

## Luttinger Liquid Behavior in Multiwall Carbon Nanotubes

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The low-energy theory for multiwall carbon nanotubes including the long-ranged Coulomb interactions, internal screening effects, and single-electron hopping between graphite shells is derived and analyzed by bosonization methods. Characteristic Luttinger liquid power laws are found for the tunneling density of states, with exponents approaching their Fermi liquid value only very slowly as the number of conducting shells increases. With minor modifications, the same conclusions apply to transport in ropes of single-wall nanotubes.

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Metallic carbon nanotubes constitute a novel and exciting realization of one-dimensional (1D) conductors [1,2]. The strong electronic correlations observed experimentally [3] pose many challenges to theorists. It is well known that electron-electron interactions invalidate the Fermi liquid description in one dimension. Often 1D conductors can instead be described as a Luttinger liquid (LL) at low-energy scales. The theoretical prediction [4,5] of LL behavior in a single-wall nanotube (SWNT) has indeed been verified in a recent transport experiment [6]. Furthermore, pronounced interaction effects have been observed for an individual multiwall nanotube (MWNT) composed of several (typically 10 to 20) concentric graphite shells [7,8], most notably a pronounced zero-bias anomaly at low applied voltage, which was tentatively interpreted in terms of LL theory. Since MWNTs are much easier to manipulate than SWNTs, interaction effects might be useful for applications, and whether a MWNT can display LL behavior could then be of practical importance. Similar questions apply to an individual bundle ("rope") of SWNTs, the system actually studied in Ref. [6].

In this Letter, the low-energy theory of an individual MWNT composed of  $N$  metallic graphite shells [9] with radii  $R_1 < R_2 < \dots < R_N$  is derived, taking into account the externally unscreened Coulomb interaction, internal screening effects, and intershell electron tunneling. Semi-conducting shells and an insulating substrate are incorporated in terms of a space-dependent dielectric constant. The theory holds for energy scales  $k_B T, eV \ll v/R_N$  [we set  $\hbar = 1$ ], where  $v \approx 8 \times 10^5$  m/sec is the radius-independent Fermi velocity; for  $R_N = 10$  nm,  $v/R_N \approx 350$  meV. One then needs to take into account only two transport bands  $\alpha = \pm$  per shell. For both bands, there is a right- and a left-moving ( $r = \pm = R/L$ ) branch with linear dispersion. By employing bosonization methods [10], pronounced LL effects are predicted for a MWNT, with only a slow crossover to Fermi liquid behavior as  $N$  increases. The relation between experimentally measurable exponents and microscopic quantities turns out to be quite different from a SWNT.

Unfortunately, little is known about the role of lattice defects and structural inhomogeneities in MWNTs. However, experiments [7,11] are consistent with elastic mean-free paths of up to a few  $\mu\text{m}$ , and molecular-mechanics simulations [12] have demonstrated that MWNTs remain rigid and structurally uniform even on a substrate. The model studied below should thus provide at least a qualitative understanding of interaction effects in MWNTs.

The construction of the low-energy theory starts by expanding the electron operator for spin  $\sigma = \pm$  on shell  $n = 1, \dots, N$  using the Bloch waves,

$$\Psi_{\sigma n}(x, y) = \sum_{r\alpha} \phi_{ran}(x, y) \psi_{r\alpha\sigma n}(x), \quad (1)$$

where  $0 < y < 2\pi R_n$ , and  $x$  is the transport direction. The Bloch waves depend on the helicity of each shell,

$$\phi_{ran}(x, y) = (2\pi R_n)^{-1/2} \exp[i\alpha(k_{F,n}x + p_{F,n}y)]. \quad (2)$$

The expansion (1) allows one to formulate the theory in terms of 1D fermion operators  $\psi_{r\alpha\sigma n}(x)$ . Without interactions and intershell tunneling, the linear dispersion implies  $N$  copies of a 1D massless Dirac Hamiltonian,

$$H_0 = -iv \int dx \sum_{r\alpha\sigma n} r \psi_{r\alpha\sigma n}^\dagger \partial_x \psi_{r\alpha\sigma n}. \quad (3)$$

In the next step, let us include the Coulomb interactions among the electrons. Backscattering is neglected for reasons explained below. Since one is normally off half filling due to the presence of external gates, umklapp scattering is also ignored. Under the expansion (1), the important forward scattering processes yield

$$H_I = \frac{1}{2} \sum_{n,m=1}^N \int dx dx' \rho_n(x) V_{nm}(x-x') \rho_m(x'), \quad (4)$$

with 1D densities  $\rho_n = \sum_{r\alpha\sigma} \psi_{r\alpha\sigma n}^\dagger \psi_{r\alpha\sigma n}$  for the  $n$ th shell. The effective 1D interaction potential is obtained from the externally unscreened 3D Coulomb potential by

integrating over the circumferential coordinates using the Bloch functions (here  $a$  is a lattice spacing),

$$V_{nm}(x) = \frac{e^2}{\kappa_{nm}} \int_0^{2\pi} \frac{d\varphi}{2\pi} \frac{d\varphi'}{2\pi} \{a^2 + (R_n - R_m)^2 + x^2 + 4R_n R_m \times \sin^2[(\varphi - \varphi')/2]\}^{-1/2}.$$

The dielectric constants  $\kappa_{nm}$  include the effect of the substrate and of semiconducting shells. The Fourier transform  $\tilde{V}_{nm}(k)$  at long wavelengths,  $|kR_N| \ll 1$ , is

$$\tilde{V}_{nm}(k) = \frac{2e^2}{\kappa_{nm}} |\ln(|k|\bar{R}_{nm})|, \quad (5)$$

with the ‘‘mean radius’’

$$\bar{R}_{nm} = \exp \int_0^{2\pi} \frac{d\varphi}{4\pi} \ln\{[(R_n - R_m)/2]^2 + R_n R_m \sin^2 \varphi\}.$$

The logarithmic singularity of  $\tilde{V}_{nm}(k \rightarrow 0)$  reflects the unscreened  $1/r$  tail of the Coulomb interaction. In order to make contact with the usual LL concept, we restrict our-

selves to a long-wavelength description by cutting off this singularity at  $k = 2\pi/L$ , where  $L$  is the MWNT length [13]. Including a factor  $4/\pi v$  for later convenience, this leads to the dimensionless coupling constants

$$U_{nm} = \frac{8e^2}{\pi v \kappa_{nm}} \ln(L/2\pi\bar{R}_{nm}). \quad (6)$$

For  $L = 1 \mu\text{m}$ ,  $\bar{R}_{nm} = 6 \text{ nm}$ , and  $\kappa_{nm} = 1.4$ , Eq. (6) yields  $U_{nm} \approx 16$ . Therefore both the intrashell and the intershell electrostatic interactions are typically very strong and of comparable magnitude. Because of the weak logarithmic dependence of  $U_{nm}$  on  $\bar{R}_{nm}$ , the approximation  $U_{nm} = U$  can already yield sensible results. Similar reasoning applies to ropes of SWNTs, where, however, geometrical considerations suggest that  $U_{nm} \approx U$  may not be justified anymore. Nevertheless, the logarithmic singularity (6) and hence the same order of magnitude for  $U_{nm}$  are also expected for ropes.

By virtue of bosonization,  $H_0 + H_I$  can now be diagonalized. The bosonized 1D fermion operator is [10]

$$\psi_{r\alpha\sigma n}(x) = \frac{\eta_{r\alpha\sigma n}}{\sqrt{2\pi a}} \exp\{iq_F r x + i(\pi/4)^{1/2}[\theta_{c+,n} + r\varphi_{c+,n} + \alpha\theta_{c-,n} + r\alpha\varphi_{c-,n} + \sigma\theta_{s+,n} + r\sigma\varphi_{s+,n} + \alpha\sigma\theta_{s-,n} + r\alpha\sigma\varphi_{s-,n}]\}, \quad (7)$$

where symmetric and antisymmetric linear combinations of the two transport bands  $\alpha = \pm$  have been formed for charge and spin degrees of freedom. The resulting four channels are labeled by the index  $\gamma = (c+, c-, s+, s-)$ . The boson phase fields obey the commutator algebra

$$[\theta_{\gamma,n}(x), \varphi_{\gamma',n'}(x')] = -(i/2)\delta_{nn'}\delta_{\gamma\gamma'} \text{sgn}(x - x'),$$

so that  $\varphi_{\gamma,n}$  has the canonical momentum  $\Pi_{\gamma,n} = -\partial_x \theta_{\gamma,n}$ . The Majorana fermions  $\eta_{r\alpha\sigma n}$  ensure anticommutation relations between 1D fermions with different indices  $r\alpha\sigma n$ . Finally,  $q_F = E_F/v$  is determined by external gate voltages, where the Fermi energy  $E_F$  and hence  $q_F$  are identical for all  $N$  shells [14]. Using Eq. (7), the density is  $\rho_n(x) = (4/\pi)^{1/2} \partial_x \varphi_{c+,n}$ , and as a consequence the Hamiltonian decouples in all four channels,  $H_0 + H_I = \sum_\gamma H_\gamma$ . The charged channel is described by

$$H_{c+} = \frac{v}{2} \sum_{n,m=1}^N \int dx \{[\Pi_{c+,n}^2 + (\partial_x \varphi_{c+,n})^2] \delta_{nm} + U_{nm} \partial_x \varphi_{c+,n} \partial_x \varphi_{c+,m}\}. \quad (8)$$

The three neutral channels correspond to Eq. (8) with  $U_{nm} = 0$ . Diagonalizing  $H_{c+}$  then leads to an eigenvalue problem similar to the one studied by Matveev and Glazman [15] for many-channel quantum wires,

$$\sum_{m=1}^N \{(1 - g_j^{-2})\delta_{nm} + U_{nm}\} \Gamma_{mj} = 0, \quad (9)$$

where  $\varphi_{c+,n}(x) = \sum_j \Gamma_{nj} \Phi_j(x)$  and  $\Gamma_{nj}$  is an orthogonal matrix. With the new fields  $\Phi_j$  and their momenta  $\tilde{\Pi}_j$ ,

$H_{c+}$  takes a standard LL form,

$$H_{c+} = \frac{v}{2} \sum_{j=1}^N \int dx [\tilde{\Pi}_j^2 + g_j^{-2} (\partial_x \Phi_j)^2]. \quad (10)$$

The LL interaction constants  $g_j \leq 1$  for the  $N$  eigenmodes measure the correlation strength.

It is then easy to determine all scaling exponents of interest. The exponents  $\eta_{b/e}$  of the tunneling density of states (TDOS),  $\rho(E) \sim E^\eta$ , for tunneling of an electron into the outermost shell ( $n = N$ ) in the bulk or close to the end of the tube are

$$\eta_b = \frac{1}{8} \sum_{j=1}^N \Gamma_{Nj}^2 (g_j^{-1} + g_j - 2), \quad (11)$$

$$\eta_e = \frac{1}{4} \sum_j \Gamma_{Nj}^2 (g_j^{-1} - 1). \quad (12)$$

These exponents govern the power laws  $G \sim T^\eta$  of the temperature-dependent linear conductance for tunneling into the MWNT and can be measured in the experimental setup of Ref. [7], where external leads contact only the outermost shell of the MWNT. Moreover, in the limit of weak disorder backscattering, the linear conductance corrections  $\delta G \sim T^{-p}$  are characterized by the exponent

$$p = \frac{1}{2} \sum_j \Gamma_{Nj}^2 (1 - g_j). \quad (13)$$

At sufficiently low temperatures, disorder in a LL always leads to a strong backscattering situation. The conductance then vanishes as  $T^\beta$  for  $T \rightarrow 0$ , where  $\beta = 2\eta_e$ .

Observation of the exponents  $p$  and  $\beta$  requires good contacts between external leads and the MWNT.

To elucidate the qualitative features, we now solve Eq. (9) for  $U_{nm} = U$ . The eigenvalues are

$$g_1 = g = (1 + NU)^{-1/2}, \quad g_{2,\dots,N} = 1. \quad (14)$$

The first has eigenvector  $\Gamma_{n1} = N^{-1/2}$  and is identified with the standard collective LL plasmon mode, now with an  $N$ -dependent LL parameter  $g$ . The remaining  $N - 1$  degenerate modes correspond to Fermi liquid quasiparticles. Using  $\sum_{j>1} \Gamma_{Nj}^2 = 1 - 1/N$ , the above exponents read  $\eta_b = (g^{-1} + g - 2)/8N$ ,  $\eta_e = (g^{-1} - 1)/4N$ , and  $p = (1 - g)/2N$ . For  $N \rightarrow \infty$ , the exponents  $\eta_{b,e}$  and  $\beta$  approach zero (the Fermi liquid value) only as  $N^{-1/2}$ , because the LL parameter  $g$  also goes to zero as  $N^{-1/2}$  (up to logarithmic corrections). For instance, for  $U = 16$  and  $N = 10$  the exponents are  $\eta_e = 0.292$  and  $\eta_b = 0.135$ , while the SWNT values are  $\eta_e = 0.780$  and  $\eta_b = 0.295$ . On the other hand, the exponent  $p$  scales as  $1/N$  for large  $N$  and hence vanishes more rapidly. For instance, for  $N = 10$  it is  $p = 0.046$ , but for  $N = 1$  we get  $p = 0.379$ . LL power laws in MWNTs or ropes of SWNTs are thus generally much more pronounced for the TDOS than for the backscattering corrections in the presence of weak disorder. This important fact may be used to obtain information about the number  $N$  of conducting shells.

The result (13) holds for defects in the  $N$ th shell. More generally, a lattice defect at  $x = 0$  in the  $M$ th shell leads to  $H_{\text{imp}} = \lambda [\prod_{\gamma} \cos[\sqrt{\pi} \varphi_{\gamma,M}(0)]]$ . The exponent  $p$  in Eq. (13) is then replaced by  $p_M = \frac{1}{2} \sum_j \Gamma_{Mj}^2 (1 - g_j)$ . In marked contrast to a Fermi liquid, a defect in an inner shell leads to a reduction of the conductance through the outermost shell,  $\delta G \sim T^{-p_M}$ , by a modification of the internal screening properties.

Next single-electron hopping between the shells is incorporated. Using Eq. (2), it can be written as

$$H_t = \sum_{n,m=1}^N T_{nm} \int dx \sum_{r\alpha\sigma} e^{-i\alpha(k_{F,n} - k_{F,m})x} \psi_{r\alpha\sigma n}^\dagger \psi_{r\alpha\sigma m}, \quad (15)$$

where the hopping matrix acts only between nearest-neighbor shells,  $T_{nm} = -t_n \delta_{n+1,m} - t_{n-1} \delta_{n,m+1}$ . Processes where  $r\alpha\sigma$ -type fermions are scattered into different  $r\alpha\sigma$  states are suppressed against Eq. (15) by momentum conservation and by higher scaling dimensions. If there is a Fermi-momentum mismatch,  $k_{F,n} \neq k_{F,n+1}$ , intershell coherence is strongly reduced by the oscillatory factor, and the respective (irrelevant) hopping  $t_n$  should not enter the matrix  $T_{nm}$ . Renormalization group (RG) arguments suggest that  $H_t$  can be relevant, and therefore we go back and first diagonalize  $H_0 + H_t$ . With the orthogonal matrix  $Q_{n\nu}$ ,

$$\psi_{r\alpha\sigma n}(x) = \sum_{\nu=1}^N Q_{n\nu} \tilde{\psi}_{r\alpha\sigma\nu}(x), \quad (16)$$

the rotated fermions  $\tilde{\psi}_{r\alpha\sigma\nu}$  again obey the 1D Dirac Hamiltonian (3). Denoting the  $N$  eigenvalues of  $T_{nm}$  as  $T_\nu$ , the respective eigenvectors span  $Q_{n\nu}$  so that  $H_t = \sum_\nu T_\nu \int dx \tilde{\rho}_\nu(x)$ , with 1D densities  $\tilde{\rho}_\nu = \sum_{r\alpha\sigma} \times \tilde{\psi}_{r\alpha\sigma\nu}^\dagger \psi_{r\alpha\sigma\nu}$ . The interactions now read

$$H_I = \frac{\pi v}{8} \sum_{\nu_1\nu_2\nu_3\nu_4} Y_{\nu_1\nu_2\nu_3\nu_4} \sum_{r\alpha\sigma,r'\alpha'\sigma'} \int dx \times \tilde{\psi}_{r\alpha\sigma\nu_1}^\dagger \tilde{\psi}_{r\alpha\sigma\nu_2} \tilde{\psi}_{r'\alpha'\sigma'\nu_3}^\dagger \tilde{\psi}_{r'\alpha'\sigma'\nu_4}, \quad (17)$$

with matrix elements

$$Y_{\nu_1\nu_2\nu_3\nu_4} = \sum_{nm} U_{nm} Q_{\nu_1 n} Q_{n\nu_2} Q_{\nu_3 m} Q_{m\nu_4}. \quad (18)$$

The complicated four-fermion interactions (17) reflect the difficulties encountered in previous studies of coupled LLs [10]. According to Eq. (6), however, in MWNTs the couplings  $U_{nm}$  are approximately equal, which allows one to make further progress.

For  $U_{nm} \approx U$ , the dominant matrix elements in Eq. (18) are  $Y_{\nu\nu\nu\nu} \equiv W_{\nu\nu} = W_{\nu'\nu}$ . In fact, if  $U_{nm} = U$ , orthogonality of  $Q_{n\nu}$  implies that  $W_{\nu\nu} = U$  and all other matrix elements vanish. Keeping only the couplings  $W_{\nu\nu}$ , Eq. (17) simplifies to  $H_I = (\pi v/8) \times \sum_{\nu\nu'} W_{\nu\nu'} \int dx \tilde{\rho}_\nu(x) \tilde{\rho}_{\nu'}(x)$ . The model  $H_0 + H_I + H_t$  can then be solved exactly by bosonization in the rotated basis. Using Eq. (7) for  $\tilde{\psi}_{r\alpha\sigma\nu}(x)$ , the neutral channels are described by the same Hamiltonian as before. The only change arises in the charged sector,

$$H_{c+} = \frac{v}{2} \sum_{j=1}^N \int dx [\tilde{\Pi}_j^2 + g_j^{-2} (\partial_x \Phi_j)^2] + \sum_j \epsilon_j N_j, \quad (19)$$

where  $\epsilon_j = \sum_\nu T_\nu \Gamma_{\nu j}$ . The “zero modes”  $N_j = (4/\pi)^{1/2} \int dx \partial_x \Phi_j$  denote the total number of particles in the  $j$ th eigenmode. The eigenvalues  $g_j$  and the matrix  $\Gamma_{\nu j}$  are determined from Eq. (9) with  $U_{nm}$  being replaced by  $W_{\nu\nu'}$ . From Eq. (19), an important effect of the hopping consists of a splitting of the  $N$  1D transport bands. Using  $\sum_j \Gamma_{\nu j} \epsilon_j = T_\nu$ , the TDOS for tunneling into the outermost shell is generally of the form

$$\rho(E) = \sum_\nu Q_{N\nu} Q_{N\nu}^\dagger \rho_\nu(E - T_\nu), \quad (20)$$

where  $\rho_\nu(E) \sim E^{\eta_{\nu,b/e}}$ . The bulk/end exponents  $\eta_{\nu,b/e}$  for tunneling into the  $\nu$ th MWNT eigenmode are given by Eqs. (11) and (12), respectively, with the replacement  $\Gamma_{Nj} \rightarrow \Gamma_{\nu j}$ . For  $U_{nm} = U$ , both exponents are independent of  $\nu$  and equal the previous expressions. For large  $N$ , and taking all  $t_n = t$ , the eigenvalues of  $T_{nm}$  are  $T_\nu = -2t \cos(2\pi\nu/N)$ , with  $Q_{N\nu} Q_{N\nu}^\dagger = 1/N$ . Hence  $\rho(E) \sim N^{-1} \sum_\nu (E - T_\nu)^{\eta_{b/e}}$ , where the threshold energies  $T_\nu$  become narrowly spaced. In effect, for large enough  $N$ , the LL singularities will then be smeared out simply because subsequent thresholds are too closely together. For  $N \approx 5-10$ , however, different  $T_\nu$  should be sufficiently well separated.

What about the other interactions in Eq. (17)? For  $U_{nm} \approx U$ , the corresponding couplings are small and can be studied using the perturbative RG. If three or four indices  $\nu_i$  are different, the resulting perturbations are always highly irrelevant since single-particle excitations for more than two modes are involved in the scattering. The contribution with  $\nu_1 = \nu_3$  and  $\nu_2 = \nu_4$  corresponds to a simultaneous transfer of two fermions from one mode to another, which is also an irrelevant process. The remaining interactions are due to the scaling fields  $B_{\nu\nu'} = B_{\nu'\nu} = -Y_{\nu\nu'\nu}$ . Again omitting irrelevant terms, they cause a weak renormalization of the LL parameters in the neutral channels [which were all equal to one before], and the marginally relevant contribution

$$H' = \frac{v}{2\pi a^2} \sum_{\nu < \nu'} B_{\nu\nu'} \int dx \times \prod_{\gamma} \cos\{\sqrt{\pi}[\varphi_{\gamma,\nu}(x) - \varphi_{\gamma,\nu'}(x)]\} + (\cos \leftrightarrow \sin). \quad (21)$$

Since marginally relevant scaling fields generally lead to exponentially small gaps for small couplings [10], the LL model indeed describes the electronic properties of a MWNT, except at exceedingly low-energy scales below this gap. The predicted pseudogap in a rope [16] may be rationalized by noting that, since there  $U_{nm} \approx U$  is not as accurate, the  $B_{\nu\nu'}$  are presumably larger.

So far, backscattering (BS) has not been incorporated. BS amounts to the coupling of  $2k_F/2q_F$  oscillatory charge densities on different shells. Here only the CDW- $\pi$  (charge-density wave) operators [4] need to be considered since the CDW-0 operator is not relevant. Since symmetries present in the noninteracting case have to be overcome, the CDW- $\pi$  operators are only generated via short-ranged interactions which in turn are reduced by a factor  $a/R_n$  due to the doughnutlike wave functions [4]. Therefore the BS coupling between shells  $n$  and  $m$  will carry a factor  $a^2/R_n R_m$ . From the bosonization analysis, BS now simply leads to a renormalization of the  $B_{\nu\nu'}$  in Eq. (21). Hence the BS gap is suppressed by a factor  $\exp(-R_n R_m/a^2) \approx \exp(-100)$  compared to standard quantum wires and can be safely neglected here.

Another concern is the ionic potential of other shells acting on electrons in the  $n$ th shell. Because of different radii and helicities, neighboring graphite lattices are generally incommensurate, and thus the ionic potential amounts to a quasiperiodic random energy landscape. The elastic mean-free path  $\ell_n$  for electrons in shell  $n$  can be estimated from a disordered tight-binding approach [17] by taking independent random on-site energies with variance  $\sigma_E^2$ . This yields

$$\ell_n = 2\pi\sqrt{3}(V_0/\sigma_E)^2 R_n. \quad (22)$$

For an order-of-magnitude estimate of  $\sigma_E$ , the nearest-neighbor hopping  $t_n$  can be used. Since  $t_n$  is typically one-

tenth of the intratube hopping  $V_0$  [18], Eq. (22) predicts  $\ell_n/R_n \approx 1000$ . For typical MWNT lengths of the order of 1–10  $\mu\text{m}$ , transport is ballistic and the neglect of the ionic potential is justified.

In conclusion, the low-energy theory of an individual MWNT on an insulating substrate has been given. Including the long-ranged Coulomb interaction, pronounced correlation effects are predicted which can be understood in the framework of the Luttinger liquid model. Clearly, there are many open questions to be addressed in future work, e.g., the effects of a parallel or perpendicular magnetic field, or the virtual or real population of higher subbands as the energy scale increases.

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