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Luther-Emery liquid in the NMR relaxation rate of carbon nanotubes

Balázs Dóra[1,*], Miklós Gulacsi[1], Ferenc Simon[2,3], Pawel Wzietek[4], and Hans Kuzmany[2]

[1] Max-Planck-Institut für Physik Komplexer Systeme, Nöthnitzer Str. 38, 01187 Dresden, Germany
[2] Fakultät für Physik, Universität Wien, Strudlhofgasse 4, 1090 Wien, Austria
[3] Budapest University of Technology and Economics, Institute of Physics and Solids in Magnetic Fields Research Group, Hungarian Academy of Sciences, P.O. Box 91, 1521 Budapest, Hungary
[4] Laboratoire de Physique des Solides, UMR 8502, Université Paris-Sud, 91405 Orsay, France

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* Corresponding author: e-mail dora@pks.mpg.de

We analyze a recent NMR experiments by Singer *et al.* [Singer *et al.* Phys. Rev. Lett. **95**, 236403 (2005).], which showed a deviation from Fermi-liquid behavior in carbon nanotubes with an energy gap evident at low temperatures. A comprehensive theory for the magnetic field and temperature dependent NMR ^{13}C spin-lattice relaxation is given in the framework of the Luther-Emery and Luttinger liquids. The low temperature properties are governed by a gapped relaxation due to a spin gap (~ 30 K), described by the Luther-Emery liquid picture, which crosses over smoothly to the Luttinger liquid behaviour with increasing temperature.

1 Introduction Low dimensional carbonaceous systems, fullerenes, carbon nanotubes (CNTs), and graphene display a rich variety of exotic states and strongly correlated phenomena. These include superconductivity in alkali doped fullerenes [1], quantized transport in single-wall carbon nanotubes (SWCNTs), and massless Dirac quasiparticles showing a halve integer quantum Hall-effect in graphene even at room temperature. A compelling correlated state of one-dimensional systems is the Tomonaga-Luttinger liquid (TLL) state. The TLL state has been suggested to describe the low energy properties of CNTs with a single shell, the single-wall carbon nanotubes [2–6]. Transport [7] and photoemission studies [8, 9] provided evidence for the existence of the TLL state in SWCNTs. In these studies, power-law behavior of temperature and bias dependent conductivity and a power-law Fermi edge was observed, respectively.

Nuclear magnetic resonance (NMR) is a powerful method to characterize correlated states of materials as it is sensitive to the density of states near the Fermi edge. For a material with a Fermi-liquid state, the temperature dependent spin-lattice relaxation time, T_1 follows the so-called Korringa temperature dependence for which $1/T_1T$ is constant. Recently, ^{13}C enriched SWCNTs were grown inside carbon nanotubes from ^{13}C enriched fullerenes [10]. This allowed a high precision measurement of T_1 in small diameter SWCNTs by Singer et al. [11]. A tentative fit of the experiments with a gapped Fermi liquid type density of states (DOS) indicated overall agreement but obvious discrepancies in detail. When the magnetic field and temperature dependent data for T_1 were fitted with this phenomenological model a gap at the Fermi surface with $2\Delta \simeq 40$ K opened already above room temperature, i.e. its T_c is larger than 300 K. This strongly violates the $2\Delta/k_BT > 3.52$ relation, thus simple mean field theories are not applicable [12]. Also, the phenomenological description can not account for the strong overshoot of $1/T_1T$ when T approaches the gap.

Here, we analyze the NMR results in the framework of the Luttinger liquid and Luther-Emery liquid pictures

(interacting one-dimensional electrons without and with a gap, respectively). At high temperatures, the former dominates, while the latter accounts for the dominance of a spin gap at low temperatures. We show that the temperature and magnetic field dependent T_1 of ^{13}C can be explained in the TLL scenario with a small spin gap of the order of 30 K.

The most striking difference between Fermi and Luttinger liquids is the anomalous power-law dependence of correlation functions at low energies in the latter (including the NMR relaxation rate), with critical exponents changing continuously with the interaction strength. This is to be contrasted to a Fermi liquid, where the quasi particle picture (electron) holds, implying critical exponents fixed to an integer. In a Luttinger liquid, low energy properties are described by critical phenomena of collective modes.

In the following, we construct the low energy theory for CNT with the use of bosonization. We determine the various contributions to the NMR relaxation rate at high and low temperatures, and fit the obtained formula to the experimental results.

2 Bosonization of SWNT The Hamiltonian describing the kinetic energy of electrons on SWNT is given by [2–6]

$$H_{kin} = -iv \int dx \sum_{r\alpha\sigma} r\Psi^+_{r\alpha\sigma}(x)\partial_x\Psi_{r\alpha\sigma}(x), \qquad (1)$$

where only two transport bands ($\alpha = \pm$) are taken into account, and for each band, there is a right and left moving branch ($r = \pm$) with linear dispersion, σ stands for the spin.

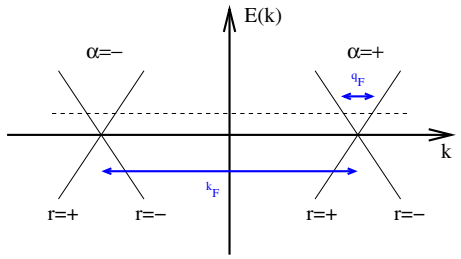

Figure 1 (online colour at: www.pss-b.com) The schematic band structure of a metallic nanotube is shown.

The forward scattering interaction reads as

$$H_{FS} = \frac{1}{2} \int dxdy\rho(x)V(x-y)\rho(y), \qquad (2)$$

where $\rho(x) = \sum_{r\alpha\sigma} \Psi^+_{r\alpha\sigma}(x)\Psi_{r\alpha\sigma}(x)$, $V(x)$ is the Coulomb potential. For the moment, we neglect the umklapp and backward scattering processes. The Hamiltonian is bosonized via

$$\Psi_{r\alpha\sigma}(x) = \frac{\eta_{r\alpha\sigma}}{\sqrt{2\pi a}} \exp\left[i(q_F r + k_F\alpha)x + i(r\theta_{\alpha\sigma} + \phi_{\alpha\sigma})\right], \qquad (3)$$

where k_F denotes the undoped Fermi surface of a graphene layer, q_F determines the band filling [2] and $|q_F| \ll k_F$ to ensure the applicability of the low energy continuum approximation. $\eta_{r\alpha\sigma}$'s are Majorana fermions, standing for the proper anticommutation relations between different branches. The phase fields are expressed by the symmetric and antisymmetric charge and spin modes ($\delta = \pm$) between the valleys of the charge and spin excitations ($j = c, s$) as $2\theta_{\alpha\sigma} = \theta_{c+} + \sigma\theta_{s+} + \alpha\theta_{c-} + \alpha\sigma\theta_{s-}$ and $2\phi_{\alpha\sigma} = \phi_{c+} + \sigma\phi_{s+} + \alpha\phi_{c-} + \alpha\sigma\phi_{s-}$. This leads to the Hamiltonian

$$H_0 = H_{kin} + H_{FS} = \\ = \sum_{j,\delta} \frac{v_{j\delta}}{2\pi} \int dx \left[K^{-1}_{j\delta}(\partial_x\theta_{j\delta})^2 + K_{j\delta}(\partial_x\phi_{j\delta})^2\right], \qquad (4)$$

$v_{j\delta} = v_F/K_{j\delta}$. For long range Coulomb interaction, $K_{c+} \leq 1$ and is usually around 0.2, while for the other modes, $K_{j\delta} \geq 1$, but only slightly greater than unity [2, 8, 9, 13].

Other interaction terms such as umklapp and backscattering have exhaustively been studied in Refs. [2, 5, 6], and we refrain from their analysis here. These parallel closely to the investigation of coupled Luttinger liquid chains. Their effect can be summarized as follows: at half filling, umklapp scattering generates a finite charge (Mott) gap in both the symmetric and antisymmetric charge sector, while spin backscattering leads to the opening of a spin gap in the spin sectors [3–5]. This is analogous to the spin ladder magnon excitations [14] found in the strong antiferromagnetic coupling limit. Thus, the ground state at half filling is a Mott insulator with all excitations gapped [15]. Away from half filling, the $c+$ sector is gapless as discussed before, while all other sectors remain massive. This means, that at low temperatures ($T \ll \Delta$), exponentially activated behaviour is expected [3, 4], while in the high temperature regime ($T \gg \Delta$), the usual Luttinger liquid behaviour is restored. Since the strength of backward and umklapp interactions is inversely proportional to the diameter of the tube, these become important (and generate a gap) only for narrow tubes [2, 3, 13], such as the one in Refs. [10, 11] with an inner tube diameter of 0.7 nm.

The above statements apply mainly to long-range Coulomb interactions [2, 3, 5], which assumes isolated nanotubes. For arrays (ropes), screening becomes important, and Hubbard-like models with short range potentials are invoked [4, 6]. Nevertheless, the previous results, including the presence of a spin gap, hold in this case as well.

3 NMR relaxation rate We start our analysis in the high temperature region to characterize the TLL behaviour first. Hence, we postpone the discussion of the gaps for the moment, and concentrate on the temperature exponents in the NMR relaxation rate, $1/T_1$. In general, $1/T_1$ measures the local dynamics of the spins, and it is related to the transverse spin susceptibility, which in the case of nanotubes,

reads as

$$\chi_\perp(i\omega_n) = -\int_0^\beta \mathrm{d}\tau e^{i\omega_n\tau}\langle T_\tau S^+(x=0,\tau)S^-(x=0,0)\rangle, \tag{5}$$

where $S^\pm(x) = \sum_{r\alpha r'\alpha'} \Psi^+_{r\alpha\pm}(x)\Psi_{r'\alpha'\mp}(x)$. From this, after analytic continuation to real frequencies, the NMR relaxation time is

$$(T_1T)^{-1} \sim \lim_{\omega\to 0} \frac{\mathrm{Im}\chi_\perp(\omega)}{\omega}. \tag{6}$$

In general, there are four different length scales [2] in $S^\pm(x)$, and the corresponding wavevectors are 1. $q_1 = 0$, 2. $q_2 = 2k_F$, 3. $q_3 = 2q_F$ and 4. $q_4 = 2(k_F \pm q_F)$. These show up in the term to $S^\pm(x)$ at 1. $\alpha = \alpha'$, $r = r'$, 2. $\alpha = -\alpha'$, $r = r'$, 3. $\alpha = \alpha'$, $r = -r'$ and 4. $\alpha = -\alpha'$, $r = -r'$, respectively.

In all cases, the corresponding contribution to the susceptibility decays at long times as $\sim 1/t^{\gamma_i/2}$ with the exponents listed in Table 1. Based on Ref. [2], these can be approximated by $\gamma_1 \approx \gamma_2 \approx 2 + 1/K_s + K_s$ and $\gamma_3 \approx \gamma_4 \approx K_c + 2 + 1/K_s$ with $K_s = K_{s+}$ and $K_c = K_{c+}$. In the most general case with broken spin rotational symmetry, all these exponents usually differ from each other. However, from realistic models [3–5], one can conjecture their values as $K_{s-} = K_{c-} = 1$. In the presence of external magnetic field, which couples to the $s+$ field and breaks the spin rotational invariance, marginal interactions change the exponents [13], hence K's can differ from their zero field value. This follows from the exact Bethe-Ansatz solution of the Hubbard model in magnetic field [16] as well. Based on all this, a simple power counting determines the temperature exponents in the NMR relaxation rate as $\sim T^{\gamma_i/2-2}$.

Having determined the high temperature Luttinger liquid behaviour, we now turn to the evaluation of the response function at low temperatures where the gaps are dominating. In general, at $T = 0$, all the correlation functions containing a gapped field acquire a factor $\sim \exp(-\Delta\sqrt{\tau^2 + (x/v)^2})$, which suppresses exponentially the long time-long distance power-law Luttinger liquid behaviour [17, 18]. This picture is further corroborated by the existing exact solutions of the attractive Hubbard model along the Luther-Emery line ($K = 1/2$), where strongly interacting gapped bosons are mapped onto non-interacting fermions [13, 19], i.e. the non-interacting massive Thirring model [20]. In some cases, this mapping also allows for an explicit evaluation of certain correlation functions, such as the finite momentum transverse spin susceptibility [17,20].

At finite temperatures, this simple picture needs to be modified. From the exact solution of gapped systems such as the sine-Gordon model [13, 19], it is well established that different type of excitations contribute to higher frequencies and temperatures [21]. The simplest ones are the solitons and antisolitons (kinks), which, when attract each other, form breathers. Their contribution can systematically be analyzed based on the "Form Factor Bootstrap Approach" [22, 23], which uses the integrability of the underlying model (in the case of the Hubbard model, for example) to determine the matrix elements of the appropriate operators between the ground state and excited states. In this case, a simple exponential term cannot account for their variety.

The explicit determination of these matrix elements is certainly beyond the scope of the present investigation. Instead, we utilize our knowledge about these excitations. At temperatures comparable to Δ, they start to contribute significantly to the response functions. This situation can be mimicked by retaining our simple exponential factor in the correlation functions, and adjusting the gap at each temperature to give reasonable contribution. In other words, this means a $\Delta(T)$ gap function. In spirit, this approach is identical to the self-consistent harmonic approximation [24], where one replaces the complicated interaction term (such as the cosine in the sine-Gordon model) by simpler quadratic ones, and optimizes its coefficient to minimize the free energy of the system [25]. Such an approach for the sine-Gordon model leads to a mass which monotonically decreases with temperature, while above certain $T \sim \Delta(T = 0)$, the potential generating a gap for a field is completely washed due to the fluctuations of the field itself [25]. Above these temperatures, the mass practically disappears. Such an approach gives very accurate results for the dependence of the gap on model parameters at zero temperature [13, 24].

At finite temperatures, contributions to the real time transverse spin correlator read as

$$\chi_\perp(t) \sim -\frac{(T2a\pi/v)^{\gamma_i/2}}{|\sinh(\pi Tt)|^{\gamma_i/2}} \exp\left(-i\frac{\gamma_i\pi}{4} - i\Delta(T)|t|\right), \tag{7}$$

which reduces to the standard Luttinger liquid correlator in the absence of the gap. Here Δ is the sum of the existing gaps. At half filling, all sectors are gapped [15], hence Δ is their sum. Away from half filling, which is presumably the case in Ref. [11], the spin sectors are fully gapped [5], and the Δ is dominated by their contribution. From this, by Fourier transformation, we can evaluate the frequency dependent retarded correlation functions, which reads as

$$\chi_\perp(\omega) \sim \left[\exp\left(\frac{iK\pi}{2}\right) B\left(-i\frac{E_+}{2\pi T} + \frac{K}{2}, 1-K\right) - \right.$$
$$\left. - \exp\left(-\frac{iK\pi}{2}\right) B\left(-i\frac{E_-}{2\pi T} + \frac{K}{2}, 1-K\right)\right] T^{K-1} i, \tag{8}$$

where $B(x,y) = \Gamma(x)\Gamma(y)/\Gamma(x+y)$ is Euler's beta function, $\Gamma(x)$ is Euler's gamma function, $E_\pm = \omega \pm \Delta(T)$ and is shown in Fig. 2. From this, after taking the $\omega \to 0$ limit,

Table 1 The long time exponents of the different contributions to the transverse spin susceptibility ($\sim t^{-\gamma_i/2}$). These also determine the temperature dependence of the NMR relaxation rate as $(T_1T)^{-1} \sim T^{\gamma_i/2-2}$.

i	q_i	α, α', r, r'	exponent (γ_i)
1	0	$\alpha = \alpha', r = r'$	$\frac{1}{K_{s+}} + K_{s+} + \frac{1}{K_{s-}} + K_{s-} \approx 4$
2	$2k_F$	$\alpha = -\alpha', r = r'$	$\frac{1}{K_{c-}} + K_{c-} + \frac{1}{K_{s+}} + K_{s+} \approx 4$
3	$2q_F$	$\alpha = \alpha', r = -r'$	$K_{c+} + K_{c-} + \frac{1}{K_{s+}} + \frac{1}{K_{s-}} \approx 3.2$
4	$2(k_F \pm q_F)$	$\alpha = -\alpha', r = -r'$	$K_{c+} + \frac{1}{K_{c-}} + \frac{1}{K_{s+}} + K_{s-} \approx 3.2$

the NMR relaxation rate follows as

$$(T_1T)^{-1} = A\left(\frac{2a\pi}{v}\right)^K T^{K-2}\mathrm{Im}F(T), \qquad (9)$$

where

$$F(T) = \exp\left(\frac{iK\pi}{2}\right)\frac{\Gamma(1-K)\Gamma\left(-\frac{i\Delta(T)}{2\pi T}+\frac{K}{2}\right)}{\Gamma\left(-\frac{i\Delta(T)}{2\pi T}-\frac{K}{2}+1\right)}\times$$
$$\times\left[\Psi\left(-\frac{i\Delta(T)}{2\pi T}-\frac{K}{2}+1\right)-\Psi\left(-\frac{i\Delta(T)}{2\pi T}+\frac{K}{2}\right)\right], \qquad (10)$$

and $K = \gamma_i/2$, A is a proportionality factor, which is determined by the contribution of the respective fermionic fields and cannot be obtained by the method used here, $\Psi(x)$ is the digamma function. At high temperatures, Eq. (9) exhibits a T^{K-2} behaviour [26], as is expected from Luttinger liquids [13, 19]. Based on the possible values of Luttinger liquid parameters, the contributions with γ_3 and γ_4 are the most divergent ones at low temperatures due to the presence of the $c+$ mode, hence we will concentrate on them in the followings. They lead to $K = 1 + (K_c + 1/K_s)/2$.

In Fig. 3, we fit our Eq. (9) to the experimental data on the inner wall of metallic double-wall carbon nanotubes as a function of temperature at magnetic fields $H = 3.6$ T and 9.3 T. To increase the strength of the NMR signal, selective enrichment of the inner shells using ^{13}C isotope was performed, hence mainly the properties of the inner tube has been probed by the magnetic resonance measurements. Here, we take $\Delta = \Delta_0(1-T/T_c)^{1/4}$, which is very close to that found in the self consistent solution of the soliton energy in the sine-Gordon model [25]. With this, we can nicely account for the various complicated excitations of our model. Obviously, we expect a smooth crossover of Δ as the temperature increases through T_c to a small value ($\Delta(T)/T \ll 1$) rather than a sharp drop to zero, but for computational purposes, we use the above sharp form. Our results are robust with respect to variations of the gap function, e.g. the quality of the fitting remains the same

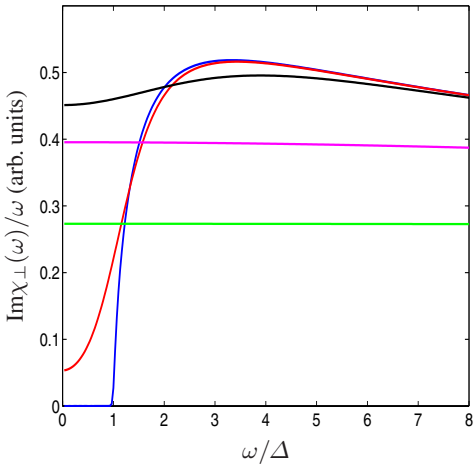

Figure 2 (online colour at: www.pss-b.com) The imaginary part of the local spin susceptibility is plotted as a function of the frequency for various temperatures ($T/\Delta = 0$ (blue), 0.2 (red), 0.8 (black), 5 (magenta) and 20 (green)) and $K = 1.7$. The presence of the spin gap is only visible at low temperatures $T \ll \Delta$. With increasing temperature, the susceptibility becomes practically frequency independent.

by changing the exponent, 1/4, within the range 0.1 - 0.7, since only a finite gap a low temperatures ($T \ll T_c$) and a tiny gap at high temperatures is required ($T \gg T_c$). For the fits, we use $T_c \approx 13$ K and $\Delta_0 \approx 32$ K. Theoretically [25], the critical temperature, at which the cosine potential practically disappears from the sine-Gordon model, is roughly determined as $T_c \approx \Delta(0)/e$, in perfect agreement with our fitting. Above T_c, the gap does not make itself felt any more. For the Luttinger liquid parameters, we assume $K_c = 0.2$, in accordance with Refs. [2] and [5], and deduce $K_s = 1.07$ for $H = 3.6$ T and $K_s = 0.87$ for $H = 9.3$ T, which vary with the field but stay close to unity as expected [13]. For the sake of completeness, we give the numerical prefactors, which are found to be $A = 1.4 \times 10^4$ K/s, while upon reinserting original units, $k_B 2a\pi/\hbar v = 10^{-4}$ 1/K, and v can be taken roughly as v_F. The latter falls into the same order of magnitude, if we identify a, as a rough estimate, to be of the order of the honeycomb lattice constant (~ 2.46 Å), and $v_F \sim 10^6$ m/s,

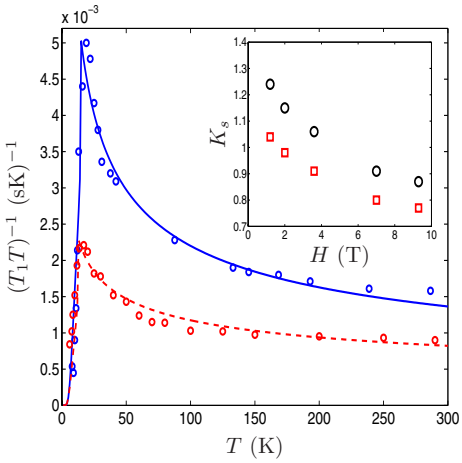

Figure 3 (online colour at: www.pss-b.com) The fit of Eq. ((9)) to the experimental data (open circles) of Ref. [11] at $H = 3.6$ T (red dashed line) and 9.3 T (blue line). The inset shows the deduced K_s exponents as a function of the field for $K_c = 0.2$ (black circles) and 0.05 (red squares).

which again certifies our approach. Since the NMR relaxation rate has been measured at 290 K for different fields [11], we can also determine the field dependence of the K_s exponent, which is shown in the inset of Fig. 3 for two different, field independent values of K_c. It stays close to 1 and decreases with field. The other exponents $K_{c\pm}$ and K_{s-} can also vary with the field, but since no exact solutions are available for the Luttinger liquid exponents of carbon nanotubes unlike for the Hubbard model [16], we choose the simplest possible approach of assigning all the field dependence to the $s+$ field. It is evident from the figure, that the larger K_c, the larger K_s, hence a suitable increase of the former in magnetic field can result in an increment of the latter. Given the simplicity of our scheme, the obtained fits are excellent and the parameters are reasonable. Finally, we mention the possibility, that upon doping, the properties of the $c+$ mode are expected to change (e.g. K_{c+}) similarly to the change of the $s+$ mode in a magnetic field.

4 Conclusion We have analyzed a recent experimental data [11] of the NMR relaxation rate of CNT. With the use of bosonization, we find that the low temperature region is well described by a spin gapped Luther-Emery liquid. Relaxation is dominated by exponentially activated behaviour with a gap ~ 30 K. This crosses over to the usual Luttinger liquid picture at high temperatures compared to the gap, with characteristic non-integer power-law exponents. Finally, we predict, that the observed spin gap can significantly be reduced and even completely suppressed through a quantum phase transition by experimentally accessible magnetic fields ($\sim 25 - 30$ T) due to its small value, similarly [13, 27] to $Cu_2(C_5H_{12}N_2)_2Cl_4$.

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