Orthogonality Catastrophes in Carbon Nanotubes

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Carbon nanotubes provide a remarkably versatile system in which to explore the effects of Coulomb interactions in one dimension. The most dramatic effects of strong electron-electron repulsion are orthogonality catastrophes. These orthogonality catastrophes come in different varieties, and can be observed both in low-bias transport and tunneling measurements on nanotubes. This article contains a review of previous work and new material on the crossover between Coulomb blockade and Luttinger behavior.

1 Introduction and Model

In this section we describe the basic influence of interactions in carbon nanotubes, following Ref. 1. The band structure of metallic nanotubes has been discussed by several authors.\textsuperscript{2,3} It is well-captured by a simple tight-binding model of $p_z$ electrons on the honeycomb lattice. For the metallic tubes, evaluating the resulting tight-binding band structure for the discrete set of allowed quantized transverse momenta $q_y$ leads to only two gapless one-dimensional metallic bands (for the simplest $(N,N)$ armchair tubes, these have $q_y = 0$).\textsuperscript{2,3} These dominate the low-energy physics, disperse with the same velocity, $v_F$, and can be described by the simple 1d free Fermion model,

$$H_0 = \sum_{i,\alpha} \int dx v_F \left[ \psi_{Ri\alpha}^\dagger \partial_x \psi_{Ri\alpha} - \psi_{Li\alpha}^\dagger \partial_x \psi_{Li\alpha} \right],$$

where $i = 1,2$ labels the two bands, and $\alpha = \uparrow, \downarrow$ the electron spin. We neglect curvature effects (which could open up small gaps in all but the armchair tubes), since these are subdominant to the Coulomb interactions. Higher sub-bands (for the armchair tube these have $q_y \neq 0$) are of course also present, and will be returned to in Sec. 3.

We will make heavy use of the bosonized representation of Eq. 1 obtained by writing $\psi_{R/Li\alpha} \sim e^{i(\phi_{i\alpha} \pm \theta_{i\alpha})}$, where the dual fields satisfy $[\phi_{i\alpha}(x), \theta_{j\beta}(y)] = -i\pi \delta_{ij} \delta_{\alpha\beta} \Theta(x - y)$. Expressed in these variables (1) takes the form

$$H_0 = \sum_{i,\alpha} \int dx v_F \left[ \frac{\psi_{Ri\alpha}^\dagger \partial_x \psi_{Ri\alpha}}{2\pi} \right].$$

The slowly varying electronic density in a given channel is given by $\rho_{i\alpha} \equiv \psi_{Ri\alpha}^\dagger \psi_{Ri\alpha} + \psi_{Li\alpha}^\dagger \psi_{Li\alpha} = \partial_x \theta_{i\alpha}/\pi$. The normal modes of $H_0$ describe long wavelength particle-hole excitations which propagate with a dispersion $\omega = v_F q$.

Turning to the interactions, a tremendous simplification occurs when $N$ is large: the only couplings which survive in this limit are forward scattering processes which involve small momentum transfer. Roughly speaking, this can be understood as follows. “Interbranch” scattering processes (such as backscattering and umklapp) involve a momentum transfer of order $2k_F \sim 1/a$, where $a$ is the carbon-carbon bond length. The matrix elements are therefore dominated by the short range part of the interaction, at distances $r \sim a$, where the interaction changes significantly from site to site. However, the electrons in the lowest sub-band are spread out around the circumference of the tube, and for large $N$ the probability of two electrons to be near each other is of order $1/N$. For the Coulomb interaction, the resulting dimensionless interaction vertices are of order $(e^2/hv_F) \times 1/N$. By contrast forward scattering processes, in which electrons stay in the same branch, involve small momentum exchange. They are dominated by the long range part of the Coulomb interaction, at distances larger than the radius, and there is no $1/N$ suppression.
For $N > 10$ it is thus appropriate to consider a Luttinger model, in which only forward scattering vertices are included. A further simplification arises because the squared moduli of the electron wavefunctions in the two bands are identical and spin independent. All the forward-scattering vertices can thus be written as a single interaction, coupling to the total charge density 

$$\rho_{\text{tot}} = \sum_{i\alpha} \partial_x \theta_{i\alpha}/\pi.$$ 

We will suppose that the Coulomb interaction is externally screened on a scale $R_s$, which is long compared to the tube radius $R$, but short compared to the length of the tube. For simplicity, we model this by a metallic cylinder of radius $R_s$, placed around the nanotube. From elementary electrostatics, the energy to charge the nanotube with an electron density $\epsilon \rho_{\text{tot}}$ is

$$H_{\text{int}} = e^2 \ln(R_s/R) \int d\rho_{\text{tot}}^2.$$  \hspace{1cm} (3)

Since $H_{\text{int}}$ only involves $\rho_{\text{tot}}$ it is convenient to introduce a spin and channel decomposition via, $\theta_{i,\rho/\sigma} = (\theta_{i\uparrow} \pm \theta_{i\downarrow})/\sqrt{2}$ and $\theta_{\mu\pm} = (\theta_{1\mu} \pm \theta_{2\mu})/\sqrt{2}$ with $\mu = \rho, \sigma$, and similar definitions for $\phi$. As defined, the new fields $\theta_a$ and $\phi_a$ with $a = (\rho/\sigma, \pm)$, satisfy the same canonical commutators $[\phi_a(x), \theta_b(y)] = -i\pi \delta_{ab} \Theta(x-y)$. In the absence of interactions the Hamiltonian is simply $H_0 = \sum_a \int_{x,\tau} H_0(\theta_a, \phi_a)$, which describes three “sectors” of neutral excitations and one charged excitation. Including the interactions only modifies the charge sector, which is described by the sum of two terms $H_\rho = H_0(\theta_{\rho+}, \phi_{\rho+}) + H_{\text{int}}(\theta_{\rho+})$ which may be written

$$H_\rho = \int dx \frac{v_\rho}{2\pi} \left[ g^{-1}(\partial_x \theta_{\rho+})^2 + g(\partial_x \phi_{\rho+})^2 \right].$$  \hspace{1cm} (4)

This describes the 1d acoustic plasmon which propagates with the velocity $v_\rho = \sqrt{v_F (v_F + (8e^2/\pi\hbar) \ln(R_s/R))}$ and is characterized by the Luttinger parameter $g = v_F/v_\rho$. Taking a screening radius of $R_s = 0.1\mu$, one estimates $g \approx 0.2$.

2 Low-Bias Transport

As evidenced in Eq. 3, the elementary excitations of the Luttinger liquid are collective modes, very different from the quasiparticles of a normal Fermi liquid. The most natural experimental measure of this non-Fermi-liquid physics are those which probe the overlap between these excitations and bare electrons. The simplest way to achieve this is via a low-bias transport experiment in which electrons are removed from a metallic source lead and injected into the nanotube (and vice versa at the drain). Surprisingly, for this purpose it is beneficial to have poor contact between the leads and the tube. In this poorly-contacted limit, transport proceeds by sequential tunneling of individual electrons into and out of the nanotube, and is thus a measure of the single-particle overlap. By contrast, with ideal (adiabatic) contacts, the charge on the tube is no longer a good quantum number, and transport occurs via a collective flow of charge. Despite the significant different between the single-particle spectrum of Luttinger and Fermi liquids, their collective mode spectra are quite similar. Indeed, as discussed by many authors, an ideally contacted pure Luttinger liquid has a two-terminal resistance which is completely unaffected by interactions. The ideal four-terminal resistance of a zero temperature Luttinger liquid is zero, a characteristic of ballistic transport.

We consider the transport through a single tunnel junction, which could be between a normal lead and the nanotube, or between two nanotubes. The former situation has been investigated experimentally and theoretically in Ref. 6 (see the paper by Bockrath for details). In general, the current through a single tunnel junction can be determined perturbatively in the tunneling amplitude $w$. Consider the tunneling Hamiltonian

$$H_{\text{tunn.}}(t) = \sum_{\alpha} w e^{iV t} c_{\alpha}^\dagger c_{\alpha} + w^* e^{-iV t} c_{\alpha}^\dagger c_{\alpha},$$  \hspace{1cm} (5)
where $c^\dagger_{r,l}/c_{r,l}$ create/annihilate electrons to the left and right of the junction. If the right-hand-side is a nanotube, there is in general some tunneling amplitude into each of the Dirac channels: $c_{r\alpha} = \sum_{i=1,2} \sum_{P=R/L} \Phi_P^{(r)} \psi_P^{i \alpha}$. We have chosen to include the voltage difference between the two leads here as a time-dependent vector potential. The current across the junction follows from gauge invariance: $I = \partial L/\partial a_x$, where $a_x$ is the component of the vector potential normal to the junction. The first non-zero contribution to the current in time-dependent perturbation theory in $\omega$ is

$$\langle I(t) \rangle \propto |\omega|^2 \int_{-\infty}^{\infty} e^{iV(t-t')} A_{\text{ret}}(t-t') \equiv A_{\text{ret}}(V),$$

where $A_{\text{ret}}$ is the standard retarded correlation function of the operator $\hat{A} = c^\dagger_{l\alpha} c_{r\alpha}$. Since the overall magnitude of the current depends on an unknown transmission probability, we will simplify various formulae by omitting constant prefactors where possible. The retarded correlator is obtained in the usual way by analytic continuation from the imaginary-time correlator $A(\tau) = \langle T_{\tau} A(\tau) A(0)^\dagger \rangle$. In particular,

$$A_{\text{ret}}(V) = \sinh(\beta V/2) \int_{-\infty}^{\infty} dt e^{-iVt} A(\frac{\beta}{2} - it),$$

where, as usual $\beta = 1/k_B T$.

### 2.1 Thermodynamic Limit

We first consider the case in which the systems on either side of the contact are semi-infinite. This is a valid assumption for a finite Luttinger liquid provided the temperature or bias voltage is large compared to its capacitive charging energy. For both the case of Fermi-liquid to nanotube and nanotube–nanotube junctions, the imaginary-time correlator above takes the form

$$A(\tau) = \left(\frac{\pi}{\sin \frac{\pi \tau}{\beta}}\right)^{2+\alpha},$$

with $\alpha > 0$, which can be obtained straightforwardly from the bosonization formulae in Sec. 1. This form obtains in many distinct cases – e.g. bulk contact between a Fermi liquid and a nanotube, bulk contact between two nanotubes, and end-to-end contact between two nanotubes. The different cases are distinguished by the value of the exponent $\alpha$. To get a feeling for the physical meaning of $\alpha$, imagine a Fermi’s golden rule estimate of the current across the junction. If the initial state is the ground state, the final state consists of e.g. a hole in the left lead and an electron in the right. Fermi’s golden rule gives a density of states factor which is the product of the electron density of states on the right and the hole density of states on the left. Indeed, a direct calculation of the tunneling density of states $\rho_{r/l}^{\text{tun}}$ for a Luttinger liquid gives the formula $\alpha = \alpha_l + \alpha_r$, where $\rho_{r/l}^{\text{tun}}(\epsilon) \sim |\epsilon|^{\alpha_r/2}$.

The density of states exponent depends crucially on the Luttinger parameter $g$, and also on the point of contact. In particular,

$$\alpha = \begin{cases} (g^{-1} - 1)/4 & \text{near cap} \\ (g + g^{-1} - 2)/8 & \text{in bulk} \end{cases}$$

Note that, since $g < 1$, the orthogonality exponent $\alpha$ is significantly larger near the end of a nanotube. This enhanced interaction effect arises essentially from the decreased ability of an added charge to spread away from the cap area relative to the bulk. A contact may be considered an end contact if the distance from the end of the nanotube is smaller than the lesser of the characteristic length scales $L_T = \hbar v_F/k_B T, L_V = \hbar v_F/eV$. It is important to realize that this
is not a small effect: for $g \ll 1$, the orthogonality exponent near the cap is nearly twice that in the bulk.

Using Eq. 8 and Eq. 9, one obtains the $I$–$V$ curve

$$I = I_0 T^{1+\alpha} \sinh(\beta V/2) \left| \Gamma \left( 1 + \frac{\alpha}{2} + i \frac{\beta V}{2\pi} \right) \right|^2,$$

where $I_0 \propto |\psi|^2$ is an unknown prefactor and $\Gamma(z)$ is the Gamma function. This form is well-known from the literature on tunneling and Caldeira–Leggett models. A slightly more complicated formula can be obtained for the differential conductance $G = dI/dV$ by differentiation; differentiating only the $\sinh(\beta V/2)$ prefactor gives a common approximate result (as the remainder is small at low biases). Eq. 10 has an important scaling property. The quantities $I/T^{1+\alpha}$ and $G/T^\alpha$ are functions only of the ratio $V/T$. This implies that IV curves taken at different temperatures should collapse if, e.g. $G/T^\alpha$ is plotted as a function of $V/T$. See the paper by Marc Bockrath in this volume for an experimental observation of this behavior.

### 2.2 Charging Effects

Coulomb blockade is a ubiquitous phenomena in mesoscopics physics. For a few micron-long nanotube, the capacitive charging energy can be of the order of tens of Kelvin, and affects the transport dramatically at low temperatures. The crossover between Luttinger liquid and Coulomb blockade behavior is a complex and relatively unstudied problem. We will discuss a simple model of charging effects for a single tunnel junction between two nanotubes. The charging behavior is, unfortunately, much less universal that the Luttinger liquid results of the previous subsection, so we must make a number of assumptions to progress.

A panoply of energy scales exist in a finite Luttinger liquid. The largest is generally the charging energy. In the Luttinger model, this is the energy of a zero mode in which each of the phase fields $\theta$ winds by $\pm \pi/2$. For $g \ll 1$, this is dominated by the charge mode $\theta_\rho$, and from Eqs. 3–4, $E_C \approx \pi \hbar v_F/8g^2L \approx (e^2/L) \ln R_\text{LS}/R_\text{LS}$. For a one-dimensional system, the level spacing also scales inversely with the length. In particular, the Luttinger liquid exhibits two such level spacings due to spin-charge separation: a plasmon energy $\varepsilon_\rho = \pi \hbar v_F/L$ and a single-particle energy $\varepsilon_0 = \pi \hbar v_F/L$. For $g \ll 1$, one has $E_C \gg \varepsilon_\rho \gg \varepsilon_0$. We will therefore consider only the largest of these energies, $E_C$. This corresponds physically to including charging effects but not individual level quantization.

Within this charging-only model, we will consider the specific case of a single tunnel-junction between two finite-length nanotubes. The treatment here is valid provided that the resistance of the nanotube-nanotube junction is much larger than the resistance to the leads. In this case, at least for not too low temperatures, transport is dominated by the internal tunnel junction. We require $R_{\text{junction}} \gg R_{\text{leads}} \gg \hbar/e^2$.

To proceed, we must specify the capacitances in the system. In general, the charging energy is given by an energy function $E_{n_1,n_2}$, where $n_1$ and $n_2$ are the charges (in units of $e$) of the left and right sides of the system. Given such a function, Eq. 9 can still be applied, but $A(\tau)$ must be recalculated. Because the charges are zero-mode quantities, the statistical average simply factors into a product of a zero-mode contribution and the thermodynamic contribution in Eq. 8.

Inserting into Eqs. 3–4, one obtains

$$I = I_0 \frac{T^{1+\alpha}}{Z} \sinh(\beta V/2) \sum_{n_1,n_2} e^{-\beta E_{n_1,n_2}} \left| \Gamma \left( 1 + \frac{\alpha}{2} + i \frac{\beta (V - \Delta E_{n_1,n_2})}{2\pi} \right) \right|^2,$$

where the “partition function” is $Z = \sum_{n_1,n_2} e^{-\beta E_{n_1,n_2}}$. We have also defined the energies $E_{n_1,n_2} = \frac{1}{2} (E_{n_1+1,n_2-1} + E_{n_1,n_2})$, $\Delta E_{n_1,n_2} = E_{n_1+1,n_2-1} - E_{n_1,n_2}$. In general, a quadratic energy
Figure 1: Ground state charge configurations in the $m, n$ plane, assuming $W < U$. Inside each hexagon the charge state is indicated by the ordered pair $(n_1, n_2)$. For $W > U$, the pattern is rotated 90 degrees and there are no horizontal segments. Some possible trajectories as the gate voltage is varied are indicated schematically by the dashed red lines.

The function $E_{n_1, n_2}$ contains five non-trivial parameters. It is helpful to consider a simplified form,

$$E_{n_1, n_2} = U(n_1 + n_2)^2 + W(n_1 - n_2 - m)^2.$$  \hspace{1cm} (12)

Here $W$ can be viewed as a capacitive charging energy for the junction itself, while $U$ gives an “bulk” contribution. A gate potential is included via $m, n$. Changing a single gate potential corresponds to motion along some straight line in the $m, n$ plane. Eqs. (11)-(12) encompass a tremendously rich behavior. Consider the Coulomb blockade regime at low temperature. At zero temperature, the ground state charge configurations of the system (for $w = 0$) are shown in Fig. 1. As $m, n$ are varied, the system makes two types of transitions between different charge states. Across the diagonal lines, one electron is added or removed from either the left or right nanotube, changing the net charge (and spin) on both tubes. Across the horizontal lines, one electron is transferred from the right to the left tube or vice versa, leaving the total charge unchanged. In the limit we are considering, transport is dominated by tunneling across the inter-tube junction. Peaks in the zero-bias conductance thus occur only across the horizontal lines.

As an external gate potential is varied, the system traces out a straight line in the $m, n$ plane. The particular slope and intercept of this line is determined by the precise geometry of the tubes and the gate. The system will thus generically undergo a sequence of ground-state transitions, some of which (crossing the diagonal lines) do not give rise to conductance peaks, but which change the charge and spin. Such “internal” transitions have indeed been observed in nanotubes.\footnote{Moreover, depending upon the particular path taken, the locations of Coulomb blockade peaks as a function of gate voltage (crossings of horizontal lines) can be quite complex, e.g. quasiperiodic or containing a fairly regular series of peaks followed by a region of zero conductance.}

Let us return to the problem of the crossover between Coulomb blockade and Luttinger behavior, contained in Eqs. (11)-(12). A general analysis is beyond the scope of these proceedings. We therefore content ourselves with a few simple situations. The most “universal” limit is the high-temperature case, basically when $T > W$ (note that $W$ and not $U$ sets the size of the energy that an electron must obtain to cross the tunnel junction). In this case, the sum in Eq. (11) can
be approximated by an integral, and
\[
I \approx I_0 2^{-\alpha} \pi \Gamma(2 + \alpha) T^{1+\alpha} \sinh(\beta V/2) e^{-\beta W} \int_0^\infty dt \frac{e^{-4 \beta W t^2} \cos \beta V t}{(\cosh \pi t)^{2+\alpha}}. \quad (13)
\]

At very high temperatures, the exponential terms in \( W \) are negligible and the result reduces to Eq. 10. An appealing feature of Eq. 13 is its independence of \( m, n \) and \( U \).

Unfortunately, the low-temperature limit is much less universal. As an illustration of the application of Eqs. 11-12, consider the behavior of the zero-bias conductance \( G(T) \) as a function of temperature when the gate voltage is tuned to a zero-bias conductance peak. In this case, one finds power-law behavior at both low and high temperatures: \( G(T) \sim G_0(W/T)^{1+\alpha} \). The prefactor, however, has the amusing property that \( G_0(W/T \gg 1) = \frac{G_0(W/T \ll 1)}{2} \). The factor of two comes physically from the fluctuations of the system between two generate ground states at low temperatures, and reflects the zero-temperature entropy of the two degenerate ground states of the system! Amusingly, a more detailed examination shows that \( G(T)/T^{\alpha} \) actually crosses non-monotonically between these limits.

### 3 High-Bias Tunneling Spectra

We have seen in the previous section how Coulomb interactions give rise to numerous interesting non-linearities in low-bias tunneling transport. Tunneling spectra also provide a powerful probe of high-energy excitations. In nanotubes, tunneling experiments have verified the presence of van Hove singularities in the density of states (at \( eV \) energies) due to the higher sub-bands. In non-interacting band theory, these singularities are of inverse square-root form, giving a contribution \( \rho_0(\epsilon) \sim \sqrt{m(\epsilon - \Delta)}^{-1/2} \Theta(\epsilon - \Delta) \) for energies just above the subband edge at \( \epsilon = \Delta \) (\( m \) is the subband effective mass).

How do interactions affect these van Hove singularities? A simplified, though unphysical model in which the mass of the higher subband is taken to be infinite provides considerable insight. In this limit the higher energy subbands can be replaced by discrete, localized levels. The “x-ray edge” problem of a localized level interacting with a conduction sea was solved by Nozieres and de Dominicis, and is one of the first demonstrations of an orthogonality catastrophe. Physically, the core hole is “dressed” through interactions with conduction electrons, which see the hole as a scattering center. This leads to a broadening and reduction of the tunneling density of states from a sharp delta-function to a power law singularity.

Remarkably, these x-ray edge effects persist even for this finite mass case, as we now proceed to demonstrate. We argue that a necessary and sufficient condition for the presence of such finite-energy singularities is a conserved quantum number distinguishing the states of the higher subband from the conduction states. In the case of the carbon nanotube, this is an angular momentum quanta. If such a distinguishing quantum number is absent (as might occur in a nanotube due to breaking of the rotational symmetry by interactions with a substrate), we expect the van Hove peak to be rounded and rendered completely nonsingular.

We describe here a simple forward-scattering model of the interaction of the conduction electrons with the higher subband – other interaction channels are discussed in Ref. 13. Near the putative van Hove singularity, the unoccupied 1d subband can be described by a non-relativistic electron operator \( d, d^\dagger \):
\[
H_0^d = \int dx d^\dagger_x \left[ -\frac{1}{2m} \frac{\partial^2}{\partial x^2} + \Delta \right] d_x. \quad (14)
\]
Here \( \Delta \) is the gap to the first subband and \( m \) is an effective mass. The electron field satisfies \( \{d_\alpha(x), d^\dagger_\beta(x')\} = \delta_{\alpha\beta} \delta(x - x') \). In the case of a carbon nanotube, there are actually multiple degenerate subbands at energy \( \Delta \). This degeneracy is unimportant within the forward-scattering model, as the tunneling DOS involves only states with a single excited electron.
The interaction Hamiltonian is \( H_{\text{int}} = \int x \mathcal{H}_{\text{int}} \), with

\[
\mathcal{H}_{\text{int}} = e^2 \ln(R_s/R) \left[ (d^\dagger d)^2 + \frac{4}{\pi} \partial_x \theta_{\rho^+} d^\dagger d \right].
\]  

(15)

To understand the effects of \( \mathcal{H}_{\text{int}} \), consider the canonical transformation

\[
\begin{align*}
\theta_{\rho^+}(x) &= \tilde{\theta}_{\rho^+}(x) - \gamma \int_{-\infty}^{x} dx' d^\dagger(x') d(x'), \\
\tilde{d}(x) &= e^{i\gamma \phi_{\rho^+}(x)/\pi} \tilde{d}(x),
\end{align*}
\]

(16) (17)

where \( \gamma = \pi (1 - g^2)/2 \). Eqs. 16-17 embody the physical process in which the conduction sea adiabatically adjusts to the heavy particle. In particular, Eq. 16 represents the depletion of the conduction electron density near the heavy particle due to Coulomb repulsion. Eq. 17 represents phase shifts of these conduction electrons when the heavy particle is introduced. Formally, the exponential of the dual \( (\phi) \) field in Eq. 17 is a Jordan-Wigner “string” operator which has been attached to the heavy particle.

Remarkably, although the canonical transformation, Eqs. 16-17, does not remove the interaction completely, it does transform the Hamiltonian into one with only irrelevant couplings in the renormalization group sense. This indicates that at long times and distances, the transformed fermion and boson correlation functions asymptotically factorize. From this result, one straightforwardly obtains a modified van Hove singularity, gives a modified van Hove singularity:

\[
\rho(\epsilon) \sim \rho_0 \left( \frac{\Delta}{\epsilon - \Delta} \right)^{\frac{2}{3} - \beta} \Theta(\epsilon - \Delta),
\]

(18)

where \( \Theta(x) \) is the heavyside step function, and the orthogonality exponent \( \beta = (1 - g^2)^2/(8g) \approx 0.3 \) for typical metallic nanotubes.

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