Effective Boundary Conditions in Electrodynamics of Nanostructures

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Abstract

The effective boundary condition method is extended to nano-scale mesoscopic systems. The EBCs appear as a result of the 2D-homogenezation procedure and have the form of two-side anisotropic impedance boundary conditions stated on the structure's surface. The surface impedance tensor has been evaluated for a set of typical nanostructures. It has been shown that, unlike to macroscopic electrodynamics, the surface impedance tensor exhibits sharp oscillations at frequencies of optical transitions. The EBC method supplemented with well-developed mathematical techniques of classical electrodynamics creates unified basis for solution of boundary-value problems in electrodynamics of nanostructures. A generalization of the EBC method to the quantum electrodynamics is also presented,

1. Introduction

Rapid progress in the synthesis of a variety of different kinds of spatially confined nanostructures with fascinating electronic and optical properties irreducible to properties of bulk media symbolizes a fundamental breakthrough in solid state physics. The key peculiarities of such structures are related to spatial confinement of the charge carrier motion and their nanoscale spatial inhomogeneity. Since the inhomogeneity scale is much less than the optical wavelength, in many cases it turns out to be possible under analysis to reduce the dimensionality of structure (low–dimensional structure). In this contribution we present a method of evaluation of the electromagnetic response of low–dimensional nanostructures formed by thin layers with intrinsic 2D periodicity with characteristic period much less than the optical wavelength, e.g., carbon nanotubes (CNs) [1], planar arrays of quantum dot (QDs) [2].

This method, conventionally referred to as the effective boundary condition (EBC) method, has been originally developed for microwaves and antenna theory [3]–[5], and has found a wide application in these fields, e. g., for the design of semi–transparent grid screens and helical sheaths in traveling wave tubes[1]. In essence, the EBC method is modification of the effective medium theory as applied to 2D–confined structures. The basic idea of the EBC method is that a smooth homogeneous surface is considered instead a periodic structure, and appropriate EBCs for the electromagnetic field are stated for this surface. These conditions are chosen in such a way that the spatial structures of the electromagnetic field due to an effective current induced on the homogeneous surface, and the electromagnetic field of the real current in the lattice turn out to be identical some distance away from the surface. The lattice parameters are included in coefficients of the EBCs. The applicability of the EBCs is restricted by the requirement that the lattice period be small compared with the free–space wavelength. The effectiveness of the

[1]Similar approaches have been developed in acoustics, hydrodynamics, elasticity theory.

EBC method is determined by a possibility its extension to more complicated situations. Such an extension is only possible when the parameters involved in the EBCs do not depend on the spatial structure of the irradiating field, or, in another words, the EBCs must be *local*, i.e., they must couple field components and their spatial derivatives at a given point of space. In the simplest case, the EBCs have the form of two–side impedance boundary conditions on the surface S:

$$[\mathbf{n}\,[\mathbf{n},\mathbf{H}^{\mathrm{I}}-\mathbf{H}^{\mathrm{II}}]] = -\frac{4\pi\widehat{\boldsymbol{\sigma}}}{c}[\mathbf{n},\mathbf{E}]\,, \tag{1}$$

$$[\mathbf{n},\mathbf{E}^{\mathrm{I}}-\mathbf{E}^{\mathrm{II}}] = 0 \tag{2}$$

where c is the speed of light. The unit vector $\mathbf{n}$ is normal to the surface S and is directed from region I to region II. The effective conductivity tensor $\widehat{\boldsymbol{\sigma}}$ contains information about geometrical configuration and constitutive parameters of the lattice. EBCs in the form given by Eqs. (1)–(2) are obtained neglecting (i) polarization of the structure in the $\mathbf{n}$ direction, and (ii) contribution of spatial dispersion into conductivity.

2. Formulation of the EBCs for Planar Nanostructures

In order to derive the EBCs, a kernel problem must be solved in each particular case. For example, this problem is formulated for grid screens as the problem of plane wave scattering by the infinite plane screen [4]. Below we consider the kernel problems for two particular cases of low–dimensional nanostructures.

A) Carbon nanotubes.
As applied to CNs, the kernel problem is to derive the EBCs for an isolated infinitely long regular CN with an arbitrary index (m,n), i. e., to derive the EBCs for a cylindrical surface of the radius R with R as the CN radius. Neglecting indirect interband transitions in the π–electrons' motion, the conductivity tensor of the CN is given by [6]

$$\widehat{\boldsymbol{\sigma}} = \begin{pmatrix} 0 & 0 \\ 0 & \sigma_{zz} \end{pmatrix}, \tag{3}$$

where σ_{zz} is the axial conductivity of CN; this quantity appears in coupling of the microscopic properties of the CN and its macroscopic electromagnetic response. The treatment of the axial dynamical conductivity of an isolated CN has been given in Refs. [7], [8]. Both semi–classical and quantum–mechanical analyses of the conductivity have been presented in the above references. In some cases, the role of spatial dispersion for CNs turns out to be essential. In that cases, Eq. (2) keeps validity while Eq. (1) is transformed to

$$\widehat{\boldsymbol{\Lambda}}[\mathbf{n}\,[\mathbf{n},\mathbf{H}^{\mathrm{I}}-\mathbf{H}^{\mathrm{II}}]] = -\frac{4\pi\widehat{\boldsymbol{\sigma}}}{c}[\mathbf{n},\mathbf{E}]\,, \tag{4}$$

where

$$\widehat{\boldsymbol{\Lambda}} = \begin{pmatrix} 1 & 0 \\ 0 & 1+\dfrac{\tilde{l}_0}{k^2(1+i\nu/\omega)^2}\dfrac{\partial^2}{\partial z^2} \end{pmatrix}, \tag{5}$$

$\tilde{l}_0$ is the spatial dispersion parameter and ν is the relaxation frequency. In a general case, the quantities σ_{zz} and $\tilde{l}_0$ are evaluated using the quantum–mechanical transport theory. Though the CN surface possesses a periodic crystalline structure, Eqs. (4)-(5) incorporate only constant coefficients (i.e., σ_{zz} and $\tilde{l}_0$), and are devoid of any periodic functions. This is because the technique of deriving the EBCs is equivalent to the averaging of microscopic fields over an infinitesimally small volume.

The conductivity was assumed to be quasi–one–dimensional while $\sigma_{z\phi}$, $\sigma_{\phi z}$ and $\sigma_{\phi\phi}$ were ignored in the derivation of the EBCs. As a consequence, we obtained conditions (2), (4) and (5) for the electromagnetic field across the CN surface. The polar conductivity $\sigma_{\phi\phi}$ is due to indirect interband transitions neglected in our model and will surely modify the stated EBCs. The role of $\sigma_{z\phi}$ and $\sigma_{\phi z}$ is expected to be important, for example, in chiral CNs in relation to the effect of natural optical activity exhibited by such CNs.

B) 2D–lattice of QDs.

In QDs, apart from the charge carrier confinement [2], there exist a class of electrodynamic effects related to light diffraction by QDs and QD ensembles, which strongly influence the electromagnetic response properties of such systems [9] –[11]. Here we consider 2D arrays of QDs to establish correlation between properties of such systems and homogeneous 2D structures like quantum wells. The key problem here is the diffraction by infinite planar quadratic lattice constituted by identical QDs imbedded in a host medium. The host medium is assumed to be dispersionless and transparent. Conventional phenomenological model of the dispersion and the gain of a single QD is as follows: $\varepsilon(\omega) = \varepsilon_h + g_0/(\omega - \omega_0 + i/\tau)$. Here ε_h is the host medium permittivity while the quantity g_0 is the phenomenological parameter proportional to the oscillator strength of the transition; $g_0 > 0$ in an inverted medium.

Let the normal $\mathbf{n}$ be directed along the z-axis and let the incident planewave be polarized along the x-axis. Further we restrict ourselves to the dipole approximation of the diffraction theory. In that case, the scattering field from an isolated QD can be expressed in terms of Hertz potentials by

$$\mathbf{E} = \sum_{l,m=-\infty}^{\infty} \left(\nabla\nabla\cdot + k_1^2\right) \mathbf{\Pi}^e_{lm} \,, \tag{6}$$

$$\mathbf{H} = -ik\varepsilon_h \sum_{l,m=-\infty}^{\infty} \nabla\times\mathbf{\Pi}^e_{lm} \,, \tag{7}$$

where

$$\mathbf{\Pi}^e_{lm} = \mathbf{e}_x \alpha_{xx} \mathcal{E}_x(0) \exp\{ik_1\rho_{lm}\}/\rho_{lm} \,, \tag{8}$$

$k_1 = k\sqrt{\varepsilon_h}$, $\rho_{lm} = [(ld-x)^2 + (md-y)^2 + z^2]^{1/2}$, α_{xx} is the QD polarizability tensor component, d is the lattice period, and $\mathcal{E}_x(0)$ is the electric field inside QD. This field is related to the mean field in the layer, $E_x(0)$, by $E_x(0) = (1 + \delta_1\alpha_{xx}/d^2)\mathcal{E}_x(0)$, where δ_1 is the lattice parameter:

$$\delta_1 = \int_{-d/2}^{d/2}\int_{-d/2}^{d/2} \frac{2x^2 - y^2}{(x^2+y^2)^{5/2}}\, dxdy \approx -\frac{8}{\sqrt{2}d} \,. \tag{9}$$

After some manipulations with Eqs. (6), (7) (see, e.g., [12]), in the limit $z \to \pm 0$ we come to Eqs. (1), (2) with the x0y plane as the S surface and

$$\hat{\boldsymbol{\sigma}} = i\frac{4\pi\omega\varepsilon_h}{cd^2}\hat{\boldsymbol{\alpha}}\left(\hat{\mathbf{I}} + \frac{\delta_1}{d^2}\hat{\boldsymbol{\alpha}}\right)^{-1} \,. \tag{10}$$

Here $\hat{\mathbf{I}}$ is the unit tensor. Second term in the brackets is due to the depolarization related to the difference between mean and acting fields.

It should be noted that the assumption (i) which neglect polarization of the structure in the $\mathbf{n}$ direction, holds true only for QDs with planar configuration in x0y plane, e. g., discs, islands, flattened pyramids, etc. For QDs with comparable extensions in all directions, the derivation of EBCs presented above should be generalized. Derivation method remains the same: scattered fields are described by Eqs. (6), (7), but Hertz potentials are given by

$$\mathbf{\Pi}^{e}_{lm} = [\mathbf{e}_x \alpha_{xx} \mathcal{E}_x(0) + \mathbf{e}_z \alpha_{zz} \mathcal{E}_z(0)] \exp\{ik_1 \rho_{lm}\}/\rho_{lm}\,, \tag{11}$$

where α_{zz} stands for the QD polarizability in the z direction. Contribution of the transverse polarizability qualitatively modifies the EBCs: tangential components of the electric field exhibit discontinuity at the S surface.

In that case, relation between z–components of internal field of QD and mean field is given by $E_z^{\mathrm{I}}(0) + E_z^{\mathrm{II}}(0) = 2(1 + \alpha_{zz}\delta_2/d^2)\mathcal{E}(0)$, where

$$\delta_1 = \lim_{z\to 0} \int_{-d/2}^{d/2} \int_{-d/2}^{d/2} \frac{2z^2 - x^2 - y^2}{(x^2+y^2+z^2)^{5/2}}\, dxdy \approx \frac{2\pi\sqrt{\pi}}{d}\,. \tag{12}$$

Then, carrying out with Eqs. (6), (7) the manipulations analogous to the described above, we come to the generalized EBCs as follows:

$$[\mathbf{n}\,[\mathbf{n}, \mathbf{H}^{\mathrm{I}} - \mathbf{H}^{\mathrm{II}}]] = -\tfrac{2\pi\hat{\boldsymbol{\sigma}}}{c}[\mathbf{n}, \mathbf{E}^{\mathrm{I}} + \mathbf{E}^{\mathrm{II}}]\,, \tag{13}$$

$$[\mathbf{n}, \mathbf{E}^{\mathrm{I}} - \mathbf{E}^{\mathrm{II}}] = -\xi[\mathbf{n}, \nabla(\mathbf{n}, \mathbf{E}^{\mathrm{I}} + \mathbf{E}^{\mathrm{II}})] \tag{14}$$

with the conductivity tensor $\hat{\boldsymbol{\sigma}}$ defined by Eq. (10) and $\xi = 2\pi\alpha_{zz}/(d^2 + \delta_2\alpha_{zz})$.

The above equations constitute the complete system of EBCs for electromagnetic field in low-dimensional nanostructures. They have been obtained in the ordinary way, by the averaging of a microscopic field over a physically infinitesimal volume. The technique of macroscopic averaging is similar to one of introducing constitutive parameters for bulk media, but differing in that the averaging occurs in boundary conditions, but not in field equations. Correspondingly, the averaging was carried out over the 2–D surface (cylindrical for CNs or plane for QD sheets) but not over the 3–D spatial element. Thus, in electrodynamics of low-dimensional structures the EBCs play the same role as constitutive relations in electrodynamics of balk media.

3. EBCs in Quantum Electrodynamics of Nanostructures

As different from macroscopic microwave lattices, in nanostructures effects become valid related to quantum nature of the electromagnetic radiation (spontaneous irradiation, Kazimir forces, electromagnetic fluctuations, etc.). Obviously, quantum electrodynamics (QED) should be applied for description of such effects. In Ref. [13] a procedure of the electromagnetic field quantization in inhomogeneous Kramers–Kronig bulk dielectrics. In the framework of this approach, the electric field operator is introduced by

$$\hat{\mathbf{E}}(\mathbf{r}) = \int_0^\infty d\omega\, \underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) + H.c. \tag{15}$$

Analogous expression can be written for the magnetic field. The Maxwell equations with a source term corresponding to the dissipation–assisted quantum noise have been formulated in Ref. [13] for the operators $\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)$, $\underline{\hat{\mathbf{H}}}(\mathbf{r},\omega)$. Physically observable quantities are found by averaging of corresponding field operators. The above mentioned source term provides necessary commutative relations for these operators. Our analysis has shown that the basic ideas presented in Ref. [13] in combination with the EBC method can be extended to the case of spatially inhomogeneous low–dimensional structures. For simplicity we neglect both the medium polarization in the z–direction and the spatial dispersion. The conductivity tensor is assumed to be given in its eigenbasis. In such a case, the field operators $\hat{\underline{\mathbf{E}}}(\mathbf{r},\omega)$, $\hat{\underline{\mathbf{H}}}(\mathbf{r},\omega)$ satisfy to the vacuum Maxwell equations and the EBCs as follows

$$[\mathbf{n}\,[\mathbf{n},\hat{\underline{\mathbf{H}}}^{\mathrm{I}}-\hat{\underline{\mathbf{H}}}^{\mathrm{II}}]]=-\frac{4\pi\hat{\boldsymbol{\sigma}}}{c}[\mathbf{n},\hat{\underline{\mathbf{E}}}]-\frac{4\pi}{c}\hat{\underline{\mathbf{J}}}_N\,, \tag{16}$$

$$[\mathbf{n},\hat{\underline{\mathbf{E}}}^{\mathrm{I}}-\hat{\underline{\mathbf{E}}}^{\mathrm{II}}]=0 \tag{17}$$

where $\hat{\underline{\mathbf{J}}}_N$ is the operator of the surface noise current density which can be presented by

$$\hat{\underline{J}}_{Ni}(\mathbf{R},\omega)=\sqrt{\frac{\hbar\omega}{\pi}\Re\{\hat{\sigma}_{ii}\}}\left[\Theta(\Re\{\hat{\sigma}_{ii}\})\hat{f}_i(\mathbf{R},\omega)+\Theta(-\Re\{\hat{\sigma}_{ii}\})\hat{f}_i^{+}(\mathbf{R},\omega)\right]\,,\quad i=1,2\,. \tag{18}$$

Here $\mathbf{R}\in S$ and $\Theta(\cdot)$ stands for the unit step function; $\hat{f}_i(\mathbf{R},\omega)$ and $\hat{f}_i^{+}(\mathbf{R},\omega)$ are the annihilation and creation operators, respectively, of the 2D bosonic field. This field satisfies to the Heisenberg equations of motions and is analogous to the 3D bosonic field described in Ref. [13]. It can easily be found that the second term in brackets in right–hand part of Eq. (18) disappears for thermodynamically equilibrium systems.

The key feature of QED EBCs (16)–(17) which distinguishes them from the classical EBCs (1)–(2) is the presence of the surface noise current $\hat{\underline{\mathbf{J}}}_N$. This current makes it possible to satisfy the commutation relations for the field operators. Corresponding proofs will be given separately elsewhere. The fundamental difference between quantum electromagnetic field and the classical one is the presence in the quantum field of zero–point vacuum oscillations. Similar to classical electromagnetic field, zero–point vacuum oscillations will diffract by spatial inhomogeneities (nanostructure). The diffraction distorts the spatial structure of zero–point oscillations as compared with the virtual photons in vacuum. This diffraction process is described by the EBC operators (16)–(17).

4. Utilization of EBCs

Potentiality of utilization of the EBCs method under consideration of particular electrodynamical problems is provided by applying the solution of the kernel problem to a variety of much more complicated situations: curved and/or bounded screens, screens placed in the vicinity of dielectric or metallic surfaces, *etc.* For example, EBCs (4)–(5) has been derived for an isolated infinitely long CN. Nevertheless, our formalism can be utilized for consideration of diffraction problems in different types of nanotubes, *viz.*, CNs of finite length (first results in this field have been presented in Ref. [14]), bent and corrugated CNs, CNs with junctions, multi-shell CNs with hexagonal cross-section, CN–based composites, etc. The derived effective boundary conditions can also serve as the basis for description of interaction of CNs with beams of electrons and other charged particles. The investigation of guided surface wave propagation in single– and multi–shell CNs carried out in [7], [8] exemplifies the application of the formalism developed, and it is of significance in its own right too. Such waves can be excited by directing laser or electron beams along a CN axis. These surface waves are characterized by strong retardation and, consequently, have large field gradients in the transverse plane. As the result, such surface waves must manifest a strong pondermotive effect.

As applied to 2D lattices of QDs, the EBC method allows us to analyze electromagnetic response of such layers (or multilayer structures) placed in microcavity: this is of importance for the design of QD–based semiconductor lasers [2]. EBCs given by Eqs. (13), (14) state mathematical equivalence of 2D periodical layer of QDs and an isolated quantum well. It should be stressed that the mechanisms of transport processes and oscillator strengths in each case are essentially different. Nevertheless, the equivalence makes it possible to extend to QD–based planar structures the well-developed mathematical formalism of investigation of quantum wells. In particular, starting conditions for QD–based lasers can be evaluated by analogy with solution of the corresponding problem for the quantum well [15].

It should be emphasized that the extension of the EBC method to deformed or complicated structures is only possible when the modification of geometrical parameters of the structure does not change the electron transport properties in it; otherwise, modification of EBC is required. For example, too close location of two planar layers with QDs will change the energy spectrum because of overlapping of exciton wave functions, tunneling, etc. Analogously, too strong bend of CN will distort quasi–free motion of π–electrons in it and, consequently, may change the conductivity character. Thus, justification of applicability of EBCs must be given in each particular case.

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