Circuit elements at optical frequencies from first principles: A synthesis of electronic structure and circuit theories

R. Ramprasad[a] and C. Tang
Department of Chemical, Materials and Biomolecular Engineering, Institute of Materials Science, University of Connecticut, 97 N. Eagleville Road, Storrs, Connecticut 06269

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A first principles electronic structure based method is presented to determine the equivalent circuit representations of nanostructured physical systems at optical frequencies, via a mapping of the effective permittivity calculated for a lattice of physical nano-elements using density functional theory to that calculated for a lattice of impedances using circuit theory. Specifically, it is shown that silicon nanowires and carbon nanotubes can be represented as series combinations of inductance, capacitance and resistance. It is anticipated that the generality of this approach will allow for an alternate description of physical systems at optical frequencies, and in the realization of novel opto- and nanoelectronic devices, including negative refractive index materials. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2226985]

I. INTRODUCTION

Circuit elements, such as capacitors, inductors and resistors, form the building blocks of a large class of electronic devices. A description of electromagnetic phenomena based on such building blocks is applicable when the electromagnetic wavelength is much larger than the typical dimensions of the building blocks. At optical and higher frequencies, however, this circuit based approach is expected to become inadequate. Metals and dielectrics are far from ideal at these high frequencies, displaying plasmonic resonances and optical absorption, respectively, preventing a clear unified description, or definition, of circuit elements at such high frequencies.

Nevertheless, a large variety of applications in the areas of plasmonics and nano-optics can benefit from a circuit based description of physical structures in the optical regime. An example would be the class of composite materials called metamaterials,[1–3] which include systems with negative effective permittivity,[4,6] negative effective permeability,[5,6] negative refractive index (NRI),[3,6,7] etc.

In recent years, circuit theory based approaches have been widely used in the creation of NRI metamaterials at GHz frequencies. A popular approach to the realization of such artificial composites has been to utilize a lattice of inductive elements—e.g., metal rods—resulting in negative effective permittivities below a cutoff frequency,[4,6] and a lattice of parallel inductor-capacitor resonators—e.g., "C" shaped metal split rings—that display negative effective permeabilities close to the resonance frequency.[5,6] Availability of a circuit based description of physical structures at optical and higher frequencies will allow for a continued application of these well established notions to the realization of optical metamaterials.

It is well known that the semiclassical formula for the dielectric function of a physical system resembles strongly the formula of the dielectric function of a collection of classical, charged, harmonic oscillators.[8] It is consequently tempting to extend this analogy, and attempt to map the physical system to a collection of circuit elements.

Such efforts aimed at the realization and representation of optical metamaterials in terms of circuit elements have met with recent successes. For instance, in a recent pioneering article, Engheta, Salandrino, and Alu[9] have shown that dielectric and metallic nanoparticles can indeed be represented in terms of circuit elements at optical frequencies. The key idea surrounding this development is the possibility of exploiting the plasmonic (nonplasmonic) behavior of certain metal (dielectric) particles in the optical regime. This allows for the definition of an equivalent particle impedance, which in general can display a resonance (much like in an *L-C* circuit).[10–12] Based on these notions, Engheta and co-workers have also proposed nanoparticle based topologies for the fabrication of optical metamaterials.[9,13] Also, Shalaev *et al.*[14] have recently successfully fabricated metamaterials in the near-infrared regime based on a periodic arrangement of pairs of parallel nanowires, with the nanowires and the gaps between them providing the necessary "optical" inductance and capacitance, respectively. In both cases, experimental complex permittivity values of the nanoparticles[9] or the bulk materials[14] at optical frequencies were used in the design/description of the structures.

Here, in the spirit of the prior work by Engheta and co-workers,[9,13] we present a paradigm to determine the circuit representations and values of a wide class of nanoscale systems (nanowires, nanotubes, nanoparticles, etc.) at optical frequencies using electronic structure theory. The novelty of the present development compared to the previous efforts is the use of electronic structure theory (in combination with circuit theory), which, in principle, can provide accurate information from first principles concerning the plasmonic and nonplasmonic behavior of a wider class of systems than is

[a]Author to whom correspondence should be addressed; electronic mail: rampi@ims.uconn.edu

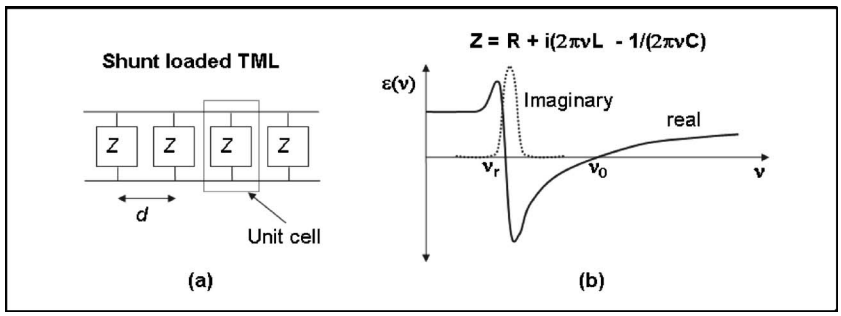

FIG. 1. (a) Periodic array of impedances shunting a transmission line (TML). A one-dimensional lattice is shown for ease of visualization; (b) the vertical component (directed from one TML plate to the other) of the frequency dependent effective permittivity [Eq. (3)] when the shunt element is a series combination of an inductor, capacitor and a resistor.

available experimentally to date. Central to this approach is our ability to calculate the frequency dependent effective permittivity of two constructs:

1. A periodic arrangement of impedances, using a classical electromagnetic theory such as transmission line (TML) theory, and
2. An ordered composite system consisting of physical nano-elements, using an electronic structure theory such as density functional theory (DFT).

In both the above cases, we are interested in the regime where the electromagnetic wavelength is much larger than the lattice parameter of the ordered medium, henceforth referred to as an effective medium, with homogeneous properties. Thus, in principle, the equivalent circuit elements at optical frequencies representing the physical nano-elements in an ordered composite can be determined by matching the effective permittivity results of DFT to TML theory. Once the nature and value of the circuit elements are determined, these circuit elements (or the actual physical nano-elements) can be "networked" such that the desired electromagnetic response results. This forms the central theme of this work.

This article is organized as follows. In Sec. II, we outline the procedure for obtaining the effective properties of a composite represented as a periodic arrangement of impedances. The electronic structure route to the determination of the effective permittivity of a periodic arrangement of physical nano-elements is then described in Sec. III. Application of these principles, and the procedure for extracting equivalent circuit representations of physical nano-elements for a few example systems, are then presented in Sec. IV. Section V contains our main conclusions.

II. EFFECTIVE PERMITTIVITY FROM CIRCUIT THEORY

Electromagnetic wave propagation through a homogeneous effective medium is analogous to that along a transmission line (TML), with suitably defined inductance per unit length and capacitance per unit length of the TML. Here, we use this analogy to obtain the frequency dependent effective permittivity of an effective medium composed of a periodic arrangement of impedances.

Let us consider a parallel plate TML periodically shunt loaded with impedances per unit length, Z, in a square lattice of side length d, as shown in Fig. 1(a). The voltage and current at one side (say, left) of the periodic unit cell, V_L and I_L, can be related to those at the opposite side (say, right), V_R and I_R, as[15]

$$\begin{pmatrix} V_L \\ I_L \end{pmatrix} = T \cdot \begin{pmatrix} V_R \\ I_R \end{pmatrix} \tag{1}$$

with the transfer matrix T of the effective medium given by

$$T = T_{\mathrm{TML},d/2} \cdot T_{\mathrm{shunt}} \cdot T_{\mathrm{TML},d/2}, \tag{2}$$

where $T_{\mathrm{TML},d/2}$ and T_{shunt} are the transfer matrices for a TML of length $d/2$ and the shunt Z element, respectively, given by standard expressions that can be found elsewhere.[15] All elements of $T_{\mathrm{TML},d/2}$, T_{shunt} and T can be written in terms of Z, d, the frequency ν, the free space permittivity, ϵ_0, the free space permeability, μ_0, and the effective permittivity, ϵ, and the effective permeability, μ, of the composite (or effective) medium. Under the assumption that $d << \lambda$ the electromagnetic wavelength ($=c/\nu$, with $c=1/\sqrt{\mu_0\epsilon_0}$ being the speed of light in vacuum), it can be shown by equating corresponding elements of the above matrix equation that the frequency dependent effective permittivity directed from one plate to the other (i.e., the vertical direction) is given by

$$\epsilon = \epsilon_0\left(1 - \frac{i}{2\pi\nu\epsilon_0 Z d^2}\right), \tag{3}$$

where $i=\sqrt{-1}$. For the special case of a square lattice of metal posts shunting the TML, $Z=i2\pi\nu L$, where L is the inductance per unit length of the metal posts (assuming that the metals are perfect with infinite conductivity), Eq. (3) reduces to the well known plasmonic equation, $\epsilon=\epsilon_0(1-\nu_0^2/\nu^2)$, suggested earlier,[4] with the cutoff frequency $\nu_0 = \sqrt{1/(4\pi^2\epsilon_0 L d^2)}$ below which ϵ is negative. When Z is composed of a series combination of an inductance and capacitance, i.e., when $Z=i[2\pi\nu L - 1/(2\pi\nu C)]$, where C has the units of capacitance times length, ϵ has the form schematically shown in Fig. 1(b). At high frequencies, the inductance L dominates, and determines ν_0; at low frequencies, the capacitance C dominates, and determines the constant positive real value of ϵ. At the resonance frequency ν_r ($=1/[2\pi\sqrt{LC}]$), the impedance changes from capacitive to inductive behavior. Inclusion of a resistance per unit length, R, in Z manifests as the imaginary part of ϵ. Equation (3) thus provides a prescription for determining the effective permittivity of a periodic arrangement of impedances. Although a simple series combination of R, L and C is presented here, this treatment allows for any combination of these basic building blocks.

III. EFFECTIVE PERMITTIVITY FROM ELECTRONIC STRUCTURE THEORY

We now move on from the classical description discussed above to a quantum mechanical one. Electronic structure methods are available to calculate the electronic part of the frequency dependent dielectric function of condensed matter systems. Since we are interested mainly in the optical regime, treatment of the electronic part alone is sufficient. For definiteness, we use a method based on density functional theory (DFT)[16] and first order perturbation theory.[16]

As this study focuses on periodic arrays of nano-elements embedded in a host medium, supercell techniques naturally lend themselves to this study. The electronic wave functions and eigenenergies obtained from supercell DFT calculations of periodic arrays of nano-elements were used to determine the imaginary part of the frequency dependent composite dielectric function, $\epsilon_{\text{imag}}(\nu)$, using the relation[17]

$$\epsilon_{\text{imag}}(\nu) = \frac{e^2 h}{2\pi m^2 \nu^2} \sum_{ij} \frac{2}{(2\pi)^3} \int_{BZ} d\mathbf{k} |M_{ij}(\mathbf{k})|^2 \delta[\nu - \nu_{ij}(\mathbf{k})], \tag{4}$$

where e, m and h are the electronic charge, electronic mass and Planck's constant, respectively, and i and j are indices that label the wave functions, ψ, and eigenenergies, E. M_{ij} is a matrix element given by $<\psi_i(\mathbf{k})|\mathbf{e}\cdot\mathbf{p}|\psi_j(\mathbf{k})>$, with $\mathbf{e}$, $\mathbf{p}$ and $\mathbf{k}$ being the polarization vector, momentum operator and a point in reciprocal space, respectively. The integration in the above equation is performed over the first Brillouin zone (BZ). The real part of the dielectric function is obtained from the imaginary part by performing a Kramers-Kronig transformation.[16]

IV. RESULTS

Calculations have been performed for three systems using the above methods using the plane-wave DFT code VASP[18] at the local density approximation level of theory: (1) bulk Si, (2) an infinite array of parallel Si nanowires, and (3) an infinite array of parallel (4,2) carbon nanotubes. These systems were chosen as their frequency dependent permittivity is reminiscent of the behavior shown in Fig. 1(b). For each case, the values of L, C, and R that result in a good fit to Eq. (3) were determined.[19]

A. Bulk Si

The equilibrium lattice constant of single crystal bulk Si as calculated here using VASP was 5.42 Å, identical to the experimental value. The optical properties were calculated using the wave function and eigenenergy results of VASP using the method described above [Eq. (4)]. Figure 2 (solid circles) shows the real and imaginary parts of the isotropic frequency (or energy) dependent electronic dielectric function.

As can be seen, the bulk dielectric constant of about 11 was recovered at low frequencies. A characteristic resonance and absorption can also be seen at about 3.7 eV, in agreement with prior work.[20] The most interesting feature to be noted is that the real and imaginary parts of the dielectric function display the behavior shown in Fig. 1(b), reminiscent of a periodic array of series L, C and R. In fact, the DFT results can be fitted remarkably well to Eq. (3), with $d = 5.42$ Å, $L = 1.2\times10^{-3}$ H/m, $C = 2.6\times10^{-29}$ F m, and $R = 1.1\times10^{12}$ Ω/m; the fit is shown as solid lines in Fig. 2. Note that the impedance comprising the L-R-C circuit is a per unit length quantity (say along the z direction) per unit cell with a cross sectional area of d^2 (say, along the x-y plane). The extracted circuit parameter values are reasonable. For instance, C corresponds to a dielectric constant of 11, and R corresponds to a resistivity of 3.2×10^{-7} Ω m ($=R\cdot d^2$) at optical frequencies. It thus appears that bulk Si can be represented at optical frequencies as a network of circuit components, with the energies corresponding to the characteristic frequencies ν_r and ν_0 at about 3.7 and 10 eV, respectively. Other dielectrics display this characteristic behavior too,[21] and could be represented as a network of L, C and R.

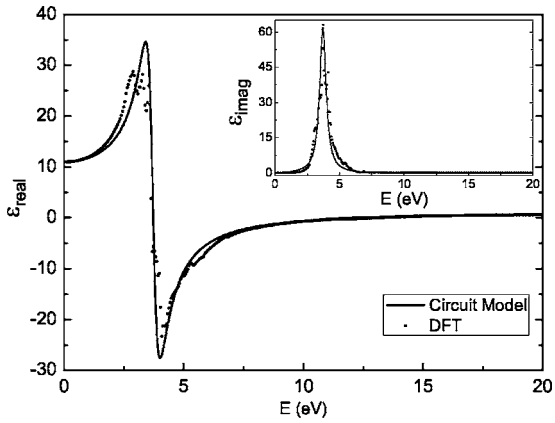

FIG. 2. Real and imaginary (inset) parts of the dielectric function of bulk crystalline silicon as calculated using DFT (solid circles) and the circuit model given by Eq. (3) (solid line). Note that $E=h\nu$.

B. Square lattice of Si nanowires

Next, we consider two square lattices of parallel (001) Si nanowires in vacuum. Each nanowire had an approximately square cross section with a side length of 8.9 Å. The surface Si atoms were passivated with H atoms to ensure tetrahedral coordination of all Si atoms. Figure 3 shows the real and imaginary parts of the effective permittivity for the lattice of nanowires with lattice constant 20 Å, as calculated within DFT along the axis of the Si nanowires, as well as the fit of the DFT results to the circuit model [Eq. (3)], with $d = 20$ Å, $L = 2.8\times10^{-4}$ H/m, $C = 8.5\times10^{-29}$ F m, and $R = 2.9\times10^{11}$ Ω/m. Again, although there is scatter in the DFT results, it appears that individual Si nanowires can be well represented as a series combination of L, C and R. Figure 4 shows the results for another array of the same nanowires, but arranged in a square lattice with lattice constant of 30 Å. The circuit model results were generated for the same values of L, C and R, but with $d = 30$ Å. As can be seen, once again the fit is reasonable, indicating that the L, C and R values are intrinsic properties of each nanowire, captured well within the circuit model. We also note that the energy corresponding

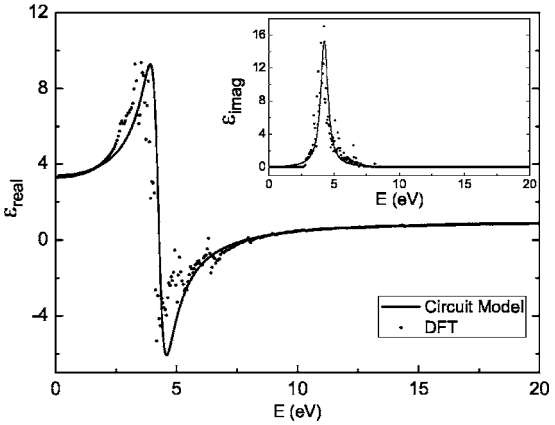

FIG. 3. Real and imaginary (inset) parts of the effective dielectric function of a periodic square lattice of (001) silicon nanowires in vacuum (lattice constant=20 Å) as calculated using DFT (solid circles) and the circuit model given by Eq. (3) (solid lines); the dielectric function is calculated along a direction parallel to the axes of the nanowires. Note that $E=h\nu$.

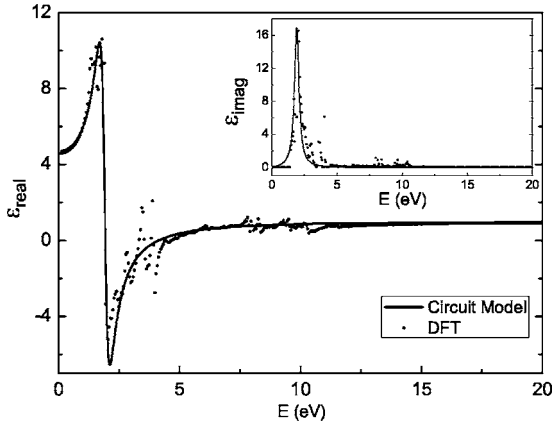

FIG. 5. Real and imaginary (inset) parts of the effective dielectric function of a square lattice of parallel (4,2) carbon nanotubes in vacuum as calculated using DFT (solid circles) and the circuit model given by Eq. (3) (solid line); the dielectric function is calculated along a direction parallel to the axes of the CNTs. Note that $E=h\nu$.

to the characteristic frequencies ν_r and ν_0 have shifted from the corresponding bulk values, with ν_0 showing the maximum deviation.

C. Square lattice of carbon nanotubes

Similarly, it appears that individual carbon nanotubes (CNTs) can be represented as a series combination of L, C and R. A DFT study of a (4,2) CNT was performed, and the equilibrium C–C bond was determined using VASP to be 1.45 Å, slightly larger than the corresponding experimental value for graphite, and in good agreement with prior DFT studies.[22] Figure 5 shows the calculated frequency (or energy) dependent effective dielectric function for a square lattice (side length=12 Å) of parallel (4,2) CNTs in vacuum, along the axis of the CNTs. Again, it is clear that this system too displays a behavior similar to that by a periodic array of series L, C and R. As before, a fit of the DFT results to Eq. (3) was performed, with $d=12$ Å, resulting in $L=2.5\times10^{-3}$ H/m, $C=4.7\times10^{-29}$ F m, and $R=4\times10^{11}$ Ω/m. The dielectric constant of an individual tube along its axis can be determined from the extracted capacitance value as $C/(\epsilon_0\pi r^2)=37$, where r(=2.1 Å) is the radius of the (4,2) CNT. This procedure also results, for the first time, in an estimate of the inductance of an individual CNT.

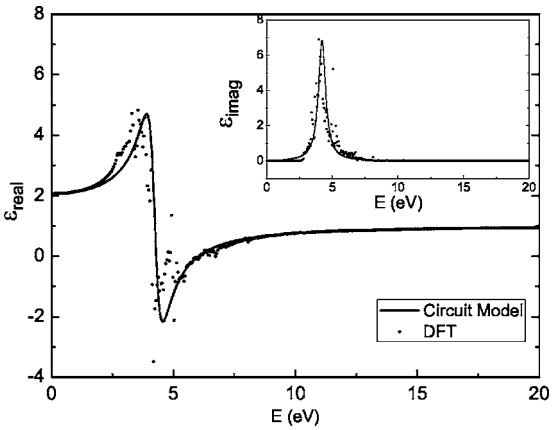

FIG. 4. Real and imaginary (inset) parts of the effective dielectric function of a periodic square lattice of (001) silicon nanowires in vacuum (lattice constant=30 Å) as calculated using DFT (solid circles) and the circuit model given by Eq. (3) (solid lines); the dielectric function is calculated along a direction parallel to the axes of the nanowires. Note that $E=h\nu$.

V. CONCLUSIONS

The above set of results indicates that it is possible to represent the optical behavior of bulk semiconductors and nanoscale systems in terms of circuit elements at optical and higher frequencies, paving the way for an alternate description of ordered physical systems in this regime. Silicon nanowires and a carbon nanotube were used to show that the dielectric function calculated using electronic structure maps to that given by circuit theory. In particular, this has the following implications for the realization of nanoscale metamaterials at optical frequencies: (1) arrays of nanoelements with appropriate circuit representations could display negative effective permittivity at a certain range of frequencies, and (2) nanoloops formed out of such nanoelements will behave as parallel L-C resonators, an array of which will display negative effective permeability close to the L-C resonance frequency. It should be noted that for the purpose of this discussion, only those systems that display an effective permittivity reminiscent of a periodic array of series L-C-R [as shown in Fig. 1(b)] were considered. Systems that display other features at optical frequencies can similarly be represented by more complicated combinations of L, C and R.

ACKNOWLEDGMENTS

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