Instability of capacitance mode in multi-walled nanotubes

K. Sasaki

Department of Physics, Tohoku University, Sendai 980-8578, Japan

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Conditions for intra and inter layer Coulomb interactions in multi-walled carbon nanotubes are derived from stability of capacitance excitations. It is pointed out, supposing the stability conditions are not satisfied, that the system has unstable modes which correspond to a charge transfer between layers or charge density oscillation in each layer depending on zero mode or non-zero modes. It is argued that the stability conditions can be broken when the vacuum polarization processes due to the massive bands are taken into account.

I. INTRODUCTION

Electrical properties of carbon nanotubes (CNTs) have attracted much attention from various points of view. They are characterized by the quantum mechanical behavior of the \( \pi \)-electrons which are interacting with each other via the Coulomb interaction. Several aspects of the Coulomb interaction are already seen in some experiments of charging \( \mathcal{J} \) and of temperature dependent resistivity \( \mathcal{R} \) which are consistent with current understanding of low energy excitations in metallic CNTs. The low energy excitations in the metallic CNTs are theoretically modeled by \textit{massless} bands (linear dispersion bands) with the Coulomb interaction \( \mathcal{I} \). On the other hand, the \textit{massive} bands (other sub-bands except the massless bands) modify the Coulomb interaction between massless fermions (electrons in the massless bands) through the vacuum polarization effect \( \mathcal{K} \). As for single-wall CNTs with small diameter, the correlation to the long wave length modes of the Coulomb interaction is negligibly small. However in case of multi-walled CNTs, because of their large diameter, the gap energy of the massive bands is small; besides, the number of the massive bands is large so that the correlation would be large also. Because of that the low energy spectrum of the multi-walled CNTs is thought to be under the influence of the vacuum polarization due to the massive bands.

The multi-walled carbon nanotubes (MWCNTs) consists of several separate single-wall CNTs and may have an unique electrical property which is not shown in each individual single-wall CNT. In this paper, we investigate a novel electrical property of a MWCNT which has two layers. Even in this simple case, there is actually a different aspect as compared with the single-wall CNT. That is the Coulomb interaction between the layers: interlayer Coulomb interaction. The Coulomb interactions in this system consist of three potentials: intra-layer Coulomb interactions in each layer and the interlayer interaction. Although it is difficult to obtain information on the Coulomb potentials as compared with the one in the single-wall CNTs, it can be thought that the intra-layer Coulomb potential for an inner layer is the same potential in the single-wall CNTs, however the intra-layer one for an outer layer is not simple because of the screening due to the presence of the electrons in the inner layer. The interlayer unscreened Coulomb potential between the layers in the MWCNTs is calculated theoretically \( \mathcal{L} \).

Since each layer of the MWCNTs has its own helical structure, we can divide the MWCNTs into three categories depending on the structural property of each layer. That are (1) metallic-metallic(M-M) , (2) metallic-semiconducting(M-S) and (3) semiconducting-semiconducting(S-S) MWCNT. If there were no interlayer interaction in the MWCNTs, analysis of those systems is the same as that of the two individual single-wall CNTs and the stability of the system is trivial. But there is the interlayer Coulomb interaction. This interaction cause a new excitation mode: charge transfer between the two layers: capacitance mode which may cause an instability of the system. For S-S and S-M types, the stability of the system is thought to be rigid. However we need to consider M-M type MWCNTs carefully. The Coulomb potentials decide the energy spectrum of the capacitance mode. Hence, for some kinds of potential, the mode could be unstable, that is, it's excitation spectrum have a negative eigenvalue. This cause the instability of the vacuum of the system and will change the vacuum into an another new vacuum state that we do not know within the model used in this paper. It is the purpose of this article to find conditions on the stability of the system and check if the condition is satisfied including the vacuum polarization correction due to the massive bands.

The paper is composed as follows. A model Hamiltonian for the low energy excitations in the M-M type MWCNTs is constructed in Sec. II. In Sec. III, we study the zero mode spectrum of the system and establish a condition for the stability of the zero mode. Then we investigate the non zero modes in Sec. IV and check if the system is stable in realistic Coulomb interactions. Summary and discussion are given in Sec. V.

II. LOW ENERGY EFFECTIVE MODEL

Because the kinetic terms and the Coulomb interactions consist of densities of each layer, we begin by specifying the densities in both layers. Let us define the density for the outer layer as \( J_{\text{out}} \) and for the inner one as \( J_{\text{in}} \). Due to the linear dispersion relation near
the Fermi points, the densities are written by the sum of left and right current as \( J_{\text{out}} = J_{L,\text{out}} + J_{R,\text{out}} \) and \( J_{\text{in}} = J_{L,\text{in}} + J_{R,\text{in}} \). Using these definitions of the left and right currents, we define a (quantum) field theoretical kinetic Hamiltonian respecting the linear dispersion as [1]

\[
H_F = \Delta \left[ \frac{L}{8} \int_D : J_{L,\text{out}}^2 + J_{R,\text{out}}^2 : dx - \frac{1}{12} \right] + \Delta \left[ \frac{L}{8} \int_D : J_{L,\text{in}}^2 + J_{R,\text{in}}^2 : dx - \frac{1}{12} \right],
\]

(1)

where \( \Delta = 2\pi \hbar v_F / L \) is the single-particle level spacing and is defined under the periodic boundary condition along a tubule axis. \( L \) is the length of the tube and \( v_F \) is the Fermi velocity. The integral region is labeled as \( D \in [0 : L] \).

The densities of each layer are expanded by the current operators as

\[
J_{\text{out}}(x) = \frac{Q_{\text{out}}}{L} + \sum_{n \in \mathbb{Z}} \left( (J_L^n)^{\dagger} + j_R^n \right) \frac{1}{L} e^{i2\pi n x / L},
\]

(2)

\[
J_{\text{in}}(x) = \frac{Q_{\text{in}}}{L} + \sum_{n \in \mathbb{Z}} \left( (J_L^n)^{\dagger} + J_R^n \right) \frac{1}{L} e^{i2\pi n x / L}.
\]

(3)

The zero modes \( Q_{\text{out}} \) and \( Q_{\text{in}} \) denote the total charges in each layer respectively. The current operators satisfy the following bosonic commutation relations:

\[
[J_L^n, (J_L^{n'})^{\dagger}] = 4n\delta_{nm},
\]

(4)

\[
[J_L^n, (J_R^{n'})^{\dagger}] = 4n\delta_{nm},
\]

(5)

and all of the other commutation relations vanish. Making use of the operator expression for the charge densities, we rewrite the kinetic Hamiltonian as the sum of zero mode and non-zero modes,

\[
H_F = H_F^0 + \sum_{n>0} H_F^n,
\]

(6)

where

\[
H_F^0 = \frac{\Delta}{16} \left( Q_{\text{out}}^2 + Q_{5,\text{out}}^2 \right) + \frac{\Delta}{16} \left( Q_{\text{in}}^2 + Q_{5,\text{in}}^2 \right),
\]

(7)

\[
H_F^n = \frac{\Delta}{4} \left( (J_L^n)^{\dagger} J_L^n + (J_R^n)^{\dagger} J_R^n \right) + \frac{\Delta}{4} \left( (J_L^n)^{\dagger} J_L^n + (J_R^n)^{\dagger} J_R^n \right).
\]

(8)

The operators \( Q_{5,\text{out}} \) and \( Q_{5,\text{in}} \) indicate the zero mode of the current in each layer, that is, they are equal to a integral of currents \( J_{L,\text{out}} - J_{R,\text{out}} \) and \( J_{L,\text{in}} - J_{R,\text{in}} \) over the nanotube length.

The vacuum of this sector is defined as a direct product of the Dirac seas of both layers (\( \mid \text{vac} \rangle = \mid \text{vac, in} \rangle \otimes \mid \text{vac, out} \rangle \)) which are labeled as \( \mid \text{vac, in} \rangle \) and \( \mid \text{vac, out} \rangle \) that satisfy the following conditions,

\[
J_L^n \mid \text{vac, in} \rangle = J_R^n \mid \text{vac, in} \rangle = 0,
\]

\[
J_L^n \mid \text{vac, out} \rangle = J_R^n \mid \text{vac, out} \rangle = 0,
\]

for positive \( n \) value. The vacuum state change into a new vacuum due to the intra and inter layer Coulomb interactions. They are given by

\[
H_C = \frac{1}{2} \int \int D J_{\text{out}}(x) V_{\text{out}}(x - x') J_{\text{out}}(x') dx dx' + \frac{1}{2} \int \int_D J_{\text{in}}(x) V_{\text{in}}(x - x') J_{\text{in}}(x') dx dx' + \int \int_D J_{\text{in}}(x) V(x - x') J_{\text{out}}(x') dx dx'.
\]

(9)

The Coulomb potentials for each layer are labeled as \( V_{\text{out}} \) and \( V_{\text{in}} \). Between the layers, there is the Coulomb interaction denoted \( V \). Explicit form of these potentials would be difficult to observe experimentally. However we can drive some information of them from the stability of the systems, that we will discuss shortly. The potentials are rewritten by the Fourier series;

\[
V_{\alpha}(x) = \sum_{n \in \mathbb{Z}} \beta_{\alpha} e^{i2\pi n x / L},
\]

(10)

where the non-zero modes are given by

\[
H_C^0 = \beta_{\text{out}} \left[ (J_L^n)^{\dagger} + j_R^n \right] \left[ (J_L^n)^{\dagger} + (J_R^n)^{\dagger} \right] + \beta_{\text{in}} \left[ (J_L^n)^{\dagger} + J_R^n \right] \left[ (J_L^n)^{\dagger} + (J_R^n)^{\dagger} \right] + \beta_n \left[ (J_L^n)^{\dagger} + J_R^n \right] \left[ (J_L^n)^{\dagger} + J_R^n \right].
\]

(12)

The Fourier components of the Coulomb interactions \( \beta_{\alpha} \) depend on system parameters; diameter and length of CNTs. Notice that \( \beta_{\text{in}} \) and \( \beta_{\text{out}} \) for \( n \neq 0 \) are modified by the vacuum polarization due to the massive bands, however the zero mode \( \beta_0^0 \) and \( \beta_0^0 \) do not receive any vacuum polarization correction. Standard Tomonaga-Luttinger theories of nanotubes take into account effects of non-linear interactions on low energy spectrum with an assumption \( \beta_0^0 = \beta_0^0 \neq 0 \). The Coulomb potentials and vacuum polarization depend on the wave number \( n \). We respect the wave number dependent nature of the Coulomb interactions and vacuum polarization effect but do not consider the non-linear interactions including a tunneling interaction between layers.

### III. Stability Condition of Zero Mode

The zero mode of the total Hamiltonian is

\[
H_F^0 + H_C^0 = \left( \frac{\Delta}{16} + \frac{1}{2} \beta_0^0 \right) Q_{\text{out}} Q_{\text{out}}
\]
Suppose that the system is open, that is, number of the electron in each layer can be changed freely, then the total charges of each layer can take any value. Therefore when there is a direction in the space spanned by \((Q_{out}, Q_{in})\) that the energy of the zero mode become lower, the system is unstable along that direction. To avoid such instability, the energy must be elliptic as a function of the charges \((Q_{out}, Q_{in})\). The condition of elliptic form corresponds to a negativity of the discriminant and results in an inequality \(G_0 < 0\) where

\[
G_0 = \beta_0^2 - \left( \frac{\Delta}{8} + \frac{1}{2} \beta_0^2 \right) \frac{1}{\Delta} Q_{in} Q_{in} + \beta_0 Q_{in} Q_{out}.
\] (13)

It should be noted that the zero mode does not receive the vacuum polarization correction. Therefore, the condition for stability must be satisfied to keep the vacuum stable. However, the non-zero modes are generally receiving the correction and depend on dispersion relations of massive bands.

IV. STABILITY CONDITION OF NON-ZERO MODES

We consider non-zero modes of the total Hamiltonian in this section. To analyze the energy spectrum of this system, first we apply the Bogoliubov transformations to the current operators of each layer separately and obtain the following expression of the Hamiltonian,

\[
H_F^\alpha + H_C^\alpha = E_{n}^{\text{out}} (\tilde{\gamma}_{L}^\alpha \tilde{\gamma}_{L}^\alpha + \tilde{\gamma}_{R}^\alpha \tilde{\gamma}_{R}^\alpha) \\
+ E_{n}^{\text{in}} ((\tilde{J}_{L}^\alpha)^{\dagger} \tilde{J}_{L}^\alpha + (\tilde{J}_{R}^\alpha)^{\dagger} \tilde{J}_{R}^\alpha) \\
+ \alpha_n (\tilde{J}_{L}^\alpha + (\tilde{J}_{R}^\alpha)^{\dagger}) \times (\tilde{\gamma}_{L}^\alpha \tilde{\gamma}_{L}^\alpha + \tilde{\gamma}_{R}^\alpha \tilde{\gamma}_{R}^\alpha),
\] (19)

where the transformed current operators are defined as follows,

\[
\left( \begin{array}{c}
\tilde{J}_{L}^\alpha \\
\tilde{J}_{R}^\alpha
\end{array} \right) = \left( \begin{array}{cc}
\cosh t_1^\alpha & \sinh t_1^\alpha \\
\sinh t_1^\alpha & \cosh t_1^\alpha
\end{array} \right) \\
\left( \begin{array}{c}
\tilde{\gamma}_{L}^\alpha \\
\tilde{\gamma}_{R}^\alpha
\end{array} \right) = \left( \begin{array}{cc}
\cosh t_2^\alpha & \sinh t_2^\alpha \\
\sinh t_2^\alpha & \cosh t_2^\alpha
\end{array} \right).
\]

From the point of view of the new current operators, the interlayer Coulomb interaction is thought to have different coupling as compared with the original coupling \(\beta_n\) that we denote a new interlayer coupling \(\alpha_n\):

\[
\alpha_n = \beta_n (\cosh t_1^\alpha - \sinh t_1^\alpha) (\cosh t_2^\alpha - \sinh t_2^\alpha).
\] (20)

The following Bogoliubov transformation angle and energy spectrum were used to rewrite the non-zero modes of the Hamiltonian,

\[
\sinh 2t^\alpha_n = \frac{\beta^\alpha}{E_n}, \quad \cosh 2t^\alpha_n = \frac{1}{E_n} \left( \frac{\Delta}{4} + \beta^\alpha_n \right), \\
E_n^\alpha = \frac{\Delta}{4} \sqrt{1 + \frac{8\beta^\alpha_n}{\Delta}}, \quad \alpha \in \{in, out\}.
\]

Second we have to diagonalize the interacting terms of both currents whose coupling is \(\alpha_n\). This can be done by shifting the current operator as

\[
\tilde{j}_{L}^\alpha = j_{L}^\alpha + \frac{\alpha_n}{E_n^{\text{out}}} (j_{L}^\alpha + (j_{R}^\alpha)^{\dagger}),
\]

\[
\tilde{j}_{R}^\alpha = j_{R}^\alpha + \frac{\alpha_n}{E_n^{\text{out}}} ((j_{L}^\alpha)^{\dagger} + j_{R}^\alpha),
\]

and the Hamiltonian leads to

\[
H_F^\alpha + H_C^\alpha = E_{n}^{\text{out}} ((\tilde{J}_{L}^\alpha)^{\dagger} \tilde{J}_{L}^\alpha + (\tilde{J}_{R}^\alpha)^{\dagger} \tilde{J}_{R}^\alpha) + \tilde{H}_{n}^{\text{in}}.
\] (23)
Here $\tilde{H}_n^{in}$ is defined as

$$\tilde{H}_n^{in} = E_n^{in} \left( (\vec{J}_L)^\dagger \vec{J}_L + (\vec{J}_R)^\dagger \vec{J}_R \right) - \frac{2a_n^2}{E_n^{out}} \left( (\vec{J}_L)^\dagger + (\vec{J}_R)^\dagger \right) \left( (\vec{J}_L)^\dagger + (\vec{J}_R)^\dagger \right) .$$

(24)

This Hamiltonian implies that an effective attractive modes that couple to the Bogoliubov transformed current operators of the inner layer appears. The coupling depends on the energy spectrum of outer layer. Suppose that the outer layer is the semiconducting tube, then the excitation spectrum $E_n^{out}$ is order of the gap energy which is very large as compared with the Coulomb energy. Therefore the effective attractive interaction weakens much.

We further diagonalize the term $\tilde{H}_n^{in}$ as

$$\tilde{H}_n^{in} = E_n \left( (\vec{J}_L)^\dagger \vec{J}_L + (\vec{J}_R)^\dagger \vec{J}_R \right) ,$$

(25)

where new current operators are defined as

$$\left( \left( \vec{J}_L^\dagger \right) \right) = \left( \begin{array}{cc} \cosh s_n & \sinh s_n \\ \sinh s_n & \cosh s_n \end{array} \right) \left( \left( \vec{J}_L^\dagger \right) \right) ,$$

(26)

with the definition of the Bogoliubov transformation angle and energy spectrum

$$\sinh 2s_n = \frac{2a_n^2}{E_n^{out}} , \quad \cosh 2s_n = \frac{1}{E_n^{in}} \left( E_n^{in} - \frac{2a_n^2}{E_n^{out}} \right) ,$$

$$\bar{E}_n = E_n^{in} \sqrt{1 - \frac{4a_n^2}{E_n^{out} E_n^{in}}} .$$

From the energy eigenvalue of the lowest excitation mode, we read a condition of stability as

$$\bar{E}_n > 0 ,$$

(27)

which results in the negativity of the discriminant for the n-th Fourier component of the Coulomb interactions:

$$G_n = \beta_n^2 - \left( \beta_n^{in} + \frac{\Delta}{8} \right) \left( \beta_n^{out} + \frac{\Delta}{8} \right) < 0 .$$

(28)

This condition of stability is similar to that of the condition of stability of the zero mode for open systems. The physical entity of the lowest excitation mode is capacitance mode between the layers. It is easy to prove this fact in the case of $\beta_n^{in} = \beta_n^{out}$. In this case, total density ($\rho = J_{in} + J_{out}$) and capacitance density ($\sigma = J_{in} - J_{out}$) decouple, so that one can calculate the energy spectrum of the capacitance modes without any difficulty. The energy spectrum is given by $4nE_n^{in}$ where $E_n^{in} = \frac{1}{4} \sqrt{1 + \frac{4}{3} (\beta_n^{in} - \beta_n)}$ and stability condition of the spectrum reduces to $G_n < 0$.

We would like to consider several cases and check if the stability condition is satisfied. First, suppose that all the Coulomb potentials are equivalent, then $\beta_n^{in} = \beta_n^{out} = \beta_n$ gives $G_n < 0$ which means the stability of the system. Second, assume the following conditions,

$$\beta_n = \frac{\beta_n^{out} + \beta_n^{in}}{2} , \quad \beta_n^{out} = \beta_n^{in} ,$$

(29)

then

$$G_n = -\frac{\Delta}{8} \times \left( \frac{\Delta}{8} + 2\beta_n^{in} \right) < 0 ,$$

(30)

which also result in the stability of the system. Finally, we analyze more realistic case. The diameter of the multi-walled nanotube is thick so that the vacuum polarization due to the massive fermions loop can be no longer negligible and each Fourier component of the intra layer Coulomb potentials is modified. Therefore the condition of instability is given by

$$\bar{G}_n = \beta_n^2 - \left( \beta_n^{in} + \frac{\Delta}{8} \right) \left( \beta_n^{out} + \frac{\Delta}{8} \right) > 0 ,$$

(31)

where

$$\beta_n^{out} = \frac{\beta_n^{out}}{1 - T_n^{out}} , \quad \beta_n^{in} = \frac{\beta_n^{in}}{1 - T_n^{in}} .$$

(32)

We denote the static response functions as $T_n^{in}$ and $T_n^{out}$ for each layer. Here we assume that the interlayer Coulomb interaction does not receive the vacuum polarization corrections. This assumption is not correct for multi-walled CNTs with more than 3 metallic layers. When the vacuum polarization due to the massive bands is huge, the effective Coulomb couplings $\beta_n^{in}$ and $\beta_n^{out}$ are close to zero. Therefore, the condition of instability reduces to the following condition:

$$\beta_n > \frac{\Delta}{8} .$$

(33)

We estimate the condition using the Fourier mode of the interlayer Coulomb potential which is given by

$$\beta_n = \frac{e^2}{2\pi L \pi} \int_0^{\pi} K_0 \left( \frac{2\pi nd}{L} \sqrt{\sin^2 x + \left( \frac{a_n}{d} \right)^2} \right) ,$$

(34)

where $K_0$ is the modified Bessel function, $a_z = 1.3[A]$ is a cutoff length which is introduced to take into account for the ionization energy of a $\pi$-electron and $e$ is charge of electron which gives $e^2/4\pi = 1.44[eV \cdot nm]$. $d$ should be understood as the mean diameter of the outer and inner layer. We plot the Fourier components of the Coulomb interaction and the energy $\Delta/8$ in Fig. Note that there are some instability regions in the spectrum in this case.

The above estimation in the case of vanishing intra-layer coupling is a rough estimation. Here we analyze the stability condition using a static response function derived at one-loop level supposing an equality $\beta_n^{in} = \beta_n^{out} = \beta_n$ and that $\beta_n^{in}$ and $\beta_n^{out}$ received the
same amount of the vacuum polarization correction. In this case, the condition of instability leads to
\[
\beta_n > \bar{\beta}_n^{\text{in}} + \frac{\Delta}{8}
\] (35)

We plot \(\beta_n\) and \(\bar{\beta}_n^{\text{in}} + \frac{\Delta}{8}\), for several MWCNTs with different diameter in Fig. 2.

Figure 2 shows that there are some unstable spectrum regions in momentum space, therefore we conclude that the system is thought to be unstable due to the vacuum polarization of the massive bands. Within the unstable region of wave number \(n \in U\), the excitation energy \(E_n\) (correctly \(4n\) times \(E_n\)) take a negative value. Hence the system energy becomes lower if the current operators of the capacitance mode have a vacuum expectation value. Although we can not calculate the value of the amplitude, this instability would cause charge density oscillations in both layers.

V. SUMMARY AND DISCUSSION

In summary, the stability conditions on the Coulomb interactions in the multi-walled CNTs consisting of two metallic layers have been given within the framework of the simple model respecting the quantum nature of the electrons in linear bands and the Coulomb interactions. We have shown that because the massive bands screen the intra-layer Coulomb interactions in each layer, there are finite unstable momentum regions in the system. Nature of the instability is governed by the capacitance mode between layers, therefore it is possible to detect the instability experimentally.

The instability conditions are divided into zero and non-zero modes conditions. If the system is unstable, we can not predict a new ground state, but, can image several candidates of a new vacuum state. For example, if the zero mode is unstable, charge transfer between the layers is expected. After that charge transfer, the electric state of each layer looks like doped or un-doped single-wall CNTs (spontaneous capacitance). As a result of the moving of sufficient amount of charge between layers, the system may turn into a stable state because the low energy excitations are affect by the massve bands. On the other hand, even in the case of stable zero mode, the non-zero modes can still be unstable because of the vacuum polarization due to the massive bands. In this case, charge density oscillation in both layers may occur.

We have not considered the tunneling interaction between two layers in the present paper. Suppose that such an interaction is present, a change of the stability condition is expected. We would like to clarify this case in future work.

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