Measuring Fractional Charge in Carbon Nanotubes

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The Luttinger model of the one-dimensional Fermi gas is the cornerstone of modern understanding of interacting electrons in one dimension. In fact, the enormous class of systems whose universal behavior is adiabatically connected to it are now deemed Luttinger liquids. Recently, it has been shown that metallic single-walled carbon nanotubes are almost perfectly described by the Luttinger Hamiltonian. Indeed, strongly non-Fermi liquid behavior has been observed in a variety of DC transport experiments, in very good agreement with theoretical predictions. Here, we describe how fractional quasiparticle charge, a fundamental property of Luttinger liquids, can be observed in impurity-induced shot noise.

I. Introduction - The study of interacting electrons in one dimension has opened a panoply of surprises which often seem counter-intuitive from the perspective of higher dimensions. The generic behavior of one-dimensional metals is that of the Luttinger liquid.\textsuperscript{[1,2]} In a Luttinger liquid, interactions conspire to host bizarre phenomena such as the separation of spin and charge, as well as anomalous power-law dependences in the resistivity and density of states. Perhaps the most fundamental difference between the Luttinger liquid and higher-dimensional metals lies in the nature of its quasiparticles. The Luttinger quasiparticles are “fractionalized”, and indeed the elementary charged quasiparticle carries not the quantum of charge ‘c’ of the electron, but instead, a fraction ‘gc’.

While the exploration of Luttinger liquid physics began more than a century ago\textsuperscript{[3]}, it has only found its way into the experimental realm in the past decade. The quantum Hall system with its chiral edge states has championed displaying Luttinger liquid features\textsuperscript{[4]}. In particular, it has provided the definitive confirmation of the existence of fractional charge through shot noise measurements\textsuperscript{[5]}. However, the challenge of finding a truly one-dimensional (1D) system of interacting electrons has persisted. A variety of transport experiments\textsuperscript{[6,7]} now convincingly demonstrate that single-walled carbon nanotubes (SWNTs) behave as Luttinger liquids, as predicted theoretically\textsuperscript{[8,9]}.

Here, we show that even the simple set-up of a clean armchair nanotube with a single weak impurity is capable of flaunting a hallmark of Luttinger liquids, namely charge fractionalization. To understand the physical meaning of charge fractionalization, consider a simple thought experiment in which electrons are sequentially transferred from a metallic electrode onto the end of a nanotube through a large contact barrier. With a sufficiently high barrier, the rate of charge addition can be made very low, so that each incident electron can be considered individually. Immediately after the addition event, the added charge e travels as a solitonic pulse away from end of the nanotube (incidentally, the added spin h/2 travels as a separate slower soliton behind the charge). This charge e soliton may be regarded as the remnant of the electron in the nanotube. Upon reaching an impurity, however, the true nature of the charge excitations of the Luttinger liquid becomes evident. In a non-interacting system, an incident particle either transmits (with probability T) or reflects (with probability R = 1 – T). In the nanotube, the charge e soliton can still transmit (with probability T), but the alternative possibility (which is the leading-order scattering process occurring with probability ≈ 1 – T for a weak impurity) is to ‘split’ into two solitons: a backscattered piece of charge ge and a transmitted piece of charge (1 – g)e. The dimensionless “Luttinger parameter” g (g < 1 for repulsively interacting Fermi systems) depends on the nature of the interactions in the system. In carbon nanotubes, theoretical estimates give g ≈ 0.2\textsuperscript{[10]}, in good agreement with transport measurements\textsuperscript{[11,12]}.

Unfortunately, the difficulty of the necessary time-resolved measurements makes the above thought experiment impractical. Nevertheless, as shown in the next section, the mathematics of Luttinger liquid theory leaves no room to doubt that such strange scattering events indeed occur. In this paper, we determine the consequences of these processes for shot noise. Strikingly, we find that at low enough temperatures, fluctuations in the net current incident on the weak barrier have the shot noise form appropriate for Poisson distributed scattering events of particles of charge ge. Measurements of such shot noise are more tractable, and can provide definitive proof of charge fractionalization.

As elucidated in what follows, to experimentally observe the described shot noise, we propose a four terminal set-up capable of measuring correlations in current CI, and the voltage drop V across an impurity placed along the nanotube. In the limit of zero temperature, we derive the relationship CI = 4g2e2hV; CI is related to the backscattered current IB via CI = geIB, while IB is given by IB = 4g2e2hV. Thus one can extract the charge...
fraction $g$. The SWNT, with its estimated value $g \approx 0.2$ far from unity, makes for an exquisite playground to test the predicted Luttinger liquid physics, specifically, the fractionalization of charge.

II. Formalism - To extract the non-equilibrium physics of the set-up described above, we formulate an effective time-dependent theory for the bulk of the nanotube and for the impurity site. The effective theory of the clean single-walled nanotube in consideration may be described by the low energy physics of the $(N, N)$ armchair tube. In the absence of interactions, this involves two gapless one-dimensional metallic bands modeled by free fermions with linear dispersion $[14]$:

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

where $v_F$ is the Fermi velocity, $R$ and $L$ label the right and left movers respectively, $i = 1, 2$ label the bands, and $\alpha = \uparrow, \downarrow$ the electronic spin. In this section, we set $\hbar = e = 1$.

The bosonized version of the fermionic operators has the form $\psi_{R/Li\alpha} \sim e^{i(\phi_{R/Li\alpha} \pm \theta_{i\alpha})}$. A more convenient basis, which we employ extensively, involves a spin and channel decomposition for $\theta$: $\theta_{i\alpha/\sigma} = (\theta_{i\uparrow} \pm \theta_{i\downarrow})/\sqrt{2}$ and $\theta_{i\alpha} = (\theta_{i\uparrow} \pm \theta_{i\downarrow})/\sqrt{2}$ with $\mu = \rho, \sigma$, and a similar one for $\phi$. The new fields obey the canonical commutation rules $[\phi_{\alpha}(x), \theta_{\beta}(y)] = -i\pi\delta_{\alpha\beta}\Theta(x-y)$, with $a, b = (\rho, \sigma, \sigma, -\sigma)$. As discussed in Ref. [10], interactions effectively involve just the charge density, $\rho = \frac{2}{\pi} \partial_t \theta_{\rho \uparrow}$.

The entire Hamiltonian density ($H = \int dx \, \mathcal{H}$), with interactions taken into account, then has the bosonized form:

$$\mathcal{H} = \sum_a \frac{v_a}{2\pi} \left( g_a^{-1} (\partial_x \phi_a)^2 + g_a (\partial_x \phi_a)^2 \right),$$

with corresponding equations of motion

$$\left( \partial_t \pm v_a \partial_x \right) n_a^{R/L} = 0.$$  \hspace{1cm} (4)

Thus, the density propagates as a one-dimensional acoustic plasmon with renormalized velocity $v_{\rho \uparrow}$. The parameter $g$ depends on the ratio of the Coulomb energy between particles and the Fermi energy, and in a SWNT has the approximate value of $0.2$ [10].

We now consider the effect of a single weak impurity at the origin ($x = 0$). In this limit, a small portion of quasi-particle backscatter, and the role of fractional charge is most transparent. In the generic case involving no spin polarization or spin flip, local weak backscattering processes may be described by:

$$H_{\text{imp}} = \sum_\alpha \left\{ \sum_{i=1,2} u_i \left[ \psi_{Ri\alpha}^\dagger \psi_{Ri\alpha} + \text{h.c.} \right] + u_3 \left[ \psi_{R\alpha}^\dagger \psi_{L\alpha} + \text{h.c.} \right] + u_4 \left[ \psi_{R\alpha}^\dagger \psi_{L\alpha} + \text{h.c.} \right] \right\}$$  \hspace{1cm} (5)

where $\alpha = \uparrow, \downarrow$, 'h.c.' denotes Hermitian conjugation, $u_1$ and $u_2$ are weak intra-subband scattering potentials, and $u_3$ and $u_4$ describe the inter-subband scattering. Processes associated with $u_1$ and $u_2$ conserve all particle numbers while the $u_3$ and $u_4$ scattering terms do not conserve $\rho$- and $\sigma$- particle numbers (these arise physically for impurities which break the sublattice-reflection symmetry of the graphene lattice).

We note that the bosonized version of Eq.(5) may be expressed in terms of right and left moving creation and annihilation operators $e^{\pm i\phi_{R/L}}$ that describe the freely propagating chiral excitations of the system; the impurity site can create, destroy, or backscatter these excitations. Most importantly, every scattering process possesses a term of the form $e^{\pm i\phi_{R/L}} e^{\mp i\phi_{L}}$, reflecting the fact that a quasiparticle characterized by the creation operator $e^{-i\phi_{R/L}}$ is always backscattered. As detailed in Ref. [12], the operator $e^{i\phi_{R/L}}$ creates a kink of magnitude $\pi g$ in $\phi_{R/L}$ at $x = 0$, or equivalently, a peak in $n_{\rho \uparrow}$ of magnitude $g$. Therefore, the magnitude of the fractional charge associated with the impurity backscattering is ‘$g$’.

Finally we consider the real time, finite temperature action applicable at the impurity site. The manner in which we employ it parallels the treatment in Ref. [13]. We integrate out the $\phi_a$ variables from the bulk Hamiltonian (though, where appropriate, we integrate out $\theta_a$ variables instead), and then integrate out fluctuations away from the impurity as in Ref. [13]. Using the Keldysh approach [13], we write the partition function in terms of time dependent backward and forward paths $\theta_{\pm} = \theta \mp \frac{1}{2} \dot{\theta}$:

$$Z = \int \prod_a D\theta_a^- D\theta_a^+ e^S,$$  \hspace{1cm} (6)

with $a = (\rho, \sigma, \sigma, -\sigma)$. The action $S = S_0 + S_1 + S_2$ is given by

$$S_0 = -\sum_a \frac{1}{\pi g_a} \int d\omega \coth \left( \frac{\omega}{2kT} \right) |\hat{\theta}_a(\omega)|^2 + \frac{2i}{\pi g_a} \int dt \hat{\theta}_a(t) \dot{\theta}_a(t),$$

$$S_1 = -i \sum_{j\sigma} \int dt [f(G_{j\sigma}^+(t)) - f(G_{j\sigma}^-(t))],$$

$$S_2 = \frac{i}{\pi} \int dt [A(t) \dot{\theta}_{\rho \uparrow}(t) + \eta(t) \theta_{\rho \uparrow}(t)].$$  \hspace{1cm} (7)
Here, $S_0$ describes the unperturbed system. $S_1$ is derived from the impurity Hamiltonian of Eq. (6). The $\Gamma_{js}$ operators are defined for $j = 1 \ldots 4$ and $s = \pm 1$:

$$\Gamma_{1s} = \theta_{\rho+} + s\theta_{\sigma+} + \theta_{\rho-} + s\theta_{\sigma-},$$

$$\Gamma_{2s} = \theta_{\rho+} + s\theta_{\sigma+} - \theta_{\rho-} - s\theta_{\sigma-},$$

$$\Gamma_{3s} = \theta_{\rho+} + s\theta_{\sigma+} + \phi_{\rho-} + s\phi_{\sigma-},$$

$$\Gamma_{4s} = \theta_{\rho+} + s\theta_{\sigma+} - \phi_{\rho-} - s\phi_{\sigma-},$$

where the $\pm$ superscripts which denote backward and forward paths are suppressed for all variables. $S_2$ originates from coupling the physical current to an external source $A$.

### III. Physical Properties

![FIG. 1. Quasiparticle transport in a nanotube with a single impurity. The current enters the nanotubes through the external contacts $A$ and $B$ while the two voltage probes $L$ and $R$ serve to measure the voltage drop across the impurity. Chiral modes are shown for clarity of expression, but they cannot be probed separately as the voltage probes couple to both right- and left-movers.](image)

An analysis of the nanotube with a single impurity, schematically shown in Fig. 1, serves to bring out striking Luttinger liquid features. However, in the absence of the impurity, the conductance measured across the external contacts $A$ and $B$, which we assume to be adiabatic, is $G = 4\frac{e^2}{h}$ [16], appropriate for a non-interacting one-dimensional system with four channels for conductance. Effectively, this is due to the fact that though an isolated nanotube would have an associated conductance $4\frac{e^2}{h}$, the external metallic contacts are three-dimensional Fermi liquids, and the observed conductance involves electrons backscattering at the interface [16].

Weak backscattering in the presence of the impurity causes a reduction in the conductance. For a temperature $kT \gg \frac{\Delta}{2}\ell$ ($\ell$ is Boltzmann’s constant), where $\ell$ is the distance from the impurity to the nearest contact ($A$ or $B$), the reduction has the form $\delta G(T) \propto -u^2T^{-2\Delta-2}$, where $\Delta = \frac{1}{2}(g + 3)$, and ‘$u$’ is the impurity strength [12]. Given the estimate $g = 0.2$ for the nanotube [16], $\delta G(T) \propto T^{-0.4}$ ought to be observable across a wide temperature range. Similar considerations for the limit of large tunneling barrier, where only few electrons tunnel through, show that an infinite wire reflects electrons completely at $T = 0$. However, at finite temperature it exhibits the temperature dependence $G \propto T^{2\lambda-2}$, where $\lambda = \frac{1}{4}(\frac{1}{2} + 3)$.

As seen in Fig. 1, the current contributing to the conductance involves right moving quasiparticles emerging from the left lead, and left movers from the right lead. As emphasized by the chiral decomposition of the previous section, the quasiparticles carry fractional charge $ge$, and a portion of them is backscattered into the lead from which they emerge. In the presence of an externally applied potential, the right and left moving chiral modes maintain a difference in chemical potential, $V_1 - V_2 - V$. The potential difference $V_{12} = V_1 - V_2$ between the chiral modes arises in the presence of an external bias voltage. In the absence of the impurity, the current $I_0$ flowing across the wire is given by $I_0 = 4ge^2\hbar V_{12}$. Unfortunately, unlike in the quantum Hall case, where the right- and left-movers are spatially separated, and $V_{12}$ is measurable [16], here the leads couple to both modes. Thus, $V_{12}$ cannot be measured, and the ideal conductance $4\frac{e^2}{h}$ cannot be extracted.

The voltage drop $V$ is caused by the backscattering of quasiparticles. The net current traversing the wire in the presence of the impurity is given by

$$I = I_0 - I_B = 4ge^2\hbar(V_{12} - V),$$

where $V$ is the voltage drop across the impurity, and $I_B = 4ge^2\hbar V$ is the backscattered current. Also, as we work in the weak backscattering limit, we have $I_B \ll I_0$. Equation (8) can be either derived from the action of Eq. (7) by calculating the average current $I = \langle \overline{2e\delta G} \rangle$ as the functional derivative $-\frac{\delta \langle \overline{2e\delta G} \rangle}{\delta g}$, or by simple consideration of chiral mode properties, as in the case of spinless fermions [12]. In our set-up, we find that $V_{12} = \frac{h}{e}A$, and $V$ is the expectation value of the voltage operator given by

$$\dot{V} = \frac{1}{4} \sum_{js} u_j \sin(\Gamma_{js} + gA),$$

with $\Gamma$’s defined in Eq. (8).

The role of fractional charge is made manifest in the shot noise generated by the quasiparticles striking the impurity. To derive the general behavior of $C_I$ at finite temperature, we define the correlation function $C_O$ in the quantity ‘$O$’;

$$C_O(\omega) = \frac{1}{2} \int dt e^{i\omega t} < \{O(t), O(0)\} >,$$

and use Eq. (7) as in Ref. [13] to obtain a relation between current and voltage fluctuations, $C_I$ and $C_V$ respectively. At low temperature, and in the limit of zero frequency, this becomes

$$C_I = \left(4ge^2\hbar\right)^2 C_V(\omega) + 2kT \frac{dI}{dV_{12}} - \left(4ge^2\hbar\right)2kT \frac{dV}{dV_{12}}.$$
We then perturbatively calculate the voltage correlations to lowest non-vanishing order in the impurity scattering potential to obtain

$$C_V(\omega \rightarrow 0) = \frac{h}{4e} \coth \left( \frac{geV_{12}}{2kT} \right) < \hat{V} > . \quad (13)$$

We observe that the noise due to voltage fluctuations is partitioned between four channels. Putting together $\text{Eq.}(9),(12)$ and $(13)$, we obtain the desired form of $C_I$:

$$C_I(\omega \rightarrow 0) = ge \coth \left( \frac{geV_{12}}{2kT} \right) I_B + 2kT \frac{dI}{dV_{12}} - 2kT \frac{dI_B}{dV_{12}} . \quad (14)$$

Setting $T = 0$, we see that $C_I$ has the celebrated shot noise form $geI_B$. It exhibits crossover from shot noise to thermal noise when the condition $geV_{12} \approx 2kT$ is satisfied. Eq. $(14)$ offers a tractable starting point for experimental data analysis.

IV. Experiment -

![Diagram](image)

FIG. 2. Experimental set-up: a nanotube with an impurity at point ‘O’ is connected in series with an ammeter and a d.c. source supply. The current enters into the nanotube through the external contacts $A$ and $B$. A voltmeter is connected across probes $L$ and $R$.

A possible experimental configuration which realizes Fig.[1] is shown in the four-probe geometry of Fig.2. As elucidated in what follows, two sets of measurements, one in the absence of the impurity and one in its presence, enable one to extract the shot noise $C_I$ of Eq.$(2)$, and the voltage $V$ of Eq.$(3)$ generated across the impurity. These two quantities in turn suffice to extract the charge fraction ‘$g$’.

In the proposed set-up, an external supply maintains a voltage bias between the ends of the nanotube and drives the current through it, while an ammeter measures this net current $I$. A voltmeter across $LR$ measures the difference in potential $V_{LR} = V_L - V_R$ (see Fig.3). As measurements involve both the absence and the presence of the impurity, this would have to be created in a controlled way such as by means of an STM tip [1].

The conditions in which the experiment ought to be performed are rather specific, but feasible. We assume that the probes $L$ and $R$ are non-invasive in their contribution to voltage drops, and that backscattering at all leads is small. The small backscattering condition is equivalent to the requirement that the two-terminal resistance of the entire wire is close to the ideal value of $\frac{\hbar}{e^2} \approx 6.25k\Omega$. We also require the further condition $eV_{12} , kT > \frac{\hbar v_F}{4}$. This ensures that the one-dimensional physics of the tube is probed as opposed to that of the three-dimensional external contacts, in essence that quasiparticle propagation is not coherent across the entire system. Finally, we make the reasonable assumption that heat generated in the external circuit is removed by phonons, hence maintaining steady temperature $T$, and that the noise in it has the current-independent Johnson-Nyquist form $C_{ext} \sim \frac{kT}{R_{ext}}$, where $R_{ext}$ is the resistance of the external circuit [17].

Determining $C_I$, the noise across the impurity, proves a tricky task due to multiple noise sources, and particularly due to possible correlations with scattering at the leads. But taking into account the conditions assumed above for $\ell$ and for the transmission at the leads, as well as the fact that the weak impurity also has large transmission, we find the noise to be additive. In other words, one can measure the current noise in the circuit in the presence and absence of the impurity, $C_I^{imp}$ and $C_I^{0}$ respectively, keeping the mean current in the circuit fixed. Then, the shot noise across the impurity would just be their difference: $C_I = C_I^{imp} - C_I^{0}$.

One could determine the voltage $V$ generated across the impurity by merely measuring the voltage $V_{LR}$ of Fig.2 across it if the leads $L$ and $R$ coupled to right- and left-movers (see Fig.[1]) symmetrically. However, as shown below, even if this coupling is asymmetric, for fixed current $I$, the voltage $V$ is simply the difference between the voltages $V_{LR}$ and $V_{LR}^{0}$ measured across $LR$ in the presence and in the absence of the impurity, respectively. To see this, let $a_R$ and $1 - a_R$ be the fractions with which the right lead couples to the right- and left-movers respectively, and similarly for the left lead. We assume $a_R$ and $a_L$ are voltage-independent, a reasonable assumption given the extremely large ($O(eV)$) electronic energy scales involved in the microscopic matrix elements which determine this coupling. With reference to the voltages shown in Fig.3, we then have

$$V_R = a_R(V_1 - V) + (1 - a_R)V_2,$$
$$V_L = a_LV_1 + (1 - a_L)(V_2 + V),$$
$$V_{LR} = V + (a_L - a_R)(V_1 - V_2 - V), \quad (15)$$

with the total current in the circuit given by

$$I = 4eV^2(V_1 - V_2) . \quad (16)$$

In the absence of the impurity, setting $V = 0$, we have
\[ V_{LR}^0 = (a_L - a_R)(V_1^0 - V_2^0), \]  

and the corresponding current

\[ I_0 = 4g \frac{e^2}{h}(V_1^0 - V_2^0). \]  

Finally, for fixed current \( I = I_0 \), Eqs. (15-18) give the required form for \( V \),

\[ V = V_{LR} - V_{LR}^0. \]  

Now, in the limit \( kT \ll V_{12} \) (which is of comparable magnitude to the actual applied voltage), Eq. (20) takes the form \( C_I = geI_B \). Since \( I_B = \frac{4e^2}{h}V \), we have

\[ C_I = 4g^2 e^3 h V. \]  

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