Self-consistent photonic band structure of dielectric superlattices containing nonlinear optical materials

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(Received 8 July 2000; revised manuscript received 11 October 2000; published 24 January 2001)

The theory of photonic crystals is extended to include the optical Kerr effect taking place in weak third-order, nonlinear materials present in the unit cell. The influence on the dispersion relations of the illumination caused by a single Bloch mode transiting through the crystal structure is examined. Special attention is given to the modification of the photonic gap width and position. Assuming an instantaneous change of refractive index with illumination, the nonlinear band structure problem is solved as a sequence of ordinary, linear band structure calculations, carried out in a plane-wave field representation.

DOI: 10.1103/PhysRevE.63.027602 PACS number(s): 42.70.Qs, 42.65.Pc, 42.70.Mp

After the work of Franken *et al.* [1], there has been steadily expanding interest in the study of nonlinear optics materials. The many effects that have been discovered, arising from the second- and third-order nonlinear polarization largely explain this enthusiasm, many of those being expected to trigger new developments in the area of photonics [2]. On the other hand, periodic dielectric structures are very effective in generating strong electromagnetic dispersions [3]. Photonic crystals are one-, two-, or three-dimensional structures which exhibit unusual optical properties. Most of these arise from strong departures in their dispersion relation from the case of homogeneous space photons, where the frequency ω is proportional to the wave-vector amplitude k. These periodic dielectric structures have their modes classified according to Bloch's theorem, involving an infinite number of branches or bands, each of which is characterized by a dispersion relation defined on the first Brillouin zone of the periodic lattice. The finite extension of each band in the frequency space can lead to frequency gaps [4] where, because of Bragg scattering, electromagnetic waves are not allowed to propagate. One of the classical structures where this effect was demonstrated is known as the ''layer-by-layer'' or ''woodpile'' structure describing a periodic face-centered cubic architecture, with dielectric rods stacked transversely [5]. This structure is apparently somewhat easier to fabricate than structures where the rods are intersecting. With an adequate adjustment of the relative position of the rod layers, the dielectric distribution can be given an ordering close to that found in the diamond crystal at atomic scales. This results, for rods with large refractive index, in the appearance of a large photonic band gap which strongly dominates the optical properties of this inhomogeneous structure. In particular, the microfabrication of structures which prohibit optical propagation in the infrared spectral range [6] are by now well mastered.

One-dimensional photonic crystals, including nonlinear materials, have recently received much interest, as it was realized that the feedback mechanisms inherent in nonlinear materials could considerably alter the photonic structure of a periodic dielectric multilayer [7]. Photonic band structure of material possessing Kerr nonlinearity have first been introduced by Tran [8] who showed the effect of the presence of electromagnetic energy on a two-dimensional photonic crystal spectrum. Tran's work shows a clear dependence of the position of the photonic band gap as a function of the incident energy. This band gap behavior is recovered in the present paper which deals with three-dimensional dielectric structures containing nonlinear optical materials. However, contrasting the mean-field finite-difference time-domain approach found in Tran, the present report introduces a polarization-dependent self-consistent field approach for the calculation of the photonic band structure of Kerr media, where the inhomogeneous refractive index depends on the light field intensity. The specific case of a nonlinear layer-by-layer structure will be examined.

The self-consistent treatment of the nonlinear band structure problem requires a series of operations involving the solution of the linear band structure. We then need to write out the essential relations which allow one to carry out those linear steps. We also develop the expressions required to compute the intensity distribution associated with each of the eigenmodes.

The wave equation, for monochromatic electromagnetic fields propagating in an inhomogeneous medium with a periodic dielectric constant $\epsilon(\vec{r})$, can be written

$$\vec{\nabla}\times\left[\sum_{\vec{G}}\left(\frac{1}{\epsilon}\right)_{\vec{G}} e^{i\vec{G}\cdot\vec{r}}\vec{\nabla}\times\vec{H}\right]=\frac{\omega^2}{c^2}\vec{H}, \tag{1}$$

where $\omega=2\pi\nu$ measures the wave frequency ν. The equivalent equation for the electric field will not be written, as, in this case, the electric field can be deduced from the magnetic field. The translation invariance permits the application of Bloch's theorem. In Fourier space the electromagnetic modes adopt the following form:

$$\vec{H}(\vec{r})=\sum_{\vec{G}}\ (h_{1\vec{G}}\vec{e}_{1\vec{G}}+h_{2\vec{G}}\vec{e}_{2\vec{G}})e^{i(\vec{k}+\vec{G})\cdot\vec{r}}, \tag{2}$$

where the Bloch vector $\vec{k}$ belongs to the crystal lattice first Brillouin zone. $\vec{e}_{1\vec{G}}$ and $\vec{e}_{2\vec{G}}$ are normalized polarization vectors both perpendicular to $\vec{k}+\vec{G}$ and normal to each other. When this is inserted in the wave equation (1), we are led to the following matrix eigenvalue problem:

$$\sum_{\vec{G}'}\sum_{\lambda'=1}^{2}\left\{\left(\frac{1}{\epsilon}\right)_{\vec{G}-\vec{G}'}[(\vec{k}+\vec{G})\times\vec{e}_{\lambda\vec{G}}]\times[(\vec{k}+\vec{G}')\times\vec{e}_{\lambda'\vec{G}'}]\right\}h_{\lambda'\vec{G}'}=\left(\frac{\omega}{c}\right)^2 h_{\lambda\vec{G}}, \quad (3)$$

$$\sum_{\vec{G}}\sum_{\lambda}|h_{\lambda\vec{G}}|^2=1. \quad (4)$$

This eigenvalue problem produces the dispersion relations of the photonic modes and the Fourier coefficients needed to reconstruct the magnetic field associated with each mode. The matrix to be diagonalized is Hermitian so that the eigenvalues are all real and the eigenvectors are orthogonal to each other. The energy flux carried by such a Bloch wave, or its associated intensity (in watt per square meter), is distributed in space according to the value of the Poynting vector. For monochromatic oscillating fields, we write

$$\vec{j}(\vec{r})=\frac{1}{2}\Re[\vec{E}\times\vec{H}^*]=\Re\sum_{\vec{G}}\vec{j}_{\vec{G}}e^{i\vec{G}\cdot\vec{r}} \quad (5)$$

with coefficients explicitly given by

$$\vec{j}_{\vec{G}}=\frac{1}{2\omega\epsilon_0}\sum_{\vec{G}'}\sum_{\vec{G}''}\left(\frac{1}{\epsilon(\vec{r})}\right)_{\vec{G}+\vec{G}'-\vec{G}''}\times\vec{H}^*_{\vec{G}'}[(\vec{k}+\vec{G}'')\times\vec{H}_{\vec{G}''}]. \quad (6)$$

The vector character of the current density implies that, in general, energy is not transported along straight lines: due to refraction effects, the energy can avoid parts of the crystal unit cell and concentrate in others. The local light intensity is given by the projection of the current density along its own direction, i.e., the Cartesian norm of the current, $I(\vec{r})=\|\vec{j}(\vec{r})\|$. This distribution has automatically the periodicity of the photonic crystal. In a Kerr medium, this quantity partially controls the refractive index

$$n(I)=n(0)+n_2^I I. \quad (7)$$

If we consider an eigenmode of the photonic crystal with a specific Brillouin zone $\vec{k}$ vector, we can evaluate its associated intensity by using Eqs. (4)–(6) by substituting there the normalized Fourier coefficients of the eigenfields. The standard normalization [Eq. (4)] amounts to occupy the mode with a specific number of photons which we will later use as a reference. Imposing the illumination level of the crystal using a single specific eigenmode means changing the number of photons occupying this mode. We will measure the illumination by specifying the ratio β between the imposed intensity and the reference intensity given by the normalization condition (4).

We now assume that the nonlinear photonic crystal receives energy in one of its eigenmodes which is then occupied by photons in such a way that the intensity distribution is given by

$$I(\vec{r})=\beta\|\vec{j}^{n\vec{k}}_{ref}(\vec{r})\|. \quad (8)$$

The occupied state $n\vec{k}$ is chosen by specifying the incident frequency ω, the direction of the propagating energy in the Brillouin zone, and the band index n appropriate to the transiting wave polarization. From these inputs, we still have to compute the Brillouin zone $\vec{k}$ vector which characterizes the excited mode by adjusting its length so that the dispersive frequency matches the given value of ω.

In practice, the computation of the nonlinear band structures is carried out as follows. First, the nonlinear effects are neglected and the linear band structure is determined without

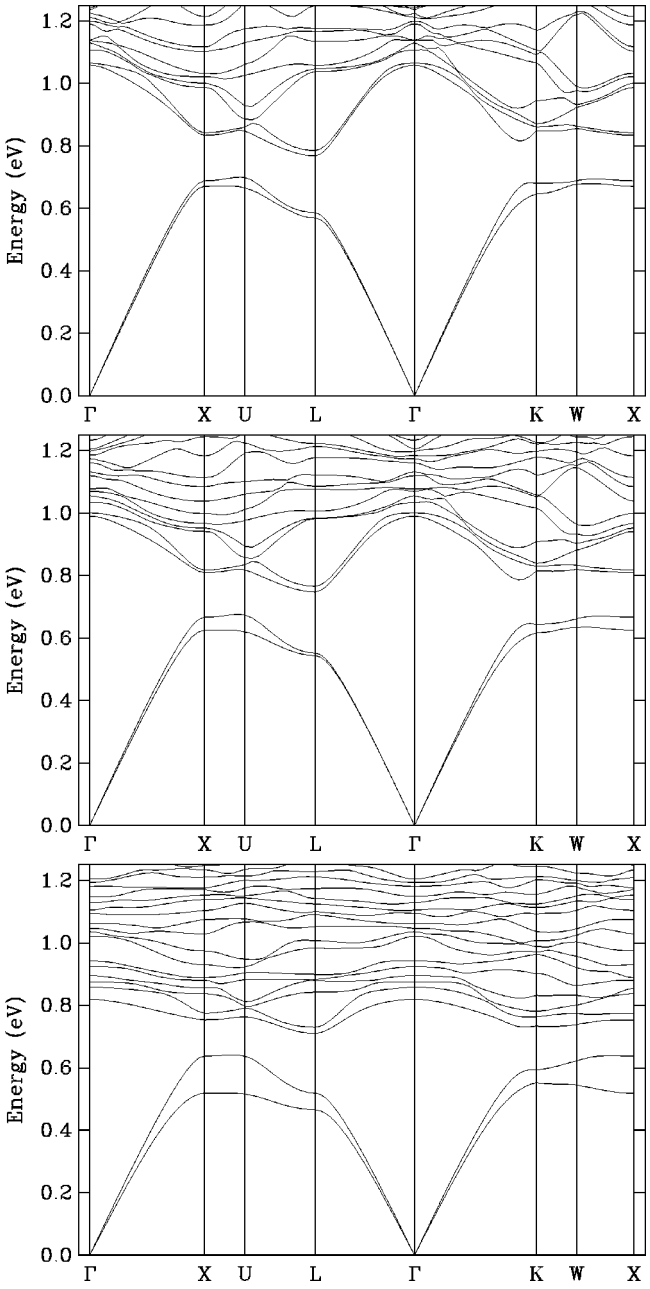

FIG. 1. Self-consistent band structure of a nonlinear woodpile photonic crystal under illumination. From top to bottom, the illumination levels are set to $\beta=5$, $\beta=10$, and $\beta=15$.

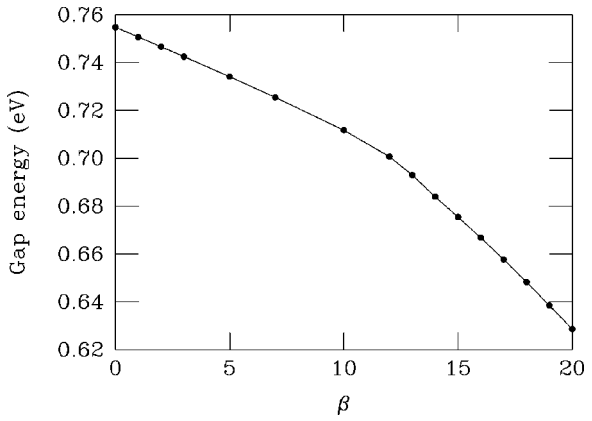

FIG. 2. Variation of the midgap frequency with illumination for a nonlinear woodpile structure.

illumination. Then we start operating with the following iterative steps: the ''pump'' photon state is determined from the chosen incident direction and the input beam frequency ω, and the corresponding intensity is determined throughout the crystal primitive cell. The resulting change of refractive index is evaluated and a new photonic band structure is computed. This procedure is repeated until the band structure is stabilized, i.e., the input refractive index matches the output refractive index.

The new aspect brought about by this iterative treatment, compared to standard linear band structure computation is the fact that, in the nonlinear case, the final band structure explicitly depends on the occupation of the photon modes. This situation is reminiscent of that found when dealing with the problem of n interacting particles in quantum mechanics where elementary treatments (Hartree, Hartree-Fock, density functional theory in the local density approximation, . . .) involve the self-consistent iterative solution of a nonlinear Schrödinger equation. There, the electronic charge density also depends on the occupation of the lower energy states, and the final energy spectrum has to be consistent with this occupation. The extension of the theory of photonic crystals from linear to weak nonlinear response can be paralleled to the extension of the quantum theory of the one-body problem to the self-consistent field approach of an n-body problem.

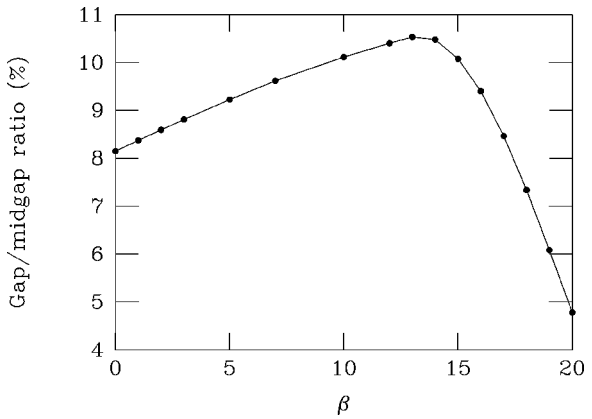

FIG. 3. Variation of the gap width to midgap frequency ratio with illumination for a nonlinear woodpile structure.

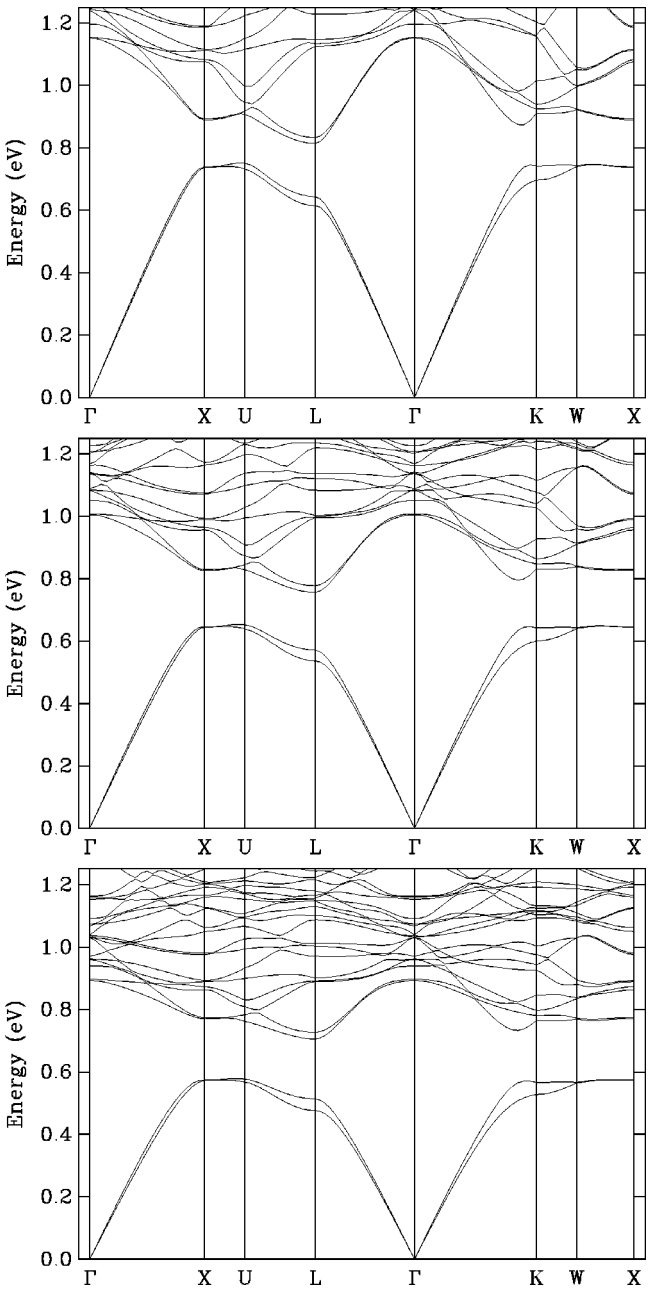

FIG. 4. Woodpile band structure computed for homogeneous rods in air (homogeneous optical density model), for different values of the refractive index: from top to bottom, $n=2.5$, $n=3.0$, and $n=3.5$.

The occupation of the modes considered here is restricted to the occupation of a single mode but this is by no means a stringent limitation: we can without conceptual difficulty populate more than one state and still proceed to the self-consistent calculation. The extension only requires to sum up the different intensities generated by all the occupied modes to obtain the global intensity which modifies the refractive index.

We now consider a photonic structure known as the ''woodpile'' structure, made of rods cut in a Kerr material, namely As_2S_3, with a dark field refractive index of 2.5, and a nonlinear coefficient n_2^I of $4.2\times10^{-19}\,m^2\,W^{-1}$ [9]. The nonlinear dielectric structure can be described as a stacking

of crossed layers of parallel rods separated by air gaps. The Bravais lattice of the photonic crystal is face-centered cubic with crystal parameter $a=1$ μm [lattice translations vectors: $\vec{a}_1=(a/2)(0,1,1)$, $\vec{a}_2=(a/2)(1,0,1)$, $\vec{a}_3=(a/2)(1,1,0)$] and the primitive-cell contents can be described as two parallelepipeds: the first one, with origin $\vec{\tau}_1=(0,0,0)$ with the edge vectors $\vec{c}_1=(a/2)(1,1,0)$, $\vec{c}_2=(a/4)(0,0,1)$, and $\vec{c}_3=(a/4)(-\sqrt{2},\sqrt{2},0)$, and the second one, with origin $\vec{\tau}_2=(a/2)(1,1,1)$ with edge vectors $\vec{c}_1=(a/2)(1,-1,0)$, $\vec{c}_2=(a/4)(0,0,1)$, and $\vec{c}_3=(a/8)(\sqrt{2},\sqrt{2},0)$. Inside these parallelepipeds, the local linear and nonlinear responses are taken to be those of arsenic trisulfide, as described above, while the rest of the primitive cell is empty with a unit refractive index and no nonlinear coefficient. Figure 1 shows the self-consistent band structure of this photonic crystal with three levels of illumination ($\beta=5$, $\beta=10$, and $\beta=15$), obtained by populating a single photon mode, chosen in the direction of the *K*-point in the fcc Brillouin zone. The exact state is chosen on the lowest band, maintaining a constant input frequency ($\omega=0.3$ eV) throughout the iterations. This computation shows a significant change of the photonic structure: except for very large values of the illumination, we essentially observe a redshift of both photonic gap edges (Fig. 2) and a significant widening of the photonic band gap (Fig. 3).

These results can be understood in terms of a significant change of the value of the refractive index of the arsenic sulfide rods. Figure 4 shows the woodpile band structure computed for homogeneous rods in air (homogeneous optical density model), with different values of their refractive index ($n=2.5$, 3.0, and 3.5). The principal effect is the overall lowering of the photonic band frequencies as the refractive index is increased. This can be easily related to the structure of the wave equation (1), where the dielectric function $\epsilon(\vec{r})$ appears at the denominator of the operator to be diagonalized. An increase of the mean value of this function will result in a decrease of the eigenfrequencies.

Returning to Fig. 1, we find that the frequency lowering is more pronounced for some of the bands: these correspond to waves whose polarization is close to that of the input excited mode. This results in a separation of the frequencies of different polarizations and a significant reduction of the effect of the photonic crystal symmetry on the polarization pairing of the bands.

The theory presented in this Brief Report is mainly designed to deal with frequencies significantly lower than atomic ionization frequencies, which roughly characterize the response of electrons responsible for the appearance of nonlinear effects. The outcome of the present work then includes the important case of infrared optical switching effects, which might be of importance in many questions relevant to optical information processing methods. Other approaches have been considered in order to generate a parametric adjustment of the photonic crystal spectrum. The use of nematic liquid crystals [10] infiltrating porous structures, for instance, has been demonstrated. Other ideas have been proposed, where parameters like the temperature [11], hydrostatic pressure, or other mechanical actions could influence the photonic gap properties. Though extremely interesting in many instances, these approaches are perhaps not optimally suited to solve the problem of optical switching, as processes which are not purely electronic in nature are bound to be excessively slow in regard to the applications requirements. By contrast, the mechanisms examined in this work are inherently fast. The main limitation on the usefulness of the effect is probably that significant changes of refractive index requires very high light intensities. The ideal material for investigating the effects is probably not known. This material should be characterized by a very strong third-order nonlinear contribution, but, at the same time, should minimize all second-order response. The centrosymmetric suppression of the second order response is known to occur in homogeneous media, but more work and careful attention is still needed to estimate this effect in the case of periodically modulated materials where some form of macroscopic symmetry lowering can be present.

This work was funded partly by the Interuniversity research project on ''Science of interfacial and mesoscopic structures'' (PAI/IUAP N. P3-49) initiated by the Belgian State Policy Programming. We acknowledge the use of Namur Scientific Computing Facility (Namur-SCF), a common project between FNRS, IBM Belgium, and the Facultés Universitaires Notre-Dame de la Paix (FUNDP). J. P. Vigneron acknowledges the hospitality of the Université de Franche-Comté in Besançon where part of this work was done. V.L. is supported by the Belgian National Fund for Scientific Research.

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