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Brillouin scattering from longitudinal acoustic phonons in superlattices with diffuse interfaces

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Abstract. The computation of the Brillouin cross section for scattering of light off longitudinal acoustic (LA) bulk phonons in a superlattice with diffuse interfaces between the layers is presented for the case of normal incidence. Unlike in previous work concerning the problem of diffuse interfaces, both the modulation of the acoustic and elasto-optic parameters, and the modulation of the refractive index are taken into account. The acoustic phonon spectrum is computed by means of numerical solution of a self-adjoint Liouville equation, and the dispersion relation is obtained using a wavevector selection rule. The transmitted (zeroth-order) electromagnetic wave, and the consequent fluctuating polarization vector field radiating Brillouin light are computed numerically for generic depth profiles of elastic, elasto-optic, and dielectric (optic) properties of the medium. The back-scattered light wave in the superlattice is obtained to first order in perturbation theory. The Brillouin differential cross section is computed by summing over the density of phonon states satisfying the wavevector selection rule. As an application, we present some results for the GaAs–AlAs superlattice, providing evidence for the differences from the case with diffuse and sharp interfaces.

1. Introduction

In a recent paper, Ghislotti and Bottani [1] described a new method for computing the cross section for Brillouin scattering of light by shear horizontal surface acoustic phonons in silicon-on-insulator (SOI) structures with sharp interfaces between the layers of the medium. In a second work, the method was generalized to treat media with *effective* diffuse interfaces (e.g. to describe real materials with inclusions between the layers [2, 3]). There it was emphasized that the method could be applied also to different systems, like superlattices or other ‘phononic crystals’ [4], where the continuous depth profiles of the physical coefficients may correspond to diffuse interfaces down to the atomic scale.

In the present paper we use our method to study the scattering of light off longitudinal acoustic phonons propagating perpendicularly to the surface of a superlattice, with arbitrary interface profiles between the layers.

A great number of papers have been devoted to the study of light scattering from acoustic and optic phonons in superlattices (for a review, see e.g. [5]); this interest has been stimulated by present and possible future applications of semiconductor heterosystems in microelectronics and non-linear optics ([6]).

Brillouin and Raman scattering are natural probes for superlattice phonons, and have been used extensively. In particular, significant experimental studies on GaAs–Ga_xAl_{1-x}As superlattices have been described by Sapriel *et al* [7], Jusserand *et al* [8], and Colvard *et al* [9].

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From a theoretical point of view, He, Djafari-Rouhani and Sapriel [10] presented a very exhaustive computation of the phonon spectrum, including the spatial modulation of both elastic and elasto-optic properties of the superlattice. Yet, their work (like the calculation by Babiker *et al* [11] using a Green function method) was limited to the case of sharp interfaces between the layers of the superlattice.

However, a remarkable number of papers have treated the case of diffuse interfaces in superlattices, because the techniques by means of which the structures are obtained, such as molecular beam epitaxy and metallo-organic chemical-vapour deposition, give rise to a natural diffusion of interfaces at the atomic scale in the medium.

In the work of Colvard *et al* [12] (treating both acoustic and optic phonon regions), the acoustic difference between GaAs and AlAs was neglected: the acoustic waves were treated as plane waves. Furthermore, the Al concentration was considered as a perturbation to that of Ga. Finally, they also assumed that the superlattice behaves like a homogeneous (effective) medium with respect to the propagation of incident and scattered light.

Subsequently, Jusserand *et al* [13, 14] presented an analysis of scattering from folded acoustic phonons, considering the case of interface broadening: they computed the folded acoustic phonon waves by taking into account the acoustic mismatch between the layers, but only for an infinite medium.

In general, we stress that all existing algorithms for computing the Brillouin cross section, in the case of superlattices with diffuse interfaces, assume that the scattering intensity is proportional to the squared modulus of the Fourier transform of the polarization field (that is, the source of the scattered light), thus neglecting the influence of refractions and reflections of the scattered waves at the interfaces in the structure.

We present here a different approach. We consider the whole medium as a thick slab with two free surfaces and depth-dependent physical properties. The system is described by giving the z-profiles of the physical coefficients. Phonons (in the acoustic range) are considered as standing waves in a thick (as compared with the period of the superlattice) slab. Furthermore, we include in our computation, in addition to the acoustic modulation, the elasto-optic and the optic modulation, so taking fully into account the reflections and the refractions of the incident and scattered electromagnetic waves at the interfaces between the layers.

Our theory is not limited to the linear zone of the dispersion curve of the longitudinal acoustic phonons, and it is applicable also when the scattering wavevector exceeds the first reduced mini-Brillouin zone. We only neglect the absorption of light (as was done in all previous work) because the skin depth of the light is large as compared with the period of the superlattice, but absorption could easily be introduced in our method [1, 3]. Of course, the wavevector selection rule typical of a periodic medium (see below) is only a good approximation in our thick-slab approach.

In section 2 we illustrate how the spectrum of bulk longitudinal phonons in the structure is obtained by means of a numerical solution of the self-adjoint Liouville equation governing the propagation of acoustic waves.

The computation of the Brillouin cross section (as proportional to the intensity of the back-scattered light at the surface of the medium) is sketched in section 3.

Finally, section 4 is devoted to the application of this method to the particular case of a GaAs–AlAs superlattice, providing evidence for the difference between a situation with sharp interfaces and one with diffuse interfaces.

2. Longitudinal elastic waves in the superlattice

We consider a superlattice made up of alternating layers of two different cubic crystals with their [001] direction perpendicular to the free surface (the $z = 0$ plane).

The semi-infinite medium is simulated by a thick slab [15] (thickness h), with two free surfaces ($z = 0$ and $z = h$), and 'periodic' depth-dependent physical properties. The superlattice is described by giving the z-profiles of the elastic coefficients, the mass density, the dielectric function, and the elasto-optic coefficients.

Though an accurate description of the phonons in the superlattice needs an atomic model, the long-wavelength acoustic phonons can be studied using an elastic continuum approach.

The wave equation for a longitudinal elastic bulk wave w_z propagating along the z-axis is [16]

$$\rho(z)\frac{\partial^2 w_z}{\partial t^2} = \frac{\partial}{\partial z}\left[C_{11}(z)\frac{\partial w_z}{\partial z}\right] \tag{1}$$

where $\rho(z)$ and $C_{11}(z)$ are the depth profiles of the mass density and of the relevant elastic coefficient in the medium, respectively.

We define the ω Fourier component of the w_z displacement field as

$$w_z(\omega, z, t) = \xi(\omega)\phi_z(\omega, z)\mathrm{e}^{-\mathrm{i}\omega t} \tag{2}$$

where $\xi(\omega)$ is the amplitude of the normal coordinate of the phonon ω. Replacing (2) in the wave equation (1) gives the self-adjoint Liouville equation [17]

$$\frac{\mathrm{d}}{\mathrm{d}z}\left[C_{11}(z)\frac{\mathrm{d}\phi_z(\omega, z)}{\mathrm{d}z}\right] + [\rho(z)\omega^2]\phi_z(\omega, z) = 0. \tag{3}$$

The mode z-profiles $\phi_z(\omega, z)$ are the real eigenfunctions of equation (3). They correspond to the real eigenvalues ω^2, the phonon eigenfrequencies.

The normalization conditions can be deduced from the statistical mean of the energy associated with the phonon of frequency ω [10]:

$$\langle\xi_\omega^2\rangle_{th}\int_0^h \rho(z)\omega^2\phi_z^2(\omega, z)\ \mathrm{d}z \propto \hbar\omega\left(n(\omega|T) + \frac{1}{2}\right) \tag{4}$$

where h is the slab thickness and $n(\omega|T)$ is the Bose–Einstein distribution function. If we take the high-temperature limit $(kT \gg \hbar\omega)$ of the Bose–Einstein distribution, equation (4) becomes

$$\langle\xi_\omega^2\rangle_{th}\int_0^h \rho(z)\phi_z^2(\omega, z)\ \mathrm{d}z \propto \frac{k_B T}{\omega^2} \tag{5}$$

from which we extract

$$\int_0^h \rho(z)\phi_z^2(\omega, z)\ \mathrm{d}z = 1 \tag{6}$$

as the normalization condition for equation (3).

For an acoustic wave propagating along the z-direction in an infinite (periodic) superlattice with sharp interfaces, the dispersion relation [18] takes the well known Kronig–Penney [19] form. If instead one considers the case of diffuse interfaces between layers, the Kronig–Penney dispersion relation is no longer correct. Defining

$$\hat{T}(z) \equiv \left\{\frac{\mathrm{d}}{\mathrm{d}z}\left[C_{11}(z)\frac{\mathrm{d}}{\mathrm{d}z}\right] + [\rho(z)\omega^2]\right\} \tag{7}$$

one can rewrite equation (3) as

$$\hat{T}(z)\phi_z(\omega, z) = 0 \tag{8}$$

where

$$\hat{T}(z) = \hat{T}(z + nD) \tag{9}$$

is a periodic operator (n is an integer and D is the period of the superlattice). Equation (8) has the Bloch-like solution

$$\phi_z(\omega, z) = u_{k_p}(\omega, z)\mathrm{e}^{\mathrm{i}k_p z} \tag{10}$$

where k_p is the phonon wavevector (defined in the extended-Brillouin-zone scheme) and

$$u_{k_p}(\omega, z + nD) = u_{k_p}(\omega, z). \tag{11}$$

In practice we used stress-free boundary conditions at both boundary surfaces of the finite slab:

$$\left(\frac{\mathrm{d}\phi_z(\omega, z)}{\mathrm{d}z}\right)_{z=0} = \left(\frac{\mathrm{d}\phi_z(\omega, z)}{\mathrm{d}z}\right)_{z=h} = 0 \tag{12}$$

and not periodic boundary conditions to obtain the spectrum of (7). In this way our eigenfunctions are standing waves and not the Bloch travelling waves (10). Equations (3), (6), and (12) constitute a well posed Sturm–Liouville eigenvalue problem [20].

The spectrum of longitudinal bulk acoustic phonons in a semi-infinite (we recover this case when our slab thickness h goes to infinity) layered medium is continuous. In our slab approximation, all of the phonon spectrum is discrete, but it becomes quasi-continuous provided that the slab is thick enough. It is then possible to compute the density of phonon states, and to simulate in this way the continuous spectrum of a semi-infinite medium.

Equation (8) allows one to obtain the eigenvalues ω for a fixed value of k_p, and gives the 'folded acoustic branches' which, when k_p is not near to the boundaries of the reduced Brillouin zones, are very close to those obtained by merely folding in the first reduced Brillouin zone the acoustic branch of a homogeneous medium with acoustic velocity V given by

$$\frac{V}{D} = \frac{V_1}{d_1} + \frac{V_2}{d_2} \tag{13}$$

where V_i and d_i $(i = 1, 2)$ are the mass density, the longitudinal acoustic velocity, and the thickness (with perfect interfaces) of layer i.

This folding method gives a zigzag succession of linear branches with the equations

$$\omega = V\left|k_p + \frac{2\pi m}{D}\right| \tag{14}$$

where k_p is the phonon wavevector along the axis z (k_p is limited to the range $-\pi/D$ to π/D—that is, to the first reduced Brillouin zone), and m is an integer which takes the values $m = 0$ for the Brillouin mode and $\pm1, \pm2, \ldots$ for the folded longitudinal acoustic modes $(\mathrm{FLA})_m$.

We numerically solved the problem of finding the whole spectrum of eigenvalues in the acoustic range using the NAG [21] routine D02KDF based on a Prufer transformation, and we found the corresponding eigenfunctions using the NAG [21] routine D02HBF.

3. The Brillouin cross section

We consider a monochromatic electromagnetic plane wave, characterized by the frequency ω_0, normally incident from the vacuum onto the top surface of the structure. The absorption of the wave is here considered negligible, as in [10–13]. Thus the medium is assumed to have a real dielectric function $\epsilon(z, \omega_0)$.

Neglecting the fluctuating part of the susceptibility in Maxwell's equations, we used a numerical method to compute $E_x^{\omega_0}$, the transmitted field in the medium.

If thermal fluctuations are now considered, acoustic phonon fields cause a stochastic variation of the properties of the medium that can be accounted for by means of an instantaneous anisotropic susceptibility.

The electromagnetic scattered field can be computed by means of first-order perturbation theory, because of the smallness of the fluctuating elastic strains produced by phonons at thermal equilibrium. That is, in our hypothesis, we can use the Born approximation in scattering theory [22].

The first step is to compute the fluctuating part of the polarization vector in the medium. Using perturbation theory, we obtain

$$P_i^{\omega_s} = \epsilon_0\,[\epsilon(z) - 1]\,E_i^{\omega_s} + \epsilon_0 \delta\chi_{ij}(\omega_0 \pm \omega_\alpha) E_j^{\omega_0}. \tag{15}$$

Above, $\epsilon(z) - 1 = \chi(z)$ is the unperturbed isotropic susceptibility when the phononic field is not taken into consideration (as a consequence of the fact that $\omega_\alpha \ll \omega_0$, we write $\epsilon(z|\omega_0 \pm \omega_\alpha) \approx \epsilon(z|\omega_0) = \epsilon(z)$).

$\delta\chi_{ij}(\omega_0 \pm \omega_\alpha)$ is the anisotropic fluctuating part of the susceptibility. It is due to the excitation of a single LA (ω_α) phonon mode. The second term in the r.h.s. of equation (15) is responsible for the radiation of Brillouin light—that is, for the scattered field $\boldsymbol{E}^{\omega_s}$ at frequencies $\omega_s = \omega_0 \pm \omega_\alpha$.

By computing the elasto-optic coupling, it is possible to verify that the part of the fluctuating polarization vector radiating the scattered field has a single component $(P_x^{\omega_s})_R$, which is written as a function of the fluctuating thermal elastic strains

$$w_{zz} = (1/2)[\partial w_z(\omega_\alpha)/\partial z] \tag{16}$$

as

$$(P_x^{\omega_s})_R = \epsilon_0 k_{13}(z) w_{zz}(\omega_\alpha) E_x^{\omega_0} \tag{17}$$

in the [001] direction. $k_{13}(z)$ is the z-profile of the relevant elasto-optic coefficient.

In the presence of the incident electromagnetic wave, the photoelastic effect leads to polarization of the superlattice along the x-axis. Therefore Brillouin scattering of a normally incident electromagnetic wave off a pure LA phonon produces a scattered electromagnetic wave.

Thus, if we consider (for the sake of brevity) just the anti-Stokes term $(P_x^{\omega_s})_R$, radiating at the circular frequency $\omega_s = \omega_0 + \omega_\alpha$ (corresponding to the annihilation of pre-existing phonons), it can be written in the following form:

$$(P_x^{\omega_s})_R = \xi(\omega_\alpha)\Psi_x(z|\omega_0, k_i; \omega_\alpha) \tag{18}$$

where

$$\Psi_x(z|\omega_0, k_i; \omega_\alpha) = \frac{1}{2} k_{13}(z) \frac{\mathrm{d}\phi_z(\omega_\alpha, z)}{\mathrm{d}z} E_x^{\omega_0}(z) \tag{19}$$

are spectral weights. They depend both on the phonon mode profiles and the zeroth-order incident electromagnetic field in the medium.

At thermal equilibrium, the thermal average of $\xi(\omega_\alpha)$ is zero, and, therefore, the average scattered-field amplitude is zero; thus the statistical properties of the Brillouin light depend on the probability density of the random variable $\xi(\omega_\alpha)$, and on its time autocorrelation function.

The inhomogeneous wave equation for the radiation of the scattered-field component $E_x^{s\alpha}(z,t) = E_x^\alpha(z)\mathrm{e}^{-\mathrm{i}\omega_s t}$ is obtained by means of Maxwell's equations as

$$\frac{\mathrm{d}^2 E_x^\alpha(z)}{\mathrm{d}z^2} + [k_s(z)]^2 E_x^\alpha(z) = -\frac{1}{2\epsilon_0}\left(\frac{2\pi}{\lambda_0}\right)^2 \xi(\omega_\alpha)\Psi_x(z|\omega_0, k_i; \omega_\alpha) \tag{20}$$

where

$$k_s(z) = \sqrt{\epsilon(z)}\frac{\omega_s}{c} \tag{21}$$

and $\lambda_0 \simeq \lambda_s$ because

$$|\omega_\alpha| \ll |\omega_0|. \tag{22}$$

The scattered-field component $E_x^\alpha(z)$ is the solution, in the vacuum, above the top surface ($z < 0$), of the homogeneous wave equation obtained from the above inhomogeneous equation by putting the index of refraction $n(z) = \sqrt{\epsilon(z)} = 1$. Its solution has a plane-wave form:

$$E_x^\alpha(z) = E_x^\alpha(0^-)\mathrm{e}^{-\mathrm{i}k_s z}. \tag{23}$$

The total scattered field in the vacuum within an infinitesimal solid angle $\mathrm{d}\Omega$ around the direction of $\boldsymbol{k}_s$, in the far-field approximation, is equal to its plane-wave (Fourier) component $(\omega_s, \boldsymbol{k}_s)$; therefore, we merely compute $E_x^\alpha(0^-) = E_x^\alpha(0^+) = E_x^\alpha(0)$.

In our model, $E_x^\alpha(0)$ is computed by means of a numerical method, using the NAG Fortran [21] routine D02HBF to obtain the form of the scattered field in the superlattice, and, therefore, also at the top surface $z = 0$.

For a semi-infinite superlattice, we make use of the wavevector selection rule [10]

$$k_p + k_i + k_s + \frac{2\pi m}{D} = 0 \tag{24}$$

where k_p is the wavevector of the phonon in the superlattice, k_i and k_s are, respectively, the wavevectors of the incident and scattered light, and m is a relative integer that defines the branches of the folded acoustic phonons.

We see that, for a given scattering wavevector $q = k_i + k_s$, a series of phonon modes are excited. As the relative integer m changes, the corresponding frequencies of the excited phonon modes in the folded longitudinal acoustic branches are obtained by folding k_p in the first reduced Brillouin zone, and using equation (8) or the simpler equation (14) (if the dispersion curve is in the linear zone) to compute a different eigenvalue, ω_α.

As a consequence of relation (22), one obtains from equation (24), in the case of backward scattering,

$$k_p + 2k_i + \frac{2\pi m}{D} = 0. \tag{25}$$

The differential scattering cross section is proportional to the thermal average of the power spectrum of the total scattered-field component, i.e. to the time Fourier transform of its time autocorrelation function.

We write the approximate scattered far field within $\mathrm{d}\Omega$ around the direction of $\boldsymbol{k}_s$ as

$$E_x^s(\boldsymbol{r}, t) = -\frac{\omega_s^2}{\epsilon_0 c^2}\mathrm{e}^{\mathrm{i}\boldsymbol{k}_s \boldsymbol{r}}\sum_\alpha A_\alpha \xi(\omega_\alpha)\mathrm{e}^{\mathrm{i}[\omega_0+\omega_\alpha]t} \tag{26}$$

where A_α is proportional to $E_x^\alpha(0)$.

The $\xi(\omega_\alpha)$ are independent random variables with mean square values fixed by the thermal equilibrium conditions

$$\langle \xi_\alpha^2 \rangle_{th} = K_B T / S \omega_\alpha^2(q_{||}) \tag{27}$$

(S is the illuminated area of the sample surface); hence finally we find that the differential scattering cross section for anti-Stokes Brillouin scattering from LA bulk phonons is proportional to

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\ \mathrm{d}\omega} \propto \sum_\alpha |A_\alpha|^2\, \delta\{\omega - [\omega_0 + \omega_\alpha(k_p)]\}. \tag{28}$$

In order to obtain the total scattering cross section, we consider only the contribution of the phonon having wavevectors k_p satisfying to the wavevector selection rule (25). Therefore we have to sum (28) over the eigenfrequencies ω_α corresponding to these k_p.

It is possible to obtain the Stokes cross section in a similar way.

4. Application to GaAs–AlAs superlattices, and discussion of the results

As an application of the theoretical description, in this section we present examples for the back-scattering in GaAs–AlAs superlattices with diffuse interfaces between the layers.

The period of the superlattice is $D = d_1 + d_2$, where d_1 and d_2 are the thicknesses of the layers: d_1 is the thickness of the GaAs layers, and d_2 that of the AlAs layers.

We assume that all of the functions describing the physical properties of the medium have a hyperbolic tangent profile in each period D of the structure. The refractive index $n(z)$ between two layers is, e.g.,

$$n(z) = \frac{1}{2}(n_1 - n_2)\left[\tanh\left(\frac{z - d_1}{\delta_1}\right) + 1\right] + n_2 + \frac{1}{2}(n_1 - n_2)\left[\tanh\left(\frac{z - d_2}{\delta_2}\right) + 1\right]$$

where:

(1) n_1 and n_2 are, respectively, the refractive indices of GaAs and AlAs; and

(2) δ_1 and δ_2 are the interface width parameters; if $\delta_1 = \delta_2$, then we write δ for both δ_1 and δ_2.

The wavelength of the light incident onto the medium is taken as $\lambda_0 = 4880$ Å. The period of the superlattice is $D = 421$ Å, and the dimension of the thick slab is 100 times the period D. The refractive indexes of GaAs and AlAs are, respectively, $n_1 = 4.39$ and $n_2 = 3.37$ (corresponding to $\lambda_0 = 4880$ Å). The velocities of the longitudinal sound waves in GaAs and AlAs are $v_1 = 4726$ m s^{-1} and $v_2 = 5630$ m s^{-1}; the mass densities are, respectively, $\rho_1 = 5314.9$ kg m^{-3} and $\rho_2 = 3745$ kg m^{-3}. Finally, the ratio of the elasto-optic coefficient of AlAs to that of GaAs is $k_{13}^2/k_{13}^1 = 0.15$. All the above numerical values are taken from references [5] and [10].

The theoretical cross section is convoluted with a Lorentzian of 15 GHz width to account for the finite experimental spectral resolution.

The behaviour of the eigenfunctions of the phononic field is shown in figures 1(a) and 1(b). When the wavelength of the mode is very large compared to the period D of the superlattice (that is, when the mode is of low order), the superlattice is 'seen' by the waves as an effectively homogeneous medium, and many authors (see, e.g., [12]), assume that the two media (GaAs and AlAs) always have identical elastic parameters. This means that the elastic waves in the infinite superlattice are approximated by plane waves insensitive

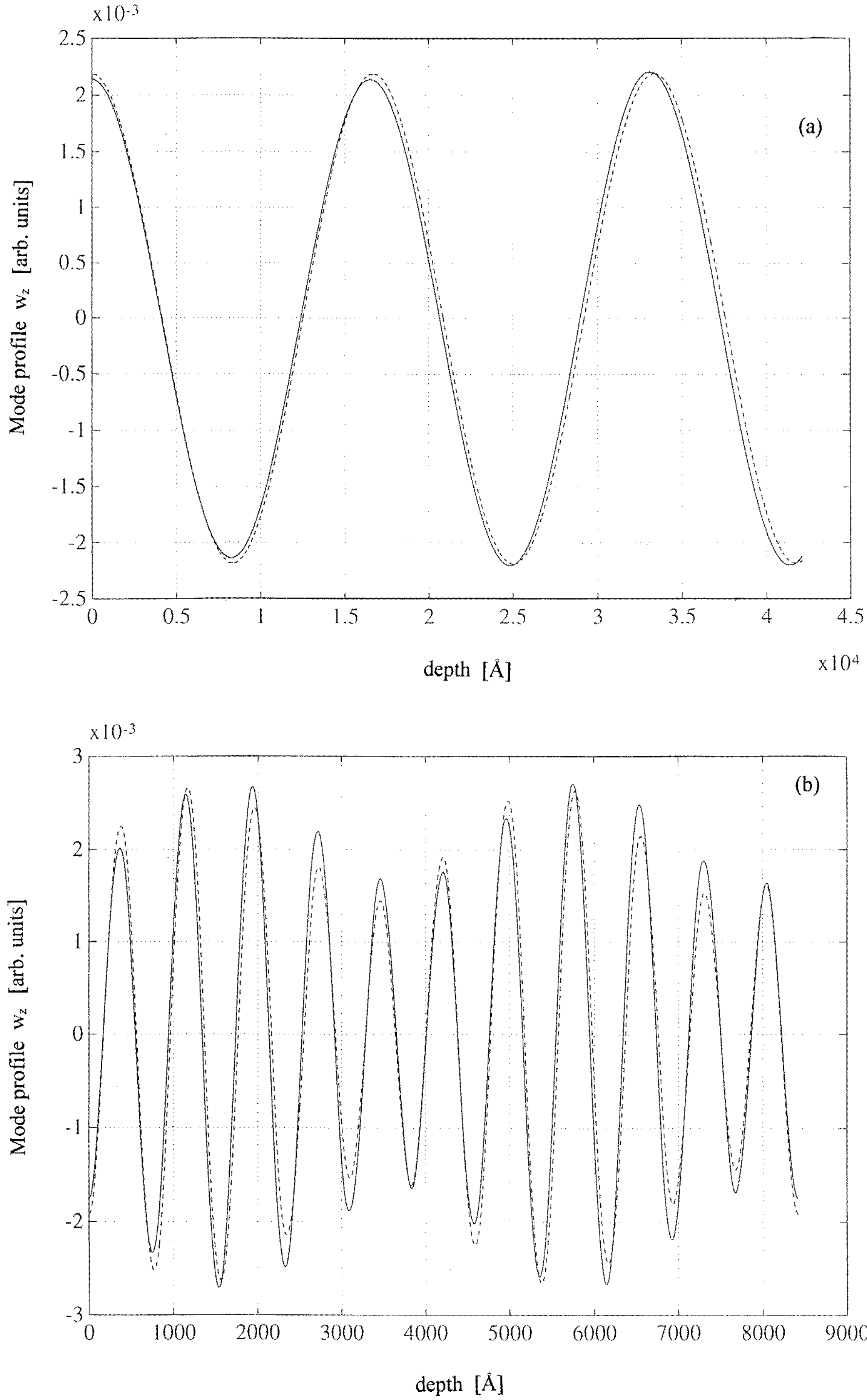

Figure 1. Mode profiles of longitudinal acoustic waves corresponding to eigenvalues of different orders. The ratio d_{AlAs}/D of the thickness of the AlAs layer to the period D of the superlattice ($D = d_{GaAs} + d_{AlAs}$) is equal to 0.73. $\delta = 5$ Å (solid line) or $\delta = 30$ Å (dashed line). (a) Eigenfunctions of order 5 for the whole thick slab. (b) Eigenfunctions of order 110 for the initial part of the thick slab.

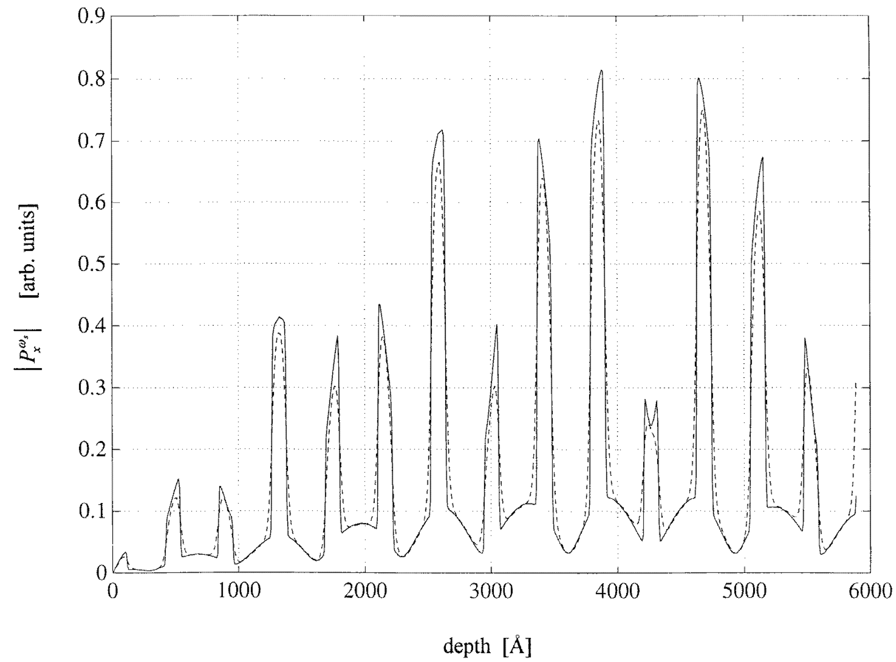

Figure 2. The mean square polarization radiating the Brillouin light versus z at the frequency of the eigenvalue of order 5, for the initial part of the superlattice, when $d_{\mathrm{AlAs}}/D = 0.73$. $\delta = 5$ Å (solid line) or $\delta = 30$ Å (dashed line).

to the nanostructure of the medium. Although we do not make such an approximation in our work, the elastic waves appear anyway as plane waves for low-order modes (see figure 1(a)). However, when the wavelength of the mode is not very large compared to the period D (that is, when the mode order is higher), the waves differ substantially from plane waves. In fact, in figure 1(b), considerable modulation of the waves, depending on the different elastic parameters of the alternating GaAs–AlAs layers—that is, depending on the nanostructure of the medium—is clearly seen. We remark that our work is the first study in which the modulation of the elastic, elasto-optic, and electromagnetic waves is fully taken into account in a superlattice with diffuse interfaces between the layers.

In figure 2 we illustrate the depth profiles of the mean square polarization radiating the Brillouin light in the superlattices with sharp and with diffuse interfaces. We see that the source of the scattered light is mainly confined to the GaAs layers because, for those layers, the value of the elasto-optic coefficient k_{13} is much higher than in the AlAs layers. This also explains why the effect of interface broadening is mainly seen within GaAs layers, where differences are amplified by the high peak intensity (and so by the high spatial frequencies) of the fluctuating polarization. The apparent doubling of the peak at around 4200 Å is probably a numerical artifact due to finite-precision effects, thus having no physical meaning. The main effect of non-sharp interfaces is to decrease the intensity of the mean square polarization, in agreement with previous results [12].

The effect of the presence of imperfect interfaces on the differential scattering cross section is presented in figure 3. In this figure, it can be seen that the peaks corresponding to the folded acoustic phonons appear as doublets: the first corresponds to $m = \pm 1$, the second to $m = \pm 2$, etc.

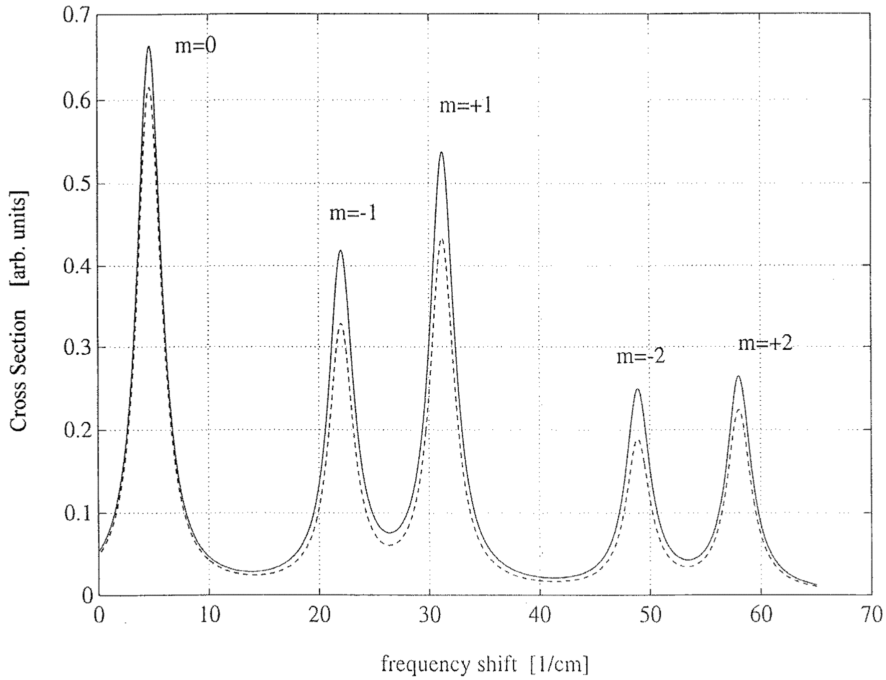

Figure 3. The theoretical cross section for the Brillouin mode and for the first folded acoustic modes when $d_{\mathrm{AlAs}}/D = 0.73$. $\delta = 5$ Å (solid line) or $\delta = 30$ Å (dashed line).

The clearest phenomenon as regards the folded longitudinal acoustic (LA) phonon modes is the decrease of intensity of their peaks as a consequence of interface broadening. A reasonable explanation is the following: if one neglects (in a first approximation) the effect of the infinite reflections and refractions of the scattered wave at the interfaces between the layers, the scattering intensity is proportional to the Fourier transform of the fluctuation of the polarization vector along the structure. For sharp interfaces, the Fourier components slowly decrease with increasing order; therefore this gives rise to many intense contributions. In contrast, for a diffuse profile, only the first orders give rise to a considerable contribution (as a limit, in the sinusoidal profile only the zero order exists). This effect is seen also for the polarization vector in figure 2.

Nevertheless it had to be considered that, for a scattering wavevector exceeding the first reduced Brillouin zone, the effect of the electromagnetic wave reflections and refractions at the interfaces cannot be neglected [10].

Figure 4 presents the dependence of the scattering cross section on the relative thickness of the layers, when the smoothness of the interfaces is fixed. In a previous study [12], it was established that, when $d_1 = d_2 = D/2$, the intensities of the folding branches with even m are zero. But, if one does not neglect (in contrast to the procedure followed by the authors of [12]) the effect of the scattered-wave reflections and refractions at the interfaces, these intensities are zero for even just a small difference between the values of d_1 and d_2. The dispersion relationships of the longitudinal phonons, in the first reduced Brillouin zone, are, in practice, equal for GaAs and AlAs crystals when the period D of the superlattice is very large compared to the lattice parameter a, as is assumed in our work [5]. Since the folded acoustic modes with $m \neq 0$ are obtained in the approximation of equation (22), and

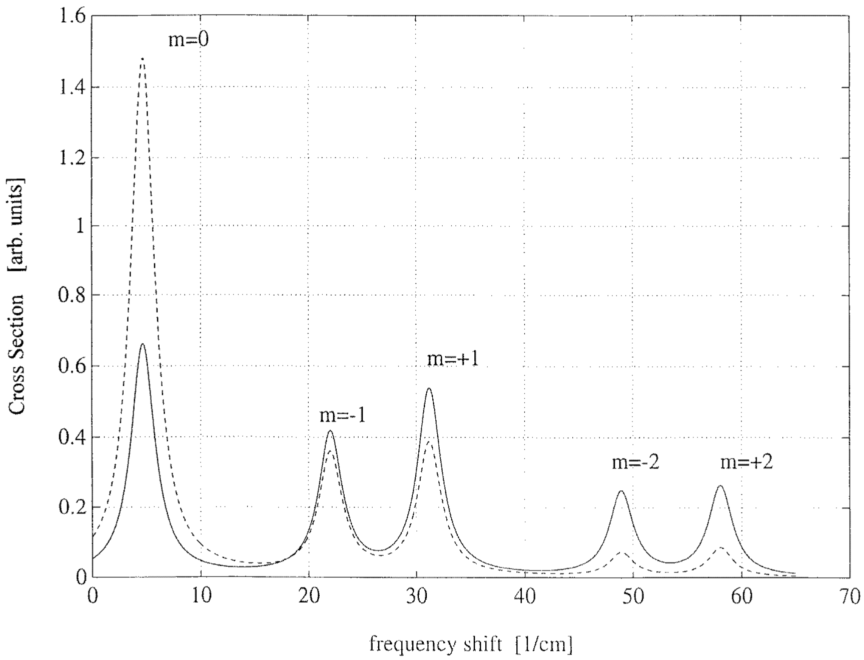

Figure 4. The theoretical cross section for the Brillouin mode and for the first folded acoustic modes when $\delta = 5$ Å. $d_{\mathrm{AlAs}}/D = 0.73$ (solid line) or $d_{\mathrm{AlAs}}/D = 0.50$ (dashed line).

their frequencies are computed using equations (13) and (14) (within the hypothesis of a perfect periodic structure), the eigenfrequencies do not exhibit a significant shift when the relative thicknesses of the GaAs and AlAs layers are changed. In fact, if one analysed a short-period superlattice ($D \simeq a$), a considerable shift of the mode frequencies as a function of the relative thicknesses would be found.

In the work of Jusserand *et al* [14], the authors observed the effect of interface broadening as a function of the growth temperature on several GaAs/AlAs short-period superlattices grown by molecular beam epitaxy. They found that only for a large increase of the growth temperature is it possible to measure a significant difference in the linewidths. Consistently, we have not found any substantial variation of the linewidths when the interface width δ changes from 0 Å to 30 Å, for a period $D = 421$ Å.

The computed relative variations of the scattering intensities for the folded LA doublets as functions of the interface width δ are presented in figures 5(a) and 5(b).

The decrease of the folded doublets can be entirely explained if one considers that, when the interface width is enhanced, the medium tends to become more and more homogeneous; therefore the folded modes (characteristic of the periodicity of the structure) tend to disappear.

We wish to stress that these curves were obtained also from Jusserand *et al* [13], in which reference the intensities of the modes were computed as proportional to the square modulus of the Fourier transform of the polarization field. In this way Jusserand *et al* found the same intensity for the two components of the folded LA doublets. In contrast, if one considers the effect that the modulation of the refractive indexes has on the scattered waves, it can be seen in figures 5(a) and 5(b) that the difference between the peaks corresponding to the same absolute value of m is not negligible. This effect is visible also in figures 3 and

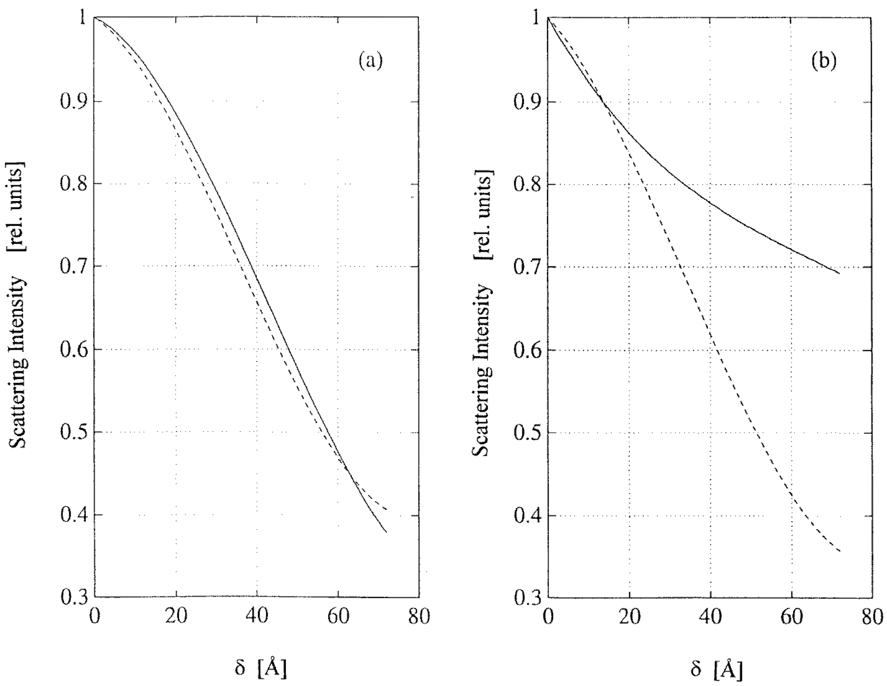

Figure 5. The computed variations of the scattering intensities for the first folded LA doublets as functions of the interface width δ, when $d_{\mathrm{AlAs}}/D = 0.73$. The intensity is assumed equal to 1 when $\delta = 0$ Å (sharp interfaces). (a) $m = +1$ (solid line) or $m = -1$ (dashed line). (b) $m = +2$ (solid line) or $m = -2$ (dashed line).

4 (for a fixed value of δ), and it is a known result that has been established in experimental work [5, 12, 14].

Finally, in figures 6(a) and 6(b) we illustrate the effect of the variation of the relative thickness of the layers on the scattering intensity of the folded longitudinal acoustic modes $(\mathrm{FLA})_m$. Here the intensity of the Brillouin mode is assumed to be equal to one. Let us note that, as the absolute value of the folding index m increases, the scattering intensity tends to decrease, and this is even more evident if the interface width parameter δ also increases. Furthermore, it is visible that the folded modes with $m = \pm 2$ vanish at a value of d_{AlAs}/D quite different from $1/2$.

For GaAs–AlAs superlattices, the fact that the scattering intensity of the even $m = \pm 2$ modes vanishes not for equal layer thicknesses of GaAs and AlAs, but for a slightly different superlattice composition, is not a new result either in the case of sharp interfaces. He *et al* [10], for example, found that the intensity of the $m = \pm 2$ modes vanishes for a relative thickness value of about 0.57. The difference between previous results given in [12] and [10] is due to the fact that in all investigations prior to [10], researchers did not take into account, in addition to acoustic and photoelastic modulation, the difference between the refractive indices of the layers. Our calculation is consistent with the results of He *et al*, but it is not limited to the case of sharp interfaces. Since even in the case of diffuse interfaces the intensity of the $m = \pm 2$ modes vanishes for a relative thickness value of about 0.57, we think that this phenomenon is independent of the diffuse character of the interfaces, and depends only on the modulation of the optical properties of the superlattice.

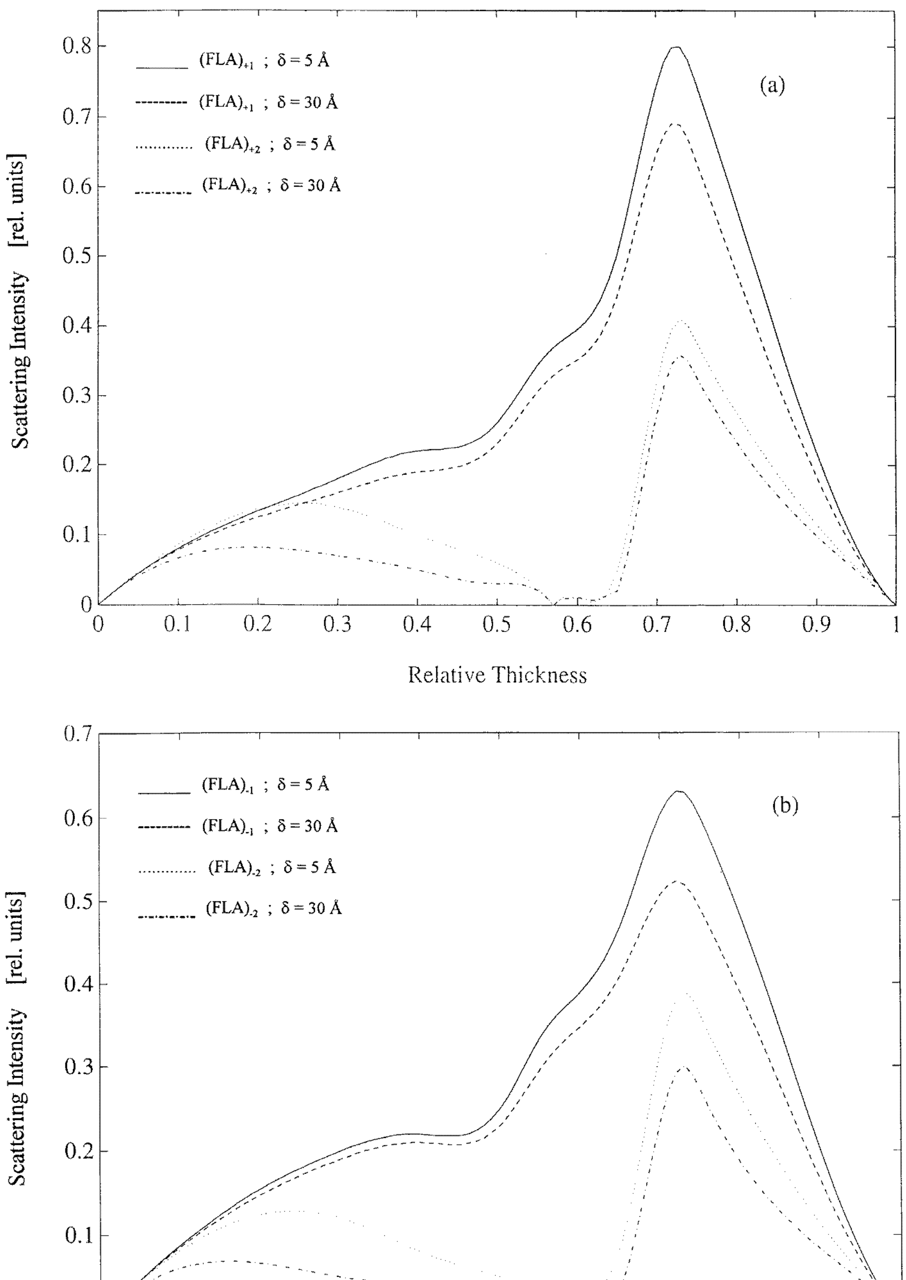

Figure 6. The variations of the scattering intensities for the folded LA doublets as functions of the relative thicknesses d_{AlAs}/D of the layers. The intensity of the Brillouin mode is normalized to one. (a) Solid line: $m = +1$, $\delta = 5$ Å; dashed line: $m = +1$, $\delta = 30$ Å; dotted line: $m = +2$, $\delta = 5$ Å; chain line: $m = +2$, $\delta = 30$ Å. (b) Solid line: $m = -1$, $\delta = 5$ Å; dashed line: $m = -1$, $\delta = 30$ Å; dotted line: $m = -2$, $\delta = 5$ Å; chain line: $m = -2$, $\delta = 30$ Å.

5. Conclusions

We have presented the computation of the cross section for Brillouin scattering of light by longitudinal acoustic waves propagating in a semi-infinite superlattice with arbitrarily diffuse interfaces between the layers.

The folded acoustic field, the transmitted zeroth-order electromagnetic field, and the scattered field are obtained by means of a numerical method using the depth profiles of the physical properties of the medium, thus taking into account the modulation of the acoustic, optic, and elasto-optic parameters. By just considering additionally the optic modulation—that is, the difference between the refractive indices of the layers—it is possible to compute exactly the spectrum of the scattered light, which is shown for the first time in the case of diffuse interfaces.

The computations are carried out by assuming that the absorption of the electromagnetic wave is negligible because the skin depth of the light is very large compared with the period of the superlattice.

Finally, as an application, the case of a superlattice made up of alternate layers of GaAs and AlAs is illustrated using a back-scattering geometry, providing evidence for the effect of the interface broadening.

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