Effective electro-optic constants of free-standing superlattices of any symmetry

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(Received 27 April 2001; revised manuscript received 5 September 2001; published 11 December 2001)

A general framework describing the effective electro-optic constants of an idealized free-standing superlattice, composed of thin alternating layers is derived, as a function of the dielectric and electro-optic constants of each of the *N* constituents. The proposed model is valid only if the energy of light is small compared to the gap of each materials. The results are applied to superlattices with layers of all classes of all symmetries (triclinic, monoclinic, orthorhombic, hexagonal, tetragonal, and cubic).

DOI: 10.1103/PhysRevB.65.033303 PACS number(s): 78.20.Jq

INTRODUCTION AND PROBLEM DEFINITION

The mastery of the molecular-beam-epitaxy process has allowed superlattices[1] (SL) to be developed. These superlattices are composed of alternating layers of many materials (N) of thicknesses l_1 to l_N, respectively. The period of such superlattices created in the direction perpendicular to the layers is defined as $L=\Sigma_{i=1}^{N} l_i$. Since the optical wavelength is large compared to the period L in a heterostructure, the superlattice can be considered as a homogeneous medium, whose physical properties are determined by the so-called effective parameters. These are usually obtained by considering some particular averages on the parameters of the constituents. The effective elastic constants have been calculated by Grimsditch for superlattices with layers of orthorhombic symmetry,[2] and for superlattices composed of layers of any symmetry.[3] The photoelastic constants[4] have been also calculated for superlattices of orthorhombic symmetry. The model proposed herein deals with an idealized free-standing superlattice: indeed the development of integrated optoelectronic SL components composed of semiconductors such as GaAs, AlAs, InP, GaP, and their alloys is becoming increasingly interesting. As some of these integrated-optic devices make use of the linear electro-optic effect (electro-optic modulators,[5,6] tunable filters,[7,8] switches, directional couplers, etc.), it is of interest to obtain analytical expressions of their electro-optic coefficients so as to predict their behavior. In telecommunications the energy of light (typically corresponding to $\lambda=1550$ nm) is small compared to the gap of the above semiconductors. Moreover, one can neglect the modification of the absorption spectra near the gap[9] due to quantum-confinement effects. In this paper, we consider an idealized free-standing superlattice with its axis oriented along the x_3 direction, x_1 and x_2 lying in the plane of the layers. Hence, all the properties are referred to orthonormal axes of reference. The thickness of each layer ($n=1$ to N) being l_1 to l_N, respectively, the related fraction of material is defined hereafter as $f_1=(l_1/L)$ to $f_N=(l_N/L)$ (Fig. 1).

The linear electro-optic (Pockels) effect, which is the basis for active waveguide device control, is often used in integrated-optic devices such as filters, modulators, etc. The change in the refractive index is proportional to the applied electric field and modifies the velocity of propagating light waves. The linear change in the coefficients of the index due to an applied electric field is $\delta(1/\varepsilon)_{ij}=\Sigma_{k=1}^{3} r_{ijk}E_k$, where ε_{ij} is the dielectric tensor of the material (i, $j=1$ to 3).[10] The differentiation of the inverse dielectric function tensor[11] leads to the constitutive relation $\delta\varepsilon_{ij}=-\varepsilon_{ii}\varepsilon_{jj}\Sigma_{k=1}^{3} r_{ijk}E_k$ which involves a 6×6 matrix κ function of the dielectric tensor. Then, in reduced notation [$\alpha=1$ to 6 represents the index-contraction of (ij)], the relation which governs the linear electro-optic effect can be expressed in matrix notation as ($n=1$ to N, and SL)

$$\delta\varepsilon^n=-\kappa^n r^n E^n \quad \text{with} \quad \kappa^n = \begin{bmatrix} (\varepsilon_{11}^n)^2 & 0 & 0 & 0 & 0 & 0 \\ 0 & (\varepsilon_{22}^n)^2 & 0 & 0 & 0 & 0 \\ 0 & 0 & (\varepsilon_{33}^n)^2 & 0 & 0 & 0 \\ 0 & 0 & 0 & \varepsilon_{22}^n\varepsilon_{33}^n & 0 & 0 \\ 0 & 0 & 0 & 0 & \varepsilon_{11}^n\varepsilon_{33}^n & 0 \\ 0 & 0 & 0 & 0 & 0 & \varepsilon_{11}^n\varepsilon_{22}^n \end{bmatrix}, \tag{1}$$

where δ_{ε^n} is the variation of the dielectric tensor in reduced notation (6×1 matrix) as an electrical field denoted E^n is applied (3×1 matrix), and r^n are the components in reduced notation (6×3 matrix) of the linear electro-optic

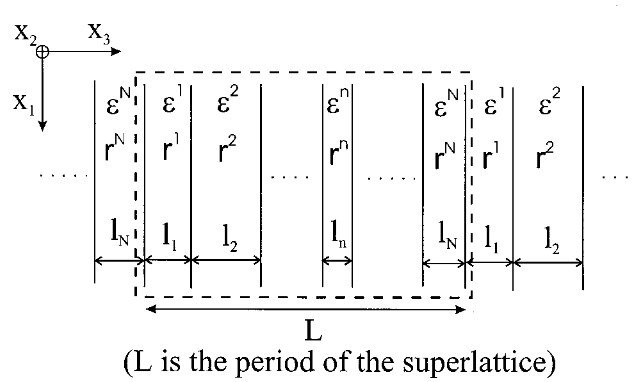

FIG. 1. Schematic diagram of a superlattice (SL) featuring N different constituents. ε^n, r^n, and l_n represent the dielectric matrix, the electro-optic matrix of the layer n and its thickness, respectively.

tensor. The matrix κ^{SL} must be calculated owing to the expressions of the effective dielectric tensor of the superlattice $\varepsilon^{\mathrm{SL}}$.

EXPRESSION OF EFFECTIVE DIELECTRIC TENSOR OF SUPERLATTICES OF ANY SYMMETRY

The basic relation between the displacement field D and the electric field E through the dielectric tensors ε can be expressed as ($n=1$ to N, and SL):

$$D^n=\varepsilon^n E^n. \tag{2}$$

The boundary conditions regarding the continuity of the tangential components of E, and the normal component of D yield the following matrix notation ($n=1$ to N):

$$E^{\mathrm{SL}}=\sum_{n=1}^{N} G^n E^n \quad \text{with} \quad G^n=\begin{bmatrix} 1/N & 0 & 0 \\ 0 & 1/N & 0 \\ 0 & 0 & f_n \end{bmatrix}, \tag{3}$$

and

$$D^{\mathrm{SL}}=\sum_{n=1}^{N} P^n D^n \quad \text{with}$$

$$P^n=\begin{bmatrix} f_n & 0 & 0 \\ 0 & f_n & 0 \\ 0 & 0 & 1/N \end{bmatrix}=\left(\frac{f_n}{N}\right)(G^n)^{-1}, \tag{4}$$

with

$$V^1E^1=\cdots=V^nE^n=\cdots=V^NE^N \quad \text{with}$$

$$V^n=\begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ \varepsilon_{31}^n & \varepsilon_{32}^n & \varepsilon_{33}^n \end{bmatrix}. \tag{5}$$

The constitutive relation (2) for the SL, with Eqs. (3), (4) entailing $E^n=(V^n)^{-1}V^1E^1$ and the basic relation (1) for each layer, yields the general matrix expressions of the effective dielectric tensor of the superlattice,

$$\varepsilon^{\mathrm{SL}}=\left[\sum_{n=1}^{N} P^n\varepsilon^n(V^n)^{-1}V^1\right]\left[\sum_{n=1}^{N} G^n(V^n)^{-1}V^1\right]^{-1}. \tag{6}$$

This general expression allows us to obtain a set of major laws for each dielectric tensor $\varepsilon_{ij}^{\mathrm{SL}}$ of the superlattice, as a function of the properties of its constituents. Assuming a superlattice involving N different materials of triclinic symmetry, we obtain the general framework of laws:

$$\varepsilon_{uv}^{\mathrm{SL}}=\sum_{i=1}^{N}\{f_i\varepsilon_{uv}^i\}-\sum_{i=1}^{N}\left\{f_i\left(\frac{\varepsilon_{u3}^i\varepsilon_{v3}^i}{\varepsilon_{33}^i}\right)\right\}+\frac{\left[\sum_{i=1}^{N}\left\{f_i\varepsilon_{u3}^i\left(\prod_{j=1\neq i}^{N}\varepsilon_{33}^j\right)\right\}\right]\left[\sum_{i=1}^{N}\left\{f_i\varepsilon_{v3}^i\left(\prod_{j=1\neq i}^{N}\varepsilon_{33}^j\right)\right\}\right]}{\left[\sum_{i=1}^{N}\left\{f_i\left(\prod_{j=1\neq i}^{N}\varepsilon_{33}^j\right)\right\}\right]\left[\prod_{i=1}^{N}\varepsilon_{33}^i\right]} \quad (\text{for } uv=11,22,12), \tag{7}$$

$$\varepsilon_{uv}^{\mathrm{SL}}=\frac{\sum_{i=1}^{N}\left\{f_i\varepsilon_{uv}^i\left(\prod_{j=1\neq i}^{N}\varepsilon_{33}^j\right)\right\}}{\prod_{i=1}^{N}\left\{f_i\left(\prod_{j=1\neq i}^{N}\varepsilon_{33}^j\right)\right\}} \quad (\text{for } uv=13,23), \tag{8}$$

$$\varepsilon_{33}^{\mathrm{SL}}=\frac{\prod_{i=1}^{N}\varepsilon_{33}^i}{\sum_{i=1}^{N}\left\{f_i\left(\prod_{j=1\neq i}^{N}\varepsilon_{33}^j\right)\right\}}. \tag{9}$$

It is then possible to define only three new general laws [Eq. (7), (8), and (9)] for the whole dielectric tensor of any idealized free-standing superlattice featuring N different components of any symmetry. Considering these laws, they just hinge on the number of occurrence of the index "3," which relates to the direction perpendicular to the layers. For a superlattice involving only two different isotropic materials ($N=2$), $\varepsilon_{11}^1=\varepsilon_{22}^1=\varepsilon_{33}^1$ and $\varepsilon_{11}^2=\varepsilon_{22}^2=\varepsilon_{33}^2$ for the first and the second constituent, respectively; then, Eqs. (7) and (9) lead to the classical results $\varepsilon_{11}^{\mathrm{SL}}=\varepsilon_{22}^{\mathrm{SL}}=f_1\varepsilon_{11}^1+f_2\varepsilon_{11}^2$ and $\varepsilon_{33}^{\mathrm{SL}}=(\varepsilon_{11}^1\varepsilon_{11}^2)/(f_1\varepsilon_{11}^2+f_2\varepsilon_{11}^1)$, as reported in Ref. 12. The three general expressions (7), (8), and (9), allow us to calculate the matrix κ^{SL} in the basic relationship (1).

In order to differentiate Eq. (7), (8), and (9), it is necessary to explain these three laws as linear combinations for the simplification of the whole calculation. Then, so as to carry on the calculation and to determinate the quantities called Ξ_{uv}^n ($n=1$ to N, and SL), it is possible to infer from equations (7), (8), and (9), the relationships known as the "Vegard rules,"

$$\Xi_{uv}^{\mathrm{SL}}=\sum_{n=1}^{N} f_n\Xi_{uv}^n. \tag{10}$$

In solid state physics such laws show off the linear variation of the lattice parameter of a primary solid solution with the atomic percentages of the solute elements. To this end, inverting Eq. (9), leads directly to the relationship $1/\varepsilon_{33}^{\mathrm{SL}}=\sum_{n=1}^{N}f_n/\varepsilon_{33}^n$. In the same way, considering both expressions (7) and (8) allowing for $\varepsilon_{33}^{\mathrm{SL}}$ from Eq. (9), the two

Vegard law type $\varepsilon_{uv}^{SL}-\varepsilon_{u3}^{SL}\varepsilon_{v3}^{SL}/\varepsilon_{33}^{SL}=\Sigma_{n=1}^{N}f_n(\varepsilon_{uv}^{n}-\varepsilon_{u3}^{n}\varepsilon_{v3}^{n}/\varepsilon_{33}^{n})$ for the group of index $uv=11$, 12, and 22, and $\varepsilon_{uv}^{SL}/\varepsilon_{33}^{SL}=\Sigma_{n=1}^{N}f_n\varepsilon_{uv}^{n}/\varepsilon_{33}^{n}$ for the group of index $uv=13$ and 23 are, respectively, obtained. Hence expressions (7), (8), and (9) may be considered as Vegard-rule-like (10) with the quantities Ξ_{uv}^{n} defined as

$$\begin{cases} \Xi_{33}^{n}=\dfrac{1}{\varepsilon_{33}^{n}}, \\ \Xi_{uv}^{n}=\dfrac{\varepsilon_{uv}^{n}}{\varepsilon_{33}^{n}} \quad \text{for} \quad uv=13,23 \\ \Xi_{uv}^{n}=\varepsilon_{uv}^{n}-\dfrac{\varepsilon_{u3}^{n}\varepsilon_{v3}^{n}}{\varepsilon_{33}^{n}} \quad \text{for} \quad uv=11,12,22 \end{cases}. \tag{11}$$

EXPRESSION OF EFFECTIVE ELECTRO-OPTIC TENSOR OF SUPERLATTICES OF ANY SYMMETRY

The voltage variation across one period of the superlattice results from the addition of the corresponding changes across the adjacent layers. Then, the electrical field in the effective medium and in the layers of the superlattice satisfies both Eqs. (3) and (5). Considering the equation of continuity (5), the first and second lines of the matrix V^n express the continuity of the tangential components of the electric vector across the surfaces, whereas the third line expresses the continuity of the normal component of the electrical displacement vector. Hence, the constitutive relationship (1) for the superlattice with Eqs. (3) and (5) yields

$$\delta_{\varepsilon^{SL}}=-\kappa^{SL}r^{SL}\left(\sum_{n=1}^{N}G^n(V^n)^{-1}V^1E^1\right). \tag{12}$$

This equation can be solved for r^{SL} only if a linear combination is defined regarding $\delta\varepsilon^{SL}$ and $\delta\varepsilon^{n}$ (for $n=1$ to N), that is,

$$\delta_{\varepsilon}^{SL}=\sum_{n=1}^{N}f_nL^n\delta_{\varepsilon}^{n} \quad \text{with} \quad \delta\varepsilon^n=\begin{bmatrix}\delta\varepsilon_{11}^{n}\\ \delta\varepsilon_{22}^{n}\\ \delta\varepsilon_{33}^{n}\\ \delta\varepsilon_{23}^{n}\\ \delta\varepsilon_{13}^{n}\\ \delta\varepsilon_{12}^{n}\end{bmatrix}. \tag{13}$$

By differentiating each linear combination expression of the six elements of the effective dielectric tensor ($uv=33$, 13, 23, 11, 22, and 12) for all symmetries of the layers which are expressed in the relations (10) and (11), Eq. (13) can be obtained with a 6×6 diagonal matrix L defined as

$$L^n=\begin{bmatrix}1&0&0&0&0&0\\0&1&0&0&0&0\\0&0&(\varepsilon_{33}^{SL}/\varepsilon_{33}^{n})^2&0&0&0\\0&0&0&(\varepsilon_{33}^{SL}/\varepsilon_{23}^{n})&0&0\\0&0&0&0&(\varepsilon_{33}^{SL}/\varepsilon_{13}^{n})&0\\0&0&0&0&0&1\end{bmatrix}$$
$$\text{for } n=1 \text{ to } N. \tag{14}$$

Considering now the basic equation (1) for the N materials and the boundary conditions (5), the expression of the electro-optic constants of the superlattice for all classes of all the symmetries, triclinic, monoclinic, orthorhombic, hexagonal, tetragonal, and cubic, is then

$$r^{SL}=[\kappa^{SL}]^{-1}\left[\sum_{n=1}^{N}f_nL^n\kappa^nr^n(V^n)^{-1}V^1\right]\times\left[\sum_{n=1}^{N}G^n(V^n)^{-1}V^1\right]^{-1}. \tag{15}$$

As the κ^{SL} matrix is diagonal, it can be noted that the linear dependence is on the inverse of the dielectric response of the superlattice, not the dielectric response itself.

DISCUSSION

Unfortunately, up to now, the number of experimental data for electro-optic coefficients of superlattices is quite limited in the literature. One of the most significant sets of experimental results reported so far deals with the measurement of the electro-optic coefficients r_{63}^{SL} and r_{41}^{SL} for a superlattice made of GaAs and ternary alloys GaAlAs.[13,14] The authors point out that such a heterostructure becomes an effective medium exhibiting a $\overline{4}2m$ symmetry, that is, $r_{41}^{SL}=r_{52}^{SL}\neq r_{63}^{SL}$. In this special case, each component of the superlattice exhibits cubic $\overline{4}3m$ symmetry, that is, $r_{41}^{n}=r_{52}^{n}=r_{63}^{n}$ and $\varepsilon_{11}^{n}=\varepsilon_{22}^{n}=\varepsilon_{33}^{n}$ for each component n of the SL. Actually, Eq. (15) highlights for this particular example that GaAs/GaAlAs SL yields an effective medium exhibiting tetragonal $\overline{4}2m$ symmetry, with

$$r_{41}^{SL}=r_{52}^{SL}=\frac{\sum_{i=1}^{N}\{f_i\varepsilon_{11}^{i}r_{41}^{i}\}}{\sum_{i=1}^{N}\{f_i\varepsilon_{11}^{i}\}}\neq r_{63}^{SL}$$
$$=\frac{\left(\prod_{i=1}^{N}\varepsilon_{11}^{i}\right)\left(\sum_{i=1}^{N}\{f_i\varepsilon_{11}^{i}r_{41}^{i}\}\right)}{\left(\sum_{i=1}^{N}\{f_i\varepsilon_{11}^{i}\}\right)^2\left(\sum_{i=1}^{N}\left\{f_i\left(\prod_{j=1\neq i}^{N}\varepsilon_{11}^{j}\right)\right\}\right)}. \tag{16}$$

This special result (16) can also be expressed as $r_{63}^{SL}=\xi^{N}(f_i,\varepsilon_{11}^{i})r_{41}^{SL}$ with the function

$$\xi^{N}(f_i,\varepsilon_{11}^{i})=\frac{\left(\prod_{i=1}^{N}\varepsilon_{11}^{i}\right)}{\left(\sum_{i=1}^{N}\{f_i\varepsilon_{11}^{i}\}\right)\left(\sum_{i=1}^{N}\left\{f_i\left(\prod_{j=1\neq i}^{N}\varepsilon_{11}^{j}\right)\right\}\right)}.$$

It is clear with this model that the function $\xi^{N=1}$ is equal to unity. That is,

$$\xi^{N=1}(f_1,\varepsilon_{11}^{1})=\frac{\left(\prod_{i=1}^{N}\varepsilon_{11}^{i}\right)}{\varepsilon_{11}^{1}\left(\prod_{j=1\neq i}^{N}\varepsilon_{11}^{j}\right)}=1$$

$$\text{when}\quad f_{i=1}=1\quad\text{and}\quad f_{j\neq i}=0.$$

Considering then homogeneous bulk constituents like GaAs, or AlAs, or a ternary alloy (GaAlAs) one find that: the cubic $\bar{4}3m$ symmetry of the constituent is characterized by $r_{41}=r_{52}=r_{63}$.

CONCLUSION

Equation (15) stands for a general formulation of the effective electro-optic constants of an idealized free-standing superlattice, when the optical wavelength is large compared to the thickness of each of the N constituents, and the energy of light is small compared to the gap of the N materials. The results are usable for superlattices with layers of all classes of all symmetries, i.e., triclinic 1 and 1, monoclinic 2, m and $2/m$, hexagonal $6mm$ and 622, tetragonal $4mm$, 422 and $\bar{4}2m$, and cubic $\bar{4}3m$ and 23, provided that appropriate simplification of the tensor components of the layers is made. The formulation also gives valuable information about the symmetry of the heterostructure considered as an effective medium.

[1] S. M. Rytov, Sov. Phys. JETP **2**, 466 (1956).
[2] M. Grimsditch, Phys. Rev. B **31**, 6818 (1985).
[3] M. Grimsditch and F. Nizzoli, Phys. Rev. B **33**, 5891 (1986).
[4] B. Djafari Rouhani and J. Sapriel, Phys. Rev. B **34**, 7114 (1986).
[5] S. Khalfallah, P. Dubreuil, L. Escotte, R. Legros, C. Fontaine, A. Munoz-Yague, B. Beche, and H. Porte, J. Lightwave Technol. **17**, 103 (1999).
[6] S. Khalfallah, P. Dubreuil, R. Legros, C. Fontaine, A. Muñoz-Yagüe, B. Beche, H. Porte, R. Warno, and M. Karpierz, Opt. Commun. **167**, 67 (1999).
[7] B. Beche, H. Porte, J. P. Goedgebuer, and C. Fontaine, J. Quant. Electron **35**, 820 (1999).
[8] B. Beche, E. Gaviot, N. Grossard, and H. Porte, Opt. Commun. **185**, 325 (2000).
[9] D. A. B. Miller, D. S. Chemla, and S. Schmitt-Rink, Appl. Phys. Lett. **52**, 2154 (1988).
[10] A. Yariv, *Quantum Electronics*, 3rd ed. (Wiley, New York, 1989).
[11] D. F. Nelson and M. Lax, Phys. Rev. Lett. **25**, 1187 (1970).
[12] M. Born and E. Wolf, *Principles of Optics*, 6th ed. (Cambridge University, Cambridge, 1980), p. 706.
[13] M. Glick, D. Pavuna, and F. K. Reinhart, Electron. Lett. **23**, 1235 (1987).
[14] M. Glick, F. K. Reinhart, and D. Martin, J. Appl. Phys. **63**, 5877 (1988).