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DERIVATION OF THE CARBON NANOTUBE SUSCEPTIBILITY TENSOR USING LATTICE DYNAMICS FORMALISM

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Abstract—We develop in this paper a theoretical approach to describe the electrodynamics of carbon nanotubes (CNTs). A lattice dynamics formalism is employed to model the mechanical response of matter to the radiation field. We start first by deriving the normal modes of the free lattice. Then, a simple and general microscopic model for light-matter interaction is proposed and the resulting mechanical equation of motion is derived using a suitable Lagrangian formalism. The symmetry group of the CNT is employed to explicitly probe the nonlocal structure of the fields and to carefully insure that higher-order Floquet modes are included in the derivation. The normal modes are then employed to perform an eigenmode expansion for the solution of the mechanical equation of motion, leading to the susceptibility tensor of the CNT medium. The final expression of this tensor describes the electrodynamics in the CNT viewed as a low-dimensional surface and is shown to be reduced effectively to a one-dimensional response function.

1. INTRODUCTION

The maturation of recent developments in measurement technology during the last 15 years has created a breakthrough in our capabilities to monitor and study structures at the nanoscale [1–3]. Of special importance is a class of low-dimensional structures called carbon nanotubes (CNT), which were discovered in 1991 by Iijima [4]. Since then, they have attracted the attention of both physicists and engineers who investigated extensively the possibility of using CNTs for various applications, ranging from mechanical shielding to nano-waveguide and transistors [5]. One of the most attractive features in CNTs is

their ability to function in either metallic or semi-conducting modes, depending on how the original graphite sheet is rolled to form the tube [6].

The electrodynamics of nanostructures differs considerably from the conventional scheme adopted in engineering electromagnetics. In this paper, we provide a theoretical study and various demonstrations of these differences by using a lattice dynamics formalism to derive the dielectric tensor of the CNT. We highlight in particular two major themes in the electrodynamics of nanometer structures.

- **Mechanics is Inevitable**: In the microscopic treatment of light-matter interaction, the mechanical response of matter represents the sole interaction with light. Therefore, any derivation of the dielectric tensor must use a mechanical model. In this paper we choose the linear lattice vibration approach for its conceptual simplicity, wide applicability, and the existence of an exact quantum mechanical analogy.
- T**he Nonlocality of the Fields**: This point is less obvious than the previous one. At the nanometer level, the periodic arrangements of atoms in crystals (like nanotubes and graphenes) creates higher-order (Floquet) mode interactions which modify the structure of the applied fields. For a satisfactory derivation of the optical characteristics of CNTs, it is important to calculate these higher-order contributions and include them directly in the final expressions. We highlight this feature in our mathematical formulation throughout the paper.

Based on the theme above, we develop in this paper a theoretical description of the carbon nanotube electrodynamics starting from suitable microscopic considerations. Previous studies of the lattice dynamics of CNTs reported applications to the mechanical and acoustic aspects of the device [17]. Here, we provide a tentative treatment of the electromagnetic coupling between an external field and the mechanical lattice vibration modes. The contribution of the electronic response to the susceptibility is treated on a phenomenological level coupled with the lattice vibration of the nanotube.

This paper is organized as follow. We start by reviewing the structure of CNTs, which is summarized using basic symmetry groups developed previously in the literature. The normal modes of the free lattice are then derived carefully. Next, we formulate a general lattice dynamics microscopic model for the light-matter interaction. The equations of motion are derived using a full Lagrangian formalism. We enforce explicitly the nonlocal structure of the fields in the derived equation of motion. This equation is then solved using an

eigenvalue expansion in terms of the free lattice normal modes obtained previously, leading to an expression for the susceptibility tensor of the nanotube. Finally, conclusions are given.

2. STRUCTURE OF CARBON NANOTUBES

In this section, we provide a compact description of the structural properties of CNTs using the language of the associated symmetry group. The details of the symmetry operations will be used in the remaining parts of this paper to find the specific mathematical form of the eigenmodes appearing in the dielectric tensor of the nanotube. The crux of our approach is that the symmetry group will determine the boundary conditions of the problem [12]. These conditions can not be derived form the Hamiltonian of the CNT but should be obtained through an independent approach. Fig. 1 illustrates the honeycomb lattice structure of graphene (graphene is defined as a 2D layer of graphite). The unit cell is specified by two atoms located at the positions $1/3\,(\mathbf{a}_1 + \mathbf{a}_2)$ and $2/3\,(\mathbf{a}_1 + \mathbf{a}_2)$, where $\mathbf{a}_1$ and $\mathbf{a}_2$ are two unit vectors defining the lattice constants and $|\mathbf{a}_1| = |\mathbf{a}_2| = a_0 = 0.2461\,\text{nm}$. The CNT is formed by rolling this sheet such that the circumference of the tube coincides with the *chiral vector* $\mathbf{c} = m\,\mathbf{a}_1 + n\,\mathbf{a}_2$. Here m and n are two integers that completely determine the structure and the properties of the CNT. If only one layer is used to form the tube, the resulting structure is called *single-wall* CNT (SWCNT). Alternatively, if the tube consists of several concentric cylinders, we call it *multi-wall* CNT (MWCNT). The chiral angle θ is defined as the angle between

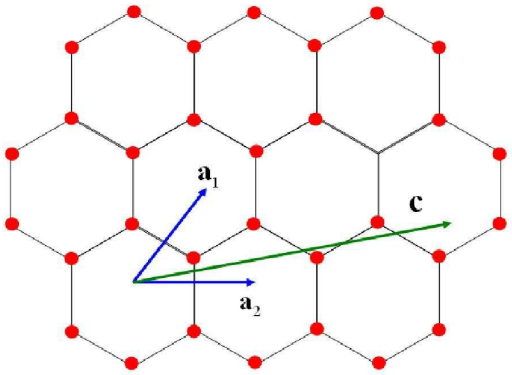

Figure 1. Graphene sheet used in forming a CNT. The dots illustrate the positions of carbon atoms.

the chiral vector $\mathbf{c}$ and $\mathbf{a}_1$. The radius of the CNT is given by

$$\rho_{\rm cn} = \frac{|\mathbf{c}|}{2\pi} = \frac{a_0}{2\pi}\sqrt{m^2+mn+n^2}. \tag{1}$$

The first symmetry group for the CNT can be derived by writing $\mathbf{c} = N\left[(m/N)\,\mathbf{a}_1 + (n/N)\,\mathbf{a}_2\right] = N\,\mathbf{c}'$, where N is the greatest common divider of m and n. It is obvious that a rotation around the tube axis by $2\pi/N$ will preserve the original structure. Equivalently, we say that each CNT possess the symmetry operation $\mathbf{C}_N$.

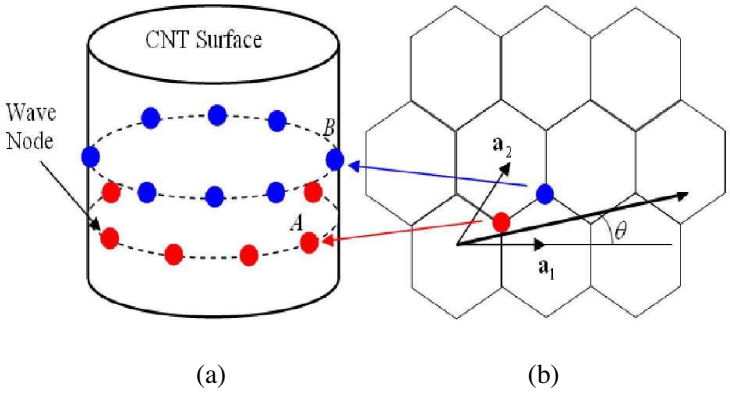

Figure 2. (a) Illustration of two atomic circles in the translational unit cell of a SWCNT. Each circle corresponds to the symmetry operation $\mathbf{C}_N$. (b) The honeycomb lattice structure of graphene. The dots represent the arbitrary first two-atom unit cell.

Through knowledge of other symmetry operations, to be discussed in a moment, it is possible by following the original work of White et al. [14] to show that the entire tube can be constructed starting from two-atom unit cell and the chiral vector. The first atom in the unit cell will be mapped to an arbitrary location in the circumference of the CNT, call it A. The second atom will be mapped to other location B obtained by rotating the first point by an angle $\gamma = 2\pi\,(\mathbf{a}_1+\mathbf{a}_2)\cdot\mathbf{c}/3\,|\mathbf{c}|^2$ and then translating the resulting point along the tube axis by a distance $d = |(\mathbf{a}_1+\mathbf{a}_2)\times\mathbf{c}|/3\,|\mathbf{c}|^2$. The two points obtained now can be used, together with the symmetry operation $\mathbf{C}_N$, to generate other $2(N-1)$ points by successive rotations around the tube axis by the angle $2\pi/N$. That is, atom A, together with $\mathbf{C}_N$, will generate other $N-1$ atoms. Atom B, again together with $\mathbf{C}_N$, will generate other set of $N-1$ atoms. Therefore, the total is

$2(N-1)+N=2N$ atoms. This set is called the *basic helical motif* of the CNT [14]. However, to complete the tube we need to use a *screw operator*, defined in [14], which is denoted here by $\mathbf{T}_h^\zeta$. Its action on a specific point consists of rotation around the tube axis by an angle ζ and then translation along the tube axis by a distance h. Therefore, the screw operation is neither pure rotation nor pure translation, but a combination of both of them. A procedure for finding ζ and h can be found in [14] and explicit closed form expressions in terms of Euler function are available as follow [15]

$$q = 2\frac{m^2+mn+n^2}{NR}, \tag{2}$$

$$\xi = \frac{q}{N}\mathrm{Fr}\left[\frac{N}{qR}\left(3-2\frac{m-n}{m}\right)+\frac{N}{m}\left(\frac{m-n}{N}\right)^{\mathrm{E}(m/N)-1}\right], \tag{3}$$

$$\zeta = \frac{2\pi}{q}\xi, \quad h = \frac{N}{\xi}a, \tag{4}$$

where $\mathrm{Fr}[x] = x - [x]$ gives the fractional part of the rational number x and $\mathrm{E}(v)$ is the Euler function giving the co-primes less than v (a is defined in (6)). By repeated application of the screw operator $\mathbf{T}_h^\zeta$ on the basic helical motif, the entire tube can be reconstructed [14]. Therefore, the two-atom unit cell that we started with is enough to determine the full structure of the CNT.

It remains to present the translational symmetry of the CNT. Depending on the chirality, each CNT will have a characteristic length a along its axis. Translations of multiples of a will leave the tube unchanged. This periodicity in the axial direction can be given by [15]

$$N_1 = -\frac{2n+m}{N\mathcal{R}}, \quad N_2 = \frac{2m+n}{N\mathcal{R}}, \tag{5}$$

$$\mathbf{a} = N_1\mathbf{a}_1 + N_2\mathbf{a}_2, \quad a = |\mathbf{a}| = \frac{\sqrt{3\left(m^2+mn+n^2\right)}}{N\mathcal{R}}a_0, \tag{6}$$

$$N_c = \frac{2\left(m^2+mn+n^2\right)}{N\mathcal{R}}, \tag{7}$$

where N_c is the number of atomic pairs in the translational unit cell. Here, $\mathcal{R} = 3$ if $(n_1-n_2)/3N$ is integer and $\mathcal{R} = 1$ otherwise. We denote here this symmetry operation by $\mathbf{T}_a$ and it means translation along the tube axis by distance a. The translational unit cell consists then of a cylindrical tube of radius ρ_{cn} and height a. The number of atoms in the unit cell is given by N_c.

To summarize, $\mathbf{C}_N$ is the symmetry operation of pure rotation (translations along the circumferential direction $\mathbf{c}$), $\mathbf{T}_a$ is the operation of pure translation, and $\mathbf{T}_h^\zeta$ is the (mixed) screw operation. It is worthy to mention that other symmetry operations were derived in [15] and [16] to fully characterize SWCNTs and MWCNTs. However, for the purpose of this paper the, previously mentioned three symmetry operations are enough to drive the basic mathematical form of the fields in the CNT environment. As will be shown in the next section, all other symmetry operations will alter only the number of terms taken in the resulted summations without changing the basic form.

3. LATTICE DYNAMICS FORMALISM

3.1. Motivation

The mathematical theory of lattice vibrations in solids is well-known since the original work of Max Born [11]. We start first in this section by providing a quick sketch of the theoretical background that motivates the application of lattice dynamics formalisms to study the optical properties in solids.

The exact solution for the problem of light-matter interaction requires applying quantum theory to describe the mechanical response of charged particles to the external fields. Such solution, however, implies dealing with the full Hamiltonian of the matter system, which complicates considerably the mathematical treatment. However, since the electron mass is much smaller than the nuclear mass, it is possible to write the Hamiltonian in the form $\mathcal{H}_0 = \mathcal{H}_{0e} + T$. Here, $\mathcal{H}_{0e}$ is the Hamiltonian of the electronic system and T is the kinetic energy operator of the nuclear parts. Without going to the mathematical details (see [13] for example), a perturbation method can be applied to the previous Hamiltonian to obtain a solution accurate to the order of the smallness of T with respect to $\mathcal{H}_0$. The approximation leading to this solution is called the *adiabatic approximation* and is one of the most fundamental tools in condensed matter physics. Within this approximation, the problem of the interaction of electromagnetic fields with CNT can be treated in terms of the lattice vibrations of the nuclei. It is as if we imagine at each moment that the nuclear motion is fixed and the electrons are responding by creating a nontrivial charge and current distribution. The nuclear motion is a *parameter* for the electronic state.

3.2. Hamiltonian of the Carbon Nanotube Free Lattice

The free lattice of carbon nanotubes is the periodic arrangement of atoms in the crystal structure as described by the exact symmetry groups of Section 2. The basic formulation of this paper will follow a classical Hamiltonian approach. Since we are going to proceed under the harmonic approximation assumption, an exact quantum-mechanical quantization of this classic solution can be obtained in a straightforward way.

Following the treatment of symmetry operations in Section 2, we write the position vector of the $\mathbf{n}s$th atom as

$$\mathbf{R}_{\mathbf{n}s} = S_{\mathbf{n}}^{\zeta,h}\mathbf{R}_{\mathbf{0}s}, \tag{8}$$

where $\mathbf{R}_{\mathbf{0}s}$ is the position vector of the sth atom ($s = 1, 2$) in the initial unit cell (atom pair) and the operator $S_{\mathbf{n}}^{\zeta,h}$ represents the repeated application of $\mathbf{C}_N$ followed by the skew operator, with parameters ζ and h, starting from the initial pair at $\mathbf{n} = \mathbf{0}$ and ending in the current pair $\mathbf{n}$. Equation (8) will be applied implicitly in all of the coming calculations.

We start with a crystal structure in the equilibrium state. Due to an external field, or any other source, the locations of the atoms will be disturbed. The resulting motion can be described as vibrations around a center defined by the undisturbed equilibrium locations, represented by a position vector $\mathbf{R}$. Let the displacement around this center be $\mathbf{u}$. By assuming that the external force is weak, the resulting displacement $\mathbf{u}$ can be considered as a small perturbation around the equilibrium position. Therefore, it is possible to expand the total potential energy of the lattice in power series as

$$\begin{aligned} U(\mathbf{R}+\mathbf{u}) \;=\; & U(\mathbf{R}) + \sum_{\mathbf{n}}\sum_{s}\frac{\partial U}{\partial \mathbf{R}_{\mathbf{n}s}}\mathbf{u}_{\mathbf{n}s} \\ & + \frac{1}{2}\sum_{\mathbf{n}}\sum_{s}\sum_{\mathbf{n}'}\sum_{s'}\mathbf{u}_{\mathbf{n}'s'}\frac{\partial U}{\partial \mathbf{R}_{\mathbf{n}s}\partial \mathbf{R}_{\mathbf{n}'s'}}\mathbf{u}_{\mathbf{n}s} + \cdots, \end{aligned} \tag{9}$$

where $\partial^2 U/\partial \mathbf{R}_{\mathbf{n}s}\partial \mathbf{R}_{\mathbf{n}'s'}$ is a 3×3 matrix and all vectors are three-dimensional. The $\alpha\beta$th element of this matrix is the second-order derivative $\partial^2 U/\partial R_\alpha \partial R_\beta$ evaluated at the equilibrium positions $R_\alpha = R_{\mathbf{n}s\alpha}$ and $R_\beta = R_{\mathbf{n}'s'\beta}$, where $\alpha, \beta = 1, 2, 3$ enumerate the three orthogonal spatial directions.

Since we assume that $\mathbf{R}$ describes the equilibrium state of the lattice, the potential energy attains a minimum at this location. Consequently, the second term in (9) will vanish. If we keep only the

second-order terms in the series expansion, we obtain the well-known linear harmonic approximation Hamiltonian [7, 11]

$$\mathcal{H} = \sum_{\mathbf{n}} \sum_{s} \frac{\mathbf{P}_{\mathbf{n}s} \cdot \mathbf{P}_{\mathbf{n}s}}{2M} + \frac{1}{2} \sum_{\mathbf{n}} \sum_{s} \sum_{\mathbf{n}'} \sum_{s'} \mathbf{u}_{\mathbf{n}'s'} \frac{\partial^2 U}{\partial \mathbf{R}_{\mathbf{n}s} \partial \mathbf{R}_{\mathbf{n}'s'}} \mathbf{u}_{\mathbf{n}s}, \quad (10)$$

where $\mathbf{P}_{\mathbf{n}s}$ is the momentum of the $\mathbf{n}s$th atom. The first term corresponds to the total kinetic energy and the second term to the potential energy of the CNT lattice. Since the entire tube is constructed from identical atoms (carbon), we introduce M to denote the atomic mass of carbon.

Once the Hamiltonian of the CNT is constructed, the dynamic equations follow directly

$$\dot{\mathbf{u}}_{\mathbf{n}s} = \frac{\partial \mathcal{H}}{\partial \mathbf{P}_{\mathbf{n}s}}, \qquad \dot{\mathbf{P}}_{\mathbf{n}s} = -\frac{\partial \mathcal{H}}{\partial \mathbf{u}_{\mathbf{n}s}}. \quad (11)$$

Thus, it follows from (10) and (11) that

$$M \ddot{\mathbf{u}}_{\mathbf{n}s} + \sum_{\mathbf{n}'} \sum_{s'} A^{ss'}_{\mathbf{n},\mathbf{n}'} u_{\mathbf{n}'s'} = 0, \quad (12)$$

where the matrix $A^{s,s'}_{\mathbf{n},\mathbf{n}'}$ is defined as

$$A^{ss'}_{\mathbf{n},\mathbf{n}'} = \frac{\partial^2 U}{\partial \mathbf{R}_{\mathbf{n}s} \partial \mathbf{R}_{\mathbf{n}'s'}}. \quad (13)$$

Equation (12) captures the dynamics of the CNT lattice under no external electromagnetic field. Therefore, its solutions should provide the free vibrations, or the *normal modes*, from which we can later expand the (driven) response to the field.

3.3. Normal Modes

Assume that the axis of the CNT coincides with the z-direction. In light of Bloch's theorem, the general solution of Equation (12) can be written as [7, 9, 11]

$$\mathbf{u}_{\mathbf{n}s} = \mathbf{u}_s e^{-j\mathbf{k}\cdot\mathbf{R}_{\mathbf{n}}} e^{j\omega t} = \mathbf{u}_s e^{-jk_\phi \phi_{\mathbf{n}}} e^{-jk_z z_{\mathbf{n}}} e^{j\omega t}, \quad (14)$$

where we have used the notation $\mathbf{R}_{\mathbf{n}} = \langle z_{\mathbf{n}}, \phi_{\mathbf{n}} \rangle$ and $\mathbf{k} = \langle k_z, k_\phi \rangle$. Here $\mathbf{u}_s$ is a constant of motion that depends on $\mathbf{k}$. We will show now that the symmetry group of the CNT leads to the result that k_z must be continuous while k_ϕ is quantized.

Assume that the CNT is finite in the longitudinal (axial) direction, in which it consists of L identical unit cells each with length a. By applying the periodic boundary condition of Born [11], we get $\mathbf{u}_{\mathbf{n}s}(z=0) = \mathbf{u}_{\mathbf{n}s}(z=La)$, which in light of (14) will give the condition $e^{-jk_zLa} = 1$. Thus, we can write $k_z = (2\pi/La)\, l'$, $l' = 0, 1, 2, \ldots, \infty$. For very large L, the ratio $2\pi/La$ becomes very small. Since the CNT structure is infinite in the z-direction, we require $L \to \infty$, and k_z therefore becomes continuous.

For the transverse (circumferential) direction, we apply the symmetry operation $\mathbf{C}_N$, which states that rotations around the z-axis will preserve the structure. Therefore, we write $l \cdot \lambda = c = 2\pi\rho_{\mathrm{cn}}$, where c is the circumference of the nanotube. This leads to the quantization rule

$$k_\phi = l, \ \forall\, l = 0, 1, 2, \ldots, N_c - 1. \tag{15}$$

We can now write the lth normal mode of the free lattice as

$$\mathbf{u}_{\mathbf{n}s} = \mathbf{u}_s e^{-jl\phi_{\mathbf{n}}} e^{-jk_z z_{\mathbf{n}}} e^{j\omega_{\mathbf{k}} t}, \tag{16}$$

where the function $\omega_{\mathbf{k}} = \omega(\mathbf{k})$ is the dispersion relation (to be found). Substituting (16) into (12), we get

$$-M\omega_{\mathbf{k}}^2 \mathbf{u}_s + \sum_{\mathbf{n}'} \sum_{s'} A^{ss'}_{\mathbf{n},\mathbf{n}'} \mathbf{u}_s e^{-jl(\phi_{\mathbf{n}'} - \phi_{\mathbf{n}})} e^{-jk_z(z_{\mathbf{n}'} - z_{\mathbf{n}})} = 0. \tag{17}$$

The above equation reduces to

$$-M\omega_{\mathbf{k}}^2 \mathbf{u}_s + \sum_{s'} \hat{A}^{ss'}(\mathbf{k})\, \mathbf{u}_s = 0, \tag{18}$$

where the dynamic matrix

$$\hat{A}^{ss'}(\mathbf{k}) = \sum_{\mathbf{m}} A^{ss'}_{\mathbf{m},\mathbf{n}} e^{j\,\mathbf{k}\cdot\mathbf{R}_{\mathbf{m}}} \tag{19}$$

can be interpreted as the discrete Fourier transform of the lattice force matrix $A^{ss'}_{\mathbf{m},\mathbf{n}}$. It can be shown that by employing the general symmetry properties of the dynamic matrix that this transform does not depend on $\mathbf{n}$ [13].

Equation (18) is an eigenvalue problem and can be set in the form

$$\left\| M\omega_{\mathbf{k}}^2 \delta_{ss'} \delta_{\alpha\alpha'} - \hat{A}^{ss'}_{\alpha\alpha'}(\mathbf{k}) \right\| = 0, \tag{20}$$

where the symbol $\|\cdot\|$ denotes the determinant and $\delta_{ss'}$ is the Kronecker delta function. Here, $\alpha = 1, 2, 3$ enumerates the three spatial direction

and $s = 1, 2$ enumerates the atom in the $\mathbf{n}$th two-atom unit cell. Therefore, Equation (20) is of order 6. Its eigenvalues will determine the dispersion relation while the eigenvectors constitute a complete set of basis function for the representation of the full space of the lattice vibrations.

Let us introduce the canonical variables $\tilde{\mathbf{u}}_{\mathbf{n}s} = M^{1/2}\mathbf{u}_{\mathbf{n}s}$ and $\tilde{A}^{ss'}(\mathbf{k}) = M^{-1}\hat{A}^{ss'}(\mathbf{k})$. Now, the fact that the force matrix (13) is conjugate symmetric with respect to the transformation $\mathbf{n}s\alpha \longleftrightarrow \mathbf{n}'s'\alpha$ makes its Fourier transform (19) Hermitian. Following the standard diagonalization procedure in linear algebra, the eigenvalue problem (20) leads to a set of real eigenvalues $\omega^i(\mathbf{k})$ and complex eigenvectors $\tilde{e}^i_{s\alpha}(\mathbf{k})$ satisfying the following equations

$$\sum_s \sum_\alpha \tilde{e}^i_{s\alpha}(\mathbf{k})\, \tilde{e}^{*i'}_{s\alpha}(\mathbf{k}) = \delta_{ii'} \quad \text{(orthonormality)}, \tag{21}$$

$$\sum_i \tilde{e}^{*i}_{s\alpha}(\mathbf{k})\, \tilde{e}^i_{s'\alpha'}(\mathbf{k}) = \delta_{ss'}\delta_{\alpha\alpha'} \quad \text{(completeness)}, \tag{22}$$

$$\sum_{s,s'} \sum_{\alpha,\alpha'} \tilde{e}^{*i}_{s\alpha}(\mathbf{k})\, A^{ss'}_{\alpha\alpha'} \tilde{e}^{i'}_{s\alpha}(\mathbf{k}) = \delta_{ii'}\omega^i(\mathbf{k})^2 \quad \text{(diagonality)}. \tag{23}$$

Therefore, the most general solution to the lattice dynamics problem takes the following form

$$u_{\mathbf{n}s\alpha} = \sum_{i=1}^{6} \sum_{l=1}^{N_c} \int_{-\pi}^{\pi} dk_z \varsigma^i(k_z, l) \tilde{e}^i_{s\alpha}(k_z, l) \exp\left(j\omega^i_l(k_z)\, t\right) \exp(-jl\varphi_{\mathbf{n}} - jk_z z_{\mathbf{n}}), \tag{24}$$

where $\varsigma^i(k_z, l)$ are the expansion weights.

3.4. Expansion of the Force Matrix

To obtain numerical values for the eigenvalue and eigenvectors derived in the previous section, an estimation of the unknown force constants in (13) must be attempted. However, although the force matrix of the lattice vibration in CNTs is phenomenological, and experimental results are needed to accurately determine its values, we will show in this section that a considerable reduction in the complexity of the problem can be achieved if the symmetry operations of the CNT are applied. Since the potential function is a system characteristic, not a wave, the periodic boundary condition can not be applied to the axial direction; pure translations in the z-direction will maintain their discrete nature. Together with the rotational periodicity, we

expand the potential function as a Fourier series in both the z- and φ-directions as follow

$$U(z,\varphi,\rho) = \sum_{\mathbf{l}} \Gamma_{\mathbf{l}}(\rho)\, e^{j\mathbf{g_l}\cdot\mathbf{t}}, \tag{25}$$

where $\mathbf{l} = (l_1, l_2)$ and

$$\mathbf{g_l} = \left\langle \frac{2\pi}{a} l_2, N l_1 \right\rangle, \qquad \mathbf{t} = \langle z, \phi \rangle . \tag{26}$$

The summation in (25) can be further restricted by applying the screw symmetry condition

$$U(z,\phi,\rho) = U(z-h, \phi-\zeta, \rho), \tag{27}$$

with ζ and h are the parameters of the screw operator defined in (4). Equation (27) gives $\exp\left(jl_1 N\zeta + j\frac{2\pi}{a} l_2 h\right) = 1$, which translates to the following condition

$$\mathbf{C}: \;\; l_1 N\zeta + l_2 \frac{2\pi h}{a} = 2\pi v, \quad v = 0, \pm 1, \pm 2, \ldots, \pm\infty. \tag{28}$$

In the coming parts, we write a summation over $\mathbf{l}$ that takes into consideration the condition $\mathbf{C}$, as defined in (28), in the form $\sum_{\mathbf{l}|\mathbf{C}}$. Therefore, using the definition of the force matrix in (13), we can arrive to the reduced expansion shown in Appendix A. The attractive feature of this expansion is that all the functions are calculated at the same radial distance $\rho = \rho_{\mathrm{cn}}$, the radius of the CNT in (1). Therefore, the problem of determining the force constants for all dimensions is reduced to knowledge of sufficient number of the coefficients $\Gamma_{\mathbf{l}}$ and $\partial\Gamma_{\mathbf{l}}/\partial\rho$ evaluated at the single point $\rho = \rho_{\mathrm{cn}}$. This is a considerable reduction of the number of degrees of freedom in the original problem that was achieved by applying the symmetry group of CNTs.

4. INTERACTION WITH THE ELECTROMAGNETIC FIELD

4.1. Microscopic Model for the Interaction

In this section, we provide a simple atomic model for the interaction between external electromagnetic fields and the lattice of CNTs. The starting assumption is that the amplitude of the external source is weak, giving rise to small distortion in the electronic clouds

surrounding the vibrating nuclei in the lattice dynamics model formulated in the previous parts. To accomplish this formally, we assume that the motion of atoms in the lattice will introduce a small distribution of non-trivial electric charge density $\delta\rho$ and current density $\delta\mathbf{J}$. Therefore, based on the first-order approximation, we retain only the linear variation of the functional dependence of these quantities in their Taylor series expansion. It is possible then to write

$$\delta\rho = \sum_{\mathbf{n}} \sum_{s} \sum_{\alpha} \Upsilon_{\mathbf{n}s\alpha}\left(\mathbf{r}\right) u_{\mathbf{n}s\alpha} \tag{29}$$

and

$$\delta J_{\alpha} = \sum_{\mathbf{n}} \sum_{s} \sum_{\beta} \Gamma_{\alpha\beta}^{\mathbf{n}s}\left(\mathbf{r}\right) \dot{u}_{\mathbf{n}s\beta}, \tag{30}$$

where $\Upsilon_{\mathbf{n}s\alpha}$ and $\Gamma_{\alpha\beta}^{\mathbf{n}s}$ are phenomenological parameters describing the "rate" of the system response to the linear variation in the displacements $u_{\mathbf{n}s\alpha}$. These can be calculated using a suitable quantum-mechanical model. However, such calculations are beyond the scope of this paper and will not be carried out here. Notice that zero-order terms in the Taylor series are exactly zero because the overall CNT is electrically neutral.

It should be noticed that the assumptions behind the expansions (29) and (30) are quite general and apply to most of the practical cases encountered in the study of material response to weak electromagnetic fields, with exceptions mainly in laser applications where the nonlinear effects must be accounted for explicitly. Moreover, a quantum-mechanical treatment of the nontrivial charge and current distributions $\delta\rho$ and $\delta\mathbf{J}$ can be included implicitly in the functions $\Upsilon_{\mathbf{n}s\alpha}$ and $\Gamma_{\mathbf{n}s}$. Therefore, our model can include for pure quantum effects, like electronic transitions, as long as the adiabatic approximation is valid.

4.2. Coupling between External Fields and the Lattice Oscillators

Based on the microscopic model of Section 4.1, we derive the equation of motion under the influence of an applied electromagnetic field described by scalar and vector potentials ϕ and $\mathbf{A}$. Within the adiabatic approximation, we start from the following non-relativistic

Lagrangian [18]

$$L = \frac{1}{2}\sum_{\mathbf{n}}\sum_{s}\sum_{\alpha} M\left(\dot{u}_{\mathbf{n}s\alpha}\right)^2 - \frac{1}{2}\sum_{\mathbf{n}}\sum_{s}\mathbf{u}_{\mathbf{n}'s'}A^{ss'}_{\mathbf{n},\mathbf{n}'}\mathbf{u}_{\mathbf{n}s} + \sum_i e_i\mathbf{v}_i\cdot\mathbf{A}\left(\mathbf{r}_i,t\right) - \sum_i e_i\phi\left(\mathbf{r}_i,t\right) + \int \mathrm{d}^3r\mathcal{L}_{EM}, \quad (31)$$

where i enumerates the fundamental charges e_i and $\mathcal{L}_{EM}$ is the field Lagrangian density given by

$$\begin{aligned}\mathcal{L}_{EM} &= \frac{\varepsilon_0}{2}\left[\left(\nabla\phi + \frac{\partial\mathbf{A}}{\partial t}\right)^2 - c^2\left(\nabla\times\mathbf{A}\right)^2\right] \\ &= \frac{\varepsilon_0}{2}\left(|\mathbf{E}|^2 - c^2|\mathbf{B}|^2\right). \end{aligned} \quad (32)$$

Due to the microscopic nature of the sources, we can write [8]

$$\begin{aligned}\delta\rho &= \sum_i e_i\delta\left[\mathbf{r} - \mathbf{r}_i\left(t\right)\right], \\ \delta\mathbf{J} &= \sum_i e_i\dot{\mathbf{r}}_i\delta\left[\mathbf{r} - \mathbf{r}_i\left(t\right)\right].\end{aligned} \quad (33)$$

Therefore, the Lagrangian in (31) takes the form

$$L = \frac{1}{2}\sum_{\mathbf{n}}\sum_{s}\sum_{\alpha} M\left(\dot{u}_{\mathbf{n}s\alpha}\right)^2 - \frac{1}{2}\sum_{\mathbf{n}}\sum_{s}\mathbf{u}_{\mathbf{n}'s'}A^{ss'}_{\mathbf{n},\mathbf{n}'}\mathbf{u}_{\mathbf{n}s} + \int \mathrm{d}^3r\,\mathbf{A}\left(\mathbf{r},t\right)\cdot\delta\mathbf{J} - \int \mathrm{d}^3r\,\phi\left(\mathbf{r},t\right)\delta\rho + \int \mathrm{d}^3r\mathcal{L}_{EM}. \quad (34)$$

The first and second terms represent the mechanical energies, kinetic and potential, respectively. The third and fourth terms capture the interaction between the external fields and the local lattice sources. The last term is a representation of the energy stored in the fields.

Applying Hamilton's principle, the equation of motion under the Lagrangian (34) is given by [8]

$$\frac{d}{dt}\left(\frac{\partial L}{\partial\dot{u}_{\mathbf{n}s\alpha}}\right) - \frac{\partial L}{\partial u_{\mathbf{n}s\alpha}} = 0. \quad (35)$$

In Appendix B, we show that the following equation of motion can be derived from the Lagrangian formalism above

$$M\ddot{u}_{\mathbf{n}s\alpha} + \sum_{\mathbf{n}'}\sum_{s'}\sum_{\alpha'} A^{ss'\alpha\alpha'}_{\mathbf{n},\mathbf{n}'}u_{\mathbf{n}'s'\alpha'} = \int \mathrm{d}^3r\,\Gamma^{\mathbf{n}s}_{\alpha}\left(\mathbf{r}\right)\cdot\mathbf{E}\left(\mathbf{r},t\right). \quad (36)$$

This is an inhomogeneous equation that governs the dynamics of the system under the influence of external electromagnetic field.

4.3. The Nonlocal Structure of the Fields

Following the general theoretical framework developed in [12], we emphasize that when applying the electromagnetic theory to problems at the nanometer scale it is of paramount importance to take into consideration the non-local relations between the fields and the induced polarization at different spacial locations. This is particularly important when dealing with crystal structures like CNTs where the prevailing characteristics are periodic, as exemplified by the associated symmetry group.

There is some confusion about non-locality in electromagnetic theory, which accompanies the nature of the field appearing in (36). Therefore, we will spend some time discussing the conceptual background of the problem. First, as we mentioned before, the correct mathematical treatment of the problem requires that we insert the *actual* field, which is different from the applied field [6, 7, 9]. Moreover, in [12] an explicit form for the field in the CNT medium was derived and showed that higher-order (Floquet) modes will emerge from the periodic structure of the problem. The main reason why such higher-order modes are sometimes ignored in literature is the fact that we usually apply conventional *macroscopic* electromagnetic theory, where the fields are averaged over spacial regions large compared to the microscopic scale but small in comparison to the macroscopic one. Therefore, all of the short-wavelength components will be washed out by the averaging in the theory. For a satisfactory derivation of the optical characteristics using microscopic models, this averaging, which amounts to loosing some of the important information in the field structure that may participate in determining the final observed value for the dielectric function must be avoided. Now, although this function itself is usually measured by a device that inherently averages the results, we should notice that such averaging in the final result is *not* equivalent to averaging the fields from the start using a macroscopic electromagnetic theory. The mathematical treatment of the incoming parts are designed to clearly demonstrate these claims in the particular example of nanotubes.

To summarize, what is needed for a correct microscopic derivation of the optical characteristics is two requirements

1. Distinction between the applied and actual (local) field.
2. Distinction between the "slow" part of the field (long-wavelength, fundamental mode, low-frequency part, etc) and the "fast" part

(short-wavelength, higher-order modes, high-frequency parts, etc).

In the present approach, we achieve the two requirements above simultaneously by using the microscopic model for the interaction in Section 4.1. We first translate the first requirement into the following equation

$$\mathbf{E} = \mathbf{E}^e + \delta\mathbf{E}, \tag{37}$$

where $\mathbf{E}$ refers here to the actual (local) field acting on the atoms in the CNT. $\mathbf{E}^e$ is the external (applied) field. The difference between the actual and the external field is denoted by $\delta\mathbf{E}$. The mechanism behind its existence is the distortion of the electronic wave functions and the nuclear displacements through the lattice vibrations that result in the build up of non-trivial charge and current distributions (29) and (30).

Without loss of generality, we assume that the external field has only long-wavelength component (averaged). The induced fields $\delta\mathbf{E}$, however, contains all the harmonics of the CNT structure due to the non-local effects [12]. Therefore, we expand the total field as

$$\mathbf{E} = \bar{\mathbf{E}} + \tilde{\mathbf{E}}, \tag{38}$$

where $\bar{\mathbf{E}}$ is the long-wavelength (slow) part and $\tilde{\mathbf{E}}$ is the short-wavelength (fast) part. We immediately obtain

$$\bar{\mathbf{E}} = \mathbf{E}^e + \delta\bar{\mathbf{E}} \tag{39}$$

and

$$\tilde{\mathbf{E}} = \delta\tilde{\mathbf{E}}. \tag{40}$$

Within the general assumption of the linear harmonic approximation of the lattice dynamics and the weakness of the external sources, we expand $\delta\mathbf{E}$ in Taylor series and retain only the linear terms

$$\delta E_\alpha = \sum_{\mathbf{n}'} \sum_{s'} \sum_{\alpha'} \Lambda_{\alpha\alpha'}^{\mathbf{n}'s'}(\mathbf{r})\, u_{\mathbf{n}'s'\alpha'}, \tag{41}$$

where $\Lambda_{\alpha\alpha'}^{\mathbf{n}s}(\mathbf{r})$ is some unknown function that represents the "rate" of the change in the system response to the linear displacements $u_{\mathbf{n}s\alpha}$. Now, this function is a characteristic of the original lattice (zero displacement). Therefore, it must obey the exact symmetry group introduced in Section 2. We can then expand it in a Fourier series like Equation (25). We write then (41) as

$$\delta E_\alpha = \sum_{\mathbf{n}'} \sum_{s'} \sum_{\alpha'} \sum_{\mathbf{l}|\mathbf{C}} \Lambda_{\alpha\alpha'}^{\mathbf{n}'\mathbf{s}'\mathbf{l}}(\rho)\, e^{j\mathbf{g}_\mathbf{l}\cdot\mathbf{t}} u_{\mathbf{n}'s'\alpha'}, \tag{42}$$

where the Fourier expansion

$$\Lambda_{\alpha\alpha'}^{\mathbf{n}'s'}(\mathbf{r}) = \sum_{\mathbf{l}|\mathbf{C}} \Lambda_{\alpha\alpha'}^{\mathbf{n}'\mathbf{s}'\mathbf{l}}(\rho)\, e^{j\mathbf{g_l}\cdot\mathbf{t}}. \tag{43}$$

has been used. Here, $\mathbf{g_l}$ and $\mathbf{t}$ are defined as in (26). Based on the previous discussion, we write the long-wavelength part as the component $\mathbf{l} = 0$ and the short-wavelength parts as all the other (higher) order modes. We find

$$\delta\bar{\mathbf{E}} = \sum_{\mathbf{n}'}\sum_{s'}\sum_{\alpha'} \Lambda_{\alpha'}^{\mathbf{n}'s'0}(\rho)\, u_{\mathbf{n}'s'\alpha'} \tag{44}$$

and

$$\delta\tilde{\mathbf{E}} = \sum_{\mathbf{n}'}\sum_{s'}\sum_{\alpha'}\sum_{\mathbf{l},\mathbf{l}\neq 0} \Lambda_{\alpha'}^{\mathbf{n}'\mathbf{s}'\mathbf{l}}(\rho)\, e^{j\mathbf{g_l}\cdot\mathbf{t}} u_{\mathbf{n}'s'\alpha'}. \tag{45}$$

Substituting Equations (37)–(45) into (36) we obtain

$$M\ddot{u}_{\mathbf{n}s\alpha} + \sum_{\mathbf{n}'}\sum_{s'}\sum_{\alpha'} A'^{ss'\alpha\alpha'}_{\mathbf{n},\mathbf{n}'} u_{\mathbf{n}'s'\alpha'} = \int \mathrm{d}^3 r\, \Gamma_{\alpha}^{\mathbf{n}s}(\mathbf{r}) \cdot \bar{\mathbf{E}}(\mathbf{r},t), \tag{46}$$

where the new dynamic matrix is given by

$$A'^{ss'\alpha\alpha'}_{\mathbf{n},\mathbf{n}'} = A^{ss'\alpha\alpha'}_{\mathbf{n},\mathbf{n}'} - \sum_{\mathbf{l}\neq 0|\mathbf{C}} \int \mathrm{d}^3 r\, \Gamma_{\alpha}^{\mathbf{n}s}(\mathbf{r}) \cdot \Lambda_{\alpha'}^{\mathbf{n}'\mathbf{s}'\mathbf{l}}(\rho)\, e^{j\mathbf{g_l}\cdot\mathbf{t}} \tag{47}$$

and represents the structure of the lattice dynamics with the short-wavelength contribution of the fields taken implicitly into consideration. This re-arrangement of the equation of motion will allow us to calculate the (measured) dielectric function due to a *macroscopic* field $\bar{\mathbf{E}}$ with the microscopic higher-order Floquet modes already handled properly by the new matrix (47). The important feature of Equation (46) is that it has exactly the same structure of (36), rendering the theory of lattice vibrations developed in Section 3 applicable. The only important difference, however, is that the eigenvalues in (21), (22), and (23) are now generally complex.

4.4. The Dielectric Tensor

We have now all the tools required to derive an expression for the dielectric function of the CNT. To accomplish this, we excite the nanotube by a wave in the form

$$\mathbf{E} = \mathbf{E}_0 e^{-j\mathbf{k}\cdot\mathbf{r}} e^{j\omega t} = \mathbf{E}_0 e^{-jk_z z - jk_\phi \phi} e^{j\omega t}. \tag{48}$$

The response of the medium consists of lattice vibrations propagating in the direction $\mathbf{k} = \langle k_z, k_\phi \rangle$. That is, we restrict ourselves to the electrodynamics in the CNT medium. Using the general formulation of Section 3.3, it is possible to expand the medium response in terms of eigenmodes of the homogenous equation of motion as follow

$$u_{\mathbf{n}s\alpha} = \sum_i \eta_i (\mathbf{k}) e^{-j\mathbf{k}\cdot\mathbf{R_n}} e^{j\omega t} e^i_{s\alpha} (\mathbf{k}) . \tag{49}$$

Substituting (49) into (46), employing the canonical transformations $\tilde{u} = \sqrt{M}u$ and $\tilde{A}^{ss'}_{\alpha\alpha'} = (1/M)\, \hat{A}^{ss'}_{\alpha\alpha'}$, we find

$$\begin{aligned} &-\omega^2 \sum_i \eta_i \tilde{e}^i_{s\alpha} (\mathbf{k}) + \sum_{s'} \sum_{\alpha'} \sum_i \tilde{A'}^{ss'}_{\alpha\alpha'} \eta_i \tilde{e}^i_{s'\alpha'} (\mathbf{k}) \\ &= \frac{1}{\sqrt{M}} \int \mathrm{d}^3 r\, \Gamma^{\mathbf{n}s}_\alpha (\mathbf{r}) \cdot \mathbf{E}_0 e^{-j\mathbf{k}\cdot\mathbf{r}} e^{j\,\mathbf{k}\cdot\mathbf{R_n}}, \end{aligned} \tag{50}$$

where $\hat{A}'$ is the modified dynamic matrix defined exactly like (19). Multiplying (50) by $\tilde{e}^{*i'}_{s\alpha}$, summing over s and α, we obtain

$$\begin{aligned} &-\omega^2 \sum_i \sum_s \sum_\alpha \eta_i \tilde{e}^i_{s\alpha} \tilde{e}^{*i'}_{s\alpha} + \sum_i \eta_i \sum_s \sum_\alpha \sum_{s'} \sum_{\alpha'} \tilde{e}^{*i'}_{s\alpha} \tilde{A'}^{ss'}_{\alpha\alpha'} \tilde{e}^i_{s'\alpha'} \\ &= \frac{1}{\sqrt{M}} \sum_s \sum_\alpha \int \mathrm{d}^3 r\, \Gamma^{\mathbf{n}s}_\alpha (\mathbf{r}) \cdot \mathbf{E}_0 e^{-j\,\mathbf{k}\cdot\mathbf{r}} e^{j\,\mathbf{k}\cdot\mathbf{R_n}} \tilde{e}^{*i'}_{s\alpha}. \end{aligned} \tag{51}$$

Applying (21) and (23) to the first and second terms in (51), respectively, we can arrive to the following expression for the expansion coefficients

$$\eta_i (\mathbf{k}) = \frac{\frac{1}{\sqrt{M}} \sum_s \sum_\alpha \int \mathrm{d}^3 r\, \Gamma^{\mathbf{n}s}_\alpha (\mathbf{r}) \cdot \mathbf{E}_0 e^{-j\,\mathbf{k}\cdot(\mathbf{r}-\mathbf{R_n})} \tilde{e}^{*i'}_{s\alpha} (\mathbf{k})}{\omega_i^2 (\mathbf{k}) - \omega^2}. \tag{52}$$

We proceed now to calculate the induced current in the medium. Using (30), the total current is written as

$$J_\alpha = \frac{j\omega}{\sqrt{M}} \sum_{\mathbf{n}} \sum_s \sum_\beta \sum_i \Gamma^{\mathbf{n}s}_{\alpha\beta} (\mathbf{r})\, \eta_i e^{-j\,\mathbf{k}\cdot\mathbf{R_n}} e^{j\omega t} \tilde{e}^i_{s\alpha}, \tag{53}$$

where (49) has been used in writing (53). In order to simplify the forthcoming calculations, we resort to a the reasonable assumption that the coefficients of the Taylor series expansion $\Gamma^{\mathbf{n}s}_\alpha (\mathbf{r})$ are centered

around the $\mathbf{n}s$th unit cell. Thus, we write $\Gamma_{\alpha}^{\mathbf{n}s}(\mathbf{r}) = \Gamma_{\alpha}^{s}(\mathbf{r} - \mathbf{R}_{\mathbf{n}s})$. The Fourier transform of the current (53) is then written as

$$J_{\alpha}(\mathbf{k}, t) = \frac{1}{V} \int d^3 r J_{\alpha}(\mathbf{r}, t)\, e^{j\,\mathbf{k}\cdot\mathbf{r}}. \tag{54}$$

Therefore, we can compute the ℓ_1th-component of the CNT medium response to the ℓ_2th-component of the field ($\ell_1, \ell_2 = 1, 2, 3$) as

$$\begin{aligned} J_{\ell_1}(\mathbf{k}, t) \;=\; & \frac{j\omega}{VM} \int d^3 r \sum_{\mathbf{n}} \sum_{s} \sum_{\beta} \sum_{i} \frac{E_{0l_2} \Xi_{\ell_2}^{i}}{\omega_i^2 - \omega^2} \Gamma_{\ell_1\beta}^{s}(\mathbf{r} - \mathbf{R}_{\mathbf{n}s}) \\ & \times e^{j\,\mathbf{k}\cdot(\mathbf{r}-\mathbf{R}_{\mathbf{n}})} e^{j\omega t} \tilde{e}_{s\ell_1}^{i}(\mathbf{k}), \end{aligned} \tag{55}$$

where we define the *effective charge* as

$$\Xi_{\ell}^{i}(\mathbf{k}) = \sum_{s} \sum_{\alpha} \int d^3 r \phi\, \Gamma_{\alpha\ell}^{s}(\mathbf{r} - \mathbf{R}_{\mathbf{n}s})\, e^{-j\mathbf{k}\cdot(\mathbf{r}-\mathbf{R}_{\mathbf{n}})} \tilde{e}_{s\ell}^{*i}(\mathbf{k}). \tag{56}$$

It is clear that the integral in (56) can be made independent of $\mathbf{n}$ by the transformation $\mathbf{y} = \mathbf{r} - \mathbf{R}_{\mathbf{n}}$. Now, re-arranging (55), it is possible to write

$$\begin{aligned} J_{\ell_1}(k, t) \;=\; & \frac{j\omega}{VM} E_{0l_2} e^{j\omega t} \sum_{\mathbf{n}} \sum_{i} \frac{\Xi_{\ell_2}^{i}}{\omega_i^2 - \omega^2} \\ & \times \left[\sum_{s} \sum_{\beta} \int d^3 r\, \Gamma_{\ell_1\beta}^{s}(\mathbf{r} - \mathbf{R}_{\mathbf{n}s})\, \tilde{e}_{s\ell_1}^{i} e^{j\,\mathbf{k}\cdot(\mathbf{r}-\mathbf{R}_{\mathbf{n}})} \right]. \end{aligned} \tag{57}$$

Using the definition of the effective charge in (56) for the term between brackets in (57), we get

$$J_{\ell_1}(\mathbf{k}, t) = \frac{j\omega}{VM} E_{0l_2} e^{j\omega t} \sum_{\mathbf{n}} \sum_{i} \frac{\Xi_{\ell_1}^{i*}(\mathbf{k})\, \Xi_{\ell_2}^{i}(\mathbf{k})}{\omega_i^2(\mathbf{k}) - \omega^2}. \tag{58}$$

The polarization density $\mathbf{P}$ is connected to the volume current density $\mathbf{J}_v$ by the relation $\mathbf{J}_v(\mathbf{k}, t) = (\partial/\partial t)\, \mathbf{P}(\mathbf{k}, t)$. For the problem under consideration here, by restricting ourselves to the surface current and polarization density, we obtain

$$P_{\ell_1}(\mathbf{k}, \omega) = \frac{1}{j\omega} \left(N_c / 2\pi a \rho_{\mathrm{cn}}\right) J_{\ell_1}(\mathbf{k}, \omega) = \chi_{\ell_1\ell_2}(\mathbf{k}, \omega)\, E_{\ell_2}(\mathbf{k}, \omega), \tag{59}$$

where $\chi_{\ell_1\ell_2}$ is the susceptibility tensor of the medium. Here, since the effective charges are independent of $\mathbf{n}$, the current density emerges

naturally from truncating the summation over $\mathbf{n}$ in (58) for a certain number of atoms in the CNT surface and then dividing over the area of this surface in order to get the density of the current. From Section 2, this ratio can be found to be $N_c/2\pi a\rho_{\rm cn}$. Applying (59) to (58), we finally arrive to the following expression for the susceptibility tensor of the CNT

$$\chi_{\ell_1\ell_2}(\mathbf{k},\omega) = \frac{N_c}{2\pi a\rho_{\rm cn}M}\sum_i \frac{\Xi^{i*}_{\ell_1}(\mathbf{k})\,\Xi^{i}_{\ell_2}(\mathbf{k})}{\omega_i^2(\mathbf{k})-\omega^2}\,. \tag{60}$$

We keep the information about the electronic contribution to the total dielectric function in the effective charges defined in (56). Strictly speaking, if electronic transitions represent the main contribution to the dielectric function at a certain frequency band in which the lattice interactions are insignificant, then a phenomenological model, or an accurate quantum calculation, can be used in the response functions in (29) and (30). Within this scheme, the total dielectric function for the CNT medium can be finally stated as

$$\varepsilon_{\ell_1\ell_2}(\mathbf{k},\omega) = \varepsilon_0 + \frac{N_c}{2\pi a\rho_{\rm cn}M}\sum_i \frac{\Xi^{i*}_{\ell_1}(\mathbf{k})\,\Xi^{i}_{\ell_2}(\mathbf{k})}{\omega_i^2(\mathbf{k})-\omega^2}\,. \tag{61}$$

This represents the key result of the paper. We have derived the electrodynamic responses of CNT media using a microscopic model of its interaction with the electromagnetic field. Since the derivation was achieved within the linear oscillator formalism, a direct quantization procedure for the above formulation is straightforward but lengthy and avoided here because of the limitation of space.

5. DISCUSSION OF THE RESULTS

There are certain interesting features in the derived expression (61) that are worthy to be mentioned here from the purely theoretical point of view.

1. We first notice that since $\mathbf{k} = \langle k_z, k_\phi\rangle$, the fields are propagating only on the tube surface. That does not mean that no actual field can impinge obliquely on the tube, but it illustrates the peculiar, although not quite surprising, fact that for low-dimensional structures the entire electrodynamics takes place in a reduced-dimensional space. The CNT medium does not exist for $\rho \neq \rho_{\rm cn}$, which prohibits mechanical vibrations to propagate in the radial direction. Notice that the displacements themselves may have three spatial components, but the vibrations can propagate only

along the tube surface. For the more general case where there is an incident field *outside* the CNT and it is desired to calculate the scattered field, one should go beyond the model developed in this paper. It is well-known that for nanometer structures, where the effect of spatial dispersion is important, the conventional boundary-condition approach to Maxwell's equations fails to solve for the unknown fields uniquely. One actually needs additional boundary-conditions (ABC) augmented to Maxwell's boundary conditions [19]. Although there has been many approaches to solve this problem [18–20], the determination of these ABCs for a general structure is still an open problem. One should resorts to microscopic models to provide a specification of how the mechanical movements of the microscopic sources behave at the interface between the two media. In the case illuminated in this paper, this means that a more sophisticated model is needed to describe the mechanical motion of the oscillators in the lattice vibrations at the boundary $\rho = \rho_{\text{cn}}$. It is not clear how to accomplish this with our formalism. One possibility would be to introduce a more detailed mathematical model for the distribution of the microscopic charges (29) and (30) that is not necessary localized over the CNT surface. Then, by describing how its radial dependences behave while the oscillations pass through the boundary $\rho = \rho_{\text{cn}}$ it may become possible to derive the required ABC. However, we leave such speculations to future work. To summarize, it is stated that the derivation of this paper is meant to deal with the electrodynamics *within* the CNT medium. An observer living on the CNT surface (the medium) simply can not "see" the space where $\rho \neq \rho_{\text{cn}}$, which explains the absence of k_ρ in (61). The remarks here refers to the new situation where a dielectric constant for a sheet or a surface was derived rigorously without approximating the sheet as a limit of very small thin film.

2. The effective charges in $\Xi_\ell^i(\mathbf{k})$ are actually vectors. The tensor product $\Xi_{\ell_1}^{i*}(\mathbf{k})\,\Xi_{\ell_2}^{i}(\mathbf{k})$ is symmetric and does not reduce to scalar because the CNT has no cubic symmetry.
3. Although the susceptibility in (60) is written in terms of both k_ϕ and k_z, the problem is actually one-dimensional. As we saw in Section 3.3, k_ϕ is effectively quantized in the form $k_\phi = n$, where $n = 1, 2, \ldots, N_c$. Therefore, it is possible to write the field as

$$\mathbf{E} = \mathbf{E}_0 \sum_{n=1}^{N_c} a_n e^{-jn\phi} e^{-jk_z z}, \tag{62}$$

where a_n is the expansion coefficients. Substituting (62) to (60),

we find

$$\mathbf{P} = \bar{\chi}\left(k_z, \omega\right) \cdot \mathbf{E}_0, \tag{63}$$

where the effective 1D susceptibility $\bar{\chi}\left(k_z, \omega\right)$ is given by

$$\bar{\chi}\left(k_z, \omega\right) = \sum_{n=1}^{N_c} a_n \bar{\chi}\left(k_\phi = n, k_z, \omega\right) . \tag{64}$$

Therefore, it may be instructive to think of the electrodynamics of the CNT as occurring effectively in one-dimensional space with the mapping from the full 2D space to the reduced 1D space accomplished by (64).

6. CONCLUSION

A general derivation for the dielectric tensor of carbon nanotubes was achieved using a lattice dynamics formalism. The derivation demonstrated the peculiar feature of CNTs as a low-dimensional structure. Specifically, we showed that the problem can be reduced to a one-dimensional case. The derivation was based on a microscopic model for the charge and current sources induced in the system because of the lattice vibrations. We developed a Lagrangian formalism to deal with the problem and solved for the resulting mechanical motion using the dynamic (retarded) interactions through the lattice sites. The final expression for the dielectric tensor rests on only three reasonable assumptions: (1) The adiabatic approximation, (2) the external field is weak so the system response is linear, and (3) the nontrivial sources induced in the lattice are localized on the surface of the CNT, each around the center of the atomic unit cell. The lattice dynamics model of this paper can be easily quantized, rendering the model quantum-mechanically complete within the three assumptions stated above.

APPENDIX A. REDUCED FORM FOR THE FORCE MATRIX ELEMENTS

The list of the matrix elements calculated in Section 3.4 using the symmetry group of the CNT are listed here as follow

$$A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha = 1, \alpha' = 1\right) = \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}} \left(\frac{2\pi}{a}\right)^2 \left(l_2 l'_2\right) \Gamma_{\mathbf{l}} \Gamma_{\mathbf{l}'} \Omega^{\mathbf{l}}_{\mathbf{n}\mathbf{n}'ss'}$$

$$A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha = 1, \alpha' = 2\right) = \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}} \left(\frac{2\pi}{a} N l_2 l'_1\right) \Gamma_{\mathbf{l}} \Gamma_{\mathbf{l}'} \Omega^{\mathbf{l}}_{\mathbf{n}\mathbf{n}'ss'}$$

$$
\begin{aligned}
A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha=1,\alpha'=3\right) &= \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}}\left(\frac{2\pi}{a}l_2\right)\Gamma_{\mathbf{l}}\frac{\partial\Gamma_{\mathbf{l}'}}{\partial\rho}\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'}\\
A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha=2,\alpha'=1\right) &= \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}}\left(\frac{2\pi}{a}Nl_1l_2'\right)\Gamma_{\mathbf{l}}\Gamma_{\mathbf{l}'}\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'}\\
A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha=2,\alpha'=2\right) &= \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}}\left(N^2l_2l_2'\right)\Gamma_{\mathbf{l}}\Gamma_{\mathbf{l}'}\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'}\\
A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha=2,\alpha'=3\right) &= \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}}\left(Nl_2\right)\Gamma_{\mathbf{l}}\frac{\partial\Gamma_{\mathbf{l}'}}{\partial\rho}\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'}\\
A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha=3,\alpha'=1\right) &= \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}}\left(\frac{2\pi}{a}l_2'\right)\frac{\partial\Gamma_{\mathbf{l}}}{\partial\rho}\Gamma_{\mathbf{l}'}\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'}\\
A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha=3,\alpha'=2\right) &= \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}}\left(Nl_2'\right)\frac{\partial\Gamma_{\mathbf{l}}}{\partial\rho}\Gamma_{\mathbf{l}'}\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'}\\
A^{ss'}_{\mathbf{n},\mathbf{n}'}\left(\alpha=3,\alpha'=3\right) &= \sum_{\mathbf{l},\mathbf{l}'|\mathbf{C}}\frac{\partial\Gamma_{\mathbf{l}}}{\partial\rho}\frac{\partial\Gamma_{\mathbf{l}'}}{\partial\rho}\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'}
\end{aligned}
\tag{A1}
$$

where we used

$$
\Omega^{\mathbf{l}}_{\mathbf{nn}'ss'} = e^{j\mathbf{g}_{\mathbf{l}}\cdot\mathbf{t}_{\mathbf{ns}}}e^{j\mathbf{g}_{\mathbf{l}}\cdot\mathbf{t}_{\mathbf{n}'\mathbf{s}'}} = e^{j\mathbf{g}_{\mathbf{l}}\cdot\left(\mathbf{t}_{\mathbf{ns}}-\mathbf{t}_{\mathbf{n}'\mathbf{s}'}\right)}. \tag{A2}
$$

APPENDIX B. DERIVATION OF THE EQUATION OF MOTION UNDER EXTERNAL FIELDS

First, we convert the Lagrangian into an explicit function of the canonical variables u and $\dot{u}$ by substituting (29) and (30) into (32) to get

$$
\begin{aligned}
L \;=\; & \frac{1}{2}\sum_{\mathbf{n}}\sum_{s}\sum_{\alpha}M\left(\dot{u}_{\mathbf{n}s\alpha}\right)^2 - \frac{1}{2}\sum_{\mathbf{n}}\sum_{s}\mathbf{u}_{\mathbf{n}'s'}A^{ss'}_{\mathbf{n},\mathbf{n}'}\mathbf{u}_{\mathbf{n}s}\\
& + \int \mathrm{d}^3r\sum_{\alpha}\sum_{\mathbf{n}}\sum_{s}\sum_{\beta}\Gamma^{\mathbf{n}s}_{\alpha\beta}\left(\mathbf{r}\right)A_{\beta}\left(\mathbf{r},t\right)\dot{u}_{\mathbf{n}s\beta}\\
& - \int \mathrm{d}^3r\sum_{\mathbf{n}}\sum_{s}\sum_{\alpha}\Upsilon_{\mathbf{n}s\alpha}\left(\mathbf{r}\right)\phi\left(\mathbf{r},t\right)u_{\mathbf{n}s\alpha}.
\end{aligned}
\tag{B1}
$$

We start by calculating the individual terms in the Euler-Lagrange

Equation (35). Simple manipulations lead to

$$\begin{aligned}\frac{\partial L}{\partial \dot{u}_{\mathbf{n}s\alpha}} &= M\dot{u}_{\mathbf{n}s\alpha} + \frac{\partial}{\partial \dot{u}_{\mathbf{n}s\alpha}} \int \mathrm{d}^3 r \sum_{\beta} \sum_{\mathbf{n}'} \sum_{s'} \sum_{\beta'} \Gamma_{\beta\beta'}^{\mathbf{n}'s'}(\mathbf{r})\, \delta J_{\beta} \dot{u}_{\mathbf{n}'s'\beta'} \\ &= M\dot{u}_{\mathbf{n}s\alpha} + \int \mathrm{d}^3 r \sum_{\beta} \Gamma_{\beta\alpha}^{\mathbf{n}s}(\mathbf{r})\, A_{\beta}(\mathbf{r},t), \end{aligned} \tag{B2}$$

and

$$\begin{aligned}\frac{\partial L}{\partial u_{\mathbf{n}s\alpha}} &= -\sum_{\mathbf{n}'} \sum_{s'} \sum_{\alpha'} A_{\mathbf{n},\mathbf{n}'}^{ss'\alpha\alpha'} u_{\mathbf{n}'s'\alpha'} \\ &\quad -\frac{\partial}{\partial u_{\mathbf{n}s\alpha}} \int \mathrm{d}^3 r \sum_{\mathbf{n}'} \sum_{s'} \sum_{\alpha'} \Upsilon_{\mathbf{n}'s'\alpha'} \phi u_{\mathbf{n}'s'\alpha'} \\ &= -\sum_{\mathbf{n}'} \sum_{s'} \sum_{\alpha'} A_{\mathbf{n},\mathbf{n}'}^{ss'\alpha\alpha'} u_{\mathbf{n}'s'\alpha'} - \int \mathrm{d}^3 r\, \Upsilon_{\mathbf{n}s\alpha} \phi(\mathbf{r},t). \end{aligned} \tag{B3}$$

Substituting (B2) and (B3) into (35), we find

$$\begin{aligned} & M\ddot{u}_{\mathbf{n}s\alpha} + \sum_{\mathbf{n}'} \sum_{s'} \sum_{\alpha'} A_{\mathbf{n},\mathbf{n}'}^{ss'\alpha\alpha'} u_{\mathbf{n}'s'\alpha'} \\ & + \int \mathrm{d}^3 r \left\{ \sum_{\beta} \Gamma_{\beta\alpha}^{\mathbf{n}s}(\mathbf{r}) \frac{\partial A_{\beta}}{\partial t} + \Upsilon_{\mathbf{n}s\alpha} \phi(\mathbf{r},t) \right\} = 0. \end{aligned} \tag{B4}$$

We need to use the equation of continuity

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{J} = 0. \tag{B5}$$

By plugging (29) and (30) into (B5), it is possible to write

$$\begin{aligned} & \sum_{\mathbf{n}} \sum_{s} \sum_{\beta} \Upsilon_{\mathbf{n}s\beta}(\mathbf{r})\, \dot{u}_{\mathbf{n}s\beta} \\ &= -\nabla \cdot \sum_{\mathbf{n}} \sum_{s} \sum_{\beta} \Gamma_{\alpha\beta}^{\mathbf{n}s}(\mathbf{r})\, \dot{u}_{\mathbf{n}s\beta} \\ &= -\sum_{\alpha} \frac{\partial}{\partial x_{\alpha}} \sum_{\mathbf{n}} \sum_{s} \sum_{\beta} \Gamma_{\alpha\beta}^{\mathbf{n}s}(\mathbf{r})\, \dot{u}_{\mathbf{n}s\beta} \\ &= -\sum_{\mathbf{n}} \sum_{s} \sum_{\beta} \left[\sum_{\alpha} \frac{\partial}{\partial x_{\alpha}} \Gamma_{\alpha\beta}^{\mathbf{n}s}(\mathbf{r}) \right] \dot{u}_{\mathbf{n}s\beta}. \end{aligned} \tag{B6}$$

Therefore, by equating like terms we obtain

$$\Upsilon_{\mathbf{n}s\beta}\left(\mathbf{r}\right)=-\sum_{\alpha}\frac{\partial}{\partial x_{\alpha}}\Gamma_{\alpha\beta}^{\mathbf{n}s}\left(\mathbf{r}\right)=-\nabla\cdot\Gamma_{\beta}^{\mathbf{n}s}, \tag{B7}$$

which transforms (B4) to

$$\begin{aligned}&M\ddot{u}_{\mathbf{n}s\alpha}+\sum_{\mathbf{n}'}\sum_{s'}\sum_{\alpha'}A_{\mathbf{n},\mathbf{n}'}^{ss'\alpha\alpha'}u_{\mathbf{n}'s'\alpha'}\\&+\int\mathrm{d}^{3}r\left\{\Gamma_{\alpha}^{\mathbf{n}s}\left(\mathbf{r}\right)\cdot\frac{\partial\mathbf{A}\left(\mathbf{r},t\right)}{\partial t}-\nabla\cdot\Gamma_{\alpha}^{\mathbf{n}s}\left(\mathbf{r}\right)\phi\left(\mathbf{r},t\right)\right\}=0.\end{aligned} \tag{B8}$$

Using the vector identity $\psi\nabla\cdot\mathbf{F}=\nabla\cdot\left(\psi\mathbf{F}\right)-\mathbf{F}\cdot\nabla\psi$, the last term in (B8) can be written as

$$\begin{aligned}\int\mathrm{d}^{3}r\,\nabla\cdot\Gamma_{\alpha}^{\mathbf{n}s}\left(\mathbf{r}\right)\phi\left(\mathbf{r},t\right)&=\int\mathrm{d}^{3}r\,\nabla\cdot\left(\phi\Gamma_{\alpha}^{\mathbf{n}s}\right)-\int\mathrm{d}^{3}r\,\Gamma_{\alpha}^{\mathbf{n}s}\cdot\nabla\phi\\&=\oint_{S}d\mathbf{s}\cdot\phi\Gamma_{\alpha}^{\mathbf{n}s}-\int\mathrm{d}^{3}r\,\Gamma_{\alpha}^{\mathbf{n}s}\cdot\nabla\phi,\end{aligned} \tag{B9}$$

where the divergence theorem was utilized in the last step. By letting the surface S goes to infinity, where the fields are asymptotically zero [8], the surface integral in (B9) will vanish and what left is

$$\int\mathrm{d}^{3}r\,\nabla\cdot\Gamma_{\alpha}^{\mathbf{n}s}\left(\mathbf{r}\right)\phi\left(\mathbf{r},t\right)=-\int\mathrm{d}^{3}r\,\Gamma_{\alpha}^{\mathbf{n}s}\cdot\nabla\phi. \tag{B10}$$

Substituting (B10) to (B8) and using $\mathbf{E}=-\partial\mathbf{A}/\partial t-\nabla\phi$, we finally arrive to (36).

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