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# Phase coherent transport in ropes of single-wall carbon nanotubes

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To study the phase breaking scattering events in single-wall carbon nanotubes (SWNTs), ropes of SWNTs are intentionally damaged by $Ar^+$ ion-milling. Due to this treatment, the average distance an electron can travel before being elastically scattered is reduced to about 10nm. This significantly increases the probability of one-dimensional localization and allows us to obtain the phase coherence length ($L_\Phi$) in ropes of SWNTs as a function of temperature. We find that Nyquist *scattering* ($\tau_\Phi \sim T^{-2/3}$) as well as another dephasing mechanism *with* a $\tau_\Phi \sim T^{-1}$ dependence are involved in limiting the phase coherent transport. We also investigate the scattering of hot electrons in the system. The results support the statement that two different scattering mechanisms dominate the phase coherence length for different rope samples.

Extensive studies of the electrical transport in single- and multi-wall carbon nanotubes (SWNTs and MWNTs) have deepened our insight into the electrical properties of one-dimensional metallic and semiconducting systems. The main impact of the reduced dimensionality on electrical transport is the decrease in the phase space available for different kinds of scattering events. In the case of metallic nanotubes it has been shown for example that the scattering of electrons with acoustic phonons follows a $\sim T$ dependence [1,2] instead of a $\sim T^5$ power law as expected for three-dimensional metallic structures in the Grüneisen regime.

While it is possible to investigate momentum changing scattering processes in a standard transport experiment, not every scattering event can be detected in this way. In particular, there are phase breaking mechanisms limiting the phase coherence length $L_\Phi$, such as Thouless scattering, which do not cause backscattering of the current carrying electrons and therefore cannot be measured in such an experiment. A common way to obtain information about $L_\Phi$ in two- and three-dimensional samples is to evaluate the magnetoresistance in a transport experiment. This approach was successfully used for MWNTs by different groups [3–6] and the analysis of the data clearly showed the two-dimensional character of electron motion in these systems. However, in one-dimensional systems such as SWNTs, no magnetic field dependence of resistance is expected, unless the fields are extremely high such that the dispersion relation of the sample is significantly changed, [7] or the tubes are used as building blocks for more complicated geometries, e. g. a ring geometry. [8] Because of this lack of a direct process for measuring $L_\Phi$, little is known about phase coherent transport in SWNTs.

This paper examines the nature of the major phase breaking scattering mechanisms in single wall nanotubes and the dependence of $L_\Phi$ on temperature. In order to perform a phase sensitive measurement, a rope of SWNTs is intentionally damaged by $Ar^+$ ion bombardment at an energy of 500eV. Simulations [9] suggest that this treatment would reduce the elastic mean free path $L_0$ from several micrometers [10] to $L_0$ = 8 – 10nm due to the creation of carbon vacancies, which are known [11,12] to act as strong scatterers in SWNTs. Because of the drastically increased probability for backscattering, electrons would tend to strongly localize within the damaged metallic tubes. Due to their band-gap, semiconducting tubes play a minor role in transport. Localization leads to an exponential increase of sample resistance with decreasing temperature according to [13,14]:

$$R_d = \frac{L}{L_\Phi} \cdot \frac{h}{2e^2} \cdot \frac{1}{2} \cdot \left(\exp\left(\frac{2L_\Phi}{ML_0}\right) - 1\right) \qquad (1)$$

where A4 is the number of one-dimensional modes, which is 2 in the present case and $L$ is the voltage probe separation provided that the entire rope is uniformly damaged. In contrast to the case of an undamaged sample, the localization length $M \cdot L_0$ is smaller than $L_\Phi$, which means that strong localization dominates the transport. Since the phase coherence length itself generally shows a strong dependence on temperature, while none of the other parameters in equation (1) do, measurement of $R_d(T)$ provides a powerful tool to study $L_\Phi(T)$.

We have prepared in total 13 nanotube rope samples in the following way. The ropes were dispersed in dichloroethane using mild sonication, followed by several stages of centrifugation. The purified nanotube ropes were dispersed on an oxidized silicon substrate and gold electrodes were subsequently fabricated on top of the ropes. The key feature of our experiment is the sputtering of the rope before deposition of the electrodes as described above. Only the areas underneath the contacts were damaged. Sputtering of the entire rope instead reduces the sample conductivity to such an extent, that even when the voltage probes are only a few hundred nanometers apart, the rope resistance is in the GΩ-range. This means that when using eq. (1), we have to keep in mind that $L$ must be identified with only the damaged areas. Contributions to the resistance from the undamaged segments of the tube are significantly smaller.

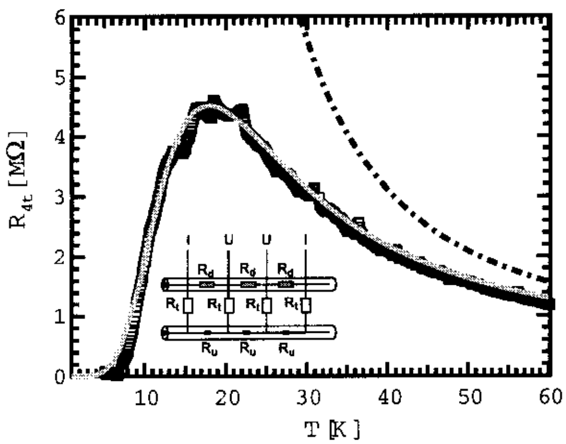


FIG. 1. Four terminal resistance vs. temperature of a sputter-damaged rope. The squares represent the measured data. The grey, solid line is a fit according to our model with $L_\Phi \sim T^{-0.5}$. The resistor network used to model the data is shown as an inset with $R_d$, $R_u$ and $R_t$ as described in the text. The dash-dotted line is the bare contribution from $R_d$ for the same phase coherence length dependence on $T$.

Fig. 1 shows a representative four-terminal measurement (squares) on a rope processed as described above. Data were taken using a standard lock-in configuration. Due to the nature of the four-terminal measurement, contributions to the resistance by the contacts are excluded. In addition, we ensured that no sample heating was caused by the current by keeping the voltage drop across the whole sample always well below $k_BT$. Compared with an undamaged rope, [10] we find an average resistance at room-temperature approximately 1000 times larger, consistent with the $\sim$ 1000 times smaller elastic mean free path in the damaged rope. In addition, we observe a drastic increase of the resistance $R_{4t}(T)$ with decreasing temperature. This is expected, since $L_\Phi$ in general increases with decreasing $T$, and thus $R_d$ increases accordingly. All 13 rope samples show a similar temperature dependence, while the actual resistance values vary from rope to rope.

Fig. 1 shows that there is also an intriguing decrease in the sample resistance at lowest temperatures, which is observed for all rope samples. Recently [15] we showed that the decrease in $R_{4t}$ below about 12K is due to direct tunneling from one metallic tube to another one inside the rope. (For a detailed discussion on the coupling between SWNTs in a rope see Stahl and co-workers [15].) A simple resistor network as sketched in the inset of Fig. 1 was found to adequately describe the damaged rope system. In this network, a damaged metallic tube (with resistance segments $R_d$) and an undamaged metallic tube (with resistance segments $R_u$) parallel to each other and linked through tunneling resistances $R_t$ carry the current through the rope. Since the Ar-sputter treatment only damages the top part of the rope there are always both damaged and undamaged tubes present in our ropes. The general idea is that the current can be switched from a damaged to an undamaged tube as the temperature is lowered. This change of current path is responsible for the non-monotonic behavior (see Ref. 15).

Using this ansatz we are able to obtain excellent fits - see grey straight line in Fig. 1 - to our measurements. The contribution to $R_{4t}$ from $R_d$, the localization in the damaged tubes, is displayed as a dash-dotted line. It is obvious that the whole network has to be taken into account to obtain a good fit to the experimental results. Let us consider the fitting parameters used in the network analysis. $R_d$ from eq. (1) has only one adjustable parameter $L_\Phi$. Our simulations give an $L_0$ of about 10nm. $R_t$ is the coupling resistance between a damaged and an undamaged metallic tube. Since tunneling was found [15] to mediate the transport from tube to tube, $R_t$ is constant and, more importantly, can easily be determined from the local maximum in the $R_{4t}$ versus temperature plot. Finally, the resistance of the undamaged tube $R_u$ is much smaller than both $R_t$ and $R_d$ and thus has almost no impact on the fit. This leaves essentially *one* fitting parameter - which is $L_\Phi$ - to account for the resistance behavior and in particular for the increase of resistance for decreasing temperature.

Using the above resistance network model we analyzed the temperature dependence of $L_\Phi$ for our samples. We assumed a power law temperature dependence of the scattering time: $\tau_\Phi(T) = \tau_\Phi(1K) \cdot T^{-\alpha}$ where $\alpha$ is determined by the dominant phase breaking mechanism. Since the introduction of defects in our experiment is artificial, Fig. 2 shows the phase coherent scattering time instead of $L_\Phi = \sqrt{D\tau_\Phi}$ (with $D$ being the diffusion constant in the damaged rope). To deduce $L_\Phi$ for an undisturbed system, $\tau_\Phi$ has to be multiplied with the Fermi-velocity $v_F$ ($\sim 10^6$m/s) assuming that the defects introduced have no impact on the phase breaking scattering mechanisms but only change the elastic mean free path. The light grey lines in Fig. 2 are the results extracted from the fits. At $T$ = 1K we find phase coherence lengths of the order of micrometers. Interestingly, the data do not support the idea of *one* specific mechanism dominating the phase coherence in all the ropes. Instead, the lines tend to cluster around $\tau_\Phi \sim T^{-0.66}$ *and* $\tau_\Phi \sim T^{-1}$ (shown in Fig. 2 as black thick lines as a guide to the eyes).

There are several possible phase breaking scattering mechanisms that may be operating.

1) Electron-phonon interaction is known to be

the dominant inelastic scattering process for classical transport between a few degrees Kelvin and room-temperature. [1,6,10] Fig. 2 shows the scattering times $\tau_{\text{el-ph}} \sim T^{-1}$ experimentally found for the scattering of electrons with acoustic phonons in undamaged ropes. [10] Obviously, the phonon scattering rate is not high enough to explain our data. However, we cannot exclude the possibility that the electron-phonon interaction is enhanced in the presence of defects and thus becomes the dominant phase breaking mechanism in ropes of damaged SWNTs.

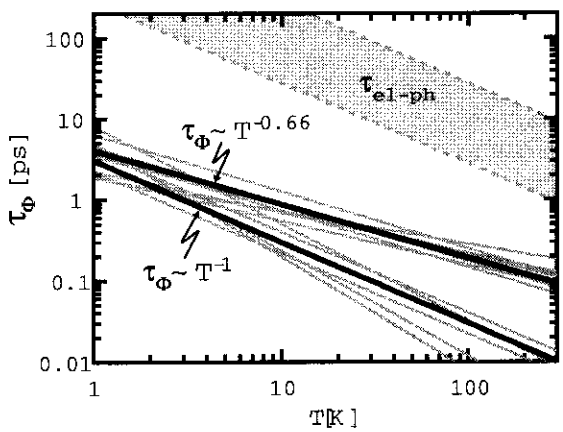


FIG. 2. Extracted phase coherence times $\tau_\Phi$ as a function of temperature (thin grey lines). The black thick lines representing $\tau_\Phi \sim T^{-0.66}$ and $\tau_\Phi \sim T^{-1}$ are guides to the eye. The grey area marked $\tau_{\text{el-ph}}$ is the measured contribution from electron-phonon scattering according to Ref. 10.

2) Electron-electron scattering is also expected to give rise to a scattering time $\tau_{\text{el-el}} \sim T^{-1}$. [16] While electron-electron (e-e) scattering is not visible in a classical transport experiment for regular metals, it can cause backscattering in metallic nanotubes due to the crossing of the one-dimensional bands at the Fermi-points. On the other hand, it was experimentally found [10] that e-e scattering is less effective in backscattering than acoustic phonons. Thus, e-e scattering is unlikely to be the dominant mechanism in limiting $\tau_\Phi$.

3) Nyquist scattering describes the scattering of an electron by the field-fluctuations of the other, surrounding electrons. Due to the statistical nature of these fluctuations such a scattering is different for each electron; thus the electronic ensemble loses its coherence. In one-dimensional systems the scattering time $\tau_{\text{N}}$ is predicted to follow a power law on temperature $\tau_{\text{N}} \sim T^{-2/3}$. [17] This theory is valid for diffusive electron motion and should be applicable in the present case. For MWNTs it was recently found from magnetic field dependent measurements that Nyquist scattering is the mechanism limiting the phase coherent transport for temperatures up to about 50K. [6,18]

4) The Thouless phase-breaking process is not exactly a scattering process. But it limits the coherence due to the thermal smearing of the energies of the electrons. The thermal energy $k_BT$ poses an inevitable uncertainty regarding the energy of the electrons in the ensemble. According to the uncertainty relation this results in a finite "lifetime" for the coherence according to $\tau_{\text{Th}} \sim T^{-1}$.

From the above discussion we conclude that processes 1), 3) and 4) can, in principle, account for the experimental observations. However, the question remains, why do we observe two sets of curves, one with $\alpha \approx 0.66$ and one with $\alpha \approx 1$? Is it just the uncertainty of the analysis of the data in our model that is responsible for the variation we observe?

To address this question, we performed a hot electron injection experiment. In combining an ac with a dc current, we can vary the excess energy of the current carrying electrons relative to $E_F$ and measure the differential resistance through the ac signal. The temperature was kept constant at 1.5K while sweeping the dc voltage.

We argue that while Nyquist scattering with $\alpha = 2/3$ should show a similar dependence on electron excess energy as on temperature, Thouless scattering and electron-phonon scattering both with $\alpha = 1$ should not. Applying a dc voltage creates additional fluctuations because of the larger electric field and thus increases the probability for Nyquist scattering. On the other hand, this is not the case for Thouless scattering and electron-phonon scattering. Although increasing the average energy of the electrons, a dc voltage has no impact on the Thouless scattering probability because the uncertainty of the Thouless energy does not increase. In the case of electron-phonon scattering the number of final states an electron can be scattered into does not scale with the excess energy as long as optical phonons do not get excited. This is due to the fact that there is exactly one final state for electron-phonon scattering available for every initial electron state if temperature smearing is neglected. [19]

Fig. 3 shows measurements for a sample with $\alpha = 0.7$ (sample 1) and one with $\alpha = 1.2$ (sample 2) belonging to the two different classes of slopes we found. For comparison the temperature dependence of both samples are shown on the left side, while the dc voltage dependence is shown on the right. The voltage plotted on the x-axis is the total externally applied voltage. Taking into account the resistance contributions from the current contacts and from the areas outside the two voltage probes, we estimate the internal voltage drop between the voltage probes to be roughly 5 times smaller than the external value of $U_{\text{dc}}$. This means excess energies are always small enough to suppress the impact of optical phonons and any transitions between one-dimensional subbands.

Comparing a) and b), both temperature and electron energy have almost the same impact on the phase coherence length and thus on the strong localization of electrons inside this rope. This behavior matches that predicted for Nyquist scattering, plus the value for $\alpha$ is close to 0.66. On the other hand, sample 2 shows a very different kind of behavior. Plots c) and d) show that applying a dc bias has almost no effect on the sample resistance, while we observe the typical dependence of $R_{4t}$ on temperature. This implies that, in contrast to sample 1, the phase coherence length does not decrease with increasing excess energy. This is expected to be the case for a sample where $L_\Phi$ is dominated by Thouless scattering and/or electron-phonon scattering as suggested by the value of $\alpha$ being close to one. The absence of a significant variation of resistance with $U_{dc}$ supports the statement made above that electron-electron scattering with $\alpha = 1$ is not the dominant phase breaking scattering mechanism. The key point is, that at least two different scattering mechanism are in fact dominating the phase coherent transport through different SWNT rope samples.

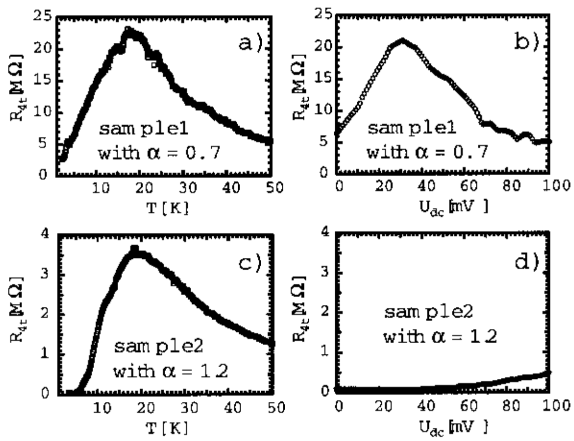


FIG. 3. Comparison between the impact of temperature and electron excess energy on the rope resistance for a sample (a) nd b)) with $\alpha = 0.7$ and for another one (c) and d)) with $\alpha = 1.2$.

We believe that the explanation for the observation of the two groups of slopes could be that metallic tubes with different chirality in the rope samples dominate the transport. The variation in chirality gives rise to dispersion relations exhibiting different values of $k_F$. Electron-phonon scattering as well as Nyquist scattering may depend on $k_F$ and thus the actual interaction strength may vary from rope to rope. On the other hand, it is unreasonable to expect any dependence of scattering time on $k_F$ for Thouless scattering. The observation that the scattering times for $\tau_\Phi \sim T^{-1}$ are always smaller than those for $\tau_\Phi \sim T^{-2/3}$ cannot be consistently explained assuming Thouless scattering since there is no reason why Thouless scattering should be absent for some tubes while being dominant for others. Taking all this into account, a combination of electron-phonon and Nyquist scattering with variable interaction strength is most likely to be responsible for our observations and consistent with the temperature dependence as well as the dependence on electron excess energy.

In conclusion, we have performed a thorough investigation on phase coherent transport in ropes of SWNTs. By intentionally damaging part of the ropes we are able to adjust transport conditions, such that strong localization becomes observable. Applying a simple resistor network enables us to quantify the dependence of phase coherence length on temperature. Two different classes of rope samples were identified. It is found that a dependence of approximately $\tau_\Phi \sim T^{-0.66}$ goes along with a significant impact of dc voltage on the sample resistance which can be explained by Nyquist scattering. On the other side, samples showing roughly a $\tau_\Phi \sim T^{-1}$ dependence do not show a significant impact of resistance on electron excess energy which can be understood in the framework of electron-phonon scattering. Most likely the different chirality of the different metallic tubes carrying the main part of the current in our rope samples is responsible for our experimental findings.

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