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Double-gap superconducting proximity effect in armchair carbon nanotubes

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We theoretically explore the possibility of a superconducting proximity effect in single-walled metallic carbon nanotubes due to the presence of a superconducting substrate. An unconventional double-gap situation can arise in the two bands for nanotubes of large radius wherein the tunneling is (almost) symmetric in the two sublattices. In such a case, a proximity effect can take place in the symmetric band below a critical experimentally accessible Coulomb interaction strength in the nanotube. Furthermore, due to interactions in the nanotube, the appearance of a BCS gap in this band stabilizes superconductivity in the other band at lower temperatures. We also discuss the scenario of highly asymmetric tunneling and show that this case too supports double-gap superconductivity.

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Graphene-based materials, due to their unique two-band structure,[1,2] have recently commanded an explosion of theoretical and experimental investigations in diverse issues such as the Kondo effect and quantum Hall systems of high SU(4) symmetry,[3,4] weak localization of Dirac fermions,[5] Luttinger liquid effects,[6–9] and Coulomb blockade[8] in nanotubes. In this Rapid Communication, we theoretically investigate the possibility of a proximity-induced effect in single-walled metallic (carbon) nanotubes (SWMNTs) due to the presence of a superconducting substrate with *s*-wave pairing (see Fig. 1) and show that in this geometry the two-band structure of the SWMNT allows for the appearance of two superconducting gaps. In the case of nearly symmetric tunneling in the two sublattices, which may be realized for metallic nanotubes with quite large radius, we predict two gaps of different origins—one due to the superconducting substrate and the other due to electron-electron interactions inside the SWMNT. For very asymmetric tunneling in the two sublattices, the two gaps emerge due to the proximity of the substrate and they become identical when only one sublattice is sensitive to the substrate.

Superconductivity in nanotubes has presented several puzzles. Experimental observations include a very strong proximity effect in suspended SWMNTs,[10] intrinsic superconductivity in ultrathin nanotubes embedded in zeolite matrix,[11] and BCS-type behavior in bundles.[12] It has been predicted that superconductivity in an isolated SWMNT would be manifest only at experimentally inaccessible temperatures and for screened Coulomb interactions.[13] However, phonon exchange might be responsible for some attractive interactions in the SWMNT.[14,15] Alternatively, theoretical questions arise as to whether external factors can also stabilize superconductivity in SWMNTs, and if so, as to the nature of this superconducting behavior. In the case of a point contact with a superconductor, the proximity effect necessarily requires attractive interactions to be stabilized,[16,17] which then favors a single-gap scenario. Below, we consider the case of bulk contact with a superconductor and Coulomb interactions which are unscreened, i.e., phonon exchange is not relevant. We show that the proximity effect is stabilized, and a double superconducting gap feature appears as a generic unconventional phenomenon.

As our starting point, we focus on a long, metallic (N,N) armchair nanotube,[18] which is known to be an ideal one-dimensional conductor.[8,9,19] As shown in Fig. 2, electronic low-energy excitations of the tube consist of two linearly dispersing bands (labeled by $i=1,2$ and associated pseudospin Pauli matrices $\vec{\eta}$). These bands are symmetric and

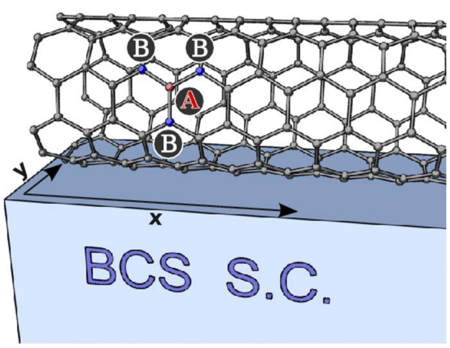

FIG. 1. (Color online) A metallic armchair SWMNT, comprising an underlying graphene structure having sublattices *A* and *B*, deposited on an *s*-wave paired BCS superconductor. The SWMNT is coupled to the substrate via electron-tunneling.

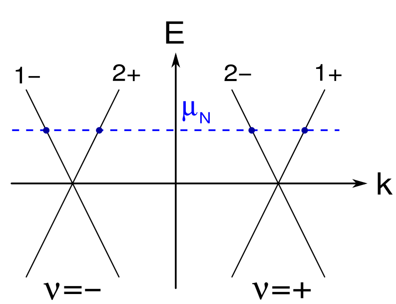

FIG. 2. (Color online) Low-energy band structure of a SWMNT. Fermi points are labeled by $\nu=\pm$ and the sublattices *A* and *B* combine to build two bands with right $(+)$ and left $(-)$ movers. Here, k represents the momentum associated with the x direction and μ_N corresponds to the Fermi energy.

antisymmetric combinations of the two sublattices of the nanotube shown in Fig. 1. In the effectively infinite system, each band has an associated right and left mover (labeled by $r=+,-$, respectively, and pseudospin Pauli matrices $\vec{\tau}$). Each of the modes also carries spin (labeled by $\alpha=\uparrow,\downarrow$ and Pauli matrices $\vec{\sigma}$), thus constituting eight degrees of freedom. The Hamiltonian governing these modes reads

$$H_0 = iv_F \sum_{ir\alpha} r \int dx\, \psi^\dagger_{ir\alpha} \partial_x \psi_{ir\alpha}; \tag{1}$$

$v_F \approx 8\times 10^5$ m/s is the Fermi velocity of each of the modes and $\psi^\dagger_{ir\alpha}$ is the operator for creating an electron of band index i, chirality r, and spin α. As shown in Fig. 1, the x axis points along the tube direction.

Below, we investigate the effects of external influences such as substrates on these low-lying SWMNT modes. As the most relevant couplings are expected to be quadratic in the fermion operators, we first consider their influence before considering interaction effects in the SWMNT.

As a realistic situation, we explore the case of a nanotube in bulk contact with a conventional BCS singlet-paired superconducting substrate via electron tunneling. The simplest such coupling is described by

$$H_{\text{tun}} = \int dx \{\chi^\dagger_\alpha(x,0)[t_A c_{A\alpha}(x) + t_B c_{B\alpha}(x)] + \text{H.c.}\}, \tag{2}$$

where an implicit sum on spin α is assumed. Electronic substrate degrees of freedom are described by the creation operator $\chi^\dagger_\alpha(x,y)$; for simplicity, in Eq. (2), we have assumed a quasi-two-dimensional substrate and the y direction is shown in Fig. 1. The nanotube degrees of freedom on sublattices A and B are related to the aforementioned low-lying modes via $c^{\nu=+}_{A/B\alpha}=(\pm\psi_{2-\alpha}+\psi_{1+\alpha})/\sqrt{2}$ and $c^{\nu=-}_{A/B\alpha}=(\pm\psi_{2+\alpha}+\psi_{1-\alpha})/\sqrt{2}$. The matrix elements $t_{A/B}$ related to the electron tunneling strengths into the two sublattices depend upon the overlap of wave functions between the substrate and nanotube degrees of freedom. Focusing on an armchair SWMNT allows us (in principle) to neglect small inhomogeneities in the tunneling amplitudes, i.e., t_A and t_B are assumed to be x independent.

An effective description of the nanotube under the influence of the substrate can be obtained by integrating out the substrate degrees of freedom $\chi_\alpha(x,y)$. Such an integration is straightforward given that the substrate can be described by the standard BCS form.[20] For singlet-paired superconductivity, which is the case of interest below, the main induced couplings stem from Andreev reflections, resulting in the Hamiltonian

$$H_{\text{ind}} = -\int dx \sum_{\text{indices}} (h_i \psi^\dagger_{ir\alpha} i\sigma^y_{\alpha\beta} \tau^x_{rw} \psi^\dagger_{iw\beta} + h_3 \psi^\dagger_{ir\alpha} i\sigma^y_{\alpha\beta} \eta^x_{ij} \psi^\dagger_{jr\beta} + \text{H.c.}). \tag{3}$$

It should be noted that the Andreev term h_3, which couples the two bands, can only be relevant if the Fermi energy μ_N of the SWMNT lies at the Dirac points; otherwise, this process does not conserve momentum and therefore becomes irrelevant at long wavelengths. So we will ignore it, considering the (general) case where μ_N does not lie at the Dirac points.[21]

The bare couplings $h_{1/2}$ take the general form $h_{1/2} = h_0(1\pm\sin 2\theta)$. For a quasi-two-dimensional substrate, the coefficient h_0 is proportional to $(t_A^2+t_B^2)$ and is inversely proportional to $\sqrt{\Delta}$, where Δ is the superconducting gap of the substrate. In general the angle θ obeys $0\leqslant\theta=\tan^{-1}(t_B/t_A)\leqslant\pi/4$.[22] However $\theta\to\pi/4$ (symmetric tunneling) should hold for large-radius armchair tubes (see Fig. 1), substrates whose underlying lattices show significant mismatch with the nanotube lattice, and for specific orientations of the tube in which the A and B sublattices are equidistant from the substrate. The situation $\theta\to 0$ corresponds to the extreme and less physical case where tunneling involves only one sublattice.

We argue that our procedure is well controlled in the weak-coupling regime, i.e., assuming that the couplings h_i are smaller than the superconducting gap Δ, which then can be used as the ultraviolet cutoff of the effective theory. It should be noted that, since an electron in the SWMNT can virtually leak into the substrate and then tunnel back into the SWMNT, in principle, the electron propagator of a given band also acquires a finite self-energy. For a superconducting substrate, we have checked that this self-energy evolves smoothly close to the quasiparticle pole. This effect is always small compared to that of electron-electron interactions on the electron self-energy[23] and thus can be safely ignored. Also, for the sake of simplicity, the superfluid phase of the superconducting substrate is set to zero.

The above arguments indicate that, depending on the microscopic details of the tunneling coupling with the substrate, the nanotube can develop two BCS-type gaps. In what follows we study the effect of Coulomb interactions on these gaps along the lines of Refs. 6 and 13. We also show that interactions have a dramatic effect when $\theta\to\pi/4$, when the coupling with the superconducting substrate only gives rise to a gap in the symmetric band. In this context interactions reinforce interband Cooper processes consisting of pair hopping from band 1 to band 2,[24–26] resulting in a superconducting gap at lower temperatures in band 2 in spite of the vanishing value of h_2. We wish to emphasize that this scenario is still likely to happen for reasonably small deviations around $\theta=\pi/4$.

To begin with, forward scattering processes, in which electrons stay in the same branch, produce a long-range interaction which involves the total charge density,[6]

$$H_{\text{ind}} = e^2 \ln(R_s/R) \int dx\, \rho^2_{\text{tot}}(x), \tag{4}$$

where $\rho_{\text{tot}}=\Sigma_{ir\alpha}\psi^\dagger_{ir\alpha}\psi_{ir\alpha}$, R being the tube radius and R_s the screening length of Coulomb interactions. In general, R_s is long compared to R but short compared to the length of the tube. In the extensively used Luttinger liquid description[27] of the SWMNT,[6,13] this term contributes to the Luttinger parameter g of the total charge "sector" as

$$g = [1 + (8e^2/v_F\pi\hbar)\ln(R_s/R)]^{-1/2}. \tag{5}$$

In this description, we find the scaling dimension of the operators $h_{1,2}$ to be $\delta_h=(3+g^{-1})/4$. We deduce that the Andreev terms $h_{1,2}$ are relevant when g exceeds the critical value g_c

$=0.2$, i.e., when $\delta_h<2$, thereby opening BCS-type gaps below the critical temperatures

$$T_{c1,2} \approx \Delta\left(\frac{h_{1,2}}{\Delta}\right)^{4/(5-g^{-1})} < \Delta. \tag{6}$$

As mentioned above, the superconducting gap Δ in the BCS-type substrate is the ultraviolet cutoff of our (effective) theory, which justifies that $T_{c2}<T_{c1}<\Delta$. For free electrons, i.e., $g=1$, we recover the critical temperatures $T_{c1,2}\sim h_{1,2}$. Coulomb interactions in the tube tend to reduce the superconducting gaps $\Delta_{1,2}\sim T_{c1,2}$ which are experimentally accessible if g is not too close to 0.2, i.e., for a screening length R_s which is smaller than 1000 Å.

Now, we investigate in depth the more unconventional, physically accessible situation $\theta\to\pi/4$ or $h_2\to 0$ below the critical temperature T_{c1}. In this situation $\Delta_2\to 0$ and apparently only band 1 is gapped. First, the superconducting gap Δ_1 in band 1 affects spectral properties such as the local density of states available for an electron to tunnel into a long tube at a given site on the tube from a metallic electrode or a scanning tunneling microscopy tip. For example, in the weak-tunneling regime, the (tunneling) current I should exhibit a prominent peak at the bias voltage $V=\pm\Delta_1/e$ reflecting the profile of the BCS density of states.[28] Whereas a BCS gap develops in band 1, close to and below T_{c1}, the band 2 charge sector still obeys the Luttinger theory,

$$H(\theta_{2\rho},\varphi_{2\rho}) = \frac{v_F}{2\pi}\int dx\left(\frac{1}{g_2^2}(\partial_x\theta_{2\rho})^2 + (\partial_x\varphi_{2\rho})^2\right), \tag{7}$$

where $\partial_x\theta_{2\rho}$ represents the charge density associated with band 2 and $\varphi_{2\rho}$ embodies the conjugate superfluid phase. The Luttinger parameter satisfies $g_2^{-2}=(g^{-2}+1)/2$.

Bands 1 and 2 are still coupled through H_{int}, and the most relevant coupling is of the form

$$H_{\text{int}}^{(1)} = \int dx\, v_{12}^{\rho}\partial_x\theta_{1\rho}\partial_x\theta_{2\rho}, \tag{8}$$

where $v_{12}^{\rho}=v_F(g^{-2}-1)/2\pi$. Given that the superfluid phase $\varphi_{1\rho}$ of band 1 is pinned below T_{c1}, thus inducing strong fluctuations in the density operator $\partial_x\theta_{1\rho}$, the coupling v_{12}^{ρ} renders the $\theta_{2\rho}$ correlation function with additional fluctuations. This potentially enhances g_2. The coupling v_{12}^{ρ} is in fact an analytic perturbation which has been thoroughly analyzed in a different context.[25] To evaluate the increase of g_2 below T_{c1} we proceed as in Ref. 25, which gives $g_2^r\approx g_2[1-g_2^4(v_{12}^{\rho})^2\pi^2/(2v_F^2)]^{-1}$,[29] and $g_2^r\approx 2g_2\approx 2\sqrt{2}g$ assuming that $g\ll 1$. The tunneling current at low bias voltage emerging from the gapless band 2 thus obeys the form $dI/dV\sim V^{\zeta}$, where $\zeta=(g_2^r+1/g_2^r-2)/4$. The exponent ζ is distinguishable from the exponent $(g+g^{-1}-2)/8$ which occurs in the absence of the substrate, i.e., above T_{c1}.[6]

The results above hold for a range of temperatures below T_{c1}. However, as shown below, at still lower temperatures, several short-range interactions become important, in particular backward scattering mechanisms and other forward scattering processes measuring the difference between intra- and intersublattice interactions. In the absence of a superconducting substrate, these momentum-conserving scattering vertices are "marginal" and become important only at an exponentially small (unreachable) energy scale.[13] However, in the presence of the substrate, below T_{c1}, these terms gain relevance as a result of the off-diagonal long-range order in band 1, i.e.,

$$\mathcal{P}_1 = \langle \psi_{1+\uparrow}^{\dagger}\psi_{1-\downarrow}^{\dagger} + \psi_{1-\uparrow}^{\dagger}\psi_{1+\downarrow}^{\dagger}\rangle \neq 0, \tag{9}$$

where we estimate $\mathcal{P}_1\sim T_{c1}/\hbar v_F$. In analogy with the two-chain Hubbard model,[24,25] we thus anticipate that interband Cooper-type processes will be reinforced below T_{c1}. We note that Eq. (3) implies $\mathcal{P}_1>0$, consistent with the superfluid phase of band 1 coinciding with that of the superconducting substrate below T_{c1}.

From Ref. 13, we identify two relevant forward (f) and backward (b) scattering processes respecting $\mathcal{P}_1\neq 0$,

$$H_{\text{int}}^{(2)} = \int dx\sum_{\alpha}\,(-f\psi_{1+\alpha}^{\dagger}\psi_{1-\bar{\alpha}}^{\dagger}\psi_{2-\alpha}\psi_{2+\bar{\alpha}} + b\psi_{1+\alpha}^{\dagger}\psi_{1-\bar{\alpha}}^{\dagger}\psi_{2-\bar{\alpha}}\psi_{2+\alpha} + \mathrm{H.c.}), \tag{10}$$

with $\bar{\alpha}=\downarrow$ if $\alpha=\uparrow$, and vice versa. Assuming the equality $b=f$, this results in the following (exact) Hamiltonian:

$$H_{\text{int}}^{(2)} = \int dx\, f\mathcal{P}_1(\psi_{2-\downarrow}\psi_{2+\uparrow} + \psi_{2+\downarrow}\psi_{2-\uparrow} + \mathrm{H.c.}), \tag{11}$$

which characterizes pair hopping mechanisms from one band to the other. Using the notations of Ref. 13, we get $f\mathcal{P}_1\approx\gamma 5.4\pi T_{c1}/\sqrt{3}N$ where γ is a constant of the order of 0.1 and N is the circumference of the tube in units of graphene periodicity. The factor $1/N$ appears because these terms stem from short-ranged contributions and the probability for two electrons to be near each other is of order $1/N$. A small deviation from $b=f$ does not affect the result because, below T_{c1}, one obtains $\langle\psi_{1+\uparrow}^{\dagger}\psi_{1-\downarrow}^{\dagger}-\psi_{1-\uparrow}^{\dagger}\psi_{1+\downarrow}^{\dagger}\rangle=0$ (as a result of $\mathcal{P}_1\neq 0$). The Cooper pair (Higgs) field associated with band 1 thus induces a superconducting gap in band 2, and the associated critical temperature T_{c3} is defined by

$$T_{c3} \approx T_{c1}\left(\frac{f\mathcal{P}_1}{T_{c1}}\right)^{2/(3-1/g_2^r)} < T_{c1}. \tag{12}$$

For $\theta\to\pi/4$, the emergent superconducting gap in band 2, $\Delta_3\sim T_{c3}$, is also accessible experimentally; for a (10,10) armchair nanotube we find $f\mathcal{P}_1/T_{c1}\sim 0.1$. $H_{\text{int}}^{(2)}$ implies that the superconducting order parameters of the two bands exhibit a relative negative sign which is consistent with repulsive interactions ($f>0$) and two-band Hubbard ladders.[24,25,27]

Thus, the band structure of the SWMNT offers two possible mechanisms for a double-gap superconducting proximity effect. For the extreme asymmetric tunneling case $\theta\to 0$, the two bands equally couple to the substrate and each exhibits a proximity-induced gap. For completely symmetric tunneling $\theta\to\pi/4$, even though only the symmetric band becomes affected by the substrate, interactions in the SWMNT still open a superconducting gap in the antisymmetric band. We conjecture that the exotic double-gap scenario occurring at $\theta \to \pi/4$ can be realized for armchair nanotubes of large radius and substrates whose underlying lattices show significant mismatch with the nanotube lattice. This situation is likely to happen as long as $T_{c3} > T_{c2}$. The two proximity effects associated with band 2 "compete," i.e., they give a different sign to the superconducting order parameter of band 2 and thus result in a quantum phase transition occurring at the value of θ for which $T_{c2} = T_{c3}$; at this special point the band 2 remains gapless until zero temperature. For temperatures smaller than T_{c2} (T_{c3}), the tunneling current exhibits a complete gap at low bias voltages and a second peak at $V = \pm\Delta_2/e$ ($V = \pm\Delta_3/e$).

In conclusion, superconducting substrates stabilize superconductivity in SWMNTs and allow for the existence of a double superconducting gap. A double-proximity effect in nanotubes is yet to be ascertained by experiment. Suggestively, several gaps have been observed in few-layer graphene coupled to superconducting leads.[30] Extensions of this work include study of the proximity effect between graphene and a (superconducting) substrate, and of possible asymmetry in the coupling with the substrate for graphene-based materials.[31]

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