Local Field Optical Effects in Two-Dimensional Quantum Confined Structures

A. V. Ghiner* and G. I. Surdutovich**

*Universidade Federal do Ceara, 60450-970, Fortaleza, CE, Brazil

**Instituto de Física de São Carlos, USP, 13560-970, São Carlos, SP, Brazil

Received July 27, 1995

By use of the generalized method of integral equations we take into account dimensions of quantum confined structures and calculate in the long-wavelength limit their optical characteristics. We show that the refractive index of such structures depends not only on a density, as in the case of Lorentz-Lorenz formula, but also on ratio of of the lattice's spatial period and the wavelength. These results are the base for an analytical description of the photonic band gap (PBG) in periodic two-dimensional semiconductor structures.

I. Introduction

The recent idea[1] that three-dimensional dielectric structures with appropriate periodicity should exhibit what is called a "photonic band gap" (PBG), by analogy with electronic band gaps in semiconductor crystals, has stimulated great activity in this field. PBG effect was first experimentally demonstrated[2] and so far has been observed only at microwave frequencies. The task to demonstrate an optical three-dimensional PBG is a considerably more difficult problem because of the submicron dimensions of photonic lattices and the nanometer-scale requisite accuracy of the fabrication. Therefore, it is of a special interest to consider the much more simple case of two-dimensional (2D) quantum-confined periodical structures when distances between dots (antidots) are comparable with their sizes. From an optical viewpoint such structures are, in fact, *rarefied media*, since in this situation the usual (for a dense medium) assumption about the great number of atoms inside the wavelength volume does not hold true any more.

Under interaction with an incident electromagnetic radiation quantum dots behave as optically interacting elementary radiators. The collective effects under propagation of radiation in such systems may be considered in the framework of the classical approach of molecular optics as a problem of the local field factors. In order to achieve a consistent description of the medium's discreteness we apply the method of integral equations (MIE)[3] of the molecular optics generalized for 2D systems[4] for finding the dielectric permittivity tensor of some quantum-confined structures. It is a first step to an analytical calculation of the two-dimensional PBGs.

Though MIE from the very outset assumes discrete structure of the medium but up to now such discreteness never has been allowed for numerically. The present work is concerned with this problem.

II. Method of integral equations as applied to a discrete medium

Further we associate a quantum dot with a point-like elementary radiator. The calculation of linear and nonlinear polarizabilities of such a radiator is a problem of quantum theory of a single quantum dot and is outside the scope of this paper. Here we assume that the size of a structure perpendicular to the surface direction is much more than the wavelength of light in the plane waveguided regime of light propagation. As a result, we come to a two-dimensional variant of the MIE[4]:

$$\vec{E}\,'(\vec{r}_l) = \vec{E}_i(\vec{r}_l) + \sum_{j(\neq 1)} \nabla\times\nabla\times\vec{d}(\vec{r}_j)G(R_{jl}) \ , \quad (1a)$$

$$\vec{H}'(\vec{r}_l) = \vec{H}_i(\vec{r}_l) + \sum_{j(\neq 1)} ik\nabla\times\vec{d}(\vec{r}_j)G(R_{jl}) \ , \quad (1b)$$

where $\vec{E}\ '(\vec{r}_l)$ and $\vec{H}\ '(\vec{r}_l)$ are the electric and magnetic fields acting on the elementary radiator placed at point $\vec{r}_l$, $\vec{E}_i(\vec{r}_l))$ and $\vec{H}_i(\vec{r}_l)$ are the strengths of the fields of the incident wave at the same point, $\vec{d}(\vec{r}_j)$ is the two-dimensional dipole, $G(kR_{jl}) = \pi i H_0^{(1)}(kR_{jl})$ is the Green function of the scalar wave equation and $H_0^{(1)}$ is the Hankel function of the first kind, zeroth order, $\vec{R}_{jl} \equiv \vec{r}_j - \vec{r}_l, k = \omega/c$. Here $\vec{r}_j$ and $\vec{r}_l$ are two dimensional vectors (in the xy plane). The gradient symbol ∇ indicates differentiation over $\vec{r}_l$. Strictly speaking, it should be noted that, just as in Ref. [4], our problem is not quite a two-dimensional one. Although all physical quantities are implied to be independent of the z-coordinate oriented along the cylinders' axes we will be interested in the determination of the z components of the electromagnetic field and electric dipole moment as well. Earlier, when the MIE was applyied to dense media (the distances b between neighbouring radiators assumed to be infinitesimal compared to the wavelength λ of the radiation) one could afford to use two principal benefits: (i) consider all the radiators inside Lorentz cavity (LC) as identical ones, i.e. assume polarization inside LC to be constant, (ii) replace a summation over out-of-cavity radiators by the integration and thus realize the passage to an integral equation[3,4].

In the case of a rarefied medium both of these benefits vanish and these difficulties arise in full measure. To overcome the first obstacle - nonidentity of the radiators inside LC - we take into account the variation of the polarization P within Lorentz cavity:

$$\vec{P}(\vec{r}_j) = \vec{P}(\vec{r}_l) - \vec{R}_{lj} \cdot \nabla \vec{P}_{r=r_l} + \vec{R}_{lj}\vec{R}_{lj} : \nabla\nabla \vec{P}_{r=r_l} + ... \tag{2}$$

The second problem is connected with out-of-LC radiators. Now it is impossible simply to change, as in the case of a dense medium, the summation to integration, since due to the finite value of the parameter kb, the phase difference between two dipoles situated outside LC remains important at any distances of these dipoles from the center. Therefore any two dipoles at points j and j' with a finite distance b between them produce at the observation point a *different field* as compared with one isolated dipole with the same total moment. To consider these dipoles as colocated it is necessary to the same satisfy not only condition as in the electrostatic approximation

$$b \ll R_{lj}, R_{lj'} \tag{3}$$

but an inequality

$$kb \ll 1, \quad \text{i.e.} b \ll \lambda \tag{4}$$

as well. Therefore, if one wants account for the discreteness of a medium one cannot simply pass from a sum to the integral, and so it is impossible to obtain an integral equation. However, it is possible to calculate *the difference* between such sum and the integral. For this purpose the special procedure of "radiator's splitting" was developed.

III. Splitting procedure and passing to the integral equations

In essence it consists in an imaginary splitting of each elementary radiator into 4 smaller ones to obtain a lattice with the same configuration but with a period $b/2$. After that we expand the field from the "splitting dipoles" at the observation point by the parameter b/R_{jl} and calculate the difference between local fields $\vec{E}\ '(r_l)$ from the lattices with the periods b and $b/2$ under the same volume density of the dipole moment (polarization $\vec{P}$). By means of the iterative repetition of such a procedure the difference of the acting fields from an initial lattice with grain size b and similar lattice with an arbirary small b may be calculated. This solves the problem of passage to the integral equation.

Thus, we proceed in the following way. The electric or magnetic field from the elementary dipole radiator may be written in the form

$$\vec{f}(\vec{r}_l) = \hat{L}(\nabla_l) G(R_{jl}) \vec{P}(r_j), \tag{5}$$

where $\hat{L}(\nabla_l)$ is a certain differential operator ($\nabla\times$ or $\nabla\times\nabla\times$) acting on $\vec{r}_l$ coordinates near the LC-center. Under the splitting procedure the displacements of these new elementary radiators must be chosen in a such way that the geometry of a new lattice would remain unchanged but the new lattice have twice lesser lattice constant. Then using the inequality

$$R_{jl} \geq a \gg b\ , \tag{6}$$

where a is the size of the LC, one can calculate the field at the LC's center. For this it is necessary substitute into (5) vector $\vec{r}_j + \vec{b}_{j'}$ for $\vec{r}_j$ and expand this expression into a Taylor series of $b_{j'}$:

$$\vec{f} = \hat{L}G\vec{P} + \frac{1}{4}\frac{\partial \hat{L}G\vec{P}}{\partial(\vec{r}_j)_s}\sum_{j'}(\vec{b}_{j'})_s + \frac{1}{8}\frac{\partial^2 \hat{L}G\vec{P}}{\partial(\vec{r}_j)_s\partial(\vec{r}_j)_t}\sum_{j'}(\vec{b}_{j'})_s(\vec{b}_{j'})_t + ... \quad (7)$$

Here j' takes values from 1 to 4 and $\vec{r}_j + \vec{b}_{j'}$ are the radius-vectors of the splitted oscillators, s, t stand for Cartesian components of the vectors. In Eq. (7) the summation is implied over all indices s and t. As is known, any lattice constituted of identical radiators has a center of symmetry and due to this fact the second term in the right-hand part of Eq. (7) is zero. Therefore, by performing the summation over "j " in both parts of Eq.(7) one obtains

$$\sum_j \hat{L}G\hat{f} = \sum_j{}' \hat{L}G\vec{f} - b^2\sum_j \Phi'_{st}\frac{\partial^2 \hat{L}G\vec{f}}{\partial(\vec{r}_j)_s\partial(\vec{r}_j)_t} \quad (8)$$

where

$$\Phi'_{st} \equiv \frac{1}{8b^2}\sum_{j'}(\vec{b}_{j'})_s(\vec{b}_{j'})_t \ , \quad (9)$$

and $\sum'_{j'}$ denotes the summation over a new lattice. By reiterating this procedure N - times we come to the conclusion that the difference between the initial sum and a sum with $b/2^N$ period is equal to a geometrical progression, each term of which is proportional to b^2. This progression may be calculated directly. As a result, we obtain an explicit expression for the difference between the initial sum and the same sum in the limit $kb \to 0$, i.e. integral over outside LC

$$\sum_j \hat{L}G\vec{P} = \int_\sigma^\Sigma [\hat{L}G\vec{P} - b^2(\hat{\Phi} : \nabla'\nabla')\hat{L}G\vec{P}]d^2\vec{r}\ ',$$

where $\hat{\Phi} \equiv \frac{4}{3}\hat{\Phi}'$. Due to presence of the operator ∇' the second volume integral may be transformed into surface integrals over the outer boundary Σ of the medium and LC surface σ. The last one can be calculated directly and gives rise to the terms denoted as $\vec{E}_b$ and $\vec{H}_b$ in the resulting integral equation

$$\vec{E}\ '(\vec{r}) = \vec{E}_i(\vec{r}) + \vec{E}_\sigma(\vec{r}) + \vec{E}_b(\vec{r}) + \int_\sigma^\Sigma \nabla\times\nabla\times\vec{P}(\vec{r}\ ')G(|\vec{r}-\vec{r}\ '|)d^2\vec{r}\ ' - b^2\int_\Sigma(\hat{\Phi} : \vec{n}_\Sigma\nabla_\Sigma)\nabla\times\nabla\times\vec{P}(\vec{r}_\Sigma)G(\vec{r}-\vec{r}_\Sigma)d\vec{r}_\Sigma \ , \quad (10a)$$

$$\vec{H}\ '(\vec{r}) = \vec{H}_i(\vec{r}) + \vec{H}_\sigma(\vec{r}) + \vec{H}_b(\vec{r}) - ik\int_\sigma^\Sigma \nabla\times\vec{P}(\vec{r}\ ')G(|\vec{r}-\vec{r}\ '|)d^2\vec{r}\ ' + ikb^2\int_\Sigma(\hat{\Phi} : \vec{n}_\Sigma\nabla_\Sigma)\nabla\times\vec{P}(\vec{r}_\Sigma)G(\vec{r}-\vec{r}_\Sigma)d\vec{r}_\Sigma \ , \quad (10b)$$

Here $\vec{E}_\sigma(\vec{r})$ and $\vec{H}_\sigma(\vec{r})$ are the contributions of the radiators inside LC, $\vec{n}_\Sigma$ is the unit vector normal to the surface Σ. Keeping the accuracy up to $(kb)^2$ terms inclusive one can write the $\vec{E}_\sigma$, $\vec{H}_\sigma$, $\vec{E}_b$, $\vec{H}_b$ in a form

$$\vec{E}_\sigma = \hat{\gamma}\cdot\vec{P} + b^2\hat{\gamma}_2\vdots(\nabla\nabla\vec{P}) \ , \quad (11a)$$

$$\vec{H}_\sigma = -ikb^2\hat{\gamma}_{M1} : (\nabla\vec{P}) \ , \quad (11b)$$

$$\vec{E}_b = (kb)^2\hat{\gamma}_b\cdot\vec{P} + b^2\hat{\gamma}_{b2}\vdots(\nabla\nabla\vec{P}) \ , \quad (11c)$$

$$\vec{H}_b = -ikb^2\hat{\gamma}_{Mb1} : (\nabla\vec{P}) \ , \quad (11d)$$

where $\hat{\gamma}$, $\hat{\gamma}_b$, $\hat{\gamma}_{Mb1}$, $\hat{\gamma}_2$, $\hat{\gamma}_{b2}$ are unitless second-, third- and forth rank tensors, respectively. They are determined by the spatial distribution of the radiators. From (9) it follows

$$(\hat{\Phi})_{st} = \Phi_s\delta_{st}, \quad (12a)$$

$$\Phi_x = \frac{b_x}{24b_y}, \quad (12b)$$

$$\Phi_y = \frac{b_y}{24 b_x}, \qquad (12c)$$

and

$$(\hat{\Phi})_{st} = \frac{\delta_{st}}{24}, \qquad (12d)$$

for rectangular and quadratic lattices, respectively.

Application of this approach to random media depends not only on the parameter kb but on the density parameter Nb^2 as well (N is the two- dimensional density of the radiators). In this way it is necessary to distinguish two different situations.

a) Gas-like medium: $Nb^2 \ll 1$

In this case, in the first order approximation in the gas parameter Nb^2, we have the same probability to find a dot in any point of space with the exception of a circle of radius b around the given dot. After an ensemble averaging from a viewpoint of a given radiator the remainder the medium looks like a continuous one. Therefore, in this special case a summation may be replaced by an integration without any "splitting" procedure at all and so the tensor $\hat{\Phi}$ must be equated to zero.

b) Jelly-like medium: $Nb^2 \leq 1$.

For a jelly-like medium we have an isotropic angular distribution, but the distance between any two adjacent particles is always approximately equal to b (an essential distinction from a gas-like case). It is evident that to guarantee the spatial isotropy after a "splitting" procedure one must rotate isotropically each 4-particle cell. After averaging over the orientations Eq. (12d) turns into scalar which to an accuracy of the $(kb)^2$ terms must be the same as for a quadratic lattice.

III. Optical properties of random and regular structures

Now return to the integral Eqs. (10): just as in Ref. [4,5] after setting the "blurred" boundary function $\vec{P}(\vec{r}_\Sigma)$ equal to zero, we can see that Σ-surface integrals vanish. Further let us introduce new variables in the form

$$\vec{E} = \vec{E}\,' + \hat{\beta}_0 \cdot \vec{P} + b^2 \hat{\beta}_2 \vdots (\nabla\nabla\vec{P}), \qquad (13a)$$

$$\vec{H} = \vec{H}\,' + ikb^2 \hat{\beta}_{M1} : (\nabla\vec{P}), \qquad (13b)$$

where $\hat{\beta}_0$, $\hat{\beta}_2$, $\hat{\beta}_{M1}$ are free parameters. Then choose these parameters in such a manner that fields $\vec{E}$ and $\vec{H}$, together with the integral equation, satisfy also the following macroscopic wave equations

$$\nabla \times \nabla \times \vec{E} - k^2 \vec{E} = 4\pi k^2 \vec{P}\ , \qquad (14a)$$

$$\nabla \times \nabla \times \vec{H} - k^2 \vec{H} = -4\pi k \nabla \times \vec{P}\ , \qquad (14b)$$

This condition totally determines the values of all free parameters in Eqs.(13a,b) and then new variables acquire in a remarkable manner physical sense of the macroscopic fields[4,5]. However, now local field factors depend not only on the medium's density but also on the lattice period. In conclusion we present the explicit expressions of the refractive index n for random (gas, jelly-like liquid) and regular (square lattice) distributions of the radiators. For random media we obtain

$$n^2 - 1 = (n_0^2 - 1)\left\{1 + \frac{n_0^2 - 1}{4}(kb)^2 \left[\left(\ln k\tilde{b} - c\right)(1 + \delta) + n_0^2 \frac{\delta - 1}{4}\right]\right\} \qquad (15)$$

where $\tilde{b} = b$, $c = 0.62$ for gas and $\tilde{b} = b/2$, $c = 0.79$ for liquid.

For a regular quadratic lattice we have

$$n^2 - 1 = (n_0^2 - 1)\left\{1 + \frac{n_0^2 - 1}{4}(k\tilde{b})^2 \left[(\ln kb - c_1)(1 + \delta) + n_0^2 \left(4c_2\,(2\sin 2\varphi - 1) - \frac{1}{4}\right)(\delta - 1)\right]\right\}\ , \qquad (16)$$

where c_1, c_2 are some constants order of unity, φ is the angle between wavevector $\vec{k}$ and vector of the lattice, $\delta = 1$ for z- direction and $\delta = 0$ for x, y- directions of the electric field vector, n_0 is determined by the two-dimensional analog of the Lorentz-Lorenz(LL) formula[4]

$$n_0^2 - 1 = 4\pi N\alpha \quad \text{for} \quad (E = E_z)\ ,$$

$$n_0^2 - 1 = \frac{4\pi N\alpha}{1 - 2\pi N\alpha} \quad \text{for} \quad (E = E_{x,,y}) \ ,$$

Thus, the value of the refractive index depends on the direction of the wavevector relatively of the lattice axis. The birefringence angle θ, which is determined as an angle between $\vec{E}$ and the induction vector, is given by the relation

$$\sin\theta = 4c_2(kb)^2 \frac{n_0^2 - 1}{n_0^2} \sin 4\varphi \qquad (17)$$

This effect manifest itself most brightly at the angle $\varphi = \pi/8$ when effective asymmetry between the lattice's sides is maximal.

IV. Conclusion

It is shown tha random medium and quadratic lattice structures, which are originally indistinguishable, demonstrate a vivid distinction with allowance for only just terms $(kb)^2$ in the first approximation. For $kb \approx 1$ it may even lead to a change of the sign for local field corrections in comparison with the LL formula. The birefringence effect in a quadratic lattice is predicted and calculated. The conditions for the birefringence and PBG observation are quite realistic. Recently lateral propagation of light through a photonic lattice structure was studied using spontaneous emission from the quantum wells[6]. The light interacted strongly with the shallow lattice and scattered along three equivalent axes of the lattice. It was interpreted as a preliminary evidence that an appropriatly fabricated (sufficiently deep) lattice would suppress the transmission of light within the PBG.

We are grateful to CNPq and FAPESP foundations for financial support.

References

1. E. Yablonovitch, Phys. Rev. Lett., **58**, 2059 (1987).
2. E. Yablonovitch and T. J. Gmitter, Phys. Rev. Lett., **63**, 1950 (1989).
3. M. Born and E. Wolf, *Principles of Optics,* (Pergamon Press, Oxford, 1964).
4. A. V. Ghiner and G. I. Surdutovich, Phys. Rev. A., **50**, 714 (1994).
5. A. V. Ghiner and G. I. Surdutovich, Clausius-Mossotti and Lorentz-Lorenz formulas: what is the difference? Seventh Rochester Conference on Coherence and Quantum Optics, June 7-10, 1995.
6. J. R. Wendt et al, J. Vac. Sci. Technol. **B 11(6)**, 2637 (1993).