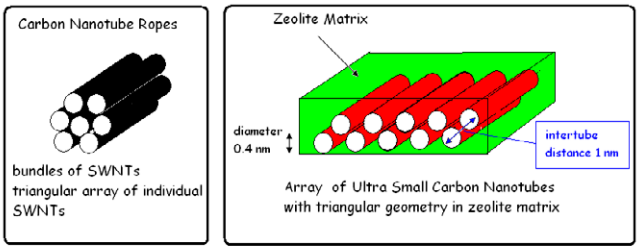
Carbon Nanotube Ropes
bundles of SWNTs
triangular array of individual SWNTs
Zeolite Matrix
diameter 0.4 nm
intertube distance 1 nm
Array of Ultra Small Carbon Nanotubes with triangular geometry in zeolite matrix

Figure 2. As already pointed out in [23] and [24], the intra-tube Coulomb repulsion at small momentum transfer (i.e. in the forward scattering channel) is efficiently screened by the presence of electronic currents in neighbouring nanotubes.

$|x - x'| \leqslant a$, because of rapidly oscillating contributions [12]. Thus we can assume g_2 to be the only relevant long-range component of the interaction. Moreover, notice that the dimensions of the CN play a central role in determining the strength of the different terms of the interaction. The radius R and the length L yield two natural cut-offs, $\approx \frac{2\pi}{R}$ and q_c, while we can classify the physical quantities according their dependence on the radius. Thus we have a long-range coupling, $g_2 = \hat{V}_0(q_c)$ weakly dependent on the radius, and two strongly dependent interaction couplings, $g_1 = \hat{V}_0(2K_F)$ and f which scale as $1/R$ [12].

After introducing the values of the couplings the effective field theory can be solved in practically the same way as was done in [12] and the bulk critical exponent has the form [20–22]

$$\alpha_{\text{bulk}} = \frac{1}{4}\left(g + \frac{1}{g} - 2\right), \tag{2}$$

where g depends just on the forward scattering as

$$\sqrt{1 + \frac{g_2}{(2\pi v_F)}} = \frac{1}{g}. \tag{3}$$

It follows that the short-range terms of the interaction can usually be neglected. However, it was predicted [12] that the effects of g_1 and f can be dominant at low temperatures (T below the crossover temperatures, $g_1 \rightarrow kT_b = De^{-\frac{2\pi v_F}{g_1}}$ and $f \rightarrow kT_f = De^{-\frac{2\pi v_F}{f}} \lesssim kT_b$) where the Luttinger liquid breaks down and a (quasi-) long-range order phase appears. For long-range interactions (which is the case for CNs in typical conditions), we have $T_f \sim T_b$, while for short-range interactions we have $T_f < T_b$. In the latter case a superconducting instability is predicted at $T \sim T_f$ if the Luttinger liquid parameter g is larger than $1/2$.

3. Breakdown of the Luttinger liquid

The above discussion shows that a pure electronic mechanism which gives superconductivity according the LL theory needs a screening of the forward scattering, g_2 (long-range effect $g > 0.5$), an increase of the backward scattering, g_1 (short-range effect T_b) and aid from the f scattering (high value of the corresponding temperature, T_f).

In typical isolated CNs of $R \gtrsim 1$ nm no SC behaviour was observed. This is in line with the above discussion, since g_1 and f are small compared to g_2 and the estimated transition temperature, $T_f \sim T_b \sim 1$ mK, is very low indeed. In this paper we argue about the drastic effects that one can achieve (1) by the interaction with other tubes and/or a matrix, (2) by an ultra-small CN radius or (3) by doping. The interactions with the surroundings can provide an effective screening of the long-range component of the interaction and strongly enhance T_c. This ia an interplay between 3D effects and the quasi 1D behaviour that can allow a SC

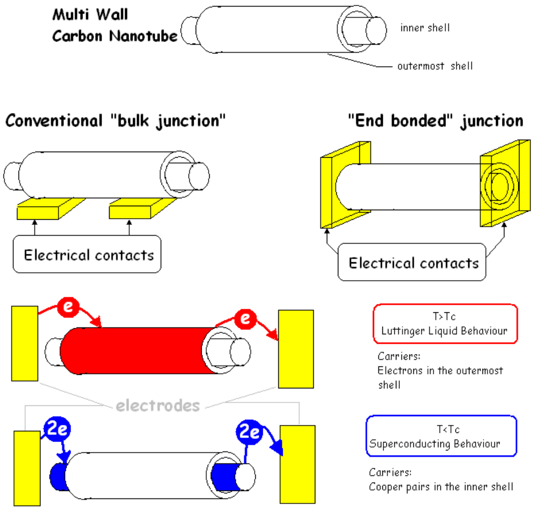
Multi Wall
Carbon Nanotube
inner shell
outermost shell
Conventional "bulk junction"
"End bonded" junction
Electrical contacts
Electrical contacts
e
e
electrodes
2e
2e
T>Tc
Luttinger Liquid Behaviour
Carriers:
Electrons in the outermost shell
T<Tc
Superconducting Behaviour
Carriers:
Cooper pairs in the inner shell

Figure 3. Electrical contacts in the systems of [6] made of Au are bonded to the tubes so they touch the top of all the shells, while conventional 'bulk junction' contacts, in contrast, touch only the outermost shell of a tube and along its length. Under usual conditions, transport measurements carried out in MWNTs reflect the electronic properties of the outer shell, which the electrodes are attached to. On the other hand, in entirely end-bonded samples the inner shells are electrically active, with relevant consequences. In particular the innermost one is able to support the transport of Cooper pairs below a temperature consistent with the measured one.

transition; further 3D effects will be pointed out below for the end-bonded MWNTs. The ultra-small radius contributes to enhancement of the short-range interaction. Although at first sight a stronger g_1 might seem to make pairing more efficient, in fact the converse is found to be true, both in the LL approach and in the Hubbard model (see below). The effects of doping will also be discussed below.

In fact in the experimental samples of [5] the CNs are arranged in large arrays with triangular geometry (with an inter-tube distance $d \approx 1$ nm, as shown in figure 2), behaving as a genuine 3D system as far as the screening properties are concerned. The presence of many CNs inside the zeolite matrix provides a large reduction of g_2 (by a factor $\approx 10^{-2}$), while the short-range components have to remain almost unchanged. This allows for the occurrence of a sizable superconducting instability within the Luttinger liquid approach.

The SC transition in end-bonded MWNTs can also be explained as an effect of the interplay between a 3D and a 1D behaviour. In fact the power law of the conductance observed for $T > 12$ K in the MWNTs of [6] is consistent with the Luttinger liquid character of the normal state. Therefore the observed sharp breakdown of the power law at 12 K is an indication that an approach based on the superconducting instability of the Luttinger liquid is well posed.

The experimental results are consistent with the idea that only the outermost shell became electrically active in the Au-bulk junction (see figure 3). In this case the conducting channel is not efficiently screened and retains a strong 1D character. The entire end-bonding of MWNTs made all the shells electrically active, while only some of the shells were electrically active in the partial Au-end junctions. Therefore, when the electrical contacts touch the top of all the shells (as in the case of the entirely end-bonded MWNTs), the activation of the internal shells gives a large dielectric effect, due to intra- and intershell screening, and at the same time it provides an incipient 3D character, which is crucial for establishing the superconducting coherence [25].

Thus we hypothesized that all contacted shells can transport the normal current as resistors in parallel connection. In the normal (Luttinger) state, a current of electrons flows in each shell while the conductance G is mainly given by the outermost shells, because the outermost shells have the smallest resistance. When the temperature decreases below T_c the superconductivity can be favoured in the inner shells of the MWNT because the short-range interactions are enhanced when the radius decreases. The long-range term of the interaction, g_2, in the end-bonded MWNTs is screened by the electronic currents located in the surrounding shells. Notice that also in this case the screening of g_2 is essentially due to the 3D nature of the MWNTs. An analogous discussion could be extended to the ropes of CNs analysed in [9].

4. Beyond the Luttinger model

The above discussion provides a consistent explanation of the presence of a phase quite different from the normal (LL) one. We could also predict the crossover temperatures as reported above by using the results of [12]. In this way we obtain values of T_c that are compatible with the experimental findings but are quite inaccurate. In order to improve our predictions we develop a different approach based on the Hubbard model which emphasizes the role of the lattice and short-range interaction.

A similar analysis was developed in [26], where the superconductivity in CNs was investigated with the renormalization group technique. By introducing the parameter calculated for the CNs analysed here in the model of [26] we find that a SC phase is supported just in the presence of a slight doping. Unfortunately also in this approach the estimate of T_c is quite inaccurate. A Hubbard-like approach which gives a prediction about the critical temperature was proposed in [19]. It is a pure electronic mechanism which leads to superconducting pairing starting from the Hubbard model on the wrapped honeycomb lattice away from half filling [27]. In this theory ultra-small radius CN are favoured for two reasons: first, the Hubbard on-site repulsion U is larger for smaller radius, and the pairing energy Δ in thereby enhanced, as we have already seen; second, at fixed U, one finds that Δ increases with decreasing radius. By using the BCS formula $\Delta = 1.76kT_c$ for the mean field transition temperature for the ultra-small CNs of [5] one estimates $T_c \approx 7$–70 K which is compatible with the measured one. We observe that the lower bound $T_c \sim 7$ K takes into account that Δ may vary by about one order of magnitude away from optimal doping. Although the theory of [19] has not yet been extended to the MWNT geometry, we may expect that inter-shell hopping should slightly enhance Δ compared to the single-shell case. Thus, we can roughly estimate the crossover temperature for the MWNTs of [6]. At optimal doping we obtain $T_c \approx 4$–40 K This value is slightly lower than that for USNTs, in qualitative agreement with the experimental findings. Also in this case the lower bound $T_c \sim 4$ K is understood in terms of possible deviation from optimal doping [28].

5. Discussion

Here we want to discuss the intrinsic 3D nature of the model and the most relevant effects that we have to take in account.

The Hubbard model keeps only the on-site repulsion U and can be safely used under the condition that the long-range component of the e–e interaction is well screened. Such screening is due to the genuine 3D nature of the systems under study.

The presence of doping deserves a specific discussion and it has to be related to some kind of external effect.

MWNTs use to be significantly doped, which leads to the presence of a large number of subbands at the Fermi level [29]. The contribution of a large number of modes at low energies then has an appreciable impact on the enhancement of observables like the DOS while the activation of several channels is also responsible for the g_2 screening. This topic was investigated in [21, 22], where the effect of doping in the suppression of tunnelling observed in MWNTs was studied. There was shown how the doping is related to the disappearance of the typical 1D behaviour by modifying the effective dimensionality of the system. The doping-induced crossover from a 1D to a 3D behaviour is analogous to the transition from the LL to the SC phase.

Concerning the doping in the SWNTs of [5], the presence of the 3D environment, surrounding matrix or nearest CNs can be assumed to be the main cause of the doping. Despite the differences, the effects of the doping (screening, dimensional crossover) are quite similar to those discussed in the case of MWNTs.

Thus we can conclude that the doping not only plays a central role in the instability of the SC in the Hubbard model, but contributes to the breakdown of the strictly 1D behaviour by adding several 1D conducting channels at the Fermi level and supporting the screening of the long-range interaction.

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