Band structure and optical properties of strained superlattices

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The electronic and optical properties of strained superlattices are considered. A similarity transformation of the strained-superlattice Hamiltonian is presented and the average crystal for this case is defined. A simple perturbative approach to compute the eigenvalues and eigenstates is introduced. The dielectric functions for strained and unstrained superlattices are compared with the two limiting cases, i.e., the corresponding average crystal and the mean value of the bulk constituents' dielectric function. Application is made for the Si/Ge and GaAs/AlAs superlattices.

I. INTRODUCTION

In the last few years, lattice-matched superlattices (SL's) have been extensively studied, because of their physical and technological importance.[1] One great success of growth techniques is that lattice-mismatched SL's can now be grown without defects, with the provision that their layers are thinner than the corresponding critical thickness.[2] In these thin lattice-mismatched SL's, the lattice constant parallel to the atomic planes (perpendicular to the SL growth axis), $a_{\parallel}$, is constant throughout the crystal. The lattice constant perpendicular to the atomic planes (parallel to the superlattice growth axis), $a_{\perp}$, changes according to the elastic theory.[3] The SL components are strained, and this type of growth is called pseudomorphic.

The presence of strain causes modification of the electronic structure of the constituent materials from that of the unstrained bulk state. Therefore, these SL's are very interesting because they provide a large range of available band gaps and different types of fundamental gaps (direct or indirect). Moreover, the strain can modify band offsets, effective masses, and carrier confinements in the strained layers. The investigation of these effects, which are important in device modeling, requires the study of the electronic and structural properties. In addition, the optical properties of SL's are of special interest with regard to optoelectronic applications, but only a few works have been reported in this field.[4-6]

The aim of this paper is to present a method for the calculation of the energy-band structure and dielectric function of the lattice-mismatched SL's. According to this method, the original problem is transformed to an equivalent one, which can be more easily studied. In particular, the Hamiltonian of the SL is transformed through a similarity transformation, and the resulting Hamiltonian consists of two parts. One part describes the average crystal and, as will be shown, it constitutes a good zeroth-order approximation of the problem, and the other part describes the interaction of the average crystal states. The use of this method gives must the opportunity to utilize perturbation theory in order to calculate the band structure of the strained SL's, avoiding the exact diagonalization of the Hamiltonian. The average crystal states can be used as a basis in order to project on them the SL states and so to interpret in a more efficient way the origin of SL effects. In addition, we investigate how well the dielectric function of the unstrained and strained SL's can be approximated by the dielectric function of the average crystal. This paper is organized as follows: The technical details of the method are presented in Sec. II. Section III describes the band structure and confinement in the Si/Ge system, where the method is applied. The dielectric function (imaginary part) of the average crystal for an unstrained superlattice (GaAs/AlAs) and a strained one (Si/Ge) is presented in Sec. IV. In Sec. V the conclusions of the paper are given.

II. DESCRIPTION OF THE METHOD

A. Lattice-matched superlattices

Before going on to the description of the method for the strained SL's, we will refer to some concepts that have been defined for the unstrained SL's (Ref. 7) and will be needed later. Let us consider a superlattice that consists of two materials with identical structures and the same lattice constant (therefore the same primitive cell can be defined). This common primitive cell produces a

lattice, which is called the underlying lattice (UL). The first Brillouin zone associates with the UL is called the first original Brillouin zone (OBZ). The SL is described in terms of a larger primitive cell, with a volume that is a multiple of the volume of the original primitive cell. The latter will be called the supercell, and the first Brillouin zone associated with it the first SL Brillouin zone (SBZ).

We suppose that the supercell contains N_0 primitive cells of the UL. The supercell primitive vectors $\{\mathbf{a}_i\}$ can be related to the primitive vectors of the UL $\{\mathbf{t}_j\}$ through the relation

$$\mathbf{a}_i=\sum_j N_{ij}\mathbf{t}_j \quad (i,j=1,2,3) , \tag{1}$$

or in matrix notation

$$\underline{A}=\underline{N}\cdot\underline{T} .$$

The different primitive cells of the UL are labeled by the triple index $l=(l_1,l_2,l_3)$, so that the origin of the lth primitive cell is given by the vector

$$\mathbf{r}_l=l_1\mathbf{t}_1+l_2+\mathbf{t}_2+l_3\mathbf{t}_3 . \tag{2}$$

The position vector of the Kth atom in the lth primitive cell of the UL lattice is

$$\mathbf{r}_{l,K}=\mathbf{r}_l+\tau_K , \tag{3}$$

where τ_K, $(K=1,2,\ldots,p)$ are the position vectors of the atoms contained in the UL primitive cell. The different supercells of the SL are labeled by the triple index $\mathbf{L}=(L_1,L_2,L_3)$, so that the origin of the Lth supercell is given by the vector

$$\mathbf{R}_L=L_1\mathbf{a}_1+L_2\mathbf{a}_2+L_3\mathbf{a}_3 . \tag{4}$$

We assume that the origin of the $l=0$ UL primitive cell coincides with that of the $\mathbf{L}=0$ supercell. Primitive cells contained in a supercell are labeled by the triple index

$$\mathbf{n}=l_0 , \tag{5}$$

where l_0 is the index of the UL primitive cell contained in the supercell at the origin ($\mathbf{L}=0$).

In reciprocal space, there are N_0 SBZ's contained in the first OBZ. For each wave vector $\mathbf{k}$ lying in the first SBZ, there exist N_0 equivalent wave vectors of the superlattice lying in the first OBZ, and these are given by

$$\mathbf{k}_i=\mathbf{k}+\mathbf{g}_i . \tag{6}$$

where $\{\mathbf{g}_i\}$ are the N_0 reciprocal-lattice vectors of the SL, lying in the first OBZ.

B. Strained superlattices

We consider now a strained superlattice that is constituted of two materials. We consider that these materials have a common primitive cell and a different lattice constant. In order to determine the relevant parameters for the strained SL, we first take under consideration a hypothetical unstrained SL consisting of the two materials with the same lattice constant, a primitive cell similar to the original materials, the same number of layers of each constituent, and the same growth axis. For this construction we can use the results of Sec. II A, i.e., from Eqs. (1) and (5) we determine the matrix N and the triple index $\mathbf{n}$. Using now the primitive vectors $\mathbf{a}_i^s$ of the strained superlattice, and the previously calculated matrix N, we define a lattice whose primitive cell is defined by

$$\mathbf{t}_i^s=\sum_j N_{ij}^{-1}\mathbf{a}_j^s \quad (i,j=1,2,3) , \tag{7}$$

or in matrix notation

$$\underline{T}^s=\underline{N}^{-1}\cdot\underline{A}^s .$$

This lattice will hereafter be called the commensurate underlying lattice (CUL), and it will be used in the process of transforming the strained SL Hamiltonian. The first Brillouin zone associated with the CUL is called the first commensurate Brillouin zone (CBZ). In reciprocal space, there are N_0 SBZ's contained in the first CBZ. For a wave vector $\mathbf{k}$ lying in the first SBZ there exist N_0 equivalent wave vectors of the superlattice lying in the first CBZ, and these are given by

$$\mathbf{k}_i^s=\mathbf{k}+\mathbf{g}_i^s , \tag{8}$$

where $\{\mathbf{g}_i^s\}$ are the N_0 reciprocal-lattice vectors of the SL lying in the first CBZ.

C. Hamiltonian matrix elements

The wave functions of the SL satisfies the Bloch theorem with respect the translations corresponding to Bravais-lattice vectors of the SL. Supposing that the original basis functions are orbitals localized around atoms, such as muffin-tin orbitals, atomic orbitals, Gaussian orbitals, etc., then it is common to use the following Bloch sums as the basis set for the superlattice:

$$\begin{aligned} Y(\mathbf{k},n,j,a)&=(1/N_s)^{1/2}\sum_m \exp[i\mathbf{k}\cdot(\mathbf{R}_m+\mathbf{r}_n^c+\tau_j^c)] \\ &\quad\times\phi_{n,j}^a(\mathbf{r}-\mathbf{R}_m-\mathbf{r}_n-\tau_j) \\ &=(1/N_s)^{1/2}\exp[i\mathbf{k}\cdot(\mathbf{r}_n^c+\tau_j^c)] \\ &\quad\times\sum_m \exp(i\mathbf{k}\cdot\mathbf{R}_m)\phi_{n,j}^a(\mathbf{r}-\mathbf{R}_m-\mathbf{r}_n-\tau_j) , \end{aligned} \tag{9}$$

where N_s is the number of supercells in the crystal, $\mathbf{k}$ is a wave vector in the first SBZ, and $\phi_{n,j}^a(\mathbf{r}-\mathbf{R}_m-\mathbf{r}_n-\tau_j)$ is the a localized orbital centered at $\mathbf{R}_m+\mathbf{r}_n+\tau_j$. $\mathbf{R}_m$ denotes the position of the mth supercell, $\mathbf{r}_n$ the position of the nth primitive cell in the supercell, and τ_j the position of the j atom relative to the nth cell. The vectors $\mathbf{r}_n^c$ and τ_j^c are the corresponding vectors that refer to the previously defined CUL. The choice of the phase factor $\exp[i\mathbf{k}\cdot(\mathbf{r}_n^c+\tau_j^c)]$ appearing in Eq. (9) is arbitrary, and it was done for later convenience.

By transforming the basis functions defined in Eq. (10) by the transformation matrix,

$$U(a,j,\mathbf{g}_i^s;\beta,j',n)=(1/N_0)^{1/2}\exp[i\mathbf{g}_i^s\cdot(\mathbf{r}_n^c+\mathcal{T}_j^c)]\delta_{\alpha\beta}\delta_{jj'}\ , \tag{10}$$

we get the new basis:

$$\psi(\mathbf{k},\mathbf{g}_i^s,j,a)=(1/N_0)^{1/2}\sum_n \exp[i\mathbf{g}_i^s\cdot(\mathbf{r}_n^c+\mathcal{T}_j^c)]Y(\mathbf{k},n,j,a)\ . \tag{11}$$

The two representations of the Hamiltonian are related by the similarity transformation:

$$\underline{H}'=\underline{U}^{-1}\cdot\underline{H}^0\cdot\underline{U}\ . \tag{12}$$

The new basis set makes the calculations easier and leads to a clearer interpretation of the results. Indeed, in the new representation, the Hamiltonian consists of diagonal blocks labeled $(\mathbf{g}_i^s,\mathbf{g}_i^s)$ and off-diagonal blocks $(\mathbf{g}_i^s,\mathbf{g}_{i'}^s)$. The elements of a diagonal block are given by

$$\langle \mathbf{k},\mathbf{g}_i^s,j,a|H|\mathbf{k},\mathbf{g}_i^s,j',\beta\rangle=\delta_{\alpha\beta}\delta_{jj'}\langle\varepsilon_{nj}^a\rangle_n+\sum_s\exp[-i(\mathbf{k}+\mathbf{g}_i^s)\cdot\mathbf{r}_s^c]\times\langle V_{nj,s}^{\alpha\beta}\rangle_n\ , \tag{13}$$

where the angular brackets $\langle\ \rangle_n$ denote averaged values with respect to n over the supercell. The symbol ε_{nj}^a denotes the on-site matrix element corresponding to an a orbital at position (n,j), and the symbol $V_{nj,s}^{\alpha\beta}$ denotes the Hamiltonian matrix element between an a, orbital at site (n,j) and a β orbital at the sth nearest-neighbor position. From Eq. (13) it is clear that the diagonal blocks of the transformed Hamiltonian contain the average interactions and therefore describe the so-called average crystal. In addition, the diagonal blocks have, in the reciprocal space, the periodicity of the reciprocal space of the CUL. This last property is a consequence of the particular choice of the phase factor in Eq. (9).

The elements of the off-diagonal blocks are

$$\langle \mathbf{k},\mathbf{g}_i^s,j,a|H|\mathbf{k},\mathbf{g}_{i'}^s,j',\beta\rangle=\delta_{\alpha\beta}\delta_{jj'}(1/N_0)\sum_n\exp[i(\mathbf{g}_i^s-\mathbf{g}_{i'}^s)\cdot(\mathbf{r}_n^c+\mathbf{r}_j^c)]\delta\varepsilon_{nj}^a+(1/N_0)\sum_s\sum_n\exp[-i(\mathbf{g}_i^s-\mathbf{g}_{i'}^s)\cdot(\mathbf{r}_n^c+\mathbf{r}_j^c)]\exp[-i(\mathbf{k}+\mathbf{g}_i^s)\cdot\mathbf{r}_s^c]\delta V_{nj,s}^{\alpha\beta}\ , \tag{14}$$

where

$$\delta\varepsilon_{nj}^a=\langle\varepsilon_{nj}^a\rangle_n-\varepsilon_{nj}^a$$

and

$$\delta V_{nj,s}^{\alpha\beta}=\langle V_{nj,s}^{\alpha\beta}\rangle_n-V_{nj,s}^{\alpha\beta}\ .$$

The last relation shows that the off-diagonal blocks depend only on the difference between the SL interactions and the average crystal values, and describe interactions between the states of the average crystal.

III. BAND STRUCTURE OF THE STRAINED Si/Ge SUPERLATTICES

In order to test the method, we have applied it on the strained system of Si/Ge, which is presently of great interest. We used an empirical tight-binding (ETB) Hamiltonian with an $\{sp^3s^*\}$ orbital basis,[8] including the spin-orbit interaction. This parametrization describes well the valence as well as the lowest conduction bands for the bulk materials. The presence of strain modifies the interatomic distances and bond angles. In an ETB model, this results in modifications of the nondiagonal Hamiltonian matrix elements. The influence of strain in our calculations has been taken into account as in Ref. 8. The valence band offset has been taken equal to 0.84 eV.[9]

The band structure of the 4:4, 3:5, and 5:3 SL's [by $n:m$ we mean $(\mathrm{Si})_n/(\mathrm{Ge})_m$] grown on a Si(001) substrate has been calculated with the above-mentioned model. In Fig. 1(a)–1(c) the energy bands along the ΓZ symmetry line in the SL Brillouin zone of these SL's are shown in comparison with the folded bands of the corresponding average crystal. We observe that the upper valence bands of the SL's are very close to those of the corresponding average crystal. Also, the lower conduction bands compare fairly well with those of the average crystal in all cases. Consequently, we can state that the average crystal for a strained SL constitutes a good zeroth-order approximation, and it can be considered as a good starting point for the description of its properties.

In addition, a great advantage of the method is the easy and efficient use of perturbation theory in order to

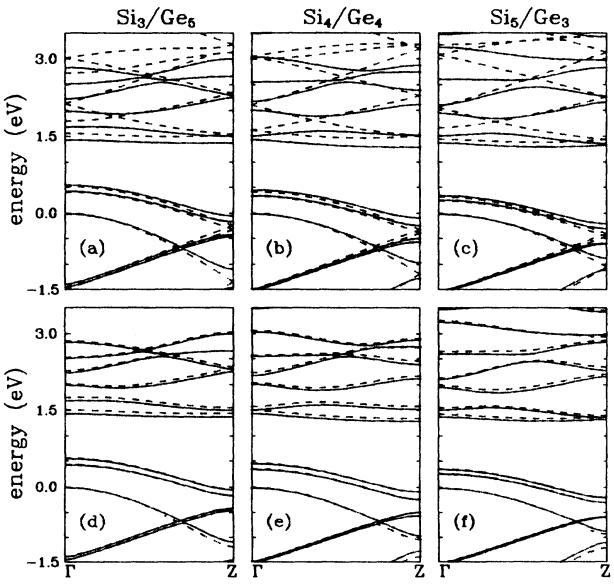

FIG. 1. Energy-band structure of the 3:5, 4:4, and 5:3 Si/Ge SL's (solid lines) compared with the corresponding average crystal bands [(a), (b), and (c), dashed lines], and with the bands that result from the perturbative Hamiltonian [(d), (e), and (f), dashed lines].

improve the band structure of the average crystal. The perturbation is applied in the following way. At first, we diagonalize the diagonal blocks of the transformed Hamiltonian. Thus, we get the band structure and the wave functions of the average crystal in the folded-zone scheme. Next, to correct the energy of an average-crystal state, we select all the states that interact appreciably with this one. The so-selected interacting average-crystal states define a truncated basis set. This set is, then, used to set up a new Hamiltonian matrix with smaller dimension than the original one. In fact, more than one state is corrected simultaneously by diagonalizing the same Hamiltonian matrix. At this point we have to notice that, because the upper valence bands of the average crystal already approach the SL bands, the inclusion of a few average-crystal states in the perturbation has the capability of improving very much the energy bands.

Figures 1(d)–1(f) show the corrected bands of the average crystal in comparison with the superlattice bands for the three SL's. It is worth noticing that the description becomes very good with the application of the perturbation, even for the bands that originally presented large deviations. The truncated basis that has been used is about 2.5 times smaller than the original one, and this fact shows the importance of the perturbation in problems where many diagonalizations of large Hamiltonians are required.

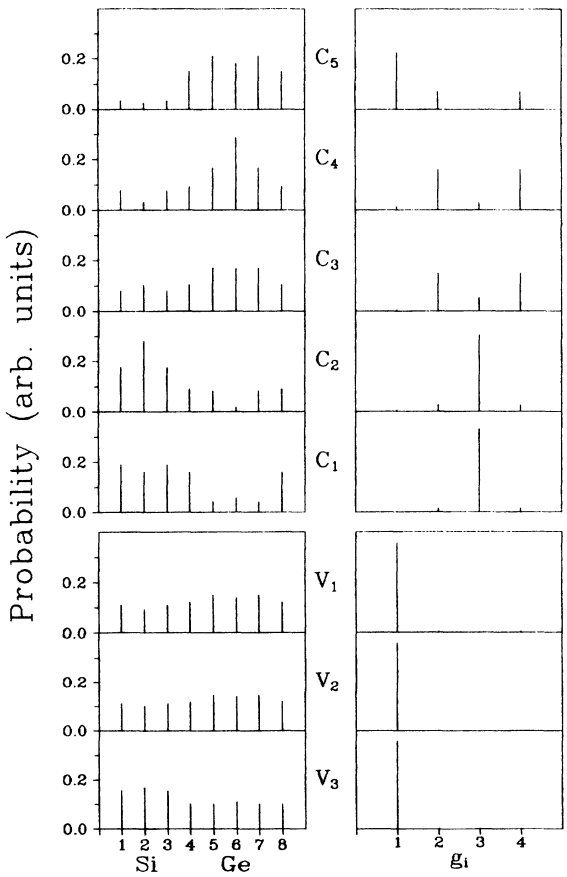

FIG. 2. Relative probabilities of finding an electron on a given site n of the $(Si)_3/(Ge)_5$ SL as a function of n (left side), and the probability for the average-crystal state g_i to appear in the SL wave function (right side).

In order to examine the nature of the SL states, we have calculated the square of the wave functions (the relative probability) of the states at the Γ point of the previous SL's as a function of atomic sites inside the supercell. We have also calculated the probability for different average-crystal eigenstates characterized by $\mathbf{g}_i$ to appear in the SL eigenfunctions. The results for the 3:5 superlattice are shown in Fig. 2. The states are ordered in increasing energy. The upper valence states (V_1, V_2, V_3) as can be seen from the figure, are extended states, while the conduction states are partially confined. The most confined state is C_5, which, as can be seen from the figure, originates from a nonfolded average-crystal state, and is localized in the Ge layers.

IV. DIELECTRIC FUNCTION

In order to calculate the dielectric function, the energies, as well as the wave functions, for a set of $\mathbf{k}$ vectors in the IBZ are needed. The momentum matrix elements are calculated using the relation

$$\mathbf{P}=\frac{m}{\hbar}\frac{dH(\mathbf{k})}{d\mathbf{k}} ,$$

which, although approximate, gives surprisingly good results and has been used in many calculations[8,10,11] previously. The necessary integration in the IBZ has been performed within the linear analytic tetrahedron method (LATM). For computational convenience, the momentum matrix elements inside each tetrahedron are taken to be equal to the mean of the values of the tetrahedron apices. This approximation has been checked by comparing the above calculation with the one in which the momentum matrix elements are assumed to vary linearly inside each tetrahedron. No significant differences have been noted. In order to obtain smooth ϵ_2, we have convoluted it with a Lorentzian of $\Gamma=50$ meV, full width at half maximum (FWHM). Results for the dielectric function will be presented only in the energy region between 0 and 3.7 eV, where the present tight-binding model gives reasonable results for the respective bulk materials.

Recently, it was shown that as the SL period increases, its dielectric function approaches the appropriate mean value of the constitutents's dielectric function.[12] Moreover, it is known that the ultrathin SL energy bands can be well approximated to the corresponding average-crystal bands.[7] It is reasonable to expect that the same will hold for the case of dielectric function. This argument will be tested by studying an unstrained SL (GaAs/AlAs) and a strained one (Si/Ge).

In Fig. 3 the imaginary part of the dielectric function (ϵ_2) for the 1:1, 2:2, 3:3, and 6:6 GaAs/AlAs SL's, as well as the ϵ_2 of the corresponding average crystal (ϵ_2^{av}) and the mean value of the constituent dielectric functions (ϵ_2^m), are presented. The energies and the momentum matrix elements have been calculated using the same model that we have used for the Si/Ge case. The tight-binding parameters have been taken from Ref. 13. One can observe that the ϵ_2 of the 1:1 and 2:2 SL's are very

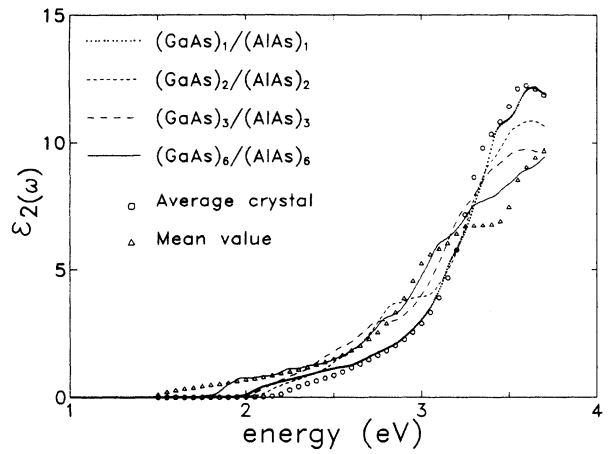

FIG. 3. Dielectric function $\epsilon_2(\omega)$ for $(GaAs)_n/(AlAs)_n$ SL's, with $n=1,2,3,6$, compared with the average-crystal dielectric function and the mean value of the bulk dielectric functions.

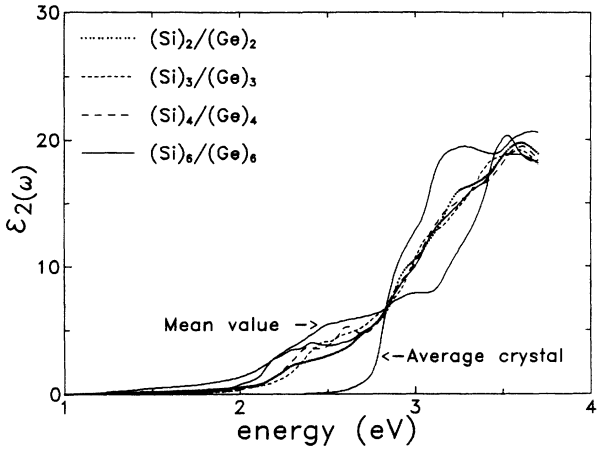

FIG. 4. Dielectric function $\epsilon_2(\omega)$ for $(Si)_n/(Ge)_n$ SL's, with $n=2,3,4,6$ compared with the average-crystal dielectric function and the mean value of the bulk dielectric functions.

close to the ϵ_2^{av}. The 3:3 case constitutes a middle case, as it lies between the ϵ_2^{av} and ϵ_2^m, while ϵ_2 of 6:6 is clearly close to ϵ_2^m. The absorption edge of all presented cases is nearer to the absorption edge of the average crystal. One should also notice that as the SL period increases, there is a continuous transfer of oscillator strength from the energy region 3.2–3.7 eV to the region 2–3.2 eV.

The cases of 2:2, 3:3, 4:4, and 6:6 strained Si/Ge SL's are presented in Fig. 4, along with the corresponding ϵ_2^{av} and ϵ_2^m dielectric functions. Contrary to the previous case, the behavior of ϵ_2 of the Si/Ge SL is rather different. Specifically, we notice that all the ϵ_2, although different from each other, are close to the ϵ_2^m in the energy region 0–2.8 eV. On the other hand, for higher energies, all ϵ_2 are surprisingly close to each other and are between ϵ_2^{av} and ϵ_2^m. The absorption edge is closer to the absorption edge of the ϵ_2^m. Comparing the GaAs/AlAs and Si/Ge cases, one can find that, while in the GaAs/AlAs case, in the presented energy range, there is a uniform transition from ϵ_2^{av} to ϵ_2^m, in the Si/Ge case a different behavior is observed for each of the above-mentioned energy regions. This is an interesting finding, which should be further investigated.

V. CONCLUSIONS

In this paper, the concept of CUL for the strained superlattices has been introduced. This lattice is useful in setting up a transformation of the SL Hamiltonian, which separates the problem into two parts. The first part describes the so-called average crystal, and it is a good starting approximation for the SL band structure. It has been shown that it is possible to calculate the SL states in an efficient way using degenerate perturbation theory. Also, the states of the average crystal can be used to project the SL states and to analyze their behavior. The average-crystal dielectric function, in conjunction with the mean value of the bulk constituents dielectric function, are two limiting cases, and can be used to analyze the SL dielectric function. We found that while the transition of the ϵ_2 for GaAs/AlAs from the ϵ_2^{av} to ϵ_2^m is uniform, the Si/Ge case is quite different.

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