Optical Forces and Light Scattering In Carbon Nanotubes

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Abstract – **The ongoing effort towards understanding the physical principles underlying optomechanical forces is an active field of research that offers diverse applications in various fields of technology. The proposed physical model for the radiation pressure exerted on an achiral carbon nanotube (CNT), is formulated using the Maxwell stress tensor. Our model consists of a system of integral equations, describing the scattering pattern of an electromagnetic field (EM) for a single, finite-length, CNT in the THz frequency range. The obtained results from the proposed model, are presented for three cases: I) The optical force exerted on a CNT subjected to a surface EM-mode. II) The optical binding of two parallel non-identical CNT's III) The excitation of a radial breathing mode by the surface EM-mode in a CNT. Our current results can be implemented in the design of CNT-based ion and gas sensors, biosensors, field emission devices, and new types of metamaterials.**

Index terms – **Carbon nanotubes; Optical forces; Stress tensor; Achiral carbon nanotubes; Integral equations;**

I. Introduction

The recent progress in research of carbon nanotubes (CNT), stimulated great activity in the design and production of new devices including commercial ones [1].CNT's show remarkable electronic, elastic, hydrodynamic and optical properties depending on their chirality. These properties can be optimally utilized, given an adequate theoretical framework. In the early stages of CNT research, it became clear that the description of their physical nature in terms of the relevant macroscopic parameters, is inconsistent (for example, permittivity). Consequently, fundamental model of their electrodynamical properties was established, with its foundations deeply rooted in the synthesis between quantum transport theory and classical electrodynamics, taking into account the effective two-side boundary conditions of the impedance type [2,3]. The scattering problem was formulated as a system of integral equations and was solved numerically [4-6]. Numerical solutions of scattering problems, are common techniques in the fields of antennas and microwave engineering.

One of the important new challenges of today's nanoscience, is investigating the coupling between nano-mechanical and nano-optical effects. The control of optical forces and the manipulation they allow in nanometer-sized particles, attracted many researchers to conduct intensive studies on the physical mechanism that controls this optomechanical coupling. In his pioneering work, Ashkin, first demonstrated the possibility of exploiting light to optically manipulate and trap microscopic objects [7]. The investigation of the trapping capabilities of a single focused laser beam, lead to the realization of the so-called optical tweezers. One of the intriguing ways of "tweezing" particles, known as optical binding, was recently discovered (including binding with chiral particles [8]).

A well-known realization of optical binding occurs when light is scattered by a pair of interacting small particles. Assuming a quasi-static regime (dipole approximation [9]), the scattering pattern can be described by the polarizability tensors of each particle. An attempt to apply this model to CNTs is given in [10]. The validity of such a model is clearly limited, since it is subjected to some constraints imposed on the particle's dimensions being small compared to the wave length λ of the exciting radiation. Nevertheless, in spite of the typical small values of CNT lengths, they do appear as unsubtle candidates for such an approximation. The main reason for this, lies in the existence of strongly retarded eigen modes in the THz frequency range, which were theoretically predicted in [4] and experimentally observed in [11,12].

The present study is a manifestation of CNTs as promising candidates for different applications of optomechanical phenomena. Therefore, we will dedicate a considerable part of the talk to the theory of radiation pressure exerted on CNTs, produced by EM-fields (so called, “optical forces”). Calculation of the optical forces, is done by means of the Maxwell stress tensor and employing integral equations technique, taking into account the effect of CNT eigen modes. In addition to dealing with the computational aspects of the problem, we also address some fundamental issues. Different formulations are known to exist for the stress tensor [13] (e.g., Maxwell, Abraham, Minkowski, Einstein-Laub, Paierls), in which different model assumptions were introduced, leading to some non-identical expressions for the field momentum and the induced force. Discussions of their correctness, have lasted for over a century [13]. From time to time these discussions seemed to converge to an overall conclusion but nevertheless some pending issues still remain.

II. Integral Equations and Stress Tensor

In this section we will formulate the system of integral equations for the structure shown in Fig. 1. The radius of the CNT is taken to be small compared with the optical forcing wavelength and its length is assumed arbitrary. The whole system is immersed in a linear medium with the common constitutive relations $\mathbf{D} = \varepsilon\mathbf{E}, \mathbf{B} = \mu\mathbf{H}$ where $\varepsilon = \varepsilon_1 + j\varepsilon_2$

and $\mu = \mu_1 + j\mu_2$ are the complex permittivity and permeability respectively of the surrounding medium. For the description of the linear electron dynamics with respect to the EM-field, the Boltzmann kinetic equation is solved in the framework of the momentum-independent relaxation time approximation. Thereafter, an analytical expression for electric current in the single CNT and its conductivity is derived.

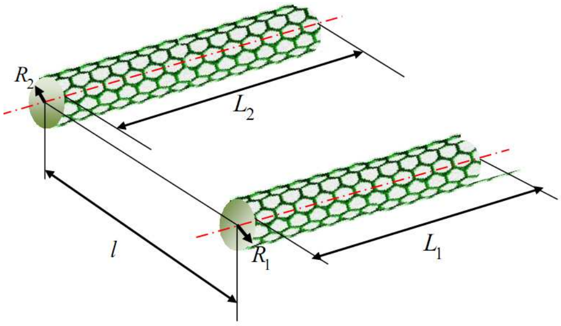

Fig.1. Configuration of the structure under consideration: A pair of parallel non-identical CNTs with corresponding radii $R_{1,2}$, lengths $L_{1,2}$ and conductivities $\sigma_{1,2}$, placed at the distance l one with respect to another.

In contrast with [3-6], the system of integral equations is written in terms of the current densities on the CNT surfaces, without averaging over the azimuthal angle. The reason for this is, the strong angle asymmetry of the currents due to the strong mutual scattering for the case of interacting CNTs. The system of integral equations for the longitudinal current densities $j_\alpha(\boldsymbol{R}_\alpha)$,evaluated on the CNT surface, can be written as:

$$j_\alpha(\boldsymbol{R}_\alpha) = \frac{1}{j\omega\varepsilon}(\partial_z^2 + \mathrm{k}^2)\sum_{\beta=1,2} R_\beta\sigma_\beta \cdot$$

$$\int_0^{2\pi}\int_{-L_\beta/2}^{L_\beta/2} j_\beta(\boldsymbol{R}'_\beta)\frac{e^{i\mathrm{k}|\boldsymbol{R}_\beta - \boldsymbol{R}'_\alpha|}}{|\boldsymbol{R}_\beta - \boldsymbol{R}'_\alpha|}d\phi_\beta dz' + E_{0z}(\boldsymbol{R}_\alpha) \qquad (1)$$

where $\alpha, \beta = 1,2$, E_{0z} is the longitudinal component of the incident field and $\mathrm{k} = \omega\sqrt{\mu\varepsilon}$ denotes the wave number of the impinging wave. The solution of (1) allows us to find the electromagnetic field in the whole space. The optical forces are next defined in terms of the Maxwell stress tensor [14], as

$$\overleftrightarrow{\mathbf{T}} = \varepsilon\mathbf{E}\otimes\mathbf{E} + \mu\mathbf{H}\otimes\mathbf{H} - \tfrac{1}{2}(\varepsilon\mathbf{E}\cdot\mathbf{E} + \mu\mathbf{H}\cdot\mathbf{H})\overleftrightarrow{\mathbf{I}} \qquad (2)$$

where $\overleftrightarrow{\mathbf{I}}$ is the unit tensor. According to the boundary conditions imposed on the surface of the CNT, some components of the stress tensor are discontinuous (note that only such components support to the optical forces). The equation for the force can then be written as:

$$\langle \boldsymbol{F} \rangle = \int \Delta\overleftrightarrow{\mathbf{T}}\boldsymbol{e}_\rho dS \qquad (3)$$

where $\Delta\overleftrightarrow{\mathbf{T}} = \langle\overleftrightarrow{\mathbf{T}}_+\rangle - \langle\overleftrightarrow{\mathbf{T}}_-\rangle$ represents the difference between the values of the stress tensor at the outside and inside the surface respectively and $\boldsymbol{e}_\rho$ is a unit radial vector (plays the role of external unit normal). The angular brackets in (3) denote averaging over the period of the optical field. The integration in (3) is conducted over the surfaces of the CNTs.

III. Optical Binding Between Two CNT's

In this section we will consider the optical binding in a system of two parallel arbitrary CNTs distant from each other at a distance of l as shown in Fig.2.

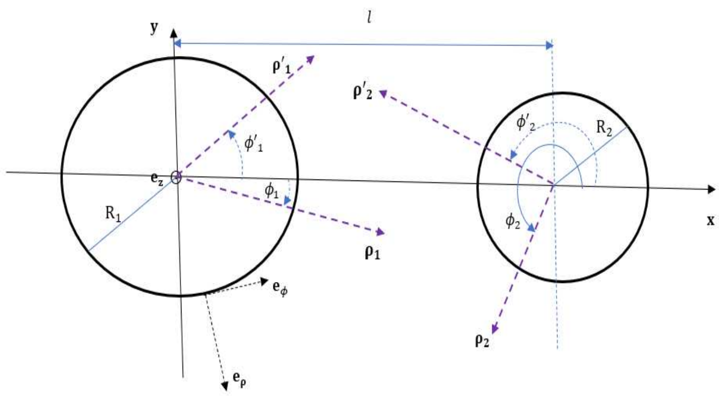

Fig.2. The coordinate system that defines the system of integral equations (5)

We assume that both CNTs are infinitely long and calculate their eigenmodes, by accounting for the electromagnetic coupling between them. The homogeneous integral equations (1) in the limiting case $L_{1,2} \to \infty$, have a non-trivial solution in the form of travelling wave e^{ihz} , where h may be considered as an eigenvalue. The averaging of the current densities over the azimuthal direction is here invalid (in spite of the smallness of CNTs radius) because of the strong azimuthal asymmetry of the field in every CNT. Integration over the z axis is done using the identity:

$$\int_{-\infty}^{\infty}\frac{e^{i\mathrm{k}|\boldsymbol{R}_\beta - \boldsymbol{R}'_\alpha|}}{|\boldsymbol{R}_\beta - \boldsymbol{R}'_\alpha|}e^{ihz'}dz' = \frac{1}{2\pi}K_0(\kappa|\boldsymbol{\rho}_\beta - \boldsymbol{\rho}'_\alpha|) \qquad (4)$$

where K_0 is the MacDonald's function [18], $\alpha, \beta = 1,2$ and

$$|\boldsymbol{\rho}_\beta - \boldsymbol{\rho}'_\alpha|^2 + |z - z'|^2, = |\boldsymbol{R}_\beta - \boldsymbol{R}'_\alpha|^2, \ \ \kappa^2 = h^2 - \mathrm{k}^2$$

The system of integral equations may then be written as

$$j_\beta(\phi_\beta) = \sum_{\alpha=1,2}\left(\frac{\sigma_\alpha\kappa^2 R_\alpha}{j\omega\varepsilon}\right)\cdot\int_0^{2\pi} G_{2D}(\phi_\beta, \phi'_\alpha)\cdot j_\alpha(\phi'_\alpha)d\phi'_\alpha \quad (5)$$

where $G_{2D}(\phi_\alpha, \phi'_\alpha) = \frac{1}{2\pi}K_0(\kappa|\boldsymbol{\rho}_\beta - \boldsymbol{\rho}'_\alpha|)$ .A simple analytical solution of the system (5), can be achieved by implementing the Born approximation, widely used in the physics of optical binding [9]. Assuming that CNTs interaction is rather small, we can take the zero approximation as

$$\begin{pmatrix} j_1(\phi_1) \approx \widetilde{\mathrm{A}} \\ j_2(\phi_2) \approx 0 \end{pmatrix} \quad (6)$$

where $\widetilde{\mathrm{A}}$ is the zero-order approximation of the surface current density on tube (2) ($[\widetilde{\mathrm{A}}] = Amp/m$). This particular choice corresponds to the value $\kappa \approx \kappa^{(0)}$, where $\kappa^{(0)}$ represents the eigenvalue of the first CNT in the absence of the second one. Substituting the above 'zero' approximation into the right-hand part of Eq. (5) and integrating over the azimuthal angle, we obtain the current density induced on the second CNT due to its interaction with the first one. The analytical integration is done using the addition theorem for cylindrical functions (see appendix A in [15]). The final result for the zero order Hertz potential Π^0 is expressed below as

$$\Pi^{(0)}(\rho_2) = \left(\dots\right) + \tilde{c} \sum_{n=-\infty}^{\infty} K_n(\kappa l) I_n(\kappa R_2)\, I_0(\kappa R_1) \cdot \begin{cases} K_n(\kappa\rho_2) I_n(\kappa R_2), & \rho_2 > R_2 \\ K_n(\kappa R_2) I_n(\kappa\rho_2), & \rho_2 < R_2 \end{cases} \quad (7)$$

where $\tilde{c} = -j k^2 R_1 R_2 \widetilde{\mathrm{A}} \sigma_2 / (\pi\omega\varepsilon)$ and the radial position vector $\boldsymbol{\rho}_2$ is described at Fig.2. The symbol $\left(\dots\right)$ in (7) denotes omitted terms, which are continuous at the surface of the second CNT and therefore do not contribute to the optical force. The optical force was calculated using (3), using the general approach considered before. Note that the optical binding force according to (7), is azimuthally dependent.

An additional mode for a pair of CNTs, can be shown to exist at the vicinity of the second tube. Its corresponding relations, can be easily determined by switching the indices $1 \leftrightarrow 2$.

IV. Optical Force On A Single CNT

In this section we evaluate the radiation pressure which is produced by the surface eigenmode propagating over the CNT [3] for the case of lossless media for which $\varepsilon, \mu, h, \kappa \in \mathcal{R}$ and σ is purely imaginary. Assuming that the Hertz vector associated with the imposed field is taken as a travelling wave in the z direction $\boldsymbol{\Pi}_\varepsilon = \Pi_\varepsilon e^{jhz} \mathbf{e_z}$,we can write the field components as: $E_\rho = (jh/\varepsilon)\, \partial_\rho \Pi_\varepsilon$, $E_z = (\omega^2\mu - h^2/\varepsilon)\Pi_\varepsilon$, $H_\phi = j\omega\, \partial_\rho \Pi_\varepsilon$.The normal and longitudinal components of the averaged stress tensor multiplied by a unit normal vector, provide the local pressure (P) exerted on the CNT surface

$$P = \frac{\sigma'' |\bar{E}_z|^2 \kappa}{2\omega} \quad (8)$$

where $|\bar{E}_z|$ is the magnitude of the total longitudinal field at the surface of the CNT, $\sigma'' = 2v_F/(\pi R_0 k R_h c)$ is the imaginary part of the surface conductivity, v_F represents the Fermi-velocity of electrons in the CNT, $Z = \sqrt{\mu/\varepsilon}$ is the wave impedance and $R_0 = \pi\hbar/e^2$ is the quantum resistance expressed in terms of electron charge and Planck's constant. The shear stress (in contrast to the pressure term P),is stipulated by the attenuation of the eigenmode and disappears in the lossless limiting case. Numerical estimations of the typical parameters of conductive CNTs in the THz frequency range, give $|P|/4\pi\varepsilon|\bar{E}_z|^2 \approx 0{,}35$, which seems to exceed the corresponding values for different types of nanostructures (e.g., dielectric nano-spheres with or without the appearance of dielectric background [16]). The considered force is determined by the field discontinuities prevailing at the CNT boundary. These discontinuities may be also considered as a proper limit of a gradient of a non-homogeneous field. Thus, from a physical point of view, the considered force may be interpreted as an analog of the gradient force [17].

V. The Excitation Of Radial Breathing Mode By The Surface EM-Wave

In order to demonstrate some possible application of the developed theory, we consider the excitation of a CNT radial breathing mode (RBM), by the surface electromagnetic mode. For simplicity, we assume that the CNT is empty and employ a classical continuum approach to model CNT elasticity [17]. The frequency of the RBM mode can be written as

$$\omega_{RBM} = \frac{1}{R}\sqrt{\frac{c_{11}}{\rho_{2D}}} \quad (9)$$

where ρ_{2D} represents the area mass density, R is the tubes radius and c_{11} denotes the elastic stiffness coefficient in the Voigt notation [17]. The radial frustration of the CNT, corresponds to the forced oscillations of an harmonic oscillator satisfying the following equation

$$\frac{d^2 r}{dt^2} + \omega_{RBM}^2 r = \frac{P}{\rho_{2D}} \quad (10)$$

where P is the magnitude of the (radial) pressure defined by eq (8) and r denotes the radial CNT displacement. Following [17], we choose $\rho_{2D} \approx 10^{-6}\mathrm{kg/m^2}$,$c_{11} \approx 352\mathrm{N/m}$, which for a CNT with radius of 1.0nm, renders a frequency $\omega_{RBM} \approx 0.6\mathrm{THz}$. This value is found to be small compared with typical frequencies (10-100THz) of surface EM-modes [12]. Therefore, an efficient excitation of RBM in CNT, is possible by using for example periodic chains of electromagnetic pulses. In this case, the optical force defined in eq. (8), becomes a periodic function in time. If the inversed value of this period is small compared with the frequency of surface EM-mode and is approximately equal to the RBM-frequency given by eq. (9), we obtain a resonant regime of RBM excitations.

A more complicated scenario, occurs in the case of a CNT filled with a compressible (inviscid) liquid. The radial frustration of the CNT is then coupled with the compressible pressure wave [20], which propagates in the liquid along the tube in the form of a traveling wave. The presence of such waves in a liquid-filled CNT, may be further enhanced by means of optical forces too.

VI. Conclusion And Outlook

A fundamental theory of optical forces acting on a system of two interacting CNTs, is presented. The system of forces exerted, on a single CNT is obtained as a degenerate case when the distance between the two tubes is very large. The analysis is based on employing the Maxwell stress tensor, which is calculated via integral equations technique. The underlying theory is free of some of the prevailing limitations of conventional methods for evaluating CNT's optical forces. The qualitative behavior of optical forces in CNTs, is defined to a considerable degree by the existence of strongly retarded surface modes, which are absent in other types of nanostructures. Our analysis shows that typical values of optical forces acting on a CNT, are generally larger or at least comparable with those exerted on other types of nanostructures. For this reason, we strongly believe that CNTs are promising candidates for diverse applications in nano-mechanics (e, g., optical traps or optical tweezers). Our analysis may also lead to a new research activity in the field of optical forces –namely the effect of interacting eigen-modes of CNTs. For future activity, we intend to examine the additional effects connected with chirality and non-homogeneity. Taking these effects into account may shed some new light on the way that eigen-modes are produced. Considering those new interference effects between nearby carbon (chiral or achiral nanotubes, make CNTs also a promising tool for controlling their surface elastic vibrational modes, liquid flows (discharge) inside them, as well as the induced fluid mixing in the surrounding liquid.

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