Intrinsic Coulomb blockade in multi-wall carbon nanotubes

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Abstract

Carbon nanotubes provide a new class of molecular wires that display new and exciting mesoscopic transport properties. We provide a detailed theoretical description for transport in multi-wall nanotubes, where both disorder and strong interactions are important. The interplay of both aspects leads to a particularly effective intrinsic Coulomb blockade for tunneling. The relation to recent experiments is discussed.

Key words: Carbon nanotubes, Coulomb blockade, Quantum wires

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1 Introduction

Transport in molecular wires has received a lot of attention during the past decade. Besides fundamental interest, much of the relevance of this field comes from potential applications in the realm of molecular electronics [1]. In this article, we will focus on one specific class of molecular wires, namely carbon nanotubes [2]. Nanotubes provide a remarkable and exciting arena for mesoscopic transport phenomena involving strong electron correlations. The primary quantity theoretically analyzed below is the energy-dependent tunneling density of states (TDOS) for tunneling into the nanotube, \( \nu(E) \sim \text{Re} \int_0^\infty dt e^{iEt} \langle \psi(t)\psi^\dagger(0) \rangle \), where energies are measured relative to the Fermi level \( E_F \) (we put \( \hbar = 1 \)). The energy dependence of the TDOS directly governs the (nonlinear) conductance of a nanotube connected to an STM tip or to metallic leads via bad contacts (tunnel junctions). It also determines the intrinsic conductance in the presence of strong impurity backscattering or weak links. From a more fundamental point of view, the TDOS provides
precious information about the importance of Coulomb interactions and electronic correlations in such a molecular wire. Recent experiments on individually contacted single-wall nanotubes (SWNTs), which are composed of a single wrapped graphite sheet, have convincingly established the ballistic (essentially defect-free) nature of electronic transport in SWNTs over distances of the order 1 µm and beyond [3,4]. Due to their small radius $R \approx 1$ to 2 nm, SWNTs are characterized by strong transverse momentum quantization, and under normal circumstances only two spin-degenerate transport bands are present at the Fermi level (assuming a metallic SWNT). In such a one-dimensional (1D) conductor, the electron-electron interactions are expected to be of crucial importance. In fact, the field-theoretical analysis [5,6] predicted the breakdown of conventional Fermi-liquid theory. On not too low energy scales, in practice meaning that the temperature should be above the sub-mK regime, a Luttinger liquid (LL) phase should emerge in SWNTs. The LL is the generic phase of interacting 1D electrons, and is characterized by the absence of Landau quasiparticles, implying a smeared Fermi surface. In addition, a LL exhibits spin-charge separation, electron fractionalization, and anomalous transport properties [7]. Recent SWNT experiments [8–12] reported clear evidence for the elusive LL behavior of 1D interacting fermions. These charge transport experiments have measured the TDOS for tunneling into the SWNT. According to LL theory, the energy dependence of the TDOS is a power law, with an exponent that explicitly depends on whether one tunnels into the end or into the middle of the SWNT. The predicted exponents have by now been observed experimentally with good precision [9–12]. For a review on the status of theory and experiment regarding electronic correlation effects in SWNTs, see Ref. [13].

The situation is more complex and controversial for multi-wall nanotubes (MWNTs), which are the focus of this article. Existing experimental observations for MWNTs do not seem to easily fit into the framework of well-established theories for disordered electrons. The reason appears to be linked to the presence of strong electron-electron interactions as will be exemplified below for the case of the TDOS. The structure of this article is as follows. In Sec. 2, we summarize basic transport properties of MWNTs and the experimental situation. In Sec. 3, phenomenological Coulomb blockade theory is reviewed, which is then given a microscopic justification based on a nonlinear $\sigma$ model calculation. The theoretical predictions for the TDOS, in particular the numerical solution of the Coulomb blockade equations is discussed in Sec. 4. Finally, some conclusions can be found in Sec. 5.
2 Electronic transport in MWNTs

Multi-wall nanotubes are composed of several (typically ten) concentrically arranged graphite shells, with outermost radius of the order $R \approx 5$ to $10$ nm and length $L$ up to several $100 \ \mu m$. It is rather obvious that the main difference between MWNTs and SWNTs, apart from the larger radii of MWNTs, should come from the presence of inner shells. Because of the large radius, the transverse quantization energy is only $v_F/R \approx 0.2$ eV, where the Fermi velocity is $v_F = 8 \times 10^5$ m/sec, and therefore one needs to be careful about the position of the Fermi level. While in an undoped MWNT, electron-hole symmetry enforces $E_F = 0$, in basically all tubes studied so far a rather strong doping effect was present, $|E_F| \approx 0.3$ to $0.5$ eV [14]. The physical origin of the doping is largely open, but may be a result of charge transfer from oxygen or from the substrate or the attached leads. This means in practice that typically 20 spin-degenerate subbands are present (instead of only two as in SWNTs), and therefore doped MWNTs correspond to multi-channel molecular quantum wires. Looking for the moment only at the outermost shell, the bandstructure of a perfect (clean) tube corresponds to a Dirac “light cone”, $E(\vec{k}) = v_F|\vec{k}|$, around each of the two gapless K points [2], with $\vec{k} = (k, k_{\perp})$ and quantized transverse momentum, $k_{\perp} = n/R$, where $n = -N, \ldots, N$ and $N = \lfloor kFR \rfloor$. The number $M = 2N + 1 \approx 10$ of spin-degenerate 1D subbands at each K point arising from periodic boundary conditions around the circumference is determined by the doping (Fermi) level via $k_F = |E_F|/v_F$. The $n$th subband is then characterized by a separate Fermi velocity, $v_n = v_F\sqrt{1 - (n/k_FR)^2}$, and Fermi momentum, $k_n = k_Fv_n/v_F$. For clarity, we focus on doped MWNTs throughout this article, where $M \gg 1$.

With one exception [15], available experiments agree that electronic transport in MWNTs is not ballistic (as in SWNTs) but diffusive [16–26]. Estimates for the mean free paths differ substantially in different studies, and seem to depend on many aspects, e.g. MWNT fabrication, purification and preparation, as well as the energy regime probed experimentally. To mention some of the experimental evidence for diffusive behaviors, there are typical weak localization features, universal conductance fluctuations and $h/2e$ oscillations in the magnetoconductance. These experiments also show that electronic transport proceeds only through the outermost shell (which is contacted by external leads), unless that shell has been intentionally damaged. There are a number of theoretical arguments supporting this observation [27–29], and we shall assume an effective single-shell model in what follows. Inner shells then cause a screening of the electron-electron interaction potential. For a computation of the TDOS, the latter effect, as well as the spin and K point degeneracy, can be absorbed by a suitable renormalization of the electron-electron interaction potential $U_0(\vec{q})$ [28]. Therefore we may simplify the computation and consider only spinless electrons at one K point. In addition, since different
shells always have incommensurate lattices due to different curvature or helicity, a quasiperiodic ionic potential from inner shells acts on outermost-shell electrons. The effect of such a potential is expected to be similar to a random potential described by a mean free path $\ell = v_F \tau$. Moreover, true disorder imposed by imperfections, substrate inhomogeneities, tube processing or defects is very likely present. Together with the inner-shell potential, this may explain the reported mean free paths, $\ell \leq 100$ nm. Estimating the localization length as $\xi \approx 4M\ell$ using the standard Thouless argument, electronic transport at low energy scales, $E\tau \ll 1$, is therefore diffusive (but not localized) for not exceedingly long doped MWNTs. However, for $E\tau \gg 1$, there is a ballistic regime, where the Luttinger liquid picture [28] is appropriate again, with minor modifications. In this article, we will mainly focus on the more complex energy regime $E\tau \ll 1$, i.e. assume a sufficiently dirty MWNT. In fact, we impose the condition $\ell \lesssim 2\pi R$ such that transport around the circumference can be considered as diffusive. The opposite limit $\ell \gtrsim 2\pi R$ has recently been addressed in Ref. [30].

At low energy scales, $E < v_F/R$, many groups have by now experimentally observed pronounced zero-bias anomalies in the TDOS of an individual MWNT [18,20–24,26]. Most of these experimental results are described by a power-law TDOS, $\nu(E) \propto E^\alpha$, just as in a LL, with exponents clustering around $\alpha \approx 0.3 \pm 0.1$ [18,21,23,24]. Remarkably, this value is of the same order of magnitude as the exponents in a SWNT and hence the interpretation in terms of a LL may seem obvious. One exception to this result has been reported in Ref. [20], where $\alpha \approx 0.04 \pm 0.02$. Occasionally, also logarithmic dependencies have been observed [26] in MWNT bundles, where probably the electron-electron interaction is externally screened. Such logarithmic dependencies could be explained by tunneling into an effectively 2D diffusive system with weak Coulomb interactions [31]. Furthermore, the TDOS at the end of the MWNT, while still of power-law form, is characterized by a doubling of the boundary exponent, $\alpha_{\text{end}} = 2\alpha$. Unfortunately, it appears to be difficult to explain these findings by Luttinger liquid theory, at least for the majority of the quoted experimental studies. Although LL theory can be extended to ballistic multi-mode wires with inner-shell screening [28], the presence of many subbands in a doped MWNT inevitably implies rather small exponents. Even optimistic estimates yield exponents that are at least one order of magnitude smaller than observed. For that reason, below we address the role of disorder for the zero-bias anomaly.

Building upon our original paper [29], we provide a theoretical description for the TDOS of MWNTs within the general framework of Coulomb blockade theory [32]. We focus on sufficiently low energy scales $E < v_F/R$, where it is sufficient to take a fixed number $M$ of subbands, and thereby ignore van Hove singularities associated with the opening of new 1D subbands as energy is varied. As is shown below, on intermediate energy scales, an apparent power law
suppression of the TDOS is found, which is distinct from the LL power laws of a ballistic system. This intrinsic Coulomb blockade phenomenon arises because of the suppression of tunneling into an strongly interacting disordered metal. Because of strong interactions, a perturbative Altshuler-Aronov-Lee (AAL) approach [31] is not possible. The corresponding nonperturbative problem for 2D systems has been studied in Refs. [33–37]. Very recently, besides our own paper [29], the 1D case has attracted considerable interest in the theory community [30,38,39].

3 Intrinsic Coulomb Blockade

The key ingredient in Coulomb blockade theory is the probability \( P(E) \) that a tunneling electron excites electromagnetic modes with energy \( E \) in the system [32]. The theory is meaningful if these modes are harmonic, and then \( P(E) \) directly determines the TDOS according to a relation first explicitly given in Ref. [38],

\[
\frac{\nu(E)}{\nu_0} = \int_{-\infty}^{\infty} dE' \frac{1 + \exp(-E/k_B T)}{1 + \exp(-E'/k_B T)} P(E - E') ,
\]

where \( \nu_0 \) is the non-interacting DOS. The probability \( P(E) \) is expressed as

\[
P(E) = \frac{1}{\pi} \text{Re} \int_0^\infty dt \exp[iEt + J(t)] .
\]

with the phase correlation function

\[
J(t) = \int_0^\infty d\omega \frac{I(\omega)}{\omega} \{ \coth(\omega/2k_B T)[\cos(\omega t) - 1] - i \sin(\omega t) \}
\]

for a given spectral density \( I(\omega) \) of electromagnetic modes. As a probability, \( P(E) \) is normalized,

\[
\int_{-\infty}^{\infty} P(E) = 1
\]

and fulfills the detailed balance relation, \( P(-E) = \exp(-E/k_B T)P(E) \).

For clarity, we now focus on the most interesting zero-temperature case. Provided \( I(\omega) \) remains finite for low frequencies, Eqs. (1) and (3) straightforwardly
lead to a power law for the TDOS with exponent $\alpha = I(\omega \to 0)$. We then should establish the harmonic nature of the electromagnetic modes and compute the low-frequency spectrum $I(\omega)$. If $I(\omega \to 0)$ is finite, a power law would directly follow. Notice that a perturbative treatment of interactions is apparently not sufficient, as a power-law TDOS is inconsistent with conventional perturbative (1D or 2D) AAL predictions [31]. In some studies [23,24], $I(\omega)$ is phenomenologically parameterized in terms of the total impedance $Z(\omega)$, i.e. the MWNT is modelled as a transmission line. Under such an approach, one obtains $\alpha = Z(0)/(h/2e^2)$. This simple transmission line model can directly explain the doubling of the end exponent, because in the bulk case one has effectively two resistances in parallel as compared to the end case. However, since this purely phenomenological approach can hardly represent a satisfactory theory, we pursue a microscopic approach. It should also be stressed at this point that the 1D pseudo-gap TDOS found for small $E$ [29,30] is apparently outside the reach of transmission line modelling. Furthermore, the doubling of the end exponent can be verified from microscopic theory as well. The derivation is discussed at length in Ref. [29], and here we focus on the bulk TDOS alone.

Recent field-theoretical developments allow to incorporate the Coulomb interactions in a nonperturbative way [33,36,37]. Adapting the Keldysh nonlinear $\sigma$ model approach for interacting disordered systems worked out in Ref. [37], the TDOS can be computed in analytical form for arbitrary interaction strength. This calculation is certainly on sound footing for long-ranged interactions (which is the case for MWNTs) in 2D. For truly 1D systems, however, the asymptotic low-energy behavior of the TDOS resulting from this approach, see Ref.[30], has been questioned recently [36]. In any event, the final result yielded by this theory indeed reproduces phenomenological Coulomb blockade theory, since the electromagnetic modes are found to be Gaussian with spectral density [29,37]

$$I(\omega) = \frac{\omega}{\pi} \text{Im} \sum_q \frac{1}{(Dq^2 - i\omega)^2} \left( U_0^{-1}(q) + \frac{\nu_0 Dq^2}{Dq^2 - i\omega} \right)^{-1}.$$  \hspace{1cm} (5)

Here the diffusion coefficient for charge diffusion on the tube surface is $D = v_F^2 \tau/2$. In Eq. (5), the $q$ summation includes an integral over the momentum parallel to the tube direction (a very long MWNT is assumed), and a summation over the discrete transverse momenta $q_\perp = n/R$ for integer $n$. For consistency, the $n$ summation is restricted to $|n| \leq N$, albeit the detailed value for the cut-off is not essential. The Fourier-transformed Coulomb interaction potential $U_0(q)$ includes the effect of external screening by nearby gates or the substrate, but not of internal screening which is fully accounted for by Eq. (5). In what follows, to keep the discussion simple, we consider an effectively short-range interaction potential characterized by a constant $U_0$; for the
case of a $1/r$ potential, see, e.g. Ref. [37]. Since the dominant contributions to $I(\omega)$ come from small $q$, it is justified to integrate over the longitudinal momentum in Eq. (5) directly (there is no UV divergence), leading to

$$I(\omega) = \frac{U_0}{2\pi(D^* - D)} \operatorname{Re} \sum_{n=-N}^{N} \left[ (-i\omega/D^* + n^2/R^2)^{-1/2} - (-i\omega/D + n^2/R^2)^{-1/2} \right]$$

with the field diffusion constant $D^* = D(1 + \nu_0 U_0)$. Although we do not present the derivation of Eq. (5) here, we feel it is important to summarize the main approximations entering this result:

1. The regime $\ell \lesssim 2\pi R$ is considered, but Eq. (6) should also yield useful results for somewhat larger $\ell$, since then the $n = 0$ mode dominates the Coulomb blockade completely. The $n = 0$ mode is unaffected by assumptions concerning transversal motion.

2. As a consequence of the assumed diffusive behavior, the spectral density (6) should only be used for $\omega \tau \ll 1$. The $I(\omega \to 0) \sim \omega^{-1/2}$ behavior in Eq. (5) due to the $n = 0$ mode implies that the dominant contribution to the Coulomb blockade indeed results from these low-energy collective modes. In Eq. (3) we therefore truncate the integration at the upper limit $1/\tau$. Since the higher energy modes are equivalent to a Luttinger liquid, which in turn only leads to comparatively weak Coulomb blockade effects in this regime, their omission is not expected to create serious problems.

3. The Coulomb interaction potential should be sufficiently long-ranged and smooth to allow for semi-classical (WKB-type) treatments. The main effect of the interaction is then to change the phase of electron wave-functions but not the amplitude. Since Coulomb blockade is a low-energy collective phenomenon and the interaction potential is rather long-ranged in MWNTs, this approximation should be justified.

4. Only the intrinsic electrodynamic modes of the MWNT are considered to be responsible for the Coulomb blockade, but not the attached external circuit. For sufficiently long MWNTs such as the ones in Ref. [23], the intrinsic resistance is large and environmental Coulomb blockade can safely be neglected.

Above the Thouless energy for diffusion around the circumference,

$$E_T \approx \frac{D}{(2\pi R)^2},$$

where charge diffusion is essentially two-dimensional, we then expect basically an exponentiated 2D AAL law. In contrast, for sufficiently low energy scales, $E < E_T$, 1D behavior takes over, where the $n = 0$ mode in Eq. (6) becomes
more and more important as the energy scale is decreased. In these two limits one can obtain the TDOS analytically. In the 2D diffusive energy regime $E > E_T$, a power law emerges by converting the $n$-summation into an integral. The bulk exponent follows directly as $\alpha = I(\omega \to 0)$, and reads [29]

$$\alpha = \frac{R}{2\pi \nu_0 D} \ln(D^*/D).$$

(8)

In effect, the AAL logarithmic correction therefore exponentiates into a power law [29]. We wish to stress that the derivation of Eq. (8) works only in the true 2D limit, characterized by a large number of bands $M$ or by $\ell \ll R$. On the other hand, for sufficiently low energy scales, only the $n = 0$ mode responsible for 1D perturbative AAL corrections is important. Keeping only the $n = 0$ term in Eq. (6) results in a divergent (“sub-Ohmic”) behavior of the low-frequency spectral density, $I(\omega) \sim \omega^{-1/2}$, and hence to the appearance of a pseudo-gap as $E \to 0$ [40],

$$\nu(E) \sim \exp(-E_0/E),$$

(9)

where we neglect a prefactor exhibiting power-law energy dependence. Alternatively, this leads to logarithmic corrections of the scale $E_0$ in Eq. (10) below. The result (9) agrees with the findings of Refs. [30,38]. Using a stationary-phase evaluation of $P(E)$ in Eq. (2), the energy scale $E_0$ in Eq. (9) follows as [40]

$$E_0 = \frac{U_0^2}{8\pi D \left( 1 + \sqrt{D^*/D} \right)^2}.$$  

(10)

For strong interactions, $D^* \gg D$, this can be simplified to $E_0 = U_0/8\pi \nu_0 D$. Remarkably, Eq. (9) representing the exponentiated 1D AAL law does not reproduce the perturbative 1D AAL $1/\sqrt{E}$ behavior under a naive direct expansion of the exponential [30,38].

4 Numerical solution of Coulomb blockade equations

To analyze the full energy dependence of the TDOS, numerical methods are mandatory. Using the spectral density (6), the phase correlation function $J(t)$ in Eq. (3) can be evaluated, which then allows for the computation of $P(E)$ according to Eq. (2). Finally, Eq. (1) yields the TDOS. For simplicity, we again focus on the $T = 0$ limit, although the finite-temperature case is also
directly accessible. A convenient check that the numerical procedure has converged is provided by the normalization condition for $P(E)$, see Eq. (4), which is accurately fulfilled for the results reported below. For convenience, the energy scale is set by $v_F/R$ throughout this section. We consider a situation with $k_F R = 5.5$ so that $N = 5$ and the number of bands is $M = 11$, and use the value $U_0/2\pi v_F = 1$ to parameterize the interaction strength. Unfortunately, it appears to be rather difficult to compute a realistic value for $U_0$ (except possibly by ab-initio methods). The above choice corresponds to rather strong interactions, but for comparison we have also carried out calculations for $U_0/v_F = 1$ (not shown, but see below).

Let us start with a rather dirty MWNT, $\ell = R$. In the mentioned units, the Thouless scale is $E_T = 0.013$, and the plot of the TDOS in Fig. 1 indeed shows an apparent power law even for energies well below $E_T$, extending up to $E \approx 0.1$ over approximately one decade. The inset shows that for $E \to 0$, the predicted pseudo-gap behavior emerges. For this parameter set, the power-law exponent $\alpha = 0.23$ predicted from the 2D limit, Eq. (8), is much smaller than the numerically observed exponent $\alpha = 1.97$. The estimate (8) is therefore too crude and really restricted to the true 2D regime, since it ignores the special role played by the $n = 0$ contribution in the spectral density.
Nevertheless, we find a clear power-law behavior at intermediate energy scales. Importantly, the regime of validity for the power law is not set by the Thouless scale, but by a smaller energy scale. Similarly, the value $E_0 = 0.015$ extracted from the slope of the Arrhenius plot in the inset of Fig. 1 is significantly smaller than the value $E_0 = 0.07$ predicted by Eq. (10). This deviation is probably linked to strong logarithmic renormalizations of the scale (10) by the power-law prefactor not written out in Eq. (9), and is always observed in our calculations. Since this renormalization of $E_0$ makes this scale quite small, the pseudo-gap TDOS is in practice difficult to distinguish from the power law except at very low energies. This may offer a (somewhat trivial) explanation for the experimental difficulties encountered in finding pseudo-gap behavior. Finally, for high energies close to $v_F/R$, the non-interacting DOS is approached.

Next we discuss the quasi-ballistic limit, taking $\ell = 10R$. The features shown in Fig. 2 are qualitatively similar as in Fig. 1, namely a pseudo-gap at very low energies turns into a power law at intermediate energies. The power law exponents become systematically smaller by increasing $\ell$, in this case $\alpha = 1.1$. The power law crosses over into the pseudo-gap as $E \to 0$, with $E_0 = 0.00025$ again much smaller than the expected value $E_0 = 0.007$. As this power law feature is definitely not linked to the Thouless scale, it is not related to the
exponent (8) for tunneling into a 2D interacting disordered metal. We have checked for this value of \( \ell \) that the power law persists for smaller \( U_0 \). In fact, the exponent \( \alpha \) then systematically decreases, and for \( U_0/v_F = 1 \) is \( \alpha \approx 0.3 \), which would be in good agreement with experiment. Since the interaction strength in the experiments conducted in Ref. [23] did probably not vary much from tube to tube, the robustness of the observed exponents with respect to changes in \( \ell/R \) is encouraging. We mention in passing that for suspended MWNTs or smaller doping levels, one may reach a regime of stronger interactions, where again power-law behavior at intermediate energies is predicted, but with larger exponents.

5 Conclusions

MWNTs represent a unique laboratory for exploring mesoscopic physics in the presence of electron-electron interactions. Here we have addressed one aspect, namely the zero-bias anomaly of the tunneling density of states due to Coulomb interactions among the electrons. Assuming a sufficiently dirty MWNT with mean free path less than the circumference, the spectral density \( I(\omega) \) of Coulomb blockade theory has been computed, and the numerical solution of the resulting equations for the TDOS was presented. The results show power-law behavior at intermediate energy scales, extending down to quite low energies over typically one to two decades. Remarkably, the power law is seen at energies less than the Thouless scale for diffusion around the circumference, i.e. it does not appear to reflect tunneling into a 2D diffusive electron liquid. At very low energies, the power-law behavior \( \nu(E) \sim E^\alpha \) crosses over into a pseudo-gap of the form \( \nu(E) \sim \exp(-E_0/E) \), although it should be stressed that in practice both are sometimes hard to distinguish because of the rather small scale \( E_0 \). According to our numerical study, the power-law exponent \( \alpha \) would be consistent with typical exponents around \( \alpha \approx 0.3 \) for interaction strength \( U_0/v_F \approx 1 \), but is significantly larger for stronger interactions. We note that the power law is very robust, and has always been observed in our calculations, regardless of the chosen values for \( \ell/R \) or \( U_0 \).

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